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CONTRIBUTION TO THE SOLUTION OF THE BATEMAN EQUATIONS FOR THE ISOTOPES TRANSMUTATION IN FISSION NUCLEAR REACTORS

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Abstract.

In several problems of nuclear engineering it is necessary to study the changes that undergo in the fuel of a nuclear reactor. Such changes are mainly due to the fission process, the radioactive decay, and neutron-nuclear reactions.

It is possible to model these changes using a system of coupled differential equations, which set a balance relationship between the gains and the losses of the isotopes that constitutes the fuel. Due to historical reasons, such equations are known as Bateman Equations, and the problems where were necessary to solve them are called burnup and activation problems.

In accordance to the methodology used, there are several solutions to these equations, which include from a matrix exponential approach to rational approximation methods.

Among these solutions, one of the most popular methodology used to solve the Bateman equation is the linear chain method, which includes a solution based on the Laplace transform, and a procedure known as linearization.

During the last three decades, the linear chain method was widely used in computational neutronic codes and, along with the exponential matrix method, was the methodology that dominate the field. Nevertheless, in 2012, a new and powerful method to solve the Bateman equations was developed by Maria Pusa and Jakko Leppänen, which is called CRAM (Chebyshev Rational Approximation Method). Such new approach is faster than the linear chain method, and can overcome some of the main difficulties related to the numerical treatment of the mentioned system of differential equations.

In spite of the development and advantages of CRAM, some authors suggest that the linear chain method continue being a complementary tool to solve burnup and activation problems, and nowadays, there are codes who continue including it among its solvers. Therefore, the study of the linear chain method, and the development of contributions related to it, is an important line of research in nuclear reactor engineering.

Among the current research about the linear chain, are the development of more general solutions, as well as the optimization of routines and the reduction of computational time. In the present thesis, contributions to the Bateman equations were developed, under these both fields.

Starting from an algorithmic level, the present work analyzes the process of linearization, which as was mentioned before, is a fundamental part of the linear chain method. From such analysis, an alternative method to carry out this procedure is proposed, which is faster than the standard methodology reported in literature.

In terms of general solutions, the present thesis contains two new approach to the study of the structures of isotopes known as cyclic chains. The first one is related to the modelling of cyclic chains through a Lagrange solution, and the second one is based on the development of a symbolic computation algorithm.

Through the present work it was possible to conclude that several aspects of the Bateman Equations and their solutions can be improved. Such aspects cover from the linearization process until the theory behind the structure of the decay and burnup chains.

Even when the Bateman equations have more than a hundred years of being developed, there are research topics that have not been studied, but which represents fields of potential developments.

In the present thesis it was concluded that the topic related to the cyclic chains can be fundamental to reduce the execution time of the linear chain method, because such chains appear in several practical problems due to the presence of heavy isotopes. Additionally, it was found that no efforts have been made to include in a practical way the general solutions to the Bateman Equation.

Furthermore, the present research showed that several procedures related with the developed burnup codes are based in empirical studies, instead of theorical analysis. A notable example of the last affirmation is the methodology of assignation of the yields, which is analyzed in detail in the present work.

These contributions and conclusions were developed in order to be included in a computational code, named Szilard, who belongs to the AZTLAN Platform project, which is a Mexican effort for developing own neutronic and thermohydraulic codes for nuclear reactors design and analysis.

Resumen

En diversos problemas de ingeniería nuclear resulta necesario estudiar los cambios que ocurren en el combustible de un reactor, los cuales se deben principalmente al proceso de fisión, al decaimiento radiactivo, y a las reacciones de los neutrones con los núcleos.

Es posible modelar estos cambios usando un sistema de ecuaciones diferenciales acopladas, las cuales establecen relaciones de balance entre las ganancias y las pérdidas de isótopos que constituyen el combustible. Debido a razones históricas, tales ecuaciones son conocidas como las ecuaciones de Bateman, y los problemas donde es necesario resolverlas se conocen como problemas de quemado o activación.

De acuerdo con la metodología usada, existen varias soluciones a estas ecuaciones, las cuales incluyen desde un tratamiento de la matriz exponencial, hasta métodos basados en aproximaciones racionales.

Dentro de esas soluciones, una de las metodologías más populares que se usa para resolver las ecuaciones de Bateman, se conoce como el método de cadena lineal, el cual incluye una solución basada en la transformada de Laplace, junto con un procedimiento conocido como "linealización".

Durante las últimas tres décadas, el método de cadena lineal fue ampliamente usado en códigos neutrónicos computacionales, y junto con el método de la matriz exponencial, fue el método dominante. Sin embargo, en el 2012, un nuevo y poderoso método para resolver las ecuaciones de Bateman fue desarrollado por Maria Pusa y Jakko Leppänen, el cual es llamado CRAM (Chebyshev Rational Approximation Method). Dicho método es más rápido que el método de cadena lineal, y puede superar algunas de las dificultades relacionadas con el tratamiento numérico del sistema de ecuaciones diferenciales mencionado antes.

A pesar del desarrollo y las ventajas que tiene el CRAM, algunos autores sugieren que el método de cadena lineal continúa siendo una herramienta complementaria para resolver problemas de activación y quemado, y actualmente hay códigos que continúan incluyéndolo dentro de sus *solvers*. Por lo anterior, el estudio del método de cadena lineal, y el desarrollo de contribuciones relacionadas con él, es una importante línea de investigación en la ingeniería de reactores nucleares.

Dentro de las actuales líneas de investigación acerca del método lineal se encuentran los desarrollos de soluciones más generales, así como la optimización de rutinas y la reducción de su tiempo computacional. En la presente tesis se desarrollan contribuciones a las ecuaciones de Bateman bajo el enfoque de estos dos campos.

Comenzando desde un nivel algorítmico, el presente trabajo analiza el proceso de linealización, el cual, como se mencionó antes, es parte fundamental del método de cadena lineal. A partir de dicho análisis se desarrolló un método alternativo para llevar a cabo este proceso, el cual es más rápido que el método estándar que se reporta en la literatura.

Con respecto a las soluciones generales, la presente tesis contiene dos nuevos enfoques relacionados con el estudio de las estructuras conocida como cadenas cíclicas. El primero de

estos enfoques se relaciona con el modelamiento de las cadenas cíclicas a través de la solución de Lagrange, y la segunda se basa en el desarrollo de un algoritmo de cálculo simbólico.

A través del presente trabajo fue posible concluir que varios aspectos relacionados con las ecuaciones de Bateman pueden mejorarse. Dichos aspectos van desde el proceso de linealización, hasta la teoría detrás de la estructura de las cadenas de decaimiento y quemado.

Aun cuando las ecuaciones de Bateman tienen más de 100 años de haber sido desarrolladas, hay líneas de investigación que no se han estudiado, pero que representan áreas potenciales de desarrollo.

En la presente tesis se concluyó que el tópico relacionado con las cadenas cíclicas puede ser fundamental para reducir el tiempo de ejecución del método de cadena lineal, porque dichas cadenas aparecen en problemas prácticos, debido a la presencia de isótopos pesados. Adicionalmente, se encontró que no han existido esfuerzos para incluir en una forma práctica las soluciones generales a la ecuación de Bateman.

Por otro lado, la presente investigación mostró que varios procedimientos relacionados con el desarrollo de códigos de quemado están basados en estudios empíricos, más que en teóricos. Un ejemplo notable de esta última afirmación es la metodología de asignación de los *yields*, que se analiza en detalle en el presente trabajo.

Estas contribuciones fueron desarrolladas con el fin de incluirse en el código llamado Szilard, que forma parte del proyecto AZTLAN platform, el cual representa un esfuerzo mexicano en el desarrollo de códigos neutrónicos y termo hidráulicos propio, para el análisis y diseño de reactores nucleares.

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Introduction

It is known that the improvement of a quality of life and the development of the countries depend, among other topics, on the use and production of energy, and particularly of electricity (NEA, 2008). Since the energy demand is strongly conditioned by the grown of population, a recurrent challenge that the countries face is to find a sustainable and economical way to produce electricity.

Unfortunately, the current world supply energy depends considerably on fossil fuels, which represent a major cause of the increasing of atmospheric concentration of greenhouse gases, particularly of CO_2, whose direct consequences include the warming of the atmosphere as well as oceans, the increase of global mean sea level, the air pollution, among other phenomena.

Nuclear power has a crucial role in the effort of reducing greenhouse gas emission, due that such plants practically do not produce such gases during their operation (IAEA, 2018), and because it can provide an economically feasible source of energy. Based on this advantage, many countries intend to include nuclear power in their energy programs for the coming decades. Therefore, the continuous improvements in technology and modelling of nuclear power, and particularly of nuclear reactors, are topics of vital importance to the progress and development of the countries.

The modelling of certain phenomena in a nuclear reactor are studied by a branch of nuclear engineering known as reactor physics. Over such discipline, and particularly over its computational methods, depends an important part of the nuclear technology's improvements and designing, because experimental work usually is difficult or even impossible (Isotalo, 2011).

Among the topics studied in reactor physics is the analysis of the changes of the fuel and material composition of a nuclear reactor, a set of phenomena known as burnup problem. Since the neutronic behavior of a nuclear reactor strongly depends on such composition, the solution of the burnup problem is very important to several applications related to design, safety, economy of a reactor core loading, evaluation of the spent fuel material, among others. Therefore, a fundamental part of the computational reactor physics is related to the development and improvement of codes that solve the burnup problems, which are called burnup codes.

In Mexico, the Aztlan Platform strategic project, sponsored by SENER-CONACYT, represents a governmental effort to develop own thermohydraulic and neutronic codes, and to meet the need to analyze and modelling nuclear power plants, as well as to develop knowledge and human resources in nuclear engineering (Gómez-Torres et. al. 2015). The present thesis is part of the outcomes of the Aztlan Platform strategic project, and contains several contributions to the computational, mathematical and physical modelling of burnup problems.

Research objectives.

The work described in this thesis focused on the development of contributions to the Bateman equations, who are the core of the burnup problems. Such contributions will be part of the novel features of the Aztlan's code Szilard.

Additionally, the present thesis addressed some topics related to the yield's assignation, a very difficult task who deserves to be discussed due to the shortage of information in the literature. Such analysis aims to facilitate the future implementation of large amount of fission products, which is a novel feature of the new-generation burnup codes.

Finally, one of the research objectives lies in the improvement of computational time related to the execution of the linear chain method, in order to make the Aztlan's code Szilard competitive in relation to the existing ones.

Main contributions.

The contributions of the present work can be divided in three levels: 1) Algorithmic, 2) Physical and 3) Mathematical. The first one, is related to the analysis of the linearization process, a fundamental part of the linear chain method, for which an alternative algorithm was proposed.

As for the physical contribution, classification and study of the cyclic chains was carried out. Starting from it, a set of solutions were proposed, which have several advantages over the linear chain method. Finally, at the mathematical level, an analysis of the general solutions of the Bateman equations was carried out, and an integral formulation was proposed.

Organization of the thesis.

The thesis is organized so that Chapter 1 provides the Fundamentals of the Bateman equation, as well as its analytical solution. This chapter describes general aspects of the Lagrange transform method, and the most important assumptions related with the solution through it. Additionally, in this chapter are discussed the general procedures behind the general solutions to facilitate wider understanding of the topic and putting the context related to the symbolical calculation algorithm.

Chapter 2 contains a detailed discussion of the definition of the type of yield, as well as a description of the procedure to assign it in a given network. Also, this chapter analyses the relationship between the topology of the networks, and the concentration of the fission products, and it provides some insights about the way in which two different burnup codes can be compared.

In Chapter 3 is described the developed algorithm for the linearization process. This chapter also contains the elements related to the analysis of the number of linear chains, and the conclusion that in a cyclic chain the linearization process can only approximate the exact solution. Finally, this chapter ends with a comparison between the proposed algorithm and the standard deep first search method.

Chapter 4 covers the theory of the cyclic chains, beginning with a classification of these structures based on their complexity, and proposing two solution to a specific kind of them named as the pure type. Additionally, this chapter includes the description of the forward and backward method, as well as an analysis of the superposition process.

In Chapter 5 the main elements of an integral formulation of the Bateman equation is discussed, which is the core for the symbolical computational algorithm. This chapter shows how through this procedure it is possible to include several of the general solutions of the Bateman equation in a burnup code. Additionally, this chapter propose a methodology to find the error involved when the effective removal coefficients are modified.

Chapter 6 provide the general conclusions, and summarize the work herein. This chapter also contains some prospects of future work.

Finally, there are two appendixes. One of them includes the pseudo-code of all the algorithms developed in the present thesis, and the other one includes some proofs and developments of theorems cited in the chapters.

Chapter 1. The Fundamentals of the Bateman Equations.

The Bateman equations are a set of coupled differential equations, which essentially describe balance relationships between the gains and the losses of the isotope's concentration. Under certain conditions, such equations can be considered linear with constant coefficients, and therefore they can be solved through a standard methodology as the Laplace transform.

In the present chapter the fundamentals of this set and its solution is discussed, beginning with a discussion of the concept of burnup, and ending with an analysis of the general formulations used when there are two or more repeated isotopes in a linear chain.

1.1 Burnup Concept.

The composition of a nuclear reactor changes during its operation. Such changes are related to three main phenomena that undergo the isotopes who compose it: radioactive decay, fission, and neutron-nucleus reactions, among others.

Since these changes are related to the production of energy, some authors suggest (Cacuci, 2010) that there is an analogy to the concept of burnup in chemical reactions, where a material change its composition in the process of energy's generation. In fact, in the same way that occurs in chemical reactions, in the nuclear ones appear new substances in such process and the fuel is depleted as burnup occurs.

Nevertheless, in addition to the level where the reactions occur, there is an important difference between the chemical burnup and the nuclear burnup. While the behavior of a chemical reactors depends slightly on the properties of the fuel, in the nuclear case there is a strongly dependency, because the neutronic properties of a nuclear reactor core are determined directly by its composition and configuration. Therefore, the study and quantification of such changes is a very important part of reactor analysis and design.

For the above discussion. it is possible to define the burnup concept in reactor physics as the study and modelling of the fuel changes in a nuclear reactor over time. Provide a definition is important, because in literature there are several others that are not necessary equivalent. For example:

i) Baratta and Lamarsh defined burnup calculations as the computation of changing reactor properties (Lamarsh and Baratta, 2001),

ii) Duderstadt and Hamilton considered as burnup the consumption of fissile nuclides (Duderstadt and Hamilton, 1975),

iii) Stacey set the concept as the cause of long-term changes in the properties of a nuclear reactor over its lifetime (Stacey, 2007),

iv) And, recently, Shigeo Ohki (Oka, 2014) provide a more complete definition considering it as the evaluation of the time-dependent power distribution and reactivity, by solving the equations for the atomic density change of nuclides contained in the fuel.

The main difference between these definitions lies in the scope of the analysis and the methodology used in the calculations. While Duderstadt and Hamilton only considered the consumption of fissile nuclides, the rest of the authors also include the study of some properties of the reactor. In this context, Ohki makes an additional step pointing out what are the main properties of interest for burnup studies: the power distribution and the reactivity.

In the present thesis the concept of burnup problem will be related, adopting a focus closer to the second part of the Ohki's definition, to the solution of the equations for atomic density change of nuclides contained in the fuel. But the present work does not include topics related to the properties of the reactor.

In other words, most of the present work is more related to the solution of the mathematical model involved in burnup calculations, than with the physical analysis that can be obtained from such a solution.

Finally, it is worth mentioning that the concept of burnup has been defined in multiple ways since the early studies about this topic. Due to this variety, some authors made attempts to provide for a formal meaning of such concept with the purpose to clarify it. At our best knowledge, the first work that tried to set a formal definition of the concept was the work of Stover and Moeller, published in 1961 (Stover and Moller, 1961). This word can be considered as the first one who studied in a deeper way the experimental and analytical method behind the burnup phenomena.

1.2 Importance of Rutherford's work.

It is possible to model the changes over time in the isotopic composition of a nuclear reactor through the following balance equation:

$$\frac{dX_i(t)}{dt} = G_i(t) - L_i(t) \tag{1.1}$$

Where $X_i(t)$ represents the concentration of the isotope i, and $G_i(t)$ and $L_i(t)$ are functions related to the gains and losses.

In order to conclude if the equation (1.1) can be solved and, if possible, finding its solution, it is necessary to determine who are the functions $G_i(t)$ and $L_i(t)$. Such solution, belonging to the physical and experimental terrain, was carried out by Ernest Rutherford in 1900 (Rutherford, 1900).

And even when Rutherford's work only considered the change due to radioactive decay, he was the first who set a differential equation system similar to (1.1), and who related the functions $G_i(t)$ and $L_i(t)$ with a relationship that nowadays is known as the exponential law of radioactive disintegration. Nevertheless, for reasons that will be discussed later, such system did not receive his name, instead it is known as the Bateman equations.

1.2.1 Rutherford and the decay process.

In a series of experiments carried out by Rutherford in 1898, he found that the thorium element emitted certain particles, whose radiation had a more penetrating character than the related to uranium (Rutherford, 1899).

He called this emission as "emanation" and, as a part of his research, he discovered that such emanation gradually lost its radioactive power. In his attempt to describe this reduction, he set the following differential equation in a paper published in 1900 (Rutherford, 1900), which can be considered as the early antecedent of (1.1):

$$\frac{dn}{dt} = q - \lambda n \tag{1.2}$$

In this equation n was the number of ions produced per second between certain plates in an experimental arrange, q was the number of ions supplied per second by the "emanation" diffusing from thorium, and λ was a constant.

Additionally, he observed that under certain experimental conditions, the rate of discharge due to the emanation reached a steady state value. He concluded that this state was reached when the loss of radiation intensity was compensated by the new radioactive particles emitted from the thorium "emanation". In other words, he confirmed his "balance" equation.

The next step of Rutherford was to remove the source of the emanation, i.e. $q = 0$ in (1.2), whereupon he deduced the following two equations

$$\frac{dn}{dt} = -\lambda n \quad \rightarrow \quad n = n(0)e^{-\lambda t} \tag{1.3}$$

Essentially, which Rutherford found was that the rate at which the number of ions decrease is proportional to number of ions presented in such time. As some authors suggest (Radvanji and Villain, 2017), (Kragh, 2012) that the most important consequence of (1.3) is the development of the radioactive law.

Nowadays, it is possible to conclude that the number of ions was proportional to the concentration of the isotope. In other words, the variable n is equivalent to the function $X_i(t)$. Therefore, through the discover of the radioactive law is possible to conclude that

$$L_i(t) = \lambda X_i(t) \tag{1.4}$$

On the other hand, the procedure to determine who is the function $G_i(t)$ was more complex, because it involved the concept of transmutation, which was a very disputed term in the early development of the nuclear energy, at such grade that several works preferred to use the term of "successive transformations".

1.2.2. The gain function G_i and the discovery of transmutation.

Nowadays, it is known that the “emanation” described by Rutherford was the ^{220}Rn isotope, which was produced by ^{224}Ra through alpha decay. Nevertheless, this conclusion was not immediate, and two years passed before Rutherford and his colleague Soddy concluded that emanation radioactivity implied that a new substance was produced by thorium, with a more penetrating property than its parent. In other words, they observed a transmutation process (Rutherford and Soddy, 1902).

Even when the term transmutation is very common in the current nuclear field, in Rutherford’s time the “transmutation” word was related to alchemy, a pseudoscience that had been discredited since the middle ages (Marshal and Marshal, 2003). Therefore, the conclusions obtained by these researches were totally revolutionary, and part of their success was due to they did not discard the transmutation hypothesis, as wrongly their French counterparts did (Malley, 1979). The discovery of the transmutation process allowed that Rutherford and Soddy observing that there were successive transformations, where a new substance was produced, which in turn produced another substance. In this way, Rutherford also discovered the first radioactive decay chains in history (Rutherford, 1904). In his work, Rutherford reported the following sequence:

$$\boxed{\text{Thorium}} \rightarrow \boxed{\begin{matrix}\text{Thorium}\\ X\end{matrix}} \xrightarrow{\text{4 days}} \boxed{\begin{matrix}\text{Thorium}\\ \text{Emanation}\end{matrix}} \xrightarrow{\substack{1\\ \text{minute}}} \boxed{\begin{matrix}\text{Emanation X}\\ \text{First change}\end{matrix}} \xrightarrow{\substack{55\\ \text{minutes}}} \boxed{\begin{matrix}\text{Second}\\ \text{Change}\end{matrix}} \xrightarrow{\substack{11\\ \text{hours}}}$$

Where the upper scripts above the arrows are the half-life reported by Rutherford, who observed that the lambda constant was different for each substance. Nowadays it is known that the actual decay chain was:

$$\boxed{^{228}\text{Th}} \xrightarrow[\alpha]{\substack{1.91\\ \text{years}}} \boxed{^{224}\text{Ra}} \xrightarrow[\alpha]{\substack{3.66\\ \text{days}}} \boxed{^{220}\text{Rn}} \xrightarrow[\alpha]{\substack{55.6\\ \text{seconds}}} \boxed{^{216}\text{Po}} \xrightarrow[\alpha]{\substack{0.145\\ \text{seconds}}} \boxed{^{212}\text{Pb}} \xrightarrow[\alpha]{\substack{10.64\\ \text{hours}}} \boxed{^{212}\text{Bi}} \rightarrow$$

Considering the conservation of mass, it was possible to conclude that in a decay chain the gain term of an element, $G_i(t)$, was the loss term of the predecessor element in the chain, i.e. $L_{i-1}(t)$. Therefore, the system (1.1) can be written as:

$$\frac{dX_i(t)}{dt} = L_{i-1}(t) - L_i(t), \qquad L_i(t) = 0, \text{for } i = 1 \tag{1.5}$$

1.2.3 The first solution for the balance equations.

From the last two sections it is clear that the Rutherford’s work was crucial to set the balance equation in the form they are currently known. In fact, using (1.3) and (1.5) in equation (1.1) it follows that:

$$\frac{dX_i(t)}{dt} = \lambda_{i-1}X_{i-1} - \lambda_i X_i \tag{1.6}$$

This differential equation is coupled, linear, with constants coefficients, and it is reduced to the radioactive law for $i = 1$. Due to this and the transmutation process, this equation has a recursive nature.

The solution for the case $i = 2$ can be obtained, among other methods, using an integral factor. Nevertheless, Rutherford proposed a physical solution, which he called "theory of the secondary changes", which deserves to be analyzed.

Using the notation given in (1.6), he began considering that after a time t_1, the number X_1 of particles remaining unchanged is given by $X_1 = X_1(0)e^{-\lambda_1 t_1}$, then the term $\lambda_1 X_1(0)e^{-\lambda_1 t_1}dt_1$ represents the amount that has changed in an interval dt_1 about t_1.

If the last term is considered as a new substance related to X_2, this concentration also will undergo for a "second" process of transmutation, characterized by another λ_2 constant. Since the radioactive decay law is also valid for this new substance, after a time t (measure from the beginning of all the transmutations) the number of particles that remaining unchanged of it can be computed using (1.3)

$$\underbrace{dX_2}_{\substack{\text{equivalent to}\\ n \text{ in (1.3)}}} = \underbrace{\lambda_1 X_1(0)e^{-\lambda_1 t_1}dt_1}_{\substack{\text{equivalent to}\\ n_0 \text{ in (1.3)}}} \cdot \underbrace{e^{-\lambda_2(t-t_1)}}_{\substack{\text{equivalent to}\\ e^{-\lambda t} \text{ in (1.3)}}} \tag{1.7}$$

Finally, it is only necessary to carry out an integration from $t_1 = 0$ to $t_1 = t$, after which it follows:

$$X_2 = \lambda_1 X_1(0)\frac{e^{-\lambda_1 t} - e^{-\lambda_2 t}}{\lambda_1 - \lambda_2} \tag{1.8}$$

This solution is notorious because it depends totally on the meaning of the radioactive law, instead of a mathematical procedure. At our best know, Rutherford found the solution to the case $i = 1$ to $i = 5$. As will be discussed in the following section, there were other authors who published other solutions, including of the general type.

Nevertheless, one of the most important part to solve the problem of the mass balance in radioactive decay was result of Rutherford's work.

1.3 Solutions before 1910.

A recurrent mistake in literature is the assumption that the mathematician Harry Bateman was the first who solved the mass balance equations given in equation (1.6). This could not be farther from the truth.

Before 1910, the date in which the Bateman's work was published, there were other authors who published general solutions to such system. At our best knowledge, J. Stark (Stark, 1904) and P. Gruner (Gruner, 1907), separately, were the first who proposed the solution for case $i = k$. It is possible that Stark and Gruner used an induction process to find their formulas, but there is not enough information about this topic.

Nevertheless, the work of these authors has a remarkable importance, to the point that late works cited them instead of the solution proposed by Harry Bateman (Bothe et. al,1933), (Meyer and Schweildler, 1927). This is significative, because it means that the Bateman solution's not necessary represented an innovation in terms of a formula for the case $i = k$. Instead, the importance of it is related to the method he developed in his work, which will be discussed in the following section.

On the other hand, it is possible to use mathematical induction to find a general solution for the case $i = k$ in the equation (1.6). Such solution is developed in Appendix A. Nevertheless, some authors suggest that mathematical induction is a type of not explanatory proof (Stylianides et. al, 2015), (Baker, 2010), and this could be a reason for which there were not authors who used as a formal proof. In this sense, the word explanatory means that a proof offers new insights in the subject, besides of providing new ways of reasoning, as well as the discover of new possible connections between different fields (Hersh, 1993).

1.4 The Bateman solution.

In 1910, the English mathematician Harry Bateman proposed a very novel solution which was based in a methodology that nowadays is known as the Laplace transform (Bateman, 1910). In fact, this was the first time where such procedure was used (Deakin, 1982). Therefore, the work of Harry Bateman was not only important to the nuclear field, but also for the mathematic filed of the differential equations and the integral transformations.

1.4.1 Procedure followed by Bateman.

Using modern notation, the Bateman solution used the following definitions.

$$\begin{cases} \mathcal{L}\{X_i\} = \tilde{x}_i = \displaystyle\int_0^\infty X_j(t)\, e^{-st} dt, \quad 1 \le j \le n \\ \mathcal{L}\left\{\dfrac{dX_j}{dt}\right\} = s\mathcal{L}\{X_j\} - X_j(0) = s\tilde{x}_j - X_j(0) \end{cases} \tag{1.10}$$

Where $\mathcal{L}$ denotes the Laplace transform and X_j is the concentration of an isotope belonging to the following decay chain:

$$X_1 \xrightarrow{r_{1,2}} X_2 \xrightarrow{r_{2,3}} \dots \xrightarrow{r_{n-1,n}} X_n \tag{1.11}$$

In the last equation the upper scripts $r_{i-1,i}$ denote the reaction who originates the transformation. This structure, where except for the first and the last isotopes all the other elements have only one father and only one daughter, is known as a linear chain. The following step is to express equation (1.6) in a matrix way:

$$\begin{cases} \dfrac{dX_1}{dt} = & -\lambda_1 X_1 \\ \dfrac{dX_2}{dt} = & \lambda_1 X_1 - \lambda_2 X_2 \\ \quad \dots & \dots \\ \dfrac{dX_n}{dt} = & \lambda_{n-1} X_{n-1} - \lambda_n X_n \end{cases} \tag{1.12}$$

Once the definitions given in (1.10) are applied to (1.12) it follows that:

$$\begin{cases} s\tilde{x}_1 - X_1(0) = & -\lambda_1\tilde{x}_1 \\ s\tilde{x}_2 - X_2(0) = & \lambda_1\tilde{x}_1 - \lambda_2\tilde{x}_2 \\ \quad\vdots & \quad\vdots \\ s\tilde{x}_n - X_n(0) = & \lambda_{n-1}\tilde{x}_{n-1} - \lambda_n\tilde{x}_n \end{cases} \tag{1.13}$$

Under the following initial conditions

$$X_i(t=0) = X_i(0) = 0 \text{ with } 2 \leq i \leq n, \; X_1(0) \neq 0 \tag{1.14}$$

And after multiple replacements it is possible to write (1.13) as:

$$\begin{cases} \tilde{x}_1 = & \dfrac{X_1(0)}{s+\lambda_1} \\ \tilde{x}_2 = & \dfrac{\lambda_1 X_1(0)}{(s+\lambda_1)(s+\lambda_2)} \\ \;\vdots & \quad\vdots \\ \tilde{x}_n = & \dfrac{\lambda_1\lambda_2 \dots \lambda_{n-1}X_1(0)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)} \end{cases} \tag{1.15}$$

It is possible to note that the last system of equations is symmetric, i.e. the equations have the same structure. Then, the solution for the last term $\tilde{x}_n$ can be applied to the other terms if the sub index is adequately used. Therefore, the problem is reduced to solve the following equation for X_n:

$$\tilde{x}_n = \int_0^\infty e^{-st}X_n(t)dt \tag{1.16}$$

A standard procedure consists of using the inverse Laplace transform, for which is valid:

$$\mathcal{L}^{-1}\{\tilde{x}_n\} = \mathcal{L}^{-1}\left\{\int_0^\infty e^{-st}X_n(t)dt\right\} = X_n \tag{1.17}$$

Nowadays there are known inverse Laplace transforms for several functions, which commonly are tabulated. Then it is necessary to express $\tilde{x}_n$ in terms of such functions with the purpose to solve equation (1.16). If it is assumed that all the lambda constants are different, it is possible to rewrite the denominator of $\tilde{x}_n$ as:

$$\frac{1}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)} = \sum_{i=1}^{n}\frac{a_i}{s+\lambda_i} \tag{1.18}$$

Where the coefficients a_i can be computed as:

$$a_i = \left[\frac{(s+\lambda_i)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)}\right]_{s=-\lambda_i} \tag{1.19}$$

The term a_i can be interpreted as a product where an index is omitted. In such way it can be written as:

$$a_i = \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} \tag{1.20}$$

Using (1.20) in (1.18):

$$\tilde{x}_n = X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} \frac{1}{s + \lambda_i} \tag{1.21}$$

Therefore:

$$X_n = \mathcal{L}^{-1}\{\tilde{x}_n\} = \mathcal{L}^{-1}\left\{ X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} \frac{1}{s + \lambda_i} \right\} \tag{1.22}$$

Considering linearity:

$$= X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} \mathcal{L}^{-1}\left\{\frac{1}{s + \lambda_i}\right\}$$

Finally, only it is necessary to use the known inverse Laplace transform:

$$\mathcal{L}^{-1}\left\{\frac{1}{s + \lambda_i}\right\} = e^{-\lambda_i t} \tag{1.23}$$

Replacing the last term, the solution is given by the following equation:

$$X_n = X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} e^{-\lambda_i t} \tag{1.24}$$

The last equation is known as the Bateman solution.

1.5 Theory behind the Bateman solution.

As was mentioned in Section 1.3, there were other solutions before the Bateman's work. Nevertheless, the main contribution of this author was not the solution but rather the methodology he used to obtain it. It is possible that this is the reason why nowadays the system given in (1.6) bears his name, and that, wrongly, some authors considered him as the first one who solved such system.

The Bateman's work was fundamental to the development of the Laplace transform methodology. About this topic, the Australian mathematician Michael Deakin carried out an historical and technical analysis about the Bateman's role and its importance as well of other authors (Deakin, 1982). Unfortunately, Harry Bateman did not receive proper credit and recognition for his work in this field, as some biographers suggest (Erdelyi, 1947).

There are thee parts of the procedure developed in the last section who deserve to be analyzed. Firstly, the equality of (1.18) is only valid when all the lambda constants are different. As will be discussed later, there are some cases when this condition is not valid. The other two parts are related to use a more formal method to find the solution for the equation (1.16), as well the uniqueness of the solution. Both topics will be described in the following sections.

1.5.1 A more formal solution based in complex theory.

Nowadays there is a more complete theory to find the solution of the equation (1.16) through the use of the Fourier-Mellin formula (Brown and Churchill, 2004), which states that:

$$X_n(t) = \frac{1}{2\pi i}\int_{\gamma - i\infty}^{\gamma + i\infty} e^{st}\tilde{x}_n(s)ds \tag{1.25}$$

Where $\tilde{x}_n$ must fulfill the following conditions: being a complex function, being analytic through the finite s plane, except for a finite number of isolated singularities. On the other hand, the positive constant γ is related with a vertical line segment between $s = \gamma - iR$ to $s = \gamma + iR$, and it must be large enough that all singularities of $\tilde{x}_n$ lie to the left of that segment, and finally t must be a real variable.

It is possible to note that $\tilde{x}_n$ given in (1.15) fulfill all these conditions. Firstly, the singularities or poles are the values at which the following function becomes zero:

$$(s + \lambda_1)(s + \lambda_2) \ldots (s + \lambda_n)$$

In other words: $-\lambda_1, -\lambda_2, \ldots, -\lambda_n$. Due to the radioactive law it is know that the lambda constants are positive, and therefore the poles are negative, for which they lie in the left of the region described in the conditions.

To find the solution, it is possible to use the residue theorem, defining a region C as an infinite semicircle in the left half plane of the complex space. It is possible to show that the path integral along such curve tends to zero as the radius tends to infinity. Therefore equation (1.25) can be written as:

$$\frac{1}{2\pi i}\oint_C e^{st}\tilde{x}_n(s)ds = \sum_{i=1}^{n} \operatorname{Res}\left(e^{st}\tilde{x}_n(s), \lambda_i\right) \tag{1.26}$$

With:

$$\operatorname{Res}(e^{st}\tilde{x}_n(s), \lambda_i) = \lim_{s \to -\lambda_i} [(s + \lambda_i)e^{st}\tilde{x}_n(s)] \tag{1.27}$$

Clearly, the constant part of the right side of equation (1.27) is equal to a_i defined in (1.20). Once this term is replaced in (1.26) a similar expression to (1.24) is obtained.

This is a more direct and formal way to obtain the Bateman's solution, being published in 2002 by Pressyanov (Pressyanov, 2002). Strictly speaking, the only difference with the procedure discussed in Section 1.4 was the way in which the inverse Laplace transform was obtained, but the other steps are identical.

1.5.2 The work of Lerch

Bateman did not use the result (1.25) to obtain his solution. In his time there was an early antecedent of this formula provided by Pincherle (Pincherle, 1887), who in fact is cited in the Bateman's publication of 1910, but whose formula was not used to obtain the inverse Laplace transform.

Instead, Bateman used a result developed by the Czech mathematician Mathias Lerch (Lerch, 1903), which set that for the Laplace transform:

$$\int_0^\infty e^{-st} f(t)dt = \phi(s) \tag{1.28}$$

The uniqueness of $f(t)$ given $\phi(s)$ is granted if there is a c such that:

$$\lim_{x\to 0} t^c f\left(\ln\left(\frac{1}{x}\right)\right) = 0 \tag{1.29}$$

Bateman used this result in his work considering that for the equation:

$$\tilde{y}(s) = \int_0^\infty e^{-st} f(t)dt$$

There is only a function $f(t)$ that will yield a function $\tilde{y}_n(s)$. In other words, he used the fact that the inverse Laplace transform is unique, and therefore through the following equation:

$$\tilde{y}(s) = \frac{1}{s+\lambda_i} = \int_0^\infty e^{-st} \cdot f(t)dt$$

He concluded that $f(t) = e^{-\lambda_i t}$, which is the same result that (1.23). Even more, the procedure that is used today in several engineering applications is more related to the Lerch's work than with the formula (1.25).

1.6 The Backward and forward methods.

It was said in Section 1.2.3 that the mass balance given in (1.6) has a recursive nature. Such recursion can be expressed in two ways: backward and forward. In the first type, it is necessary to know the solution of X_n in order to obtain the solution of X_{n-1}. For the second type is the contrary situation.

Both recursive methods are very important because there are some cases where there is not symmetry on a differential equation system related to burnup problems. For example, in Section 1.4.1 it was only necessary to find the solution for $\tilde{x}_n$, and then extending it to other isotopes changing the sub index in an adequate way. Nevertheless, as it will be study in Chapter 4, this situation is not always true, and there are cases where it will be necessary to use the recursion of the solution.

Since the recursion formulations usually are slow or consume a lot of computational time, one of the contributions of the present thesis consist of finding a procedure which transform the recursive formulation to a general one. On the other hand, an additional contribution is related to a general methodology of using one of the recursion methods through the convolution theorem. Both topics are discussed below.

1.6.1 The backward method.

For the index $i = n$, the equation (1.6) can be written as:

$$\frac{dX_n(t)}{dt} = \lambda_{n-1}X_{n-1} - \lambda_n X_n \tag{1.30}$$

Once the term X_{n-1} is isolated:

$$X_{n-1} = \frac{1}{\lambda_{n-1}}\left(\frac{dX_n(t)}{dt} + \lambda_n X_n\right) \tag{1.31}$$

On the other hand, for X_{n-1} there is a mass balance equation given by (1.6) under the index $i = n - 1$:

$$\frac{dX_{n-1}(t)}{dt} = \lambda_{n-2}X_{n-2} - \lambda_{n-1}X_{n-1} \tag{1.32}$$

Again, it is possible to isolate the term X_{n-2} from (1.32) :

$$X_{n-2} = \frac{1}{\lambda_{n-2}}\left(\frac{dX_{n-1}(t)}{dt} + \lambda_{n-1}X_{n-1}\right) \tag{1.33}$$

If (1.31) is replace in (1.33) it follows that:

$$X_{n-2} = \frac{1}{\lambda_{n-2}}\left(\frac{d}{dt}\frac{1}{\lambda_{n-1}}\left(\frac{dX_n(t)}{dt} + \lambda_n X_n\right) + \lambda_{n-1}\frac{1}{\lambda_{n-1}}\left(\frac{dX_n(t)}{dt} + \lambda_n X_n\right)\right)$$

$$= \frac{1}{\lambda_{n-2}\lambda_{n-1}}\left(\frac{d^2}{dt^2} + (\lambda_n + \lambda_{n-1})\frac{d}{dt} + \lambda_n\lambda_{n-1}\right)X_n \tag{1.34}$$

In a similar way, for X_{n-3}

$$\begin{aligned} X_{n-3} = \frac{1}{\lambda_{n-3}\lambda_{n-2}\lambda_{n-1}}\Bigg(&\frac{d^3}{dt^3} + (\lambda_n + \lambda_{n-1} + \lambda_{n-2})\frac{d^2}{dt^2} \\ &+ (\lambda_{n-1}\lambda_n + \lambda_{n-2}\lambda_n + \lambda_{n-2}\lambda_{n-1})\frac{d}{dt} + \lambda_{n-2}\lambda_{n-1}\lambda_n\Bigg)X_n \end{aligned} \tag{1.35}$$

And for the case of X_{n-4}:

$$\frac{1}{\lambda_{n-4}\lambda_{n-3}\lambda_{n-2}\lambda_{n-1}} \times (\frac{d^4}{dt^4} + (\lambda_n + \lambda_{n-1} + \lambda_{n-2} + \lambda_{n-3})\frac{d^3}{dt^3}$$

$$+(\lambda_n\lambda_{n-1} + \lambda_{n-2}\lambda_n + \lambda_{n-2}\lambda_{n-1} + \lambda_{n-3}\lambda_n + \lambda_{n-3}\lambda_{n-1} + \lambda_{n-3}\lambda_{n-2})\frac{d^2}{dt^2} \qquad (1.36)$$

$$+(\lambda_{n-2}\lambda_n\lambda_{n-1} + \lambda_{n-3}\lambda_n\lambda_{n-1} + \lambda_{n-3}\lambda_{n-2}\lambda_n + \lambda_{n-3}\lambda_{n-2}\lambda_{n-1})\frac{d}{dt}$$

$$+\lambda_{n-3}\lambda_{n-2}\lambda_{n-1}\lambda_n)X_n$$

Through the last terms it is possible to propose a general formula to X_{n-k}, it is only necessary to infer the patron in the coefficients before the derivatives. Such task was carried out using combinatorics and represents one of the contributions of this thesis.

1.6.2 Combinatorics

A combination of n elements of a set A, will be defined as a sequence of n elements belonging to A where the order and position is not important. For example, if $A = \{d_1, d_2, d_3, d_4\}$ the following sequences are all the different combinations of two elements of such set:

$$d_1d_2, d_1d_3, d_1d_4, d_2d_3, d_2d_4, d_3d_4$$

After an analysis of the terms inside of (1.34)-(1.36) it is possible to infer that the general formula has the following structure:

$$X_{n-k} = \prod_{j=1}^{k}\frac{1}{\lambda_{n-j}}\left(\frac{d^k}{dt^k} + c_1\frac{d^{k-1}}{dt^{k-1}} + c_2\frac{d^{k-2}}{dt^{k-2}} + \cdots + c_{k-2}\frac{d}{dt} + \prod_{i=0}^{k-1}\lambda_{n-i}\right)X_n \qquad (1.37)$$

The coefficients $c_1, c_2, \dots, c_{k-2}$ are related to combinations of the elements $\lambda_1, \lambda_2, \dots, \lambda_{k-1}$. For example, for c_1 it is possible to note from (1.34) – (1.36):

$$\begin{cases} c_1 = \lambda_n\lambda_{n-1} & \text{for } k = 1 \\ c_1 = \lambda_n + \lambda_{n-1} & \text{for } k = 2 \\ c_1 = \lambda_n + \lambda_{n-1} + \lambda_{n-2} & \text{for } k = 3 \\ c_1 = \lambda_n + \lambda_{n-1} + \lambda_{n-2} + \lambda_{n-3} & \text{for } k = 4 \end{cases} \qquad (1.38)$$

In each case c_1 is the sum of the combinations of one element of the set A_k, with:

$$A_k = \{\lambda_n, \lambda_{n-1}, \dots, \lambda_{n-k+1}\}$$

For c_2, the following relationship is true:

$$\begin{cases} \lambda_n\lambda_{n-1} & \text{for } k = 2 \\ \lambda_{n-1}\lambda_n + \lambda_{n-2}\lambda_n + \lambda_{n-2}\lambda_{n-1} & \text{for } k = 3 \\ \lambda_n\lambda_{n-1} + \lambda_{n-2}\lambda_n + \lambda_{n-2}\lambda_{n-1} + \lambda_{n-3}\lambda_n + \lambda_{n-3}\lambda_{n-1} + \lambda_{n-3}\lambda_{n-2} & \text{for } k = 4 \end{cases} \qquad (1.39)$$

Again, it is possible to note that c_2 is the sum of all combinations of two elements of the set A_k. For c_3 and k_4:

$$\lambda_{n-2}\lambda_n\lambda_{n-1} + \lambda_{n-3}\lambda_n\lambda_{n-1} + \lambda_{n-3}\lambda_{n-2}\lambda_n + \lambda_{n-3}\lambda_{n-2}\lambda_{n-1}$$

Which is equal to the sum of all combinations of three elements of the set A_k. Following a similar reasoning, it is possible to conclude that c_j is the sum of all the combinations of j elements of the set A_k.

1.6.3 An efficient way to express combinations.

Even when the coefficients c_j were found in the last section, it was not provided an efficient way to compute them. In fact, until now it is necessary to carry out at least three steps to build such coefficients: 1) build the set A_k, 2) compute the combinations, 3) perform a sum.

Possibly the most difficult step is related to build the sequences of combinations. It is very interesting that most books deal with the computation of the numbers of combinations, instead that with the procedure to build such sequences. Even more, the author of this thesis was unable to find a reference where the procedure to find the sum of the combinations were discussed. Therefore, it was necessary to develop it.

The idea behind such procedure depends on positions. For example, the following set will be considered:

$$U = \{u_1, u_2, \dots, u_n\}$$

The combinations of r elements, with $1 \leq r$, can be built using the sub index. Firstly, the element u_1 is chosen, and $r-1$ places can be occupied by other elements:

$$u_1\underbrace{\boxed{}\boxed{}\dots\boxed{}}_{r-1\text{ places}} \tag{1.40}$$

In (1.40) the potential places were denoted by squares. Using the sub index, the next element that needs to be chosen is u_2. Clearly u_1 cannot be selected because the combinations are without replacement. Therefore, the follow stage of the sequence is equal to:

$$u_1u_2\underbrace{\boxed{}\boxed{}\dots\boxed{}}_{r-2\text{ places}} \tag{1.41}$$

Once such procedure is repeated r times, the first combination is complete, and it is equal to the following sequence:

$$u_1u_2u_3 \dots u_r \tag{1.42}$$

In order to build the following $n-r$ combinations, it is necessary to replace the last term, u_r, by the next terms $u_{r+1}, u_{r+2}, \dots, u_n$:

$$u_1u_2u_3 \dots u_{r-1}u_r, \qquad u_1u_2u_3 \dots u_{r-1}u_{r+1}, \dots, \qquad u_1u_2u_3 \dots u_{r-1}u_n \tag{1.43}$$

The numbers of sequences given in (1.43) are equal to $(n-r)$. For the next step, it is necessary to replace the last two terms of (1.41):

$$u_1u_2u_3\dots u_{r-2}\boxed{}\boxed{} \tag{1.44}$$

To build a new set of sequences, the terms u_{r-1} will not be included. Therefore, it is necessary to use the next item u_r in (1.44):

$$u_1u_2u_3\dots u_{r-2}u_r\boxed{} \tag{1.45}$$

From this point, the following sequences are built:

$$\begin{cases} u_1u_2u_3\dots u_{r-2}u_ru_{r+1} \\ u_1u_2u_3\dots u_{r-2}u_ru_{r+2} \\ \qquad\vdots \\ u_1u_2u_3\dots u_{r-2}u_ru_{n-1} \\ u_1u_2u_3\dots u_{r-2}u_r\,u_n \end{cases} \tag{1.46}$$

Clearly, there are $(n-r-1)$ sequences this time. Since the element u_{r-1} was not included, all the combinations in (1.46) are different from (1.43). If this procedure is repeated considered as base the sequence given in (1.44), then the following sets of sequences will be built:

$$\underbrace{\boxed{\begin{matrix} u_1u_2\dots u_{r-2}u_{r+1}u_{r+2} \\ u_1u_2\dots u_{r-2}u_{r+1}u_{r+3} \\ \vdots \\ u_1u_2\dots u_{r-2}u_{r+1}u_{n-1} \\ u_1u_2\dots u_{r-2}u_{r+1}\,u_n \end{matrix}}}_{\substack{(n-r-2)\\ \text{sequences}}}\quad \underbrace{\boxed{\begin{matrix} u_1u_2\dots u_{r-2}u_{r+2}u_{r+3} \\ u_1u_2\dots u_{r-2}u_{r+2}u_{r+4} \\ \vdots \\ u_1u_2\dots u_{r-2}u_{r+2}u_{n-1} \\ u_1u_2\dots u_{r-2}u_{r+2}\,u_n \end{matrix}}}_{\substack{(n-r-3)\\ \text{sequences}}}\quad \dots\quad \underbrace{\boxed{\begin{matrix} u_1u_2\dots u_{r-2}u_{n-2}u_{n-1} \\ u_1u_2\dots u_{r-2}u_{n-2}u_n \end{matrix}}}_{\substack{2\\ \text{sequences}}}\quad \underbrace{\boxed{u_1\dots u_{r-2}u_{n-1}u_n}}_{\substack{1\\ \text{sequence}}}$$

All these sequences are different because they differ in at least one element. Until this point there are:

$$1+2+\dots+(n-r)=\frac{(n-r)(n-r+1)}{2}\ \text{sequences} \tag{1.47}$$

For the next step, the following sequence will be considered as base:

$$u_1u_2\dots u_{r-3}\underbrace{\boxed{}}_{1}\underbrace{\boxed{}}_{2}\underbrace{\boxed{}}_{3} \tag{1.48}$$

In the position denoted by 1 it is possible to put the following elements $u_{r-1}, u_r, u_{r+1}, \dots, u_{n-2}$. Once one of these elements is selected, in the position denoted by 2, only the terms with a lager index will be considered. For example, if the term u_{r+1} is chose:

$$u_1u_2\dots u_{r-3}\underbrace{\boxed{u_{r+1}}}_{1}\underbrace{\boxed{}}_{2}\underbrace{\boxed{}}_{3} \tag{1.49}$$

Then, the following terms can be considered for the position denoted by 2: $u_{r+2}, u_{r+3}, \dots, u_{n-2}$. In other words, once that element is selected for the rest of positions, only the terms with a larger index can be considered.

Therefore, the total number of sequences will be given by the following summa:

$$\sum_{i=r-1}^{n-2}\sum_{j=i+1}^{n-1}\sum_{k=j+1}^{n} 1 = \sum_{j=r}^{n-1}\sum_{k=j+1}^{n} 1 + \sum_{j=r+1}^{n-1}\sum_{k=r+2}^{n} 1 + \cdots + \sum_{j=n-2}^{n-1}\sum_{k=n-1}^{n} 1 \qquad (1.50)$$

It is very interesting to note that each double sum of the right side is equal to the number of combinations of two elements of a total $(n-1-j)$. In fact, it is possible to write for the right side:

$$= \underbrace{\frac{(n-r+1)(n-r)}{2}}_{\substack{\text{Number of combinations}\\ \text{of 2 elements of a total}\\ \text{of } n-r}} + \underbrace{\frac{(n-r)(n-r-1)}{2}}_{\substack{\text{Number of combinations}\\ \text{of 2 elements of a total}\\ \text{of } n-r-1}} + \cdots + \underbrace{\frac{(3)(2)}{2}}_{\substack{\text{Number of combinations}\\ \text{of 2 elements of a total}\\ \text{of 3}}}$$

Considering the following relationship (Bartle and Sherbert, 2011):

$$\sum_{k=2}^{n} \frac{1}{2}(k-1)k = \frac{1}{6}(n-1)n(n+1)$$

The sum of (1.49) can be computed as:

$$\frac{1}{6}(n-r)(n-r+1)(n-r+2) \qquad (1.51)$$

The results discussed before can be expressed in terms of combinations of 2 and 3 elements, respectively. First, it is necessary to note that:

$$C(n-r+1,2) = \binom{n-r+1}{2} = \text{eq (1.47)}$$

And:

$$C(n-r+2,3) = \binom{n-r+2}{3} = \text{eq (1.48)}$$

The above discussion shows how the combinations are built using the sub-indexes, and how the number of sequences is related to the number of combinations of 2 and 3 elements of a total of $(n-r+1)$ and $(n-r+2)$.

In this last step, all the sequences have the same root given by (1.49). Then, the only elements that change are $u_{r-1}, u_r, u_{r+1}, \dots, u_{n-2}$, and the number given by the sum in (1.50) can be interpreted as the number of different combinations of 3 elements of the set:

$$\Omega = \{u_{r-1}, u_r, u_{r+1}, \dots, u_{n-2}, u_{n-1}, u_n\}$$

If the argument 1 is replaced by the sequence $u_i u_j u_k$ in such sum, instead of having the number of different combinations, such equation will be equal to the sum of all the combinations of 3 elements of Ω. In other words:

$$\sum_{i=r-1}^{n-2}\sum_{j=i+1}^{n-1}\sum_{k=j+1}^{n} u_i u_j u_k = u_{r-1}u_r u_{r+1} + u_{r-1}u_r u_{r+2} + \cdots$$

$$+u_{r-1}u_r u_n + u_r u_{r+1} u_{r+2} + \cdots + u_r u_{r+1} u_n + \cdots + u_{n-2} u_{n-1} u_n$$

Therefore, for the general case of combinations of r elements of a total of n, the associated sum will be equal to:

$$\mathcal{S}(n,r) = \sum_{l_r=r}^{n-r+1} \sum_{l_{r-1}=l_r+1}^{n-r+2} \dots \sum_{l_2=l_3+1}^{n-1} \sum_{l_1=l_2+1}^{n} \prod_{j=1}^{r} \lambda_{l_j} \qquad (1.52)$$

1.6.4 The backward formulation of the balance equation.

Using the discussion of the last section it is possible to write the coefficients $c_1, c_2, \dots, c_{k-2}$ of the equation (1.37) as

$$\begin{cases} c_1 = & \displaystyle\sum_{j=0}^{k-1} \lambda_{n-k} \\ c_2 = & \displaystyle\sum_{j=0}^{k-2} \sum_{i=j+1}^{k-1} \lambda_{n-j}\lambda_{n-i} \\ c_3 = & \displaystyle\sum_{l=0}^{k-3} \sum_{j=l+1}^{k-2} \sum_{i=j+1}^{k-1} \lambda_{n-l}\lambda_{n-j}\lambda_{n-i} \\ \vdots & \vdots \\ c_{k-2} & \displaystyle\sum_{l_{k-2}=0}^{2} \sum_{l_{k-1}=l_{k-2}+1}^{3} \dots \sum_{l_2=n-2}^{n-1} \sum_{l_1=n-1}^{n} \prod_{j=1}^{k-1} \lambda_{l_j} \end{cases} \qquad (1.53)$$

After a modification of the sub-indexes of (1.52), it is possible to write the coefficients as of (1.53) as:

$$c_i = S^*(i,k) = \sum_{l_i=n-k+i}^{n} \sum_{l_{i-1}=n-k-1}^{l_i-1} \dots \sum_{l_2=n-k+2}^{l_3-1} \sum_{l_1=n-k+1}^{l_2-1} \prod_{j=1}^{i} \lambda_{l_j} \qquad (1.54)$$

And through this last equation, the backward formulation of the balance equation will be equal to:

$$X_{n-k} = \prod_{j=1}^{k} \frac{1}{\lambda_{n-j}} \left(\frac{d^k}{dt^k} + \sum_{i=1}^{k-2} S^*(i,k) \frac{d^i}{dt^i} + \prod_{i=0}^{k-1} \lambda_{n-i} \right) X_n \qquad (1.55)$$

This last equation is one of the contributions of the present thesis, and it represents an important development to the Bateman Equations, because it allows to remove the recursion nature of the equation (1.30). Additionally, it is only necessary to know the solution of X_n to compute all the other solutions, and even when this procedure involves the calculation of higher derivatives of X_n, in Chapter 4 a general formula will be developed to it.

From this point onwards the equation (1.55) will be called the Backward Bateman Equation.

1.6.5 The forward method.

If the equation (1.6) is multiplied by the integral factor given by $e^{\lambda_i t}$, the differential equation can be solved as:

$$e^{\lambda_i t}\left[\frac{dX_i(t)}{dt}+\lambda_i X_i\right]=e^{\lambda_i t}[\lambda_{i-1}X_{i-1}]$$

$$\frac{d}{dt}\left[e^{\lambda_i t}X_i(t)\right]=e^{\lambda_i t}[\lambda_{i-1}X_{i-1}]$$

$$\int_0^t \frac{d}{dt'}\left[e^{\lambda_i t'}X_i(t')\right]dt'=\lambda_{i-1}\int_0^t e^{\lambda_i t'}X_{i-1}(t')dt'$$

$$X_i=e^{-\lambda_i t}\left(\lambda_{i-1}\int_0^t X_{i-1}(t')e^{\lambda_i t'}\,dt'+X_i(0)\right) \tag{1.56}$$

In this last method it is necessary to know the solution for the isotope X_{i-1} to obtain the solution of X_i, therefore it will be called the forward method equation. Considering the initial conditions given in (1.14), it is possible to write X_i as:

$$X_i=e^{-\lambda_i t}\lambda_{i-1}\int_0^t X_{i-1}(t')e^{\lambda_i t'}\,dt', \qquad 2\le i\le n \tag{1.57}$$

Clearly, this equation has a recursive nature. As in the previous case, for the backward method, it is possible to write equation (1.57) as a general formula. Again, it is necessary to make multiple replacements, being convenient for this case considering:

$$X_1(t)=X_1(0)e^{-\lambda_1 t} \tag{1.58}$$

Using equation (1.57) for $i=2$, and replacing (1.58):

$$X_2=e^{-\lambda_2 t}\lambda_1\int_0^t X_1(t')e^{\lambda_2 t'}\,dt'=X_1(0)e^{-\lambda_2 t}\lambda_1\int_0^t e^{-(\lambda_1-\lambda_2)t'}dt' \tag{1.59}$$

Repeating this procedure for $i=3$:

$$X_3=X_1(0)\lambda_1\lambda_2 e^{-\lambda_3 t}\int_0^t e^{-(\lambda_2-\lambda_3)t_2}dt_2\int_0^{t_2} e^{-(\lambda_1-\lambda_2)t_1}dt_1 \tag{1.60}$$

For the case $i=k, k\le i$:

$$X_k=X_1(0)\lambda_{k-1}\lambda_{k-2}\ldots\lambda_2\lambda_1 e^{-\lambda_k t}\times$$

$$\int_0^t e^{-(\lambda_{k-1}-\lambda_k)t_{k-1}}dt_{k-1}\int_0^{t_{k-1}} e^{-(\lambda_{k-2}-\lambda_{k-1})t_{k-2}}dt_{k-2}\ldots\int_0^{t_3} e^{-(\lambda_2-\lambda_3)t_2}dt_2\int_0^{t_2} e^{-(\lambda_1-\lambda_2)t_1}dt_1$$

Using the Fubini's theorem, it is possible to write the last equation as:

$$X_k = X_1(0)e^{-\lambda_k t}\prod_{u=1}^{k-1}\lambda_u \times \int_0^t\int_0^{t_{k-1}}\dots\int_0^{t_3}\int_0^{t_2} e^{-(\lambda_{k-1}-\lambda_k)t_{k-1}} \times e^{-(\lambda_{k-2}-\lambda_{k-1})t_{k-2}} \times \dots \times e^{-(\lambda_1-\lambda_2)t_1} d\tau \tag{1.60}$$

Where $d\tau = dt_{k-1}dt_{k-2} \dots dt_1$. This last equation has some advantages over the Bateman solution and the backward method, but also some disadvantages. Firstly, there are not assumptions about the lambda constants, as it was the case in equation (1.18). In fact, equation (1.60) allows the possibility that two or more lambda constants were equal. In such case it is only necessary to perform the subtractions where necessary, reducing terms and finally carry out the integral. Therefore, this is a more general solution for the linear chain given in (1.11). Besides, this analysis about the lambda constant is performed once and the integrals involved are of two types:

$$\mathcal{J} = \int_0^t x^n e^{(\lambda_i-\lambda_j)x}dx\,, \quad \mathcal{M} = \int_0^t e^{(\lambda_i-\lambda_j)x}dx \tag{1.61}$$

Such integrals can be computed in a fast way, using a symbolical algorithm, and therefore they do not involve any difficulty. In Chapter 5 such integrals will be studied in detail. Finally, several operations involved in (1.60) can be reduced through recursive functions.

The main disadvantage of this equation is its lack of generality. In fact, such equation is only valid for a linear chain, because due to the use of the equation (1.58) is implicit that this isotope has the following balance equation:

$$\frac{d}{dt}X_1 = -\lambda_1 X_1 \tag{1.62}$$

Nevertheless, as it will be discussed in Chapter 4, it is possible to find decay and transmutations structures where this balance equation can have a gain function. For such cases, equation (1.60) is not valid, and therefore it is necessary to use (1.56) or (1.57). Unfortunately, in several cases the involved integration can be very difficult and, therefore, the forward method cannot be used.

In the present thesis, the forward method is fundamental because it is used to develop a symbolical computation algorithm. But it is also important because there are several possible improvements to the Bateman equations that can be carried out through it. The most important aspects of the forward method are discussed in the following sections.

1.6.6. The forward method as an integral transformation.

It is possible to find analogies between the forward method and the equations related to diffusion and transport of neutrons. Such analogies have their origin in the concept of integral transform and the kernel, who are powerful mathematical tools widely used in Physics. The former concept can be defined through the following operation:

$$\int_0^\infty K(s,t)f(t)dt \tag{1.63}$$

Where $f(t) \geq 0$, and $K(s,t)$ is a special function whose main task is transforming, through the integral, a function f of the variable t into a function F of the variable s (Zill, 2009), (Buschman, 1996). This last function is called kernel. Clearly the definitions provided in (1.10) are integral transformations.

In order to analyze the first analogy, between the forward method and the neutron diffusion equation, it is necessary to set the following integral transform to find de neutron flux for a neutron diffusion problem:

$$\phi(\vec{r},t) = \int_{\text{all space}} s\left(\vec{r'},t\right) G\left(\vec{r},\vec{r'},t\right) dV' \tag{1.64}$$

In this case $G\left(\vec{r},\vec{r'},t\right)$ is the kernel, $s\left(\vec{r'},t\right)$ is the source distribution, and $s\left(\vec{r'},t\right) dV'$ are the neutrons emitted per second from the volume dV' located at $\vec{r'}$ (Lamarsh, 2002) .

The analogy begins considering that both, this equation and (1.57), are alternative methodologies to solve the corresponding differential equations. In fact, instead of using a differential formulation, they used an integral approach.

In the neutron diffusion case, this task involves finding the kernel, $G\left(\vec{r},\vec{r'},t\right)$, which is built according to physical and geometric properties. This function has two main roles, firstly, as it was mentioned before, it carries out the task of transforming, through the integral, a function of the vector $\vec{r'}$ into a function of the vector $\vec{r}$. Its second role is related to the following property:

$$\mathbb{L}G\left(\vec{r},\vec{r'},t\right) = \delta(\vec{r}-\vec{r'}) \tag{1.65}$$

Where $\mathbb{L}$ is the operator defined through the neutron diffusion equation:

$$\left(\nabla^2 - \frac{1}{L_D^2}\right) \cdot \phi\left(\vec{r},\vec{r'},t\right) = \mathbb{L} \cdot \phi\left(\vec{r},\vec{r'},t\right) = -\frac{s\left(\vec{r'},t\right)}{D}$$

Where L_D is the diffusion length, and D is the diffusion coefficient. Clearly, property (1.65) allows to carry out a special integration over the position, including where the source is located. The kernels defined by this last property are related to the Green's function, which is widely used in engineering and physical problems (Duffy,2001) (Stakgold and Holst, 2003).

To continue analyzing the analogy between the equations, it is necessary to observe that the formulation given in (1.57) also can be built defining a kernel, as in the neutron diffusion case. Firstly, the equivalent term to $s\left(\vec{r'},t\right)$ will be a function of generation, $R(t')$, which represents the number of atoms or isotopes that are created at time t'. For the kernel function it is necessary to find a function G_r, who models the change that the atoms generated by $R(t')$ undergo. This function will have two arguments, t and t'. It is possible to conclude from the law of radioactive decay that:

$$G_r(t',t) = e^{-\lambda(t-t')} \tag{1.66}$$

Therefore, the concentration of a substance $X(t)$ who is created through the function of generation $R(t')$ can be written as:

$$X_n = \int_0^t R(t')G_r(t',t)dt' = \int_0^t R(t')e^{-\lambda(t-t')}dt' \tag{1.67}$$

Finally, from the transmutation relationship, it will possible to conclude that $R(t') = \lambda_{n-1}X_{n-1}$:

$$X_n = \int_0^t R(t')e^{-\lambda(t-t')}dt' = \lambda_{n-1}e^{-\lambda_n t}\int_0^t X_{n-1}(t')e^{-\lambda_n t'}\,dt' \tag{1.68}$$

Where $\lambda = \lambda_n$. This is the same equation given in (1.57), but this time it was obtained from a point of view of an integral transformation, and following the analogy used for the neutron diffusion case. Additionally, the way in which the kernel was built is similar to the reasoning used by Rutherford to solve the balance equation for $i = 2$, and that was described in Section 1.2.3.

At our best knowledge, the early antecedent of the equation (1.67) appeared in the work of Meyer and Schweidler published in 1927 (Meyer arid Schweidler, 1927), but a formal discussion of it was developed by Rubinson in 1949 (Rubinson, 1949), who analyzed the way in which the integral equation is built.

1.6.7 Generalization of the Integral transformations.

The analogy between the neutron integral transport equation and the forward method is more complex. The author M'Backe Diop made a deep analysis of such analogy in 2007 (M'Backe Diop, 2007), where he used the Monte Carlo formulation for the neutral particle transport.

Through the notation used in the present thesis, and omitting the variable dependence on position $\vec{r}$, the integral equation for the forward method proposed by Diop can be expressed as:

$$X_i(t) = \int_0^t \sum_j X_{j\to i}(t')\cdot p_{j,i}\cdot e^{-\lambda_{j\to i}(t-t')}dt' + \int_0^t U_i(t')e^{-\lambda_{j\to i}(t-t')}dt' \tag{1.69}$$

There are some differences between this equation and (1.57). Firstly, (1.69) considers the possibility that several isotopes, denoted by $X_{j\to i}$, produce the isotope $X_i(t)$. For such reactions, a constant probability density, $\rho_{j,i}$, is defined, being equivalent to the lambda constant in the case of radioactive decay, but with a more general meaning that will be discussed in detail in the following chapter. Additionally, the Diop's equation considers the possibility of a source term, $U_i(t')$, which represents the contribution to X_i, whose origin is not related to the transformation of other isotopes. Clearly, these differences become the equation (1.69) more general than (1.57).

An important contribution of Diop's work is the way in which he transforms the recursive equation (1.69) to a general equation similar to (1.60). Firstly, he considered that transmutation reactions can be divided in generations. For example, in order to obtain the final concentration of the isotope X_i, first it is necessary to sum the concentration of initial isotopes, i.e. the isotopes that were at the beginning of the process of transmutation: $X_i^{(1)}(t)$, where the super index denotes the generation. The next step is to add the concentration of the isotope X_i, which was generated after a first process of transmutation, i.e. $X_i^{(2)}(t)$. Clearly, this function is different from $X_i^{(1)}$. Following this procedure, it is possible to show that:

$$X_i(t) = \sum_{p=1}^{P} X_i^{(p)}(t) \tag{1.70}$$

Where P is the last generation to be considered. In order to compute $X_i^{(p)}$, it is necessary to divide the time's domain in set of disjoint intervals, where the transmutations generation can be located through:

$$\overbrace{\underbrace{0 < t_0 < t_1}_{\substack{\text{first interval}\\ \text{where occurs the}\\ \text{first transmutation}}}}^{p_2}\ ,\ \overbrace{\underbrace{t_1 < t_2 < t_3}_{\substack{\text{second interval}\\ \text{where occurs the}\\ \text{second transmutation}}}}^{p_3}\ \ldots,\ \overbrace{\underbrace{t_{p-1} < t_p < t}_{\substack{\text{final interval}\\ \text{where ocurrs the}\\ \text{final transmutation}}}}^{P} \tag{1.71}$$

Then, to model each generation, Diop used an equation similar to the following expression[1]:

$$X_i^{(p)}(t) = \prod_{j=1}^{p-1} \rho_j \int_0^t \int_{t_0}^t \int_{t_1}^t \ldots \int_{t_p}^t U_i(t_0)\, e^{-\lambda_1(t_1-t_0)} \times e^{-\lambda_2(t_1-t_0)} \times \ldots \times e^{-\lambda_p(t-t_p)} dt_0 dt_1 \ldots dt_p \tag{1.72}$$

Where $U_i(t_0) = X_i(0)$. After brief inspection, it is possible to conclude that this equation is identical to (1.60). In fact, the apparent differences are explained due to the limit's integration. Diop interpreted equation (1.72) as a Neumann series, where an operator is applicated p times. In fact, such interpretation is a generalization of equation (1.63) (Arfken, 1985).Based on this formulation, Diop found the analogies between the forward method and the Monte Carlo formulation of the neutron transport equation, which are listed in Table 1.1.

1.7 Other interpretations and applications of the forward method.

In addition to the interpretations of the forward method given in the last section, there are three important studies who used as a base the integral formulation of the balance equation given in (1.60). The first one has a probability nature, and it is related with the Markov process; the following consist of a moment function that can be used to overcome some numerical difficulties; and the final is related to recursive expressions that allow to reduce the execution time of the Bateman equation.

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[1] In such equation a sum was omitted, but there is not lack of generality for the present analysis.

Table 1. 1. Analogies between integral formulation for neutron transport, and the forward formulation.

Concept	Transport equation	Forward equation
Phase space	$\vec{r}$	t
Phase space	E	type of nuclide
Direction	$\vec{\Omega}$	time irreversibility $t > 0$
Phase particle flux	$\phi(\vec{r},E,\vec{\Omega})$	Isotopic concentration: $X_i(\vec{r},t)$
Process	Nuclear interactions	Nuclear transmutation process
Reaction parameter	$\Sigma_t(\vec{r},E)\ [cm^{-1}]$	$\lambda\ [s^{-1}]$
Reaction density	$\Sigma_t(\vec{r},E)\phi(\vec{r},E,\vec{\Omega})$	Activity $A_i(\vec{r},t) = \lambda X_i(\vec{r},t)$
Source	$S(\vec{r},E,\vec{\Omega})$	$U_i(\vec{r},t)$
Transport operator	$\Sigma_t(\vec{r},E)\int e^{-\int_0^s \Sigma_t(\vec{r}-s'\vec{\Omega},E,\vec{\Omega})ds'}ds$	$\lambda\int_0^t e^{-\lambda(t-t')}dt'$

1.7.1 The interpretation of transition probabilities.

An important interpretation of the forward equation was provided by Raykin and Shlyakhter in 1988 (Raykin and Shlyakhter, 1988), through the concept of transition probabilities and the matricant. The work of these authors is also important to justify a fundamental procedure used to solve the Bateman equation: the linearization process, a topic that will be discussed in detail in Chapter 3. To use transition probabilities, it is necessary to express equation (1.6) in a matrix form:

$$\frac{d}{dt}\boldsymbol{X} = \boldsymbol{AX}$$

Where:

$$\frac{d}{dt}\underbrace{\begin{bmatrix} X_1(t) \\ X_2(t) \\ X_3(t) \\ \vdots \\ X_n(t) \end{bmatrix}}_{\boldsymbol{X}} = \underbrace{\begin{bmatrix} -\lambda_1 & & & & & \\ \lambda_1 & -\lambda_2 & & & & \\ & \lambda_2 & -\lambda_3 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ & & & & \lambda_{n-1} & -\lambda_n \end{bmatrix}}_{\boldsymbol{A}} \underbrace{\begin{bmatrix} X_1(t) \\ X_2(t) \\ X_3(t) \\ \vdots \\ X_n(t) \end{bmatrix}}_{\boldsymbol{X}} \tag{1.73}$$

In their work, the authors called $\boldsymbol{A}$ the reaction rate matrix. After this, it is introduced a transition probability matrix, $\boldsymbol{P}(t)$, where its elements are defined as the concentration of the isotope i at time t, with the initial conditions where only the isotope X_j has a unity initial concentration. Using the Kronecker delta, it is possible to write the mathematical definition of P_{ij} as:

$$P_{ij} = X_i(t)|_{X_k(0)=\delta_{kj}} \tag{1.74}$$

In order to provide clarity for the present analysis, the Bateman equation will be used to sketch the matrix $\boldsymbol{P}(t)$, even when Raykin and Shlyakhter did not use it in their work. According to (1.70), $P_{i=1,j=1}$ will be the solution of the isotope $X_{i=1}$, under the initial conditions where only the isotope $X_{j=1}$ has a unity initial concentration. In order words:

$$\frac{d}{dt}X_1 = -\lambda_1 X_1 \tag{1.75}$$

Under the initial conditions given by:

$$X_1(0) = 1\left[\frac{\text{atoms}}{\text{b·cm}}\right], X_i(0) = 0\ \forall i = 2,3, \dots, n \tag{1.76}$$

Through the Bateman equation it is possible to solve (17.5) with these initial conditions as follows:

$$P_{11} = X_1(0)e^{-\lambda_1 t} = e^{-\lambda_1 t}\left[\frac{\text{atoms}}{\text{b·cm}}\right] \tag{1.77}$$

For the case of $1 < k \leq n$, P_{1k} will be the solution of the isotope X_1 for a time t, where only the isotope X_k, who is after X_1, has an initial concentration different from zero and equal to the unity. Since the balance equation for this term is equal to (1.75), and due that the isotope X_1 do not receive any contribution from other isotopes, because it is at the beginning of the decay chain, depending totally in its initial concentration, it is possible to conclude that:

$$P_{12} = P_{13} = \cdots = P_{1n} = 0$$

Then, the first row of the transition probability matrix is equal to:

$$\begin{pmatrix} e^{-\lambda_1 t} & 0 & 0 & \dots & 0 \end{pmatrix}\left[\frac{\text{atoms}}{\text{b·cm}}\right] \tag{1.78}$$

The term $P_{i=2,j=1}$ is equal to the concentration of the isotope $X_{i=2}$ for a time t, where only the isotope $X_j = 1$ has an initial concentration different from zero, and equal to the unity. For this case it is necessary to solve the balance equation:

$$\frac{d}{dt}X_2 = \lambda_1 X_1 - \lambda_2 X_2 \tag{1.79}$$

Under the same initial conditions given in (1.76).

$$X_1(0) = 1\left[\frac{\text{atoms}}{\text{b·cm}}\right], X_i(0) = 0\ \forall i = 2,3, \dots, n$$

Such solution is equal to:

$$X_2(t) = \frac{\lambda_1\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)}{\lambda_2 - \lambda_1}\left[\frac{\text{atoms}}{\text{b·cm}}\right] \tag{1.80}$$

Following with the process, $P_{i=2,i=2}$ represents the concentration of the isotope $X_{i=2}$, where only $X_{j=2}$ has an initial concentration different from zero, equal to the unity. For this case it is necessary to solve the system (1.79), but with the following initial conditions:

$$X_1(0) = 0, X_2(0) = 1 \left[\frac{\text{atoms}}{\text{b·cm}}\right], X_i(0) = 0 \ \forall i = 3,4,\dots,n$$

Since $X_1(0) = 0$, then $X_1(t) = 0$, the system (1.79) is reduced to:

$$\frac{d}{dt}X_2 = -\lambda_2 X_2 \tag{1.81}$$

Whose solution is equal to $X_2(t) = e^{-\lambda_2 t}\left[\frac{\text{atoms}}{\text{b·cm}}\right]$. For the rest of the terms in this row, $P_{i=2,j=k}$ is the concentration of the isotope $X_2(t)$ with the initial conditions where always, $X_1(0) = X_2(0) = 0$. Since the concentration of X_2 depends only of X_1 and its concentration itself, it is possible to conclude that the second row of the transition probability matrix is:

$$\begin{pmatrix} \dfrac{\lambda_1\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)}{\lambda_2 - \lambda_1} & e^{-\lambda_2 t} & 0 & \dots & 0 \end{pmatrix}\left[\frac{\text{atoms}}{\text{b·cm}}\right] \tag{1.82}$$

As it can be observed, the definition provided in (1.74) can be interpreted as a redefinition of the decay chain given in (1.11), where a shift of positions is included. For example, for P_{22} such chain is redefined as:

$$X_2 \rightarrow X_3 \rightarrow \cdots \rightarrow X_n$$

In other words, the first isotope in (1.11) is removed from the chain. Using another example, for the term $P_{5,3}$, it is necessary to find the concentration of the isotope $i = 5$, considering that only the isotope $j = 3$ has an initial concentration. Under such conditions $X_1(t) = X_2(t) = 0$, and, therefore, the following reduction or shift of the decay chain is obtained:

$$X_1 \rightarrow X_2 \rightarrow \overbrace{\boxed{X_3 \rightarrow X_4 \rightarrow X_5 \rightarrow \ldots \rightarrow X_n}}^{\substack{\text{the redefined}\\ \text{chain}}} \tag{1.83}$$

From the above discussion, the transition matrix can be built as:

$$\boldsymbol{P}(t) = \begin{bmatrix} e^{-\lambda_1 t} & & & & & \\ f_{2,1}(t) & e^{-\lambda_2 t} & & & & \\ f_{3,1}(t) & f_{3,2}(t) & e^{-\lambda_3 t} & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ f_{n,1}(t) & f_{n,2}(t) & f_{n,3}(t) & \dots & f_{n,n-1}(t) & e^{-\lambda_n t} \end{bmatrix} \tag{1.84}$$

Where it is possible to proof that:

$$f_{i,j}(t) = \prod_{u=j}^{i-1}\lambda_u \sum_{r=j}^{i} e^{-\lambda_r t} \prod_{\substack{h=j\\ h\neq r}}^{i} \frac{1}{\lambda_h - \lambda_r}, \text{ with } f_{i,i}(t) = e^{-\lambda_i t} \tag{1.85}$$

This last equation, based on the Bateman solution, models the shift who was described before through the index i and j who appear in the summa and the product. In other words, such equation redefines the decay chain through the index.

1.7.2 Some properties of the probability transition matrix.

There are some important properties that it is necessary to analyze, in order to conclude that the transition probability matrix given in (1.84) has an important relationship with the system (1.73). One of these properties analyses the behavior of $\boldsymbol{P}$ when $t = 0$. The other property is related to $d\boldsymbol{P}(t)/dt$.

For the first property, it is possible to note that all the elements on the diagonal of (1.84) are equal to one when $t = 0$. On the other hand, since the functions $f_{i,j}$ models the concentration of an isotope i, who is produced by previous isotopes, denoted by j, in a system where the condition sets that this last isotope is the only that has an initial concentration different from zero; then it is clear that for time $t = 0$, there is not concentration of the isotope i, because it has not been yet created . Therefore, $f_{i,j}(t = 0) = 0\ \forall i = 1,2,..,n$.

From these facts it is possible to conclude that:

$$\boldsymbol{P}(0) = \begin{bmatrix} 1 & & & & \\ & 1 & & & \\ & & 1 & & \\ & & & \ddots & \\ & & & & 1 \end{bmatrix} = \boldsymbol{I} \tag{1.86}$$

From the second property it is necessary to show that:

$$\frac{d\boldsymbol{P}(t)}{dt} = \boldsymbol{AP}(t) \tag{1.87}$$

To make such proof it is possible to express $\boldsymbol{P}(t)$ as a sum of matrix:

$$\boldsymbol{P}(t) = \boldsymbol{M}(t) + \boldsymbol{T}(t) \tag{1.88}$$

Where:

$$\boldsymbol{M} = \begin{bmatrix} e^{-\lambda_1 t} & & & & \\ & e^{-\lambda_2 t} & & & \\ & & e^{-\lambda_3 t} & & \\ & & & \ddots & \\ & & & & e^{-\lambda_n t} \end{bmatrix}$$

And:

$$\boldsymbol{T}(t) = \begin{bmatrix} 0 & & & & & \\ f_{2,1}(t) & 0 & & & & \\ f_{3,1}(t) & f_{3,2}(t) & 0 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ f_{n,1}(t) & f_{n,2}(t) & f_{n,3}(t) & \cdots & f_{n,n-1}(t) & 0 \end{bmatrix}$$

It follows that:

$$\frac{d\boldsymbol{M}(t)}{dt} = \begin{bmatrix} -\lambda_1 e^{-\lambda_1 t} & & & & \\ & -\lambda_2 e^{-\lambda_2 t} & & & \\ & & -\lambda_3 e^{-\lambda_3 t} & & \\ & & & \ddots & \\ & & & & -\lambda_n e^{-\lambda_n t} \end{bmatrix}$$

Furthermore, from the balance equation (1.6) it is possible to note that:

$$\frac{d}{dt} f_{i,j}(t) = \lambda_{i-1} f_{i-1,j} - \lambda_i f_{i,j} \tag{1.89}$$

Therefore, $d\boldsymbol{T}(t)/dt$ will be equal to:

$$\begin{bmatrix} 0 & & & & & \\ \lambda_1 f_{1,1} - \lambda_2 f_{2,1} & 0 & & & & \\ \lambda_2 f_{2,1} - \lambda_3 f_{3,1} & \lambda_2 f_{2,2} - \lambda_3 f_{3,2} & 0 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_{n-1} f_{n-1,1} - \lambda_n f_{n,1} & \lambda_{n-1} f_{n-1,2} - \lambda_n f_{n,2} & \lambda_{n-1} f_{n-1,3} - \lambda_n f_{n,3} & \cdots & \lambda_{n-1} f_{n-1,n-1} - \lambda_n f_{n,n-1} & 0 \end{bmatrix}$$

If this last term is added to $d\boldsymbol{M}(t)/dt$ the outcome is:

$$\begin{bmatrix} -\lambda_1 e^{-\lambda_1 t} & & & & & \\ \lambda_1 f_{1,1} - \lambda_2 f_{2,1} & -\lambda_2 e^{-\lambda_2 t} & & & & \\ \lambda_2 f_{2,1} - \lambda_3 f_{3,1} & \lambda_2 f_{2,2} - \lambda_3 f_{3,2} & -\lambda_3 e^{-\lambda_3 t} & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_{n-1} f_{n-1,1} - \lambda_n f_{n,1} & \lambda_{n-1} f_{n-1,2} - \lambda_n f_{n,2} & \lambda_{n-1} f_{n-1,3} - \lambda_n f_{n,3} & \cdots & \lambda_{n-1} f_{n-1,n-1} - \lambda_n f_{n,n-1} & -\lambda_n e^{-\lambda_n t} \end{bmatrix}$$

Finally, considering that $f_{i,i} = e^{-\lambda_i t}$, the last matrix can be written as:

$$\begin{bmatrix} -\lambda_1 e^{-\lambda_1 t} & & & & & \\ \lambda_1 e^{-\lambda_1 t} - \lambda_2 f_{2,1} & -\lambda_2 e^{-\lambda_2 t} & & & & \\ \lambda_2 f_{2,1} - \lambda_3 f_{3,1} & \lambda_2 e^{-\lambda_2 t} - \lambda_3 f_{3,2} & -\lambda_3 e^{-\lambda_3 t} & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_{n-1} f_{n-1,1} - \lambda_n f_{n,1} & \lambda_{n-1} f_{n-1,2} - \lambda_n f_{n,2} & \lambda_{n-1} f_{n-1,3} - \lambda_n f_{n,3} & \cdots & \lambda_{n-1} e^{-\lambda_{n-1} t} - \lambda_n f_{n,n-1} & -\lambda_n e^{-\lambda_n t} \end{bmatrix}$$

Which is equal to $d\boldsymbol{P}(t)/dt$.

Furthermore, it is necessary to study the following product:

$$\boldsymbol{AM} = \begin{bmatrix} -\lambda_1 & & & & & \\ \lambda_1 & -\lambda_2 & & & & \\ & \lambda_2 & -\lambda_3 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ & & & & \lambda_{n-1} & -\lambda_n \end{bmatrix} \begin{bmatrix} e^{-\lambda_1 t} & & & & \\ & e^{-\lambda_2 t} & & & \\ & & e^{-\lambda_3 t} & & \\ & & & \ddots & \\ & & & & e^{-\lambda_n t} \end{bmatrix}$$

$$= \begin{bmatrix} -\lambda_1 e^{-\lambda_1 t} & & & & \\ \lambda_1 e^{-\lambda_1 t} & -\lambda_2 e^{-\lambda_2 t} & & & \\ & \lambda_2 e^{-\lambda_2 t} & -\lambda_3 e^{-\lambda_3 t} & & \\ & & & \ddots & \\ & & & \lambda_{n-1} e^{-\lambda_{n-1}} & -\lambda_n e^{-\lambda_n t} \end{bmatrix}$$

And:

$$\boldsymbol{AT} = \begin{bmatrix} -\lambda_1 & & & & & \\ \lambda_1 & -\lambda_2 & & & & \\ & \lambda_2 & -\lambda_3 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ & & & & \lambda_{n-1} & -\lambda_n \end{bmatrix} \begin{bmatrix} 0 & & & & & \\ f_{2,1}(t) & 0 & & & & \\ f_{3,1}(t) & f_{3,2}(t) & 0 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ f_{n,1}(t) & f_{n,2}(t) & f_{n,3}(t) & \dots & f_{n,n-1}(t) & 0 \end{bmatrix}$$

$$= \begin{bmatrix} 0 & & & & & \\ -\lambda_2 f_{2,1} & 0 & & & & \\ \lambda_2 f_{2,1} - \lambda_3 f_{3,1} & -\lambda_3 f_{3,2}(t) & 0 & & & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_{n-1} f_{n-1,1} - \lambda_n f_{n,1} & \lambda_{n-1} f_{n-1,2} - \lambda_n f_{n,2} & \lambda_{n-1} f_{n,3} - \lambda_n f_{n,3} & \dots & \lambda_{n-1} f_{n-1,n-1} - \lambda_n f_{n,n-1} & 0 \end{bmatrix}$$

From the above analysis it is possible to conclude that:

$$\frac{d\boldsymbol{P}(t)}{dt} = \frac{d}{dt}\big(\boldsymbol{M}(t) + \boldsymbol{T}(t)\big) = \boldsymbol{AM} + \boldsymbol{AT} = \boldsymbol{A}(\boldsymbol{M} + \boldsymbol{T}) = \boldsymbol{AP}(t) \qquad (1.90)$$

If a matrix fulfills (1.86) and (1.90), then it is said that such matrix is a matricant of system (1.73) (Pease, 1965).

1.7.3 Reaction rate Matrix as a matricant.

Once it was concluded that the matrix $\boldsymbol{P}$ defined in (1.73) is a matricant, it is possible to apply some of the properties that such matrices have. Particularly, it follows that:

$$\boldsymbol{P}(t - t_1) \cdot \boldsymbol{P}(t_1) = \boldsymbol{P}(t) \qquad (1.91)$$

This property is very important, because it is a matrix analogy to the kernel concept mentioned in (1.63), which shows that this interpretation is equivalent to use an integral transformation. In fact, as some authors suggest, the Green's function can be regarded as the matricant extended to infinitely dimensioned Hilbert spaces (Pease, 1965). On the other hand, the equation (1.91) contains a Markov process nature, because it is possible to determine a certain state of a system, depending only on the immediate previous state.

Even when the coefficients $P_{i,j}$ were computed in the last section using the Bateman equation, it is possible to compute them using probability's properties:

$$P_{i,j}(t) = \int_0^t dt_{i-1} \int_0^{t_{i-1}} dt_{i-2} \dots \int_0^{t_{j+1}} dt_j$$

$$\times \prod_{l=j}^{i-1} \lambda_l \cdot \exp\left[-\int_{t_{l-1}}^{t_l} \lambda_l(\xi)\, d\xi\right] \cdot \exp\left[-\int_{t_{i-1}}^{t_i} \lambda_i(\xi) d\xi\right]$$

Where t_l is the instant of transition of the l'th isotope into the $(l+1)$'th one. and the factor:

$$\lambda_l \cdot \exp\left[-\int_{t_{l-1}}^{t_l} \lambda_l(\xi)\, d\xi\right] \tag{1.92}$$

is the probability of this transition occurring over the interval from t_l to $t_l + dt_l$. And the term:

$$\exp\left[-\int_{t_{i-1}}^{t_i} \lambda_i(\xi) d\xi\right]$$

represents the probability that the i'th nuclide does not pass to the next one over the interval t_{i-1} to t.

Before concluding this section, it is necessary to note that the discussed interpretation has had little impact in the literature and in the developing of burnup codes. In addition to the work of Raykin and Shlyakhter, there has been few publications who address this topic, being the most recent the developed by Hálász and Szieberth (Hálász and Szieberth, 2018).

On the other hand, it was considered convenient to include such discussion in the present thesis, because it represents a little explored field where the Bateman equation can be improved. In addition, there has been pedagogical reasons, because this interpretation is one of the most sophisticated and harder to follow, and all the developments carried out in the last three Sections, represented in the Raykin and Shlyakhter's work a few lines. Therefore, the above discussion represents an introduction to the study of the forward method under a Markov process view.

1.7.4 The forward method as a function of moments.

Other study related to the forward method is described in Harr's master thesis (Harr, 2007) as the exponential moments function. Originally developed in the context of the discrete ordinates transport by Mathews and his colleagues (Mathews et. al., 1994) such function can be defined as:

$$M_n(\lambda_1, \lambda_2, \dots, \lambda_k) = \int_0^1 dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{k-1}} dt_k (1-t_1)^n \tag{1.93}$$

$$\times\, e^{-\lambda_1 t_1} e^{(\lambda_1 - \lambda_2) t_2} \dots e^{(\lambda_{k-1} - \lambda_k) t_k}$$

Considering $n = 0$, the last equation can be used to eliminate overflow errors. For such task, Harr begins considering a very similar equation to (1.60)[2], then he proposed the following change of variable:

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[2] The only difference between (1.60) and the formulation of Harr are the branches ratios.

$$u_j = \frac{t_j}{t} \tag{1.94}$$

Through which:

$$\int_0^{t_j} \frac{dt_{j-1}}{t} = \int_0^{u_j} du_{j-1} \tag{1.95}$$

Using (1.95) in (1.60), the argument of the exponential can be expressed as:

$$\begin{gathered}\int_0^1 \int_0^{u_{k-1}} \dots \int_0^{u_3} \int_0^{u_2} e^{-((\lambda_{k-1}-\lambda_k)t)u_{k-1}} \times e^{-((\lambda_{k-2}-\lambda_{k-1})t)u_{k-2}} \times \dots \\ \times e^{-((\lambda_1-\lambda_2))u_1} d\tau^*\end{gathered} \tag{1.96}$$

Where:

$$d\tau^* = du_{k-1} du_{k-2} \dots du_2 du_1 \tag{1.97}$$

Additionally, the argument of the exponentials can be rewritten including the term $0 = (\lambda_k - \lambda_k)$:

$$\begin{gathered}\int_0^1 \int_0^{u_{k-1}} \dots \int_0^{u_3} \int_0^{u_2} e^{-((\lambda_{k-1}-\lambda_k)t)u_{k-1}} \\ \times e^{((\lambda_{k-1}-\lambda_k)-(\lambda_{k-2}-\lambda_k)t)u_{k-2}} \times \dots \times e^{((\lambda_2-\lambda_k)-(\lambda_1-\lambda_k)t)u_1} d\tau^*\end{gathered} \tag{1.98}$$

Using (1.93), it is possible to write (1.98) as:

$$X_k = X_1(0) e^{-\lambda_k t} \prod_{u=1}^{k-1} \lambda_u \times M_0[(\lambda_{k-1} - \lambda_k)t, (\lambda_{k-2} - \lambda_k)t, \dots, (\lambda_1 - \lambda_k)t] \tag{1.99}$$

By a property of the Bateman equation that will be discussed in Section 1.8, the function M_0 is symmetrical in its arguments (Harr described this as an invariant property):

$$M_0(\beta_1, \beta_2, \dots, \beta_i, \dots, \beta_j, \dots, \beta_n) = M_0(\beta_1, \beta_2, \dots, \beta_j, \dots, \beta_i, \dots, \beta_n) \tag{1.100}$$

The last equation guarantees that the arguments of M_0 can be ordered in any possible way, particularly from the largest to the smallest. Then, using a new numeration:

$$\begin{gathered}\Lambda = \{\lambda_1, \lambda_2, \dots, \lambda_n\} \\ \downarrow \\ \Lambda^* = \{\lambda_{1^*}, \lambda_{2^*}, \dots, \lambda_{n^*}\}\end{gathered}$$

Where λ_1^* is the largest element in Λ , λ_n^* the smallest, and $\lambda_1^* < \lambda_2^* < \cdots < \lambda_n^*$. With this new numeration and the symmetry described in (1.100), it is possible to write:

$$M_0[(\lambda_{n-1}^* - \lambda_n^*)t, (\lambda_{n-2}^* - \lambda_n^*)t, \dots, (\lambda_1^* - \lambda_n^*)t] \tag{1.101}$$

In this form, equation (1.101) ensures that all the exponential arguments are greater than or equal to zero, and therefore it removes several overflow errors. As it was discussed in Chapter

4, these last errors are very important, because due to numerical precision, the Bateman equation fails and provides negative concentrations.

1.7.5 Recursive integral formulas.

The forward method can be used to develop recursive formulas, which allows reducing the complexity and the number of operations in the original Bateman equations. In this topic, the work of John N. Hamawi was one of the most popular from the 70's to the 90's (Hamawi, 1971).

The idea behind the recursive formulas proposed by this author is using the operations that are repeated when the following operation is carry out:

$$e^{-\lambda_k t}\int_0^t\int_0^{t_{k-1}}\dots\int_0^{t_3}\int_0^{t_2} e^{-(\lambda_{k-1}-\lambda_k)t_{k-1}}\times e^{-(\lambda_{k-2}-\lambda_{k-1})t_{k-2}}\times\dots\times e^{-(\lambda_1-\lambda_2)t_1}d\tau \tag{1.102}$$

Where $d\tau = dt_{k-1}dt_{k-2}\dots dt_1$. The recurrence begins with the following definition:

$$E(\lambda_1) = e^{-\lambda_1 t} \tag{1.103}$$

Using this, the following equation is valid:

$$\int_0^t e^{-(\lambda_1-\lambda_2)t}dt = \int_0^t \frac{e^{-\lambda_1 t}}{e^{-\lambda_2 t}}dt = \int_0^t \frac{E(\lambda_1)}{E(\lambda_2)}dt = \frac{e^{-(\lambda_1-\lambda_2)t}-1}{\lambda_2-\lambda_1}$$

Therefore:

$$e^{-\lambda_2 t}\int_0^t e^{-(\lambda_1-\lambda_2)t}dt = e^{-\lambda_2 t}\cdot\frac{e^{-(\lambda_1-\lambda_2)t}-1}{\lambda_2-\lambda_1} = \frac{E(\lambda_1)-E(\lambda_2)}{\lambda_2-\lambda_1} \tag{1.104}$$

Defining $E(\lambda_1,\lambda_2)$ as (1.104), the above calculations can be summarized as follows:

$$E(\lambda_1,\lambda_2) = E(\lambda_2)\int_0^t \frac{E(\lambda_1)}{E(\lambda_2)}dt = \frac{E(\lambda_1)-E(\lambda_2)}{\lambda_2-\lambda_1} \tag{1.105}$$

For the next step, it is necessary to carry out the following integral:

$$\int_0^t\int_0^{t_2} e^{-(\lambda_2-\lambda_3)t_2}\cdot e^{-(\lambda_1-\lambda_2)t_1}\,dt_1dt_2 = \int_0^t e^{\lambda_3 t}\cdot\frac{e^{-\lambda_1 t}-e^{-\lambda_2 t}}{\lambda_2-\lambda_1}dt$$

$$= \int_0^t \frac{E(\lambda_1,\lambda_2)}{E(\lambda_3)}dt = \frac{e^{-(\lambda_1-\lambda_3)t}-1}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} - \frac{e^{-(\lambda_2-\lambda_3)t}-1}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)}$$

Therefore:

$$E(\lambda_1,\lambda_2,\lambda_3) = e^{-\lambda_3 t}\int_0^t\int_0^{t_2} e^{-(\lambda_2-\lambda_3)t_2}\cdot e^{-(\lambda_1-\lambda_2)t_1}\,dt_1dt_2$$

$$= E(\lambda_3)\int_0^t \frac{E(\lambda_1,\lambda_2)}{E(\lambda_3)}dt = \frac{E(\lambda_1)-E(\lambda_3)}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} - \frac{E(\lambda_2)-E(\lambda_3)}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)}$$

Again, the above calculations can be summarized as:

$$E(\lambda_1,\lambda_2,\lambda_3) = E(\lambda_3)\int_0^t \frac{E(\lambda_1,\lambda_2)}{E(\lambda_3)}dt = \frac{E(\lambda_1,\lambda_3)}{(\lambda_3-\lambda_1)} - \frac{E(\lambda_2.\lambda_3)}{(\lambda_3-\lambda_2)} \qquad (1.106)$$

After several algebraic manipulations it is possible to show that:

$$\frac{E(\lambda_1,\lambda_3)}{(\lambda_3-\lambda_1)} - \frac{E(\lambda_2.\lambda_3)}{(\lambda_3-\lambda_2)} = \frac{E(\lambda_1,\lambda_2)}{(\lambda_2-\lambda_1)} - \frac{E(\lambda_2,\lambda_3)}{(\lambda_3-\lambda_2)} \qquad (1.107)$$

In this way:

$$E(\lambda_1,\lambda_2,\lambda_3) = E(\lambda_3)\int_0^t \frac{E(\lambda_1,\lambda_2)}{E(\lambda_3)}dt = \underbrace{\frac{E(\lambda_1,\lambda_2)}{(\lambda_2-\lambda_1)}}_{(1.105)} - \frac{E(\lambda_2,\lambda_3)}{(\lambda_3-\lambda_2)} \qquad (1.108)$$

In the last equation the term given by equation (1.105) has been underlined, in order to show that it is only necessary to compute $E(\lambda_2,\lambda_3)$.

Following this reasoning it is possible to show (Hamawi, 1971):

$$\begin{aligned}(\lambda_1,\lambda_2,\dots,\lambda_i,\lambda_{i+1}) &= E(\lambda_{i+1})\int_0^t \frac{E(\lambda_1,\lambda_2,\dots,\lambda_{i-1,}\,\lambda_i)}{E(\lambda_{i+1})}dt \\ &= \frac{E\big(\lambda_1,\lambda_2,\dots,\lambda_{i-1,}\,\lambda_i\big) - E(\lambda_2,\dots,\lambda_{i+1})}{\lambda_{i+1}-\lambda_i}\end{aligned} \qquad (1.109)$$

Such recursive equations are very useful to reduce the number of calculations, because they identified the repeated operations in the integral (1.102), and therefore the numbers of steps is smaller.

1.7.6 Final considerations of the recursive formulas.

One of the contributions in the present thesis is the development of recursive formulas, which are equivalent (but not identical) to those proposed by Hamawi. In order to show this procedure, it is necessary to begin with the Bateman solution (1.24), for the case $n = 2$:

$$X_2(t) = X_1(0)\lambda_1\left[\frac{e^{-\lambda_1 t}}{\lambda_2-\lambda_1} + \frac{e^{-\lambda_2 t}}{\lambda_1-\lambda_2}\right] \qquad (1.110)$$

When such equation is used in a code, it is convenient to store the following terms:

$$\alpha_1 = \frac{e^{-\lambda_1 t}}{\lambda_2-\lambda_1}, \alpha_2 = \frac{e^{-\lambda_2 t}}{\lambda_1-\lambda_2} \qquad (1.111)$$

Then, for $n = 3$, the Bateman equation for $X_3(t)$ is given by:

$$X_1(0)\lambda_1\lambda_2\left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right] \qquad (1.112)$$

For this case, it is only necessary to calculate the following values:

$$\beta_1 = \frac{1}{\lambda_3 - \lambda_1}, \quad \beta_2 = \frac{1}{\lambda_3 - \lambda_2}, \qquad \gamma^{(3)} = e^{-\lambda_3 t} \tag{1.113}$$

Then, to compute (1.112), it is possible to use (1.111) and carry out the following operations:

$$\alpha_1 \cdot \beta_1 + \alpha_2 \cdot \beta_2 + \gamma^{(3)} \cdot \beta_1 \cdot \beta_2 \tag{1.114}$$

Clearly (1.114) is equal to (1.112). Now, for the term $n = 4$, it is necessary to define the following values:

$$\alpha_1^{(2)} \leftarrow \alpha_1 \cdot \beta_1, \quad \alpha_2^{(2)} \leftarrow \alpha_2 \cdot \beta_2, \quad \alpha_3^{(2)} \leftarrow \gamma_1 \cdot \beta_1 \cdot \beta_2 \tag{1.115}$$

As well as the following calculations:

$$\beta_1^{(2)} = \frac{1}{\lambda_4 - \lambda_1}, \quad \beta_2^{(2)} = \frac{1}{\lambda_4 - \lambda_2}, \qquad \beta_3^{(2)} = \frac{1}{\lambda_4 - \lambda_3}, \quad \gamma^{(4)} = e^{-\lambda_4 t} \tag{1.116}$$

Using the last terms, the Bateman's equation for $n = 4$ is equal to:

$$\alpha_1^{(2)} \cdot \beta_1^{(2)} + \alpha_2^{(2)} \cdot \beta_2^{(2)} + \alpha_3^{(2)} \cdot \beta_3^{(2)} - \beta_1^{(2)} \cdot \beta_2^{(2)} \cdot \beta_3^{(2)} \cdot \gamma \tag{1.117}$$

This procedure can be generalized through the following equation:

$$X_n(t) = X_1(0) \prod_{k=1}^{n-1} \lambda_k \left[\sum_{i=1}^{n-1} \alpha_i^{(n-2)} \beta_i^{(n-2)} + \prod_{j=1}^{n-1} \beta_j^{(n-2)} \gamma^{(n)} \right] n \geq 3 \tag{1.118}$$

Where:

$$\alpha_i^{(n)} = \begin{cases} \alpha_i^{(n-1)} \cdot \beta_i^{(n-1)} & i < n \\ (-1)^{n+1} \gamma^{(n)} \prod_{j=1}^{n-1} \beta_j & i = n \end{cases} \tag{1.119}$$

In computational terms, the recursive formulas require to store a very large amount of data, but the number of calculations decrease. For example, for the case when $n = 4$, with the recursive formula it is only necessary to carry three division given in (1.116), four multiplications and one sum given in (1.117), one exponential evaluation, $\gamma^{(4)}$, and six stored operations.

Whereas with the standard Bateman solution without recursive operations (1.24) it is necessary to perform four exponential evaluations, twelve multiplications, four sums, and twelve divisions. To end this section, it is necessary to mention that Hamawi's work was reconsidered by Robert E. Miles in 1981 (Miles, 1981), who implemented a similar recursive formula in order to avoid singularities. Such work will be discussed in detail in Section 5. Finally, the topic of recursive formulas continues being studied, and in recent years Kai Huang (Huang et. al, 2015) and his colleagues proposed a novel formula that depends only on the original chain less the last isotope, nevertheless the study of recursive formulas will be part of future research.

1.8 Two important theorems.

There are two important theorems that will be used later in the present thesis. One of them is related to a relationship that appears in several problems, and the other one is related to a property of symmetry. Both theorems are discussed in the present section.

1.8.1 Reduction Theorem.

The Reduction Theorem states that:

$$\prod_{j=1, j\neq n}^{n} \frac{1}{d_j - d_n} + \sum_{i=1}^{n-1} \prod_{j=1, j\neq i}^{n} \frac{1}{d_j - d_i} = 0 \qquad (1.120)$$

Where all the numbers $d_1, d_2, \dots, d_n$ are different. Such theorem is very useful, because it reduces a sum of terms, expressing it as a single product. It is necessary to apply (1.120) to express in a symmetric way the Bateman equation. An example will be analyzed in order to show the application. It will assume the solution of $X_3(t)$ using the Bateman equation, and starting from it, the solution of $X_4(t)$ will be computed. If the forward method is used:

$$\begin{aligned} X_4(t) = e^{-\lambda_4 t}\lambda_3 \int_0^t X_3(t) e^{\lambda_4 t} dt = X_1(0)\lambda_1\lambda_2\lambda_3 e^{-\lambda_4 t} \\ \times \int_0^t \left[\frac{e^{-(\lambda_1-\lambda_4)t}}{(\lambda_3-\lambda_1)(\lambda_2-\lambda_1)} + \frac{e^{-(\lambda_2-\lambda_4)t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-(\lambda_3-\lambda_4)t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right] dt \end{aligned} \qquad (1.121)$$

The last integral is equal to:

$$\begin{aligned} \frac{e^{-(\lambda_1-\lambda_4)t} - 1}{(\lambda_3-\lambda_1)(\lambda_2-\lambda_1)(\lambda_4-\lambda_1)} + \frac{e^{-(\lambda_2-\lambda_4)t} - 1}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)} + \frac{e^{-(\lambda_3-\lambda_4)t} - 1}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)} \\ = \sum_{i=1}^{3} \prod_{\substack{j=1 \\ j\neq i}}^{4} \frac{e^{-(\lambda_i-\lambda_4)t}}{\lambda_j - \lambda_i} - \sum_{\substack{j=1 \\ j\neq i}}^{3} \prod_{\substack{j=1 \\ j\neq i}}^{4} \frac{1}{\lambda_j - \lambda_i} \end{aligned} \qquad (1.122)$$

The second term of the last expression is equal to:

$$= \frac{1}{(\lambda_3-\lambda_1)(\lambda_2-\lambda_1)(\lambda_4-\lambda_1)} + \frac{1}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)} + \frac{1}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}$$

After many algebraic manipulations, such expression is equal to:

$$-\frac{1}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)} \qquad (1.123)$$

Nevertheless, instead of carrying out such manipulations it is possible to use (1.120), with $n = 4$, from which:

$$\sum_{i=1}^{3}\prod_{j=1,j\neq i}^{4}\frac{1}{\lambda_j-\lambda_i}=-\prod_{j=1,j\neq 4}^{4}\frac{1}{\lambda_j-\lambda_4} \tag{1.124}$$

Replacing the last expression in (1.122), it follows that:

$$\sum_{i=1}^{3}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{e^{-(\lambda_i-\lambda_4)t}}{\lambda_j-\lambda_i}-\sum_{\substack{j=1\\ j\neq i}}^{3}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{1}{\lambda_j-\lambda_i}=\sum_{i=1}^{3}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{e^{-(\lambda_i-\lambda_4)t}}{\lambda_j-\lambda_i}+\prod_{j=1,j\neq 4}^{4}\frac{1}{\lambda_j-\lambda_4}$$

And, if again the last expression is replaced in (1.121):

$$X_4(t)=X_1(0)\lambda_1\lambda_2\lambda_3e^{-\lambda_4 t}\left\{\sum_{i=1}^{3}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{e^{-(\lambda_i-\lambda_4)t}}{\lambda_j-\lambda_i}+\prod_{j=1,j\neq 4}^{4}\frac{1}{\lambda_j-\lambda_4}\right\} \tag{1.125}$$

$$=X_1(0)\lambda_1\lambda_2\lambda_3\left\{\sum_{i=1}^{3}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{e^{-\lambda_i t}}{\lambda_j-\lambda_i}+\prod_{j=1,j\neq 4}^{4}\frac{e^{-\lambda_4}}{\lambda_j-\lambda_4}\right\}$$

Finally, it is possible to reduce the term inside the curvy brackets under the same index i, extending it to 4:

$$X_4(t)=X_1(0)\lambda_1\lambda_2\lambda_3\sum_{i=1}^{4}\prod_{\substack{j=1\\ j\neq i}}^{4}\frac{e^{-(\lambda_i-\lambda_4)t}}{\lambda_j-\lambda_i} \tag{1.126}$$

Which is the same solution that can be obtained through the Bateman Equation. The importance of the above analysis is related to the useful of the Reduction Theorem, which allowed to simplify a sum of fractions and expressed it as a single term. This property will be fundamental to developments related with cyclic chains, which will be discussed in Chapter 5.

1.8.2 Lagrange Interpolation.

It is possible to make a comparison between the interpolation polynomial in the Lagrange form and the Bateman solution, which is very useful to prove equation (1.120). At our best knowledge, the first authors who provided such proof were Slodička and Balážová (Slodička and Balážová, 2010). The Lagrange's interpolation polynomial for a set of data is given by:

$$(\lambda_1,f_1),(\lambda_2,f_2),\ldots,(\lambda_n,f_n)$$

is equal to (Burden and Faires, 2011):

$$L(x)=\sum_{i=1}^{n}f_i\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{(x-\lambda_j)}{(\lambda_i-\lambda_j)} \tag{1.127}$$

Considering the last equation, it is possible to define the following function:

$$B(x,t) = \sum_{i=1}^{n} f_i(t) \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{(x-\lambda_j)}{(\lambda_i-\lambda_j)} = L(x,t) \tag{1.128}$$

With:

$$f_i(t) = \prod_{k=1}^{n-1} \lambda_k \, e^{-\lambda_i t} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\lambda_j} \tag{1.129}$$

Clearly, $B(0,t)$ is equal to the Bateman equation defined in (1.24). In other words, the Bateman equation can be related to a Lagrange's interpolation polynomial valuated in $x = 0$, and whose coefficients are defined in (1.129). In order to proof the Reduction Theorem given in (1.120), it is necessary to build the Lagrange's interpolation polynomial of the following points:

$$(\lambda_1, g_1), (\lambda_2, g_2), \dots, (\lambda_n, g_n) \tag{1.130}$$

Then, the values $g_1, g_2, \dots, g_n$ will be built using the following function:

$$g_i = g(\lambda_i), \;\; \text{with} \quad g(x) = x \cdot \prod_{j=1}^{n} \frac{1}{\lambda_j} \tag{1.131}$$

The Lagrange's interpolation polynomial has a property of uniqueness, which means that it is the only polynomial, with a degree $< n$, who fulfills that: $L(\lambda_1) = g_1, L(\lambda_2) = g_2, \dots, L(\lambda_n) = g_n$ (Burden and Faires, 2011). Therefore, the Lagrange's interpolation polynomial, built with the conditions (1.130) and (1.131), must to be equal to the function $g(x)$. Using (1.127) and (1.130) and (1.131):

$$L_1(x) = \sum_{i=1}^{n} g_i \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{(x-\lambda_j)}{(\lambda_i-\lambda_j)} = \sum_{i=1}^{n} \prod_{j=1}^{n} \frac{\lambda_i}{\lambda_j} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{(x-\lambda_j)}{(\lambda_i-\lambda_j)} \tag{1.132}$$

At $x = 0$:

$$L_1(0) = \sum_{i=1}^{n} \prod_{j=1}^{n} \frac{\lambda_i}{\lambda_j} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{(0-\lambda_j)}{(\lambda_i-\lambda_j)} = \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{(\lambda_j-\lambda_i)}$$

Then, due to the uniqueness discussed before: $L_1(0) = g(0) = 0$. Therefore:

$$L_1(0) = \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{(\lambda_j-\lambda_i)} = 0$$

Finally, the last sum can be expresses as:

$$\sum_{i=1}^{n}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{1}{(\lambda_j-\lambda_i)}=\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{1}{(\lambda_j-\lambda_i)}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{(\lambda_j-\lambda_n)}$$

And finally:

$$\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{1}{(\lambda_j-\lambda_i)}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{(\lambda_j-\lambda_n)}=0$$

Which is equal to the proposition (1.120). Therefore, the Reduction Theorem is proved.

1.8.3 The symmetry property.

It is possible to define a symmetric function of n arguments, as a function whose result or value does not depend on the order of such arguments (Bartle and Sherpert, 2010). In Section 1.7.4 the symmetry property was discussed through the following equation:

$$M_0(\beta_1,\beta_2,\dots,\beta_i,\dots,\beta_j,\dots,\beta_n)=M_0(\beta_1,\beta_2,\dots,\beta_j,\dots,\beta_i,\dots,\beta_n) \qquad (1.133)$$

Where it is clearly shown that the function M_0 has the same value, whatever the order of its arguments. Such property also appears in some part of the Bateman Equation. Considering equation (1.24):

$$X_n=\prod_{k=1}^{n-1}\lambda_k\sum_{i=1}^{n}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{1}{\lambda_j-\lambda_i}e^{-\lambda_i t}=\prod_{k=1}^{n-1}\lambda_k\ F(\lambda_1,\lambda_2,\dots,\lambda_n) \qquad (1.134)$$

Where the function:

$$F(\lambda_1,\lambda_2,\dots,\lambda_n)=\sum_{i=1}^{n}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{1}{\lambda_j-\lambda_i}e^{-\lambda_i t} \qquad (1.135)$$

Is symmetric in all these arguments. In other words:

$$F(\lambda_1,\lambda_2,\dots,\lambda_i,\dots,\lambda_j,\dots\lambda_n)=F(\lambda_1,\lambda_2,\dots,\lambda_j,\dots,\lambda_i,\dots\lambda_n) \qquad (1.136)$$

This part of the Bateman equation was widely study by the authors Shlyakhter (Shlyakhter, 1983) and Vukadin (Vukadin, 1991), (Vukadin, 1998). In fact, the function (1.135) were called "depletion functions", and it is possible to propose recursive formulas for them, and in the same way in which the moment functions, described in Section 1.7.4, avoid round errors. This property will be very useful in Chapter 4, where several properties of a superposition process will be discussed.

1.9 A more general solution of the balance equations.

One of the most important conditions in the Bateman solution is related to the lambda coefficients, expressed in (1.18), where it was assumed the following identity

$$\frac{1}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)} = \sum_{i=1}^{n} \frac{a_i}{s+\lambda_i} \tag{1.137}$$

Where the coefficients a_i can be computed as:

$$a_i = \left[\frac{(s+\lambda_i)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)}\right]_{s=-\lambda_i} \tag{1.138}$$

Both equations only have sense if all the lambda coefficients are different. Nevertheless, in several burnup problems this condition is not fulfilled, and it is possible that more than one lambda will be equal. Of course, there are several physical implications of such cases, which will be discussed in Chapter 4, because the solution of the structures, where this situation occurs, has several issues related with the execution time and the numerical precision. In the present section a more general solution will be developed, using the Laplace transform.

1.9.1 Decomposition in partial fractions.

The most direct way to obtain a general solution is using a more general partial decomposition when there are repeated roots in the denominator. This is a well-studied topic in disciplines like Pharmacokinetics (Popovíc, 1999), Dynamic Systems (Cochin and Cadwallender, 1997), Signal and Systems (Boyd, 1993), among others. Firstly, it will be considered the following rational function:

$$A(s) = \frac{P(s)}{D(s)}$$

where $P(s) = 1$, and $D(s) = (s+\lambda_r)^n K(s)$, being $K(s)$ a polynomial whose roots are different from $s = -\lambda_r$. Then, it is possible to assume that $A(s)$ can be expresses as (Cochin and Cadwallender, 1997):

$$A(s) = \frac{1}{(s+\lambda_r)^n K(s)} = \frac{A_1}{(s+\lambda_r)} + \frac{A_2}{(s+\lambda_r)^2} + \dots + \frac{A_n}{(s+\lambda_r)^n} + \frac{A_k(s)}{K(s)} \tag{1.139}$$

Where $A_k(s)/K(s)$ is a partial decomposition that does not depend on the root $-\lambda_r$, and $A_1, A_2, \dots, A_n$ are constants. To find A_n it is possible to multiply that expression by $(s+\lambda_r)^n$:

$$A(s)(s+\lambda_r)^n = A_1(s+\lambda_r)^{n-1} + \dots + A_n + \frac{A_k(s)}{K(s)}(s+\lambda_r)^n \tag{1.140}$$

Therefore:

$$A_n = \lim_{s\to-\lambda_r} A(s)(s+\lambda_r)^n \tag{1.141}$$

To find A_{n-1}, it is possible to derivate (1.140):

$$\frac{d}{ds}A(s)(s+\lambda_r)^n = (n-1)A_1(s+\lambda_r)^{n-2} + (n-2)A_2(s+\lambda_r)^{n-3}$$

$$+2(s+\lambda_r)A_{n-2}+A_{n-1}+(s+\lambda_r)^n\frac{d}{dt}\left(\frac{A_k(s)}{K(s)}\right)+n(s+\lambda_r)^{n-1}\frac{A_k(s)}{K(s)}$$

Through the derivation process the term A_1 in (1.140) disappears, and clearly:

$$\lim_{s\to-\lambda_r}\frac{d}{ds}A(s)(s+\lambda_r)^n=A_{n-1}$$

Following such reasoning:

$$A_{n-k}=\frac{1}{k!}\lim_{s\to-\lambda_r}\frac{d^k}{ds^k}[A(s)(s+\lambda_r)^n] \tag{1.142}$$

Considering a more general function:

$$A(s)=\frac{P(s)}{(s+\lambda_1)^{k_1}(s+\lambda_2)^{k_2}\dots(s+\lambda_n)^{k_n}} \tag{1.143}$$

From the above discussion, it is clear that (1.143) can be expressed as:

$$\frac{A_{k_1,k_1}}{(s+\lambda_1)^{k_1}}+\frac{A_{k_1,k_1-1}}{(s+\lambda_1)^{k_1-1}}+\dots+\frac{A_{k_1,1}}{(s+\lambda_1)}+\frac{A_{k_2,k_2}}{(s+\lambda_2)^{k_2}}+\dots+\frac{A_{k_2,1}}{(s+\lambda_2)}$$
$$+\dots+\frac{A_{k_n,k_n}}{(s+\lambda_n)^{k_n}}+\frac{A_{k_n,k_n-1}}{(s+\lambda_n)^{k_n-1}}+\dots+\frac{A_{k_n,1}}{(s+\lambda_n)} \tag{1.144}$$

$$=\sum_{u=1}^{n}\sum_{q=1}^{k_u}\frac{A_{k_u,k_{u,q}}}{(s+\lambda_u)^u}$$

Where:

$$A_{k_i,k_i-j}=\frac{1}{j!}\lim_{s\to-\lambda_i}\frac{d^j}{ds^j}[A(s)(s+\lambda_i)^n] \tag{1.145}$$

1.9.2 General solution.

A linear chain with n different isotopes will be considered, each one appearing in the structure several times given by the numbers $k_1,k_2,\dots,k_n$, respectively, and with the lambda coefficients denoted by $\lambda_1,\lambda_2,\dots,\lambda_n$. Repeating similar steps from (1.12) to (1.15) it follows:

$$\tilde{x}_n=X_1(0)\prod_{k=1}^{n-1}\lambda_k\sum_{u=1}^{n}\sum_{q=1}^{k_u}\frac{A_{k_u,k_{u,q}}}{(s+\lambda_u)^u} \tag{1.146}$$

Considering the inverse Laplace transform of (1.146), it follows:

$$X_n = \mathcal{L}^{-1}\{\tilde{x}_n\} = X_1(0)\prod_{k=1}^{n-1}\lambda_k \sum_{u=1}^{n}\sum_{q=1}^{k_u} A_{k_u,k_{u,q}}\mathcal{L}^{-1}\left\{\frac{1}{(s+\lambda_u)^u}\right\} \tag{1.147}$$

Using that:

$$\mathcal{L}^{-1}\left\{\frac{1}{(s+\lambda_u)^u}\right\} = \frac{e^{-\lambda_u t}t^{u-1}}{(u-1)!} \tag{1.148}$$

Therefore, the Bateman solution to the general case is equal to:

$$X_n = \mathcal{L}^{-1}\{\tilde{x}_n\} = X_1(0)\prod_{k=1}^{n-1}\lambda_k \sum_{u=1}^{n}\sum_{q=1}^{k_u} A_{k_u,k_{u,q}}\frac{e^{-\lambda_u t}t^{u-1}}{(u-1)!} \tag{1.149}$$

1.9.3 Final considerations of the general Bateman solution.

As it can be noticed from the previous section, the development of the modified Bateman solution, that deals with repeated elements, is a straightforward task using Laplace transform.

At our best knowledge, the first author who published a general solution was Michael J. Newman, who followed the research line of Clayton (Clayton et. al., 1961), and published the exact solution of the s-process or slow neutron capture process (Newman, 1978). In his work, Newman developed a very similar expression to (1.149) using the same procedure: the Laplace method. The only difference between his equation and the one developed in the past section is that the removal coefficients only considered capture cross sections in his work.

Nevertheless, after Newman, about 15 different works who contains general solutions have been published. These works do not necessary use the Laplace Inversion Method, and they belong to a wide range of disciplines, from astrophysics to transport theory, including nuclear engineering.

This is a very interesting fact, because some authors developed their own solutions practically ignoring the previous works, and it seems that some of them did not know about the publications of authors that belonged to other disciplines. Nevertheless, this situation did not prevent that their solutions were obtained in many ingenious and original ways.

About a third part of the developed work in the present thesis is related to the general solutions, because one of the most important contributions consist of developing a Symbolical Computational Algorithm who implements one of such solution to a burnup code.

Therefore, the discussion about the general solutions among other topics, will be cover in detail in Chapter 5.

1.10 Conclusions of the Chapter 1.

In the present chapter, the fundamentals of the Bateman Equation were discussed. Through a detailed review of the literature, it was possible to conclude that Harry Bateman neither proposed the balance equations nor was the first who solve it, even when several authors make this affirmation in the open literature. Instead, in the present chapter was concluded that his main contribution was related with the development of the Laplace method to solve differential equations.

In terms of the balance equation, in the present chapter the backward and the forward method were discussed. For the first methodology it was possible to transform a recursive formula, to a general one, which represents an important step in the analysis of the equations, because starting from a given solution it is possible to find others.

In addition, in the present chapter several interpretations and applications of the forward method were analyzed. As it will be discussed in Chapter 5, such methodology is the core of the symbolical algorithm developed in the present thesis.

Finally, through the Laplace method developed by Bateman it was possible to find a general solution, which admits the case with repeated lambda constants. Such solution was obtained in a straightforward way, which becomes very interesting because it was the first solution to be published, but there at least 15 others reported in literature. The last fact can only be explained considering that several authors, who developed their formulas, ignored the work of other authors, or maybe they want to show another procedure to obtain them.

The general solution described in the present chapter, as long with the main theorems and properties discussed here, represents the main results who will be used in the rest of the thesis.

Chapter 2. Branching Ratios and Fission Yields.

Among the nuclear data that is used in burnup and activation problems, the branching ratios and the fission yields deserve a special discussion, because during the research related with the present thesis some issues emerged, which represented an important challenge to the development of the code Szilard.

The present chapter covers the most important topics related with these nuclear parameters, from the way in which they can be found in nuclear data libraries, until the way in which it can be implemented. Additionally, this chapter also contains a formal analysis about the yields in terms of the topology of the decay and transmutation networks, which is an important contribution to the state of the art.

2.1 Branching ratios.

As it was discussed in the past chapter, the Bateman's equation was used to model successive transformations originated by radioactive decay. Originally, there was not a full understanding about the radioactive decay process and, therefore, the lambda constants that were used represent the total decay constants. This implies that such system does not consider the possibility that the isotopes have more than one decay mode, or that they could undergo other reactions as the fission. Today it is known that the isotopes can have several decay modes, each one characterized by a specific decay constant.

Certainly, Rutherford identified in the study of the Thorium emanations, that some isotopes emitted several types of radioactivity (Rutherford, 1904). In fact, he described three types of reactions, α, β and γ, for the stage that he named as "second change", which was discussed in Section 1.2.2.

Nowadays it is known that such stage was the isotope $^{212}\mathrm{Bi}$, for which:

$$^{212}\mathrm{Bi} \overset{r}{\rightarrow} \begin{cases} ^{208}\mathrm{Tl}, & r = \alpha \\ ^{212}\mathrm{Po}, & r = \beta \\ ^{212}\mathrm{Bi}^{*}, & r = \gamma \end{cases} \tag{2.1}$$

Therefore, it is clear that Rutherford discovered the possibility of several types of decay. Nevertheless, this was not included in the formulation of the mass balance (1.6). Considering that the different decays process are independent events, it is possible to define the total lambda constant for a given isotope as:

$$\lambda_i = \sum_g \lambda_i^{(g)} \tag{2.2}$$

Where the $\lambda_i^{(g)}$ is the lambda constant of the isotope i related with the process or decay mode g. As it will be discussed later in this chapter, the equation (2.2) can be deduced from probabilistic considerations. Since there is a uniqueness between the reaction that an isotope undergoes and the product of such reaction, then it is possible to use the following notation for $\lambda_i^{(g)}$:

$$\lambda_i^{(g)} = \lambda_{i,j} \tag{2.3}$$

Where again the sub index i denotes the isotope that is decaying, and the sub index j is the product of such reaction. Clearly the relationship between (2.2) and (2.3) is given by:

$$\lambda_{i,j} = b_{i,j}\lambda_i \tag{2.4}$$

Where the constant $b_{i,j}$ is known as the branching ratio, i.e., the fraction of decays of the isotope i that produces the isotope j. In order to provide generality to the balance equation given in (1.6), it is necessary to include these branching ratios, which can be made as follows:

$$\begin{cases} \dfrac{dX_1}{dt} = & -\lambda_1 X_1 \\ \dfrac{dX_2}{dt} = & b_{1,2}\lambda_1 X_1 - \lambda_2 X_2 \\ \cdots & \cdots \\ \dfrac{dX_n}{dt} = & b_{n-1,n}\lambda_{n-1}X_{n-1} - \lambda_n X_n \end{cases} \tag{2.5}$$

Following similar steps as in the case of (1.24), it is possible to show that the Bateman equation for a linear chain, who consider the branching ratios, is equal to:

$$X_n = X_1(0)\prod_{k=1}^{n-1} b_{k,k+1}\lambda_k \sum_{i=1}^{n}\prod_{\substack{j=1 \\ j\neq i}}^{n}\frac{1}{\lambda_j - \lambda_i}e^{-\lambda_i t} \tag{2.6}$$

2.2 Branching ratios and the ENDF/B-VII.1 Library.

As with other parameters, it is possible to consult the branching ratios in several libraries of nuclear data. In the present work the ENDF/B-VII.1 library was used. At the date in which the present thesis is written there is a new actualization of such library, which was published in 2018: the ENDF/B-VIII.0. Nevertheless, the study of the branching ratios was cover in the early stages of the thesis work, when such library had not yet been published, and therefore some of the following conclusions related with missed data and other issues are referring only to ENDF/B-VII.1. In terms of the format, both versions of the library share the same structure.

2.2.1 Structure of the file

The ENDF/B-VII.1 can be downloaded from the web site of the National Nuclear Data Center[3] as a file with extension .gz. There is a folder named “decay” inside such file, who contains 3821 text files with an extension “.endf”. Each file has an identification name given by the following format:

$$\underbrace{\text{dec}}_{\boxed{1}}\underbrace{\text{-}}_{\boxed{2}}\underbrace{092}_{\boxed{3}}\underbrace{\_}_{\boxed{4}}\underbrace{\text{U}}_{\boxed{5}}\underbrace{\_}_{\boxed{6}}\underbrace{235}_{\boxed{7}}.\text{endf} \tag{2.7}$$

Where $\boxed{1}$ is the identifier of the “decay” sub library, $\boxed{2}$, $\boxed{4}$ and $\boxed{6}$ are separators, $\boxed{3}$ is the atomic number, $\boxed{5}$ is the symbol’s element, and $\boxed{7}$ is the mass number. Clearly, through the names of the text files it is possible to build a list of the isotopes, their atomic and mass number, and their symbols. Among the decay information that is contained in each text-file, it is possible to find the following lines:

$$\begin{gathered}\text{Parent half life:}\\ \text{Decay mode:}\end{gathered} \tag{2.8}$$

The information about the half-life is showed after the first line, and the type of decay is given after the second one. Figure 2.1 contains two examples of these structure for two files corresponding to the ^{235}U and the metastable^{133}Pm isotopes.

For the half-life information and the decay modes, the ENDF format uses a set of capital letters and acronyms that are listed in Table 2.1 and Table 2.2 respectively.

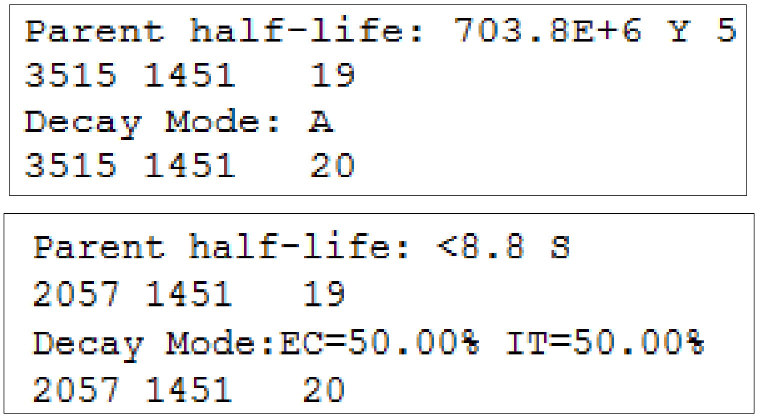
For isotopes that have more than one decay modes, each type is listed following by the branching ratio expressing as a percentage value. Figure 2.1 shows that for the metastable^{133}Pm isotope there are two decay modes: EC (electronic capture), and IT (isomeric transition), each of them with a branching ratio of 50%.

It is worth mentioning that some half-lives are given in energy units of electron-volts instead of time unities, for example for the ^{4}H isotope the ENDF reports a half-life of 4.6 MeV. For such cases it is possible to convert the values to units of time through the following equation (Duderstadt, 1974):

$$\Gamma = \frac{h\lambda}{2\pi} \tag{2.9}$$

Where h is the Planck’s constant and λ is the decay constant. For some isotopes the half-life is give as an upper bound, as it can be observed in Figure 2.1 for the metastable^{133}Pm isotope. In other cases, the value of the half-life is not known and a label of “unknown” or “?” is used instead.

The information described above is fundamental to a burnup code, because it allows to build the mass balance equation for the decay reaction. Firstly, it is necessary to build a database with such data, after which an algorithm to follow the successive transformation is required.



```
Parent half-life: 703.8E+6 Y 5
3515 1451    19
Decay Mode: A
3515 1451    20
```

```
Parent half-life: <8.8 S
2057 1451    19
Decay Mode:EC=50.00% IT=50.00%
2057 1451    20
```

Figure 2. 1. Two examples of the structure of the text files of the sub library of decay for the lines given in (2.8). The upper one is corresponding to ^{235}U and the lower one is related to ^{133}Pm.

Table 2. 1 Capital letters and acronyms that are used by the ENDF/B-VII.1 Library to express the half-lives.

Symbol	Unity or prefix	Symbol	Unity or prefix
Y	Year	M	Milli
D	Day	U	Micro
H	Hour	N	Nano
M	Minute	P	Pico
S	Second	K	Kilo
EV	Electron-volt	M	Mega

Table 2. 2a. Capital letters and acronyms that are used in the ENDF/B-VII.1 library to represent the decay modes.

Decay mode	Capital Letter	Reaction	Decay mode	Capital Letter	Reaction
α	A	${}_{Z}^{A}X \rightarrow {}_{Z-2}^{A-4}Y$	β^{+}	B+	${}_{Z}^{A}X \rightarrow {}_{Z-1}^{A}Y$
β^{-}	B-	${}_{Z}^{A}X \rightarrow {}_{Z+1}^{A}Y$	n	N	${}_{Z}^{A}X \rightarrow {}_{Z}^{A-1}X$
$2n$	2N	${}_{Z}^{A}X \rightarrow {}_{Z}^{A-2}X$	$3n$	3N	${}_{Z}^{A}X \rightarrow {}_{Z}^{A-3}X$
Electronic Capture	EC	${}_{Z}^{A}X \rightarrow {}_{Z-1}^{A}Y$	p	P	${}_{Z}^{A}X \rightarrow {}_{Z-1}^{A}Y$

Table 2.2b. (Continuation) Capital letters and acronyms that are used in the ENDF/B-VII.1 Library to represent the decay modes.

Decay mode	Capital Letter	Reaction	Decay mode	Capital Letter	Reaction
$2p$	2P	${}^{A}_{Z}X \to {}^{A}_{Z-2}Y$	$\beta^{+} + n$	B+N	${}^{A}_{Z}X \to {}^{A-1}_{Z-1}Y$
$\beta^{-} + 2n$	B2N	${}^{A}_{Z}X \to {}^{A-2}_{Z-1}Y$	$\beta^{-} + 3n$	B3N	${}^{A}_{Z}X \to {}^{A-3}_{Z-1}Y$
$\beta^{-} + 4n$	B4N	${}^{A}_{Z}X \to {}^{A-4}_{Z-1}Y$	Isomeric transition	IT	${}^{A}_{Z}X^{*} \to {}^{A}_{Z}X$

2.2.2 Some issues with the decay data.

A recurrent issue with the decay sub library of the ENDF/B-VII.1 is that the format is not uniformly followed for certain text files.

For example, in some cases a branching ratio is reported even when the isotope has only one decay mode, but in other text files this is not the case. Additionally, sometimes the equality symbol between the decay mode and the branching ratio is omitted, as well as the colon symbol. Besides the blank spaces between the values are not always respected.

Nevertheless, the most important issues are related with the lack of information. For example, the isotopes ^{40}Si, ^{28}P, and ^{36}K have more than one mode of decay, but the text files do not contain the corresponding branching ratios. Therefore, it is necessary to complete such information from other libraries in order to include them in the solution of a given chain. Through the present study, it was found that nearly other 90 isotopes have the same situation about lack of information.

Another interesting issue is related with the following summa:

$$\sum_{k=1}^{K} b_{i,k} = 1 \tag{2.10}$$

Where k is related with X_k, an isotope of a total of K isotopes who are produced by the decay of X_i. Clearly (2.10) must to be fulfilled.

Nevertheless, as some authors affirm, such sum can be greater than 1 for some isotopes. In fact, according to Isotalo (Isotalo, 2013), “the sum of branching ratios coming from a single nuclide can be greater than one as some reactions…”. , This can be considered as a contradiction, because it would imply that the mass balance is not conserved.

This issue represented a mystery on the first stage of the present research, and it was possible to confirm it finding several isotopes in ENDF/B-VII.1 for which this was true. In Table 2.3 30 examples of this are listed. However, this is not an error or contradiction. In fact, it is related with the way in which the branching rations are computed and the probabilistic nature of some reactions.

Table 2. 3. Examples of 30 isotopes whose sum of branching ratios is greater than 1, according to data obtained from ENDF/B-VII.1.

Isotope	$\sum_j b_{i,j}$	Isotope	$\sum_j b_{i,j}$	Isotope	$\sum_j b_{i,j}$	Isotope	$\sum_j b_{i,j}$
^{27}F	1.77	^{29}Ne	1.219	^{30}Na	1.32	^{135}Gd	1.18
^{110}I	1.11	^{113}Xe	1.071	^{114}Ba	1.29	^{15}B	1.97
^{145}Dy	1.5	^{143}Er	2	^{152}Lu	1.15	^{43}S	1.4
^{16}C	1.99	^{22}N	1.49	^{38}P	1.12	^{35}Ca	1.957
^{45}Cl	1.24	^{31}Ar	1.69	^{49}K	1.86	^{71}Co	1.026
^{39}Ti	2	^{43}Cr	1.23	^{46}Mn	1.22	^{101}Sn	1.26
^{57}Zn	1.65	^{61}Ge	1.8	^{75}Sr	1.052		
^{109}Te	1.09	^{114}Cs	1.09	^{104}Sb	1.08		

In order to explain it, the isotope ^{27}F will be analyzed. According to ENDF/B-VII.1, such isotope has the following decays with the respective branching ratios:

$$\beta^- = 100\%, \quad \beta^- n = 77.00\% \tag{2.11}$$

Clearly the sum is equal to 1.77 or 177%. This does not mean that the mass conservation is not valid, instead it means that a beta decay always is presented each time that the ^{27}F isotope decays. But, only in the 77% of such decays a neutron is emitted.

Clearly this is a conditional probability, because the emission of the neutron is a dependent event: always that a neutron is emitted, a beta decay is presented. A similar analysis can be carried out by all the isotopes whose sum is greater than one. Therefore, this is not an error or a contradiction, and it is very important to be considered for a detailed analysis of the decay process. Finally, in order to describe the reactions, it follows that the daughters of the isotope ^{27}F, the isotopes ^{27}Ne and ^{26}Ne are produced with a 23% and 77%, respectively.

2.3 The Bateman equation and the neutron flux.

Over the years, and with the discovery of neutron-nuclei reactions, another type of successive transformations appeared in addition to the decay process, and therefore it was necessary to model them. In 1949 Rubinson (Rubinson, 1949) observed that these reactions can be treated by similar balance equations to the radioactive decay's type, with an effective transformation constant given by:

$$\lambda_i^{\text{eff}} = \lambda_i + \sigma_i \phi \tag{2.12}$$

Where σ_i is the microscopic cross section of the nuclide i, and ϕ is the neutron flux. This expression can be extended to consider different reactions of type m, and multigroup neutron fluxes denoted by g:

$$\lambda_i^{\text{eff}} = \lambda_i + \sum_g \sum_m \phi^g \ \sigma_{m,i}^g \tag{2.13}$$

If a multigroup methodology is not used, then it is possible to analyze the neutron reaction rates in order to define the effective lambda constant. It is possible to write the volumetric reaction rate m of nuclide i, as:

$$R_{i,m}(E) = X_i \sigma_{i,m}(E)\phi(E) \tag{2.14}$$

Where X_i is the atomic density of nuclide i. This volumetric reaction rate has its origin in a beam of incident neutrons of energy E. The total volumetric reaction rate can be obtained through the integral over all the neutron energies (Isotalo, 2013):

$$\begin{aligned} R_{i,m} &= \int_0^{\infty} R_{i,m}(E)dE = X_i \int_0^{\infty} \sigma_{i,m}(E)\phi(E)dE \\ &= X_i \underbrace{\left(\frac{\int_0^{\infty} \sigma_{i,m}(E)\phi(E)dE}{\int_0^{\infty} \phi(E)dE}\right)}_{\sigma_{i,m}} \underbrace{\left(\int_0^{\infty} \sigma_{i,m}(E)\phi(E)dE\right)}_{\phi_h} \\ &= X_i \sigma_{i,k}^h \phi_h \end{aligned} \tag{2.15}$$

In the last expression ϕ_h and $\sigma_{i,m}^h$ are the homogenized one group-flux and one-group cross section. Using this definition, it is possible to write the effective removal lambda coefficient as:

$$\lambda_i^{\text{eff}} = \lambda_i + \phi_h \sum_k \sigma_{i,k}^h \tag{2.16}$$

Starting with the definitions provided in (2.13) and (2.17) it is possible to define a new branching ratio who includes the neutron-nucleus reactions, and therefore to generalize the Bateman equation. Nevertheless, firstly it is necessary to briefly discuss about the fission process and the concept of yield.

2.3.1 The fission process and the yield concept.

Essentially, the fission can be defined as the division or split of a heavy nucleus in two or more lighter nuclei, in which energy and neutrons are produced. This process was discovered a few years after the discovery of the neutron, and the term has its origin in biology, where it is used to call a division of an organelle o entity into small parts or organelles (Corcho Orrit, 2013).

There are two ways in which the fission occurs: the induced way and the spontaneous one. In the first case it is necessary a neutron who interacts with the nucleus, while the second one can occur in absence of it, because it happens as result of the nucleons interactions inside the nucleus.

The isotopes that are produced as result of this process are called “fission products”. In the case of the induced fission, such products depend on the energy of the incident neutron, and they are not always the same. Instead, they follow a statistical distribution, who is unique according to the isotope that undergoes fission. In other words, for a given heavy isotope and a given neutron incident energy, there is a unique distribution in which the fission products appear. A percentual ratio is used to describe the amount of fission product that appear in each fission, which can be defined as:

$$\gamma_{i,j}(E) = \overline{N}(X_i, X_j, E)\left(\sum_j \overline{N}(X_i, X_j)\right)^{-1} \tag{2.17}$$

Where $\overline{N}(X_i, X_j)$ is the average number of isotopes X_j that are produced as a fission of X_i for a statistical sample who was measured at the energy E, and the summa is carry out considering all the fission products that can be generated as result of the fission of X_i. The number defined in (2.17) is called as the fission yield, and it is analogue to the branch ratio defined in the Section 2.1.

2.3.2 The generalization of the branching ratio.

Using (2.17), it is possible to define the effective branching ratio, for a multigroup formulation, by the following expression (Isotalo, 2013):

$$b_{i,j}^{\text{eff}} = \frac{b_{i,j}\lambda_i + \omega}{\lambda_i^{\text{eff}}}, \qquad \omega = \sum_g \sum_m f_{i,i+1,m}^g \phi^g \, \sigma_{m,i}^g \tag{2.18}$$

Where:

$$f_{i,i+1,m}^g = \begin{cases} \gamma_{i,i+1,m}^g & \text{for fission proccess} \\ 1 & \text{for other neutron-nucleus reaction} \end{cases} \tag{2.19}$$

In this case, $\gamma_{i,i+1,m}^g$ is the fission yield of the isotope i, related with the production of the isotope j and the energy group g. For the case of one group formulation, such generalization is given as:

$$b_{i,j}^{\text{eff}} = \frac{b_{i,j}\lambda_i + \omega}{\lambda_i^{\text{eff}}}, \qquad \omega = \phi_h \sum_m f_{i,j,m}^h \sigma_{i,m}^h \tag{2.20}$$

Where $\sigma_{i,k}^h$ and ϕ_h were defined in equation (2.15) and $f_{i,j,m}^h$ is defined as:

$$f_{i,j,m}^g = \begin{cases} \dfrac{\int_0^\infty \gamma_{i,j}(E)\phi(E)dE}{\int_0^\infty \phi(E)dE} & \text{for fission proccess} \\ 1 & \text{for other neutron-nucleus reaction} \end{cases} \tag{2.21}$$

From this point forward, we will call the term λ_i^{eff} as the effective removal coefficient. An interesting fact about the definitions provided in (2.18) and (2.20) is that, for practical cases, only one term in the numerator of the branching ratio is different. In other words:

$$b_{i,j}^{\text{eff}} = \begin{cases} \dfrac{b_{i,j}\lambda_i}{\lambda_i^{\text{eff}}} & \text{for decay reactions} \\ \dfrac{\omega}{\lambda_i^{\text{eff}}} & \text{for neutron-nucleus reactions} \end{cases} \tag{2.22}$$

This is due that the unique scenario where both terms, $b_{i,j}\lambda_i$ and ω, are different from zero is when an isotope j can be produced as result of a decay reaction and a neutron-nucleus reaction, that occur in the isotope i. For example, if an isotope X_i undergoes decay neutron emission as well as the reaction $(n, 2n)$, in both cases the following equation is valid:

$$ {}^{A}_{Z}X_i \rightarrow {}^{A-1}_{Z}X_j $$

And, therefore, the numerator of $b^{\text{eff}}_{i,j}$ for this example is equal to $b_{i,j}\lambda_i + \omega$ and (2.22) cannot be used. Nevertheless, as it was mentioned before, for practical cases only, one of these terms is different from zero. Using the elements that were discussed before, the original Bateman's equation (2.6) can be written as:

$$ X_n(t) = X_1(0) \prod_{k=1}^{n-1} b^{\text{eff}}_{k,k+1}\lambda^{\text{eff}}_k \sum_{i=1}^{n} e^{-\lambda^{\text{eff}}_i t} \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\left(\lambda^{\text{eff}}_j - \lambda^{\text{eff}}_i\right)} \tag{2.23} $$

This last equation will be called the modified Bateman's equation.

2.4 Decay and transmutation networks.

An important consequence of the branching ratios and the fission yields is related to the topology and structure of the decay chains. Until this moment it was considered the following structure of successive transformation:

$$ X_1 \xrightarrow{r_{1,2}} X_2 \xrightarrow{r_{2,3}} \ldots \xrightarrow{r_{n-1,n}} X_n \tag{2.24} $$

Which was called in Section 1.4 as a linear chain. Nevertheless, since isotopes can undergo several transmutations, it is more probable to find structure as the one that is showed in Figure 2.2, which are called transmutation and decay networks. The modified Bateman's equation can be used to solve these networks, but it is necessary to apply a procedure called linearization, which essentially search all the possible paths in a network, which will be the linear chains of it. Such search is equivalent to a procedure of *tree traversal* in data structures (Cormen et. al., 2003). Figure 2.3 shows the set of linear chains that are produced after a linearization process of the network described in Figure 2.2.

In Chapter 3 the linearization process will be discussed in detail, but for the moment it is necessary to consider that, at first, almost all the decay and transmutation networks can be reduced to a set of linear chains, which can be solved using the modified Bateman equation. Nevertheless, it is necessary to carry out a mass balance of these linear chains, and considering some issues related with the structure of the networks, and the definition of two types of fission yields. Such topics will be discussed in the following sections.

2.4.1 Mass balance of a Decay and Transmutation network.

Once a network is reduced to a set of linear chains, the total concentration of a given isotope can be obtained through the solution of each of these linear chains and using a mass balance analysis.

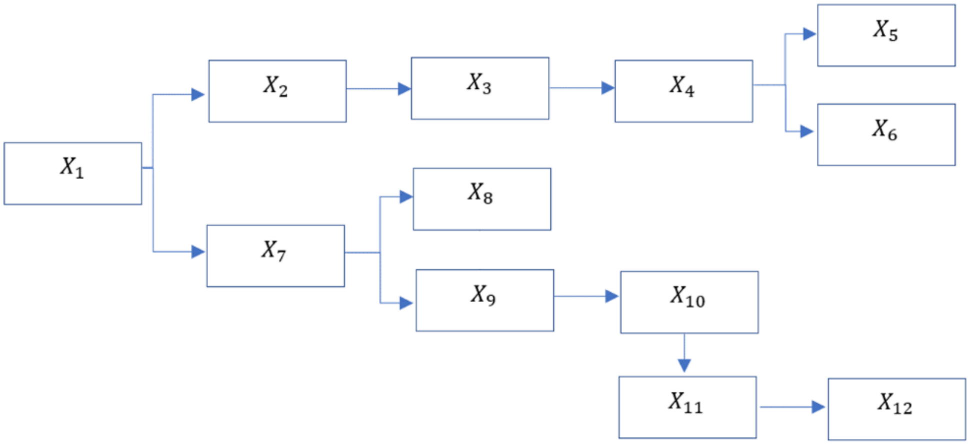


Figure 2. 2. An example of a transmutation and decay network.

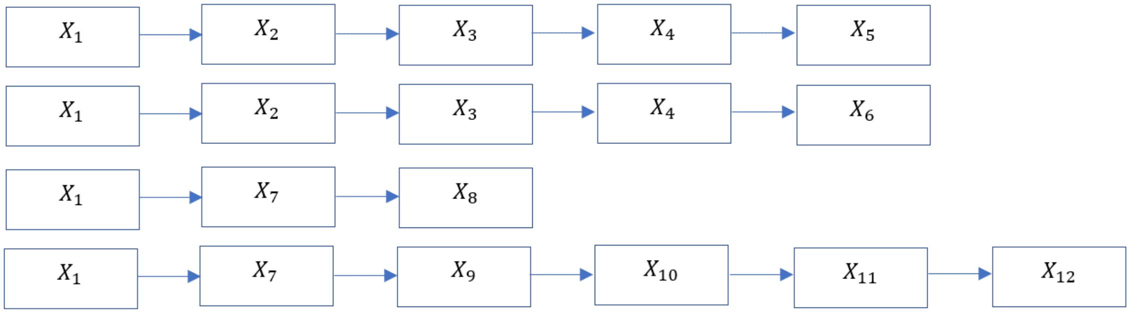


Figure 2. 3. Set of linear chains generated by a linearization process applied to the network of Figure 2.2.

It is possible to explain such mass balance analysis using the example given in Figure 2.2 and Figure 2.3, and considering that only the first isotope of the decay and transmutation network has an initial concentration different from zero. Firstly, the set of linear chains of Figure 2.3 can be expressed as the following matrix:

$$\begin{bmatrix} X_1 & X_2 & X_3 & X_4 & X_5 & \\ X_1 & X_2 & X_3 & X_4 & X_6 & \\ X_1 & X_7 & X_8 & & & \\ X_1 & X_7 & X_9 & X_{10} & X_{11} & X_{12} \end{bmatrix} \tag{2.25}$$

Analyzing this matrix from the left to the right and from the top to the bottom, a set of indexes will be assigned in order to determine which concentration will be computed for the total concentration of the isotopes. In first place, the index given by “1” will be assigned to the first time that an isotope is found when the matrix is analyzed, as it was described before. In (2.26) such assignation is showed.

The concentration of each isotope, whose index is equal to 1, will be considered for the final concentration, because it represents the elements of the original structure, before the linearization process was carried out.

$$\begin{bmatrix} 1 & 1 & 1 & 1 & 1 & \\ X_1 & X_2 & X_3 & X_4 & 1 & \\ X_1 & 1 & 1 & & & \\ X_1 & X_7 & 1 & 1 & 1 & 1 \end{bmatrix} \tag{2.26}$$

As can be noted, the second row of the matrix has only one "1" index, because the segment given by $X_1 \to X_2 \to X_3 \to X_4$, is repeated in the first and the second row. Therefore, it has already been considered for the final concentration, because these elements have an index "1", which was assigned in the first row. In other words, this is a segment that is repeated due to the linearization of the following segment of the network:

$$X_1 \to X_2 \to X_3 \to X_4 \to \left\{\begin{matrix} X_5 \\ X_6 \end{matrix}\right. \tag{2.27}$$

The index "2" will be assigned to the isotopes who appear more than one time, but whose previous elements are not the same or they are not in the same order. In (2.25) this case is not presented, but it can be exemplified if the following linear chain is considered to be part of the set given in Figure 2.3:

$$X_1 \to X_{13} \to X_{14} \to X_5$$

For this case, the isotope X_5 would appear for the second time. Nevertheless, the concentration for X_5 need to be considered, because this isotope was produced through a different succession of isotopes. In other words, this case can be represented by the following structure:

$$\left.\begin{matrix} X_1 \to X_2 \to X_3 \to X_4 \\ X_1 \to X_{13} \to X_{14} \end{matrix}\right\} \to X_5 \tag{2.28}$$

Finally, the index "3" will be assigned in the other cases. Using these rules, the final matrix of indexes will be equal to:

$$\begin{bmatrix} 1 & 1 & 1 & 1 & 1 & \\ 3 & 3 & 3 & 3 & 1 & \\ 3 & 1 & 1 & & & \\ 3 & 3 & 1 & 1 & 1 & 1 \end{bmatrix} \tag{2.29}$$

Only the elements who have an index of "1" or "2" need to be solved through the Bateman equation.

2.4.2 Mass balance as a function of segments.

The rules described in the above section can be generalized in a mathematical way using functions of segments. The function $L(X_i, k)$ can be defined as:

$$L(X_i, A) = \text{Sequence of isotopes before } X_i \text{ for a given linear chain } A \tag{2.30}$$

Clearly, each sequence that will be produced by $L(X_i, A)$ will be a segment of the original linear chain A. For example, if $A = X_1 \to X_2 \to \cdots \to X_{i-1} \to X_i \to X_{i+1} \to \cdots \to X_n$, then:

$$L(X_i, A) = X_1 \to X_2 \to \cdots \to X_{i-1}$$

Now, a second function χ will be defined as:

$$\chi(X_i, A) = \begin{cases} 3 & \text{if } L(X_i, A) \in \mathcal{K} \\ 2 & \text{if } L(X_i, A) \notin \mathcal{K} \end{cases} \tag{2.31}$$

Where the set $\mathcal{K}$ will be empty at the beginning of the analysis, but then it will store segments $L(X_i, A)$ based in the result of the function χ. Particularly, for a given isotope X_i in a linear chain A:

$$\begin{cases} \text{store } L(X_i, A) \text{ in } \mathcal{K} & \chi(X_i, A) = 2 \\ \qquad \text{nothing} & \chi(X_i, A) = 3 \end{cases} \tag{2.32}$$

Clearly, through (2.30)-(2.32) it is possible to assign the index "2" to the elements of a matrix.

2.4.3 Superposition process.

Until this moment it was considered that only the first isotope in a linear chain or in a decay and transmutation network has an initial concentration different from zero. Clearly this situation is very rare in practical burnup problems, and therefore it is necessary to consider the more general case.

It is possible to start from equation (1.13), and to ignore the initial conditions given in (1.14), considering the general case where all the isotopes have an initial concentration different from zero. In such case the resultant analogue matrix to (1.15) will be:

$$\begin{cases} \tilde{x}_1 = & \dfrac{X_1(0)}{s+\lambda_1} \\ \tilde{x}_2 = & \dfrac{\lambda_1 X_1(0)}{(s+\lambda_1)(s+\lambda_2)} + \dfrac{X_2(0)}{(s+\lambda_2)} \\ \tilde{x}_3 = & \dfrac{\lambda_2 \lambda_1 X_1(0)}{(s+\lambda_1)(s+\lambda_2)(s+\lambda_3)} + \dfrac{\lambda_2 X_2(0)}{(s+\lambda_2)(s+\lambda_3)} + \dfrac{X_3(0)}{(s+\lambda_3)} \\ \vdots & \vdots \\ \tilde{x}_n & \dfrac{\lambda_1 \lambda_2 \dots \lambda_{n-1} X_1(0)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)} + \dfrac{\lambda_2 \dots \lambda_{n-1} X_2(0)}{(s+\lambda_2)(s+\lambda_3)\dots(s+\lambda_n)} + \cdots + \dfrac{X_n(0)}{(s+\lambda_n)} \end{cases}$$

It is possible to note from the last system, that the equation for $\tilde{x}_n$ can be interpreted as the sum or superposition of the equations of the following linear chains:

$$\underbrace{\frac{\lambda_1 \lambda_2 \dots \lambda_{n-1} X_1(0)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)}}_{\substack{\text{Solution of:} \\ X_1 \to X_2 \to X_3 \to \cdots \to X_n}} + \underbrace{\frac{\lambda_2 \dots \lambda_{n-1} X_2(0)}{(s+\lambda_2)(s+\lambda_3)\dots(s+\lambda_n)}}_{\substack{\text{Solution of:} \\ X_2 \to X_3 \to X_4 \to \cdots \to X_n}} + \cdots + \underbrace{\frac{X_n(0)}{(s+\lambda_n)}}_{\substack{\text{Solution of} \\ X_n}}$$

Where in each case only the first isotope of the linear chains has an initial concentration different from zero. In other words, the general solution for X_n can be interpreted as the sum of the solutions of the following set of linear chains, with their respective initial conditions:

$$\begin{cases} \boxed{X_1 \to X_2 \to X_3 \to \cdots \to X_n} & X_1 = X_1(0), \quad X_i = 0, \;\; i \neq 1 \\ \boxed{X_2 \to X_3 \to \cdots \to X_n} & X_2 = X_2(0) \quad X_i = 0, \;\; i \neq 2 \\ \boxed{X_3 \to \cdots \to X_n} & X_3 = X_3(0) \quad X_i = 0, \;\; i \neq 3 \\ \vdots & \quad\vdots \qquad\qquad\quad \vdots \\ \boxed{X_n} & X_n = X_n(0) \quad X_i = 0, \;\; i \neq n \end{cases} \tag{2.33}$$

From (2.33) it is possible to note that the original linear chain is resized in each case. Therefore, instead of using a more general equation, it is possible to continue using the equation (2.23) and to resize the linear chain following the scheme given in (2.33). This procedure is known as superposition (Isotalo, 2013), and the final concentration of the isotopes is the sum of the concentration found in all the superposition scenarios.

It is worth mentioning that certain codes, like Helios (Studsvik, 2008), do not use the superposition process, and instead they use a more general formulation of the system (1.15). Nevertheless, in the Szilard code, the author of the present thesis uses the methodology described in this section, because the development of such code was based, mainly, in the Isotalo's description of the linear chain method. Finally, in Chapter 4 a similar superposition process will be developed for a special structure known as cyclic chains, which is based on the symmetry functions described in the past chapter.

2.5 A detailed study of the yields.

At first glance it seems that the definition of the yields given in Section 2.3.1 is sufficient to include such parameters in the Bateman equation, but nothing is further from reality. Firstly, there are two types of fission yields in the ENDF/B-VII.1 library, and it is necessary to determine which of these will be used in a burnup problem. Secondly, the type of yield that is used strongly depends on the isobaric structures of the decay and transmutation network. Finally, the assignation of a given type of yield is related with the capture reactions that are included in a given structure of isotopes. All these topics will be discussed in the following sections.

2.5.1. A brief analysis of the fission yields' graphs.

In order to understand some of the properties of the yields, it is necessary to analyze their graphs and the symmetry contained in them. As a first example, the yield distribution of the fission products issued in the fission of the ^{235}U due to an incident neutron of an energy of 0.0253 eV is showed in Figure 2.4. Most of the graphs of the fission yields have a two camel humps shape, as can be observed in that figure.

This shape is the result of the lack of symmetry in terms of the mass number of the fission products. In other words, when a fission occurs it is more probable that the two fission products have different mass number A, which implies that the original nuclei does not split in two exact half parts. On the other hand, the peaks or maximum values in the Figure 2.4 are related with the nuclear magic numbers, which represented mass numbers of nuclei whose nucleons are arranged into complete shells, and therefore they have greater stability.

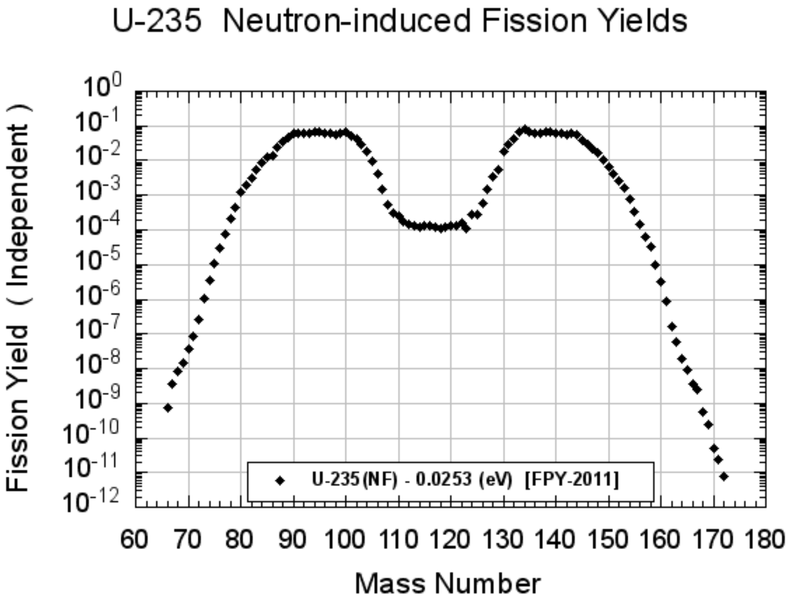


Figure 2. 4. Fission yield as a function of the mass number for ^{235}U isotope, for an incident neutron energy of 0.0253 eV. The image was obtained from the web site wwwndc.jaea.go.jp.

According to this theory, the mass numbers given by 2, 8, 20, 28, 50, 82 y 126 represented stabled nucleons configuration, and it is more probable to find fission reactions close to these numbers. A similar example of the symmetry can be found in the graph of the isotope ^{255}Fm, which is showed in Figure 2.5. In such image it is possible to note that the two camel humps shape almost has disappeared, and the fission products with a major probability are quasi-symmetric in terms of their mass numbers. In other words, the symmetry is greater.

It is possible to define the fission distribution yield function as $Y(X, E, A)$, where X is related to the heavy isotope that undergoes fission, E is the energy of the incident neutron, and A is the mass number of the fission product. Therefore, the symmetry property can be expressed as, for a given mass number A_i, there is another mass number A_j for which

$$Y(X, E, A_1) = \ Y(X, E, A_2) \tag{2.34}$$

In other words, the distribution function Y is not injective. Another interesting fact related with the symmetry is the following distribution sum:

$$\sum_{A=60}^{180} Y(X, E, A) \tag{2.35}$$

Whose result is equal to 2. This can be strange at first, because it seems that the sum must be equal to 1, since the fission yields are fractions. Nevertheless, it is necessary to remember that in a fission process two isotopes are produced, and the mass numbers of such products will have the same fission yield. In other words, two mass values A_i and A_j are characterized through a same value of yield. Therefore, when the summa is carried out, the yield value is multiplied by two.

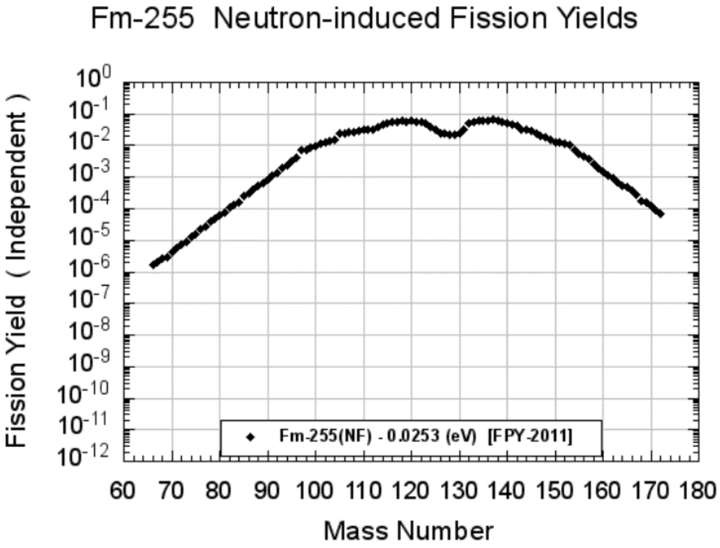


Figure 2. 5. Fission yield as a function of the mass number for the ^{255}Fm isotope, for an incident neutron energy of 0.0253 eV. The image was obtained from the web site wwwndc.jaea.go.jp.

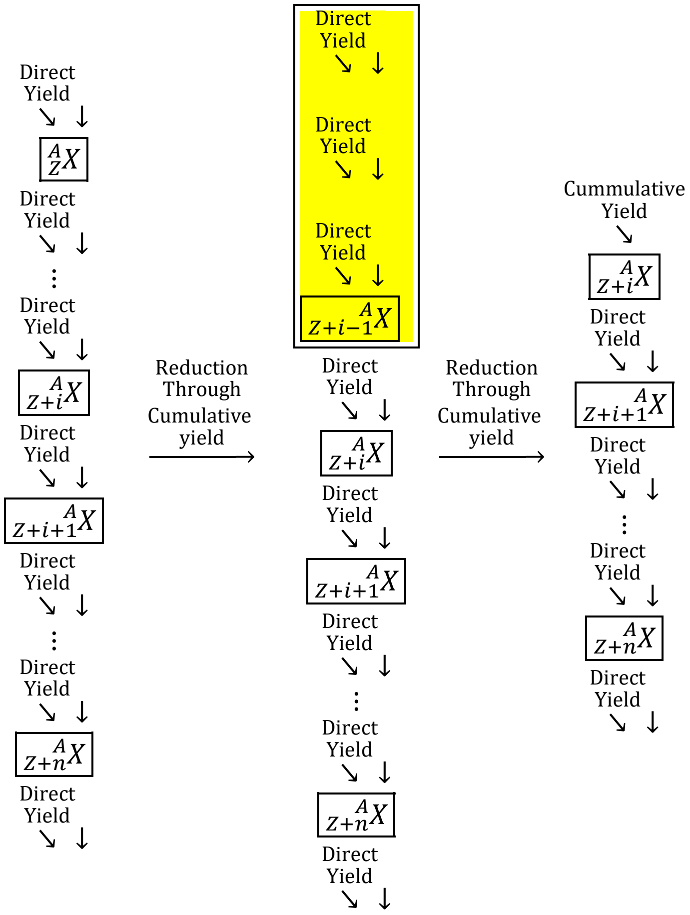
2.5.2 Cumulative and independent yield.

As it is known, fission products usually have a higher number of neutrons than protons, and therefore they are unstable, undergoing a β^- decay. This process is not instantaneous, and it takes a given time that can be related to the half-life of the fission product. Due to this behavior, it is possible to find isobaric structures in fission products networks, which have the following form:

$$\begin{array}{c} \searrow\downarrow \\ \boxed{{}^{A}_{Z}X} \\ \searrow\downarrow \\ \boxed{{}^{A}_{Z+1}X} \\ \searrow\downarrow \\ \vdots \\ \searrow\downarrow \\ \boxed{{}^{A}_{Z+n}X} \\ \searrow\downarrow \end{array} \tag{2.36}$$

In (2.36), the diagonal arrows indicate that the isotopes are produced by fission and the vertical arrows represent the β^- decay. As was mentioned before, these structures are not generated immediately after the fission process, because it is necessary that a given time t elapses, in order that the beta decay undergoes. Clearly, all the isotopes ${}^{A}_{Z}X$, ${}_{Z+1}^{\;\;A}X$,..., ${}_{Z+n}^{\;\;A}X$ are produced as result of the fission process. One of them have their origin in the fission itself in a direct way, but they also have contributions due to the beta decay from other isotopes that were generated also by fission. Therefore, if someone measure the number of isotopes that appear after a time t after the fission process, then it will be necessary to consider all the elements that belong to (2.36).

This is the idea behind the definitions of the direct fission yield, and the cumulative fission yield. The first kind is related to the amount of the fission products that appear directly by the fission process. The other kind involves both contribution: one due to the fission and another one due to the beta decay of the isobaric structures. The main advantage of these definitions is related to the reduction of the decay and transmutation networks. For example, for an isotope ${}_{Z+i}^{A}X$ that belongs to the structure (2.36), it is possible to make the following reduction:



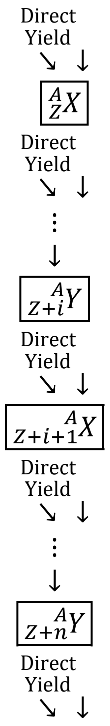
(2.37)

In (2.37) the isobaric chain given by the sequence of isotopes ${}_{Z}^{A}X, {}_{Z+1}^{A}X, \dots, {}_{Z+i-1}^{A}X$ was compacted or reduced, through a cumulative yield, which represented all the process and contributions to the isotope ${}_{Z+i}^{A}X$. Since not all the isotopes are important in several burnup problems, the cumulative yield allows reducing the isobaric chains, and therefore it is very useful to decrease the involved time in the solution of the Bateman equation.

Strictly speaking the cumulative yield is a function of time, because the production of a given isotope in an isobaric chain strongly depends on the time that has passed after the fission process, therefore it is necessary a more detailed study of this parameter, which will be carried out in the following section. It is possible to obtain some conclusions from the above discussion. Firstly:

$$\gamma_{\text{cumulative}} \geq \gamma_{\text{direct}}$$

On the other hand, there are some isotopes whose direct yield is equal to zero, while the cumulative yield is not. This means that such isotope is not directly produced by fission, but belongs to an isobaric chain that has its origin by the fission process. For example, in the following scheme, the isotopes denoted by ${}_{Z+i}^{A}Y$ and ${}_{Z+n}^{A}Y$ will have a direct yield equal to zero, but a cumulative value different from it:



(2.38)

In other words, these isotopes do not have a diagonal arrow in (2.38). An important question that can be answered from the above discussion is: In which case the cumulative and the direct yields have the same value? It is possible to solve this question through the following equation:

$$\gamma_{\text{cumulative}}(t) = \gamma_{\text{direct}} + C(t) \tag{2.39}$$

Where $C(t)$ is a contribution function that represents the yellow block in the scheme showed in (2.37). Clearly $\gamma_{\text{cumulative}}(t) = \gamma_{\text{direct}}$ only when $C(t) = 0$. Particularly, this happens for isotopes that do not have ancestors through beta decay, i.e. isotopes whose yellow block does not have elements. In Table 2.4 there are listed eight fission products of the ^{235}U, for an energy of 0.0253 eV, whose cumulative yield is equal to the direct yield.

2.5.2 Formal definitions.

The definitions provided by Mills for the direct (also called independent) yield is (Mills, 1995):

> "...the number of atoms of an isotope ${}_{Z}^{A}X^{*}$ produced directly from one fission, but after the emission of prompt neutrons, but before any radioactive decay and hence the emission of delayed neutrons".

From the definition of cumulative yield, it is possible to define:

Table 2. 4. Some examples of isotopes for which the cumulative yield is very close to the indirect yield. The percentual error was computed considering as base the direct yield.

Isotope	Direct Yield	Cumulative Yield	Percentual Error
Sb-135	$1.45527\text{x}10^{-3}$	$1.46028\text{x}10^{-3}$	0.343084888
In-131	$6.49993\text{x}10^{-5}$	$6.54135\text{x}10^{-5}$	0.633202626
In-133	$1.71443\text{x}10^{-6}$	$1.71446\text{x}10^{-6}$	0.001749822
Te-136	$1.36706\text{x}10^{-2}$	$1.32087\text{x}10^{-2}$	3.496937624
Sn-134	$1.77403\text{x}10^{-4}$	1.774150E-04	0.006763802
Cu-79	$1.31738\text{x}10^{-8}$	1.317380E-08	0
Zn-77	$2.36950\text{x}10^{-5}$	2.525490E-05	6.17662315
Kr-95	$7.20629\text{x}10^{-5}$	7.208490E-05	0.030519568

The cumulative yield is the total number of atoms of that an isotope produces over all time after one fission. There are three cases that depends on the value of the half-life of the isotopes:

Case 1: “If the isotope is stable, the cumulative yield is the total number of atoms of that isotope remaining per fission after all precursor decays (ignoring the effects of other nuclear reactions e.g. neutron capture).”

Case 2: “Similarly, for an isotope with a much longer half-life than any of its precursors, the cumulative yield is nearly equal to the amount of it produced at a time short compared to its half-life, but long compared to those of its precursors.”

Case 3: “For the rest of the isotopes, some atoms will have decayed before all have been produced, so that at no time there will actually be a cumulative yield for atoms per fission present.”

For the first case, the cumulative yield can be found as:

$$\gamma = \lim_{t\to\infty} \gamma_{\text{cumulative}}(t) = \gamma_{\text{direct}} + \lim_{t\to\infty} C(t) \qquad (2.40)$$

For practical cases, the condition $t \to \infty$ can be interpreted as a time t, from which all the precursors have been decayed. For the second case, the cumulative yield can be analyzed in a similar way in which a transient equilibrium problem is solved. In such case an isotope with a longer half-life T_n is produced, with $T_n > T_i, 1 \leq i < n$, and where $T_1, T_2, \dots, T_{n-1}$ are the half-lives of the precursors of T_n. In order to use this cumulative yield, it is necessary to consider a time t, which must to fulfill:

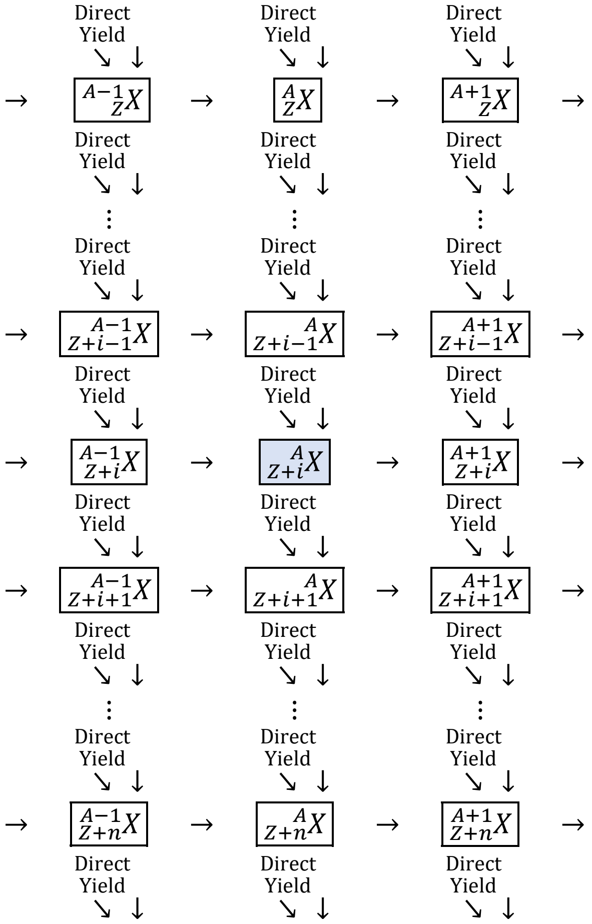
$$T_i \ll t \ll T_n \qquad (2.41)$$

Clearly this second case is similar, but not equal to the first case. Since $t \ll T_1$, it is possible to consider as a good approximation that for a given time interval the isotope X_n remains stable. Nevertheless, due to the first part of the inequality, $T_i \ll t$, it is possible to consider that most of the precursors have been decayed. Clearly the use of cumulative yield depends on the analysis of (2.40) and (2.41). Finally, the third part of the definition provided by Mills suggests that if neither (2.40) and (2.41) are valid, then it is not possible to use the cumulative yield.

From the above discussion it is possible to conclude that the assignation of the yield's type is a very hard task, because it requires a study of the isobaric chains as well as an analysis of the half-lives of the isotopes that belong to them. It is possible to summarize it using the words of Knott and Yamamoto: "… the estimation of the fission yields … is a very complicated task". (Cacuci, 2010).

2.5.4 The importance of the neutron captures and a practical solution.

Actually, the structure given in (2.36) is not complete, because it only considers beta decay and fission process. Therefore, it is also necessary to include the reaction captures, with which the final network will be:



(2.42)

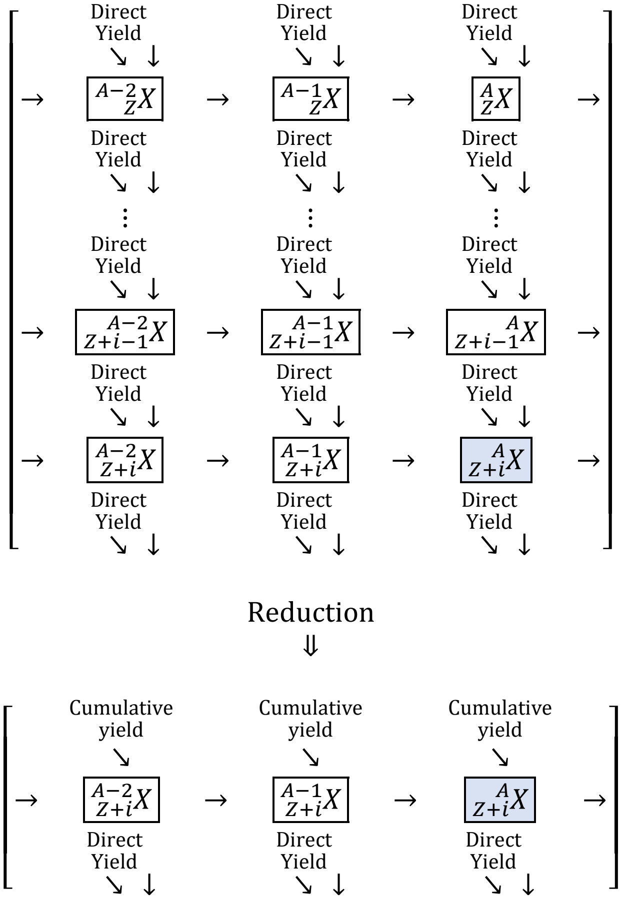
According to the definitions provided in Section 2.5.3, the cumulative yield does not consider neutron captures, and therefore it cannot be used to reduce (2.52) as in the scheme (2.37). This difficulty can be explained through concentration of the isotope $^{A}_{Z+i}X$. Firstly, the cumulative fission yield can simulate the direct fission yield and the contribution due to the precursors or ancestors of it.

Nevertheless, since the cumulative yield does not consider capture reactions, the concentration of the precursors of $^{A}_{Z+i}X$ are underestimated. For example, for the precursor $^{A}_{Z-i}X$, there are several contributions that are not considered, among which is the contribution due to the following linear chains:

$$\ldots \rightarrow \boxed{{}^{A-2}_{Z+i-1}X} \rightarrow \boxed{{}^{A-1}_{Z+i-1}X} \rightarrow \boxed{{}^{A}_{Z+i-1}X}$$

$$\ldots \rightarrow \boxed{{}^{A-1}_{Z+i-2}X} \rightarrow \boxed{{}^{A-1}_{Z+i-1}X} \rightarrow \boxed{{}^{A}_{Z+i-1}X}$$

There is a similar case for each of the precursors. Since the cumulative yield simulates the contribution to a certain isotope due to its precursors, it is clear that if the concentration of such precursors is underestimated, then the final concentration of the isotope will be lower than its actual value. Then, the cumulative yield cannot be used to reduce the complete structure. For some cases, where the capture reactions of the precursors are not very significative it is possible to use the following approximation for the concentration of ${}^{A}_{Z+i}X$:



(2.43)

In this last structure the precursors of the isotopes ${}^{A-2}_{Z+i}X$, ${}^{A-1}_{Z+i}X$ and ${}^{A}_{Z+i}X$ were simplified through the cumulative yields, and the only capture reactions that are included are those who belong to the following linear chain:

$$\ldots \rightarrow \boxed{{}^{A-2}_{Z+i}X} \rightarrow \boxed{{}^{A-1}_{Z+i}X} \rightarrow \boxed{{}^{A}_{Z+i}X}$$

This is a better approximation instead of using only a cumulative yield for ${}_{Z+i}^{A}X$, however all the relationships denoted by the horizontal arrows in the upper part of (2.43) are ignored, which can lead to incorrect outcomes for decay and transmutation networks where the reaction capture are significative. As it can be observed, the problem of the yield assignation becomes more complex when capture reactions are included.

It is possible that the most practical solution consists of using only direct or independent yield, and avoiding the cumulative one. This implies that it is necessary to use the full decay and transmutation networks and not to use simplifications. Even when this procedure is more demanding in terms of computational storage and execution time, it solves the problem in a straightforward way.

Additionally, modern codes, including SERPENT (Leppänen et al., 2015), follow this procedure, and nowadays it is common to find that some of them include more than 1000 fission products in their analysis.

2.5.5 Extraction of the Fission Yields data from ENDF/B-VII.1

A set of fission and cumulative yields from the WIMS project was used in an early stage of the development of the code Szilard (Leszczynski et. al., 2007). Such set corresponds to the decay and transmutation network that is used in the package of software that belong to that project. Nevertheless, once the difficulties discussed before were identified, it was considered more convenient to use the data directly from ENDF/B-VII.1 library.

As in the case of the branching ratios, the data related to the fission products yield is contained in a folder whose name is “nfy” (neutron fission yields). Inside such folder there are 31 data files, whose name has the following structure:

$$\underbrace{\text{nfy}}_{\boxed{1}}\ \underbrace{\text{-}}_{\boxed{2}}\ \underbrace{092}_{\boxed{3}}\ \underbrace{\_}_{\boxed{4}}\underbrace{\text{U}}_{\boxed{5}}\underbrace{\_}_{\boxed{6}}\ \underbrace{235}_{\boxed{7}}\text{.endf} \tag{2.44}$$

Except for the element denoted by 1, that represents the header of the neutron fission yields, all the other elements have the same meaning that in (2.7). In Table 2.5 the isotopes that are contained in the folder are listed. The information of each file has the structure showed in Figure 2.6. Each line has 72 characters (including blank spaces) and there are four digits related to the type of yield according to the following rule:

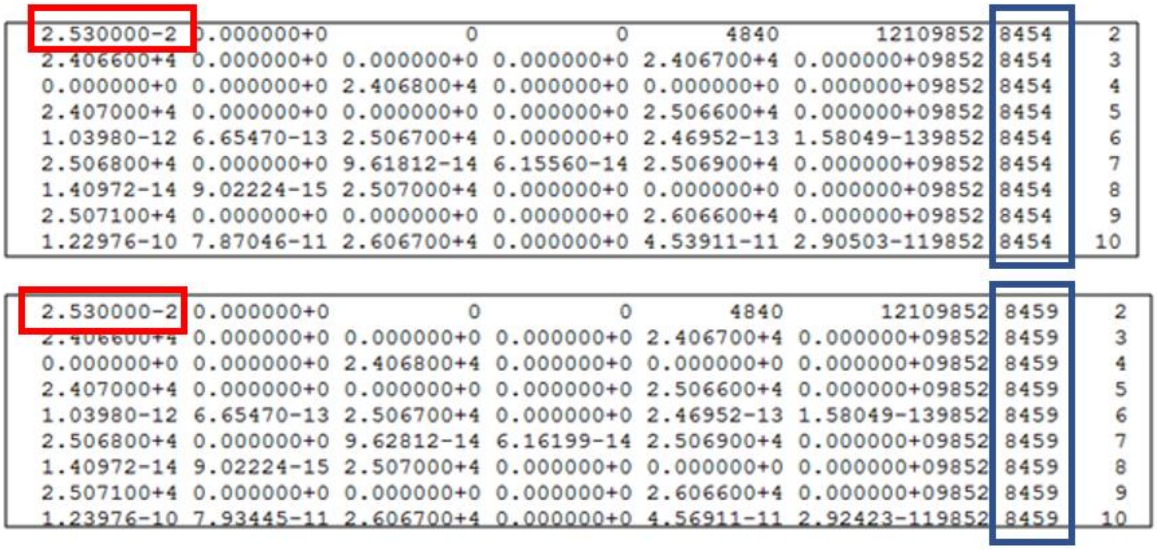
$$\begin{array}{rc} 8454 \rightarrow & \text{direct yield} \\ 8559 \rightarrow & \text{cumulative yield} \end{array} \tag{2.45}$$

Such digits are near to the end of the line. Therefore, the information of a file can be divided in two parts in accordance to these numbers. Each of this part, in turn, is divided in 4 segments that are related to the energy spectrum, inside of them there are energy identifiers, through which it is possible to cluster the information. In Table 2.6 the energy identifiers are listed, and one of them is showed in Figure 2.6.

As in the case of the branching ratios, there is a lack of uniformity in the format for the file related to ^{251}Cf. In this case, instead of using the identifier 2.530000-2 for the energy of $2.53x10^{-2}$ eV as in the rest of the isotopes, the identifier 2.520000-2 has been used.

Table 2. 5. Isotopes whose information related to the fission product yields is contained in the ENDF/B-VII.1 library.

^{227}Th	^{236}U	^{241}Pu	^{245}Cm
^{229}Th	^{237}U	^{242}Pu	^{246}Cm
^{232}Th	^{238}U	^{241}Am	^{248}Cm
^{231}Pa	^{237}Np	^{242}Am*	^{249}Cf
^{232}U	^{238}Np	^{243}Am	^{251}Cf
^{233}U	^{238}Pu	^{242}Cm	^{254}Es
^{234}U	^{239}Pu	^{243}Cm	^{255}Fm
^{235}U	^{240}Pu	^{244}Cm	



```
 2.530000-2 0.000000+0          0          0       4840   12109852 8454    2
 2.406600+4 0.000000+0 0.000000+0 0.000000+0 2.406700+4 0.000000+09852 8454    3
 0.000000+0 0.000000+0 2.406800+4 0.000000+0 0.000000+0 0.000000+09852 8454    4
 2.407000+4 0.000000+0 0.000000+0 0.000000+0 2.506600+4 0.000000+09852 8454    5
 1.03980-12 6.65470-13 2.506700+4 0.000000+0 2.46952-13 1.58049-139852 8454    6
 2.506800+4 0.000000+0 9.61812-14 6.15560-14 2.506900+4 0.000000+09852 8454    7
 1.40972-14 9.02224-15 2.507000+4 0.000000+0 0.000000+0 0.000000+09852 8454    8
 2.507100+4 0.000000+0 0.000000+0 0.000000+0 2.606600+4 0.000000+09852 8454    9
 1.22976-10 7.87046-11 2.606700+4 0.000000+0 4.53911-11 2.90503-119852 8454   10

 2.530000-2 0.000000+0          0          0       4840   12109852 8459    2
 2.406600+4 0.000000+0 0.000000+0 0.000000+0 2.406700+4 0.000000+09852 8459    3
 0.000000+0 0.000000+0 2.406800+4 0.000000+0 0.000000+0 0.000000+09852 8459    4
 2.407000+4 0.000000+0 0.000000+0 0.000000+0 2.506600+4 0.000000+09852 8459    5
 1.03980-12 6.65470-13 2.506700+4 0.000000+0 2.46952-13 1.58049-139852 8459    6
 2.506800+4 0.000000+0 9.62812-14 6.16199-14 2.506900+4 0.000000+09852 8459    7
 1.40972-14 9.02224-15 2.507000+4 0.000000+0 0.000000+0 0.000000+09852 8459    8
 2.507100+4 0.000000+0 0.000000+0 0.000000+0 2.606600+4 0.000000+09852 8459    9
 1.23976-10 7.93445-11 2.606700+4 0.000000+0 4.56911-11 2.92423-119852 8459   10
```

Figure 2. 6. Structure of the information for the fission product yields contained in the ENDF/B-VII.1 library. In the purple rectangle the identifiers for the type of yield are showed: 8454 for the direct type and 8459 for the cumulative one. In the red rectangle is showed the identifier for the energy of 2.53x10^{-2} eV.

Table 2. 6. Identifiers for the energy that are used in the ENDF/B-VII.1 library.

Energy	Identifier
2.53x10^{-2} eV	2.530000-2
5x10^{5} eV = 50 keV	5.000000+5
1.4x10^{7} eV = 14 MeV	1.400000+7

The identifiers for the fission products are given as a float number of 7 digits, which is followed by an exponent equal to 4:

$$a_{z_1}.\,a_{z_2}b_{A_1}b_{A_2}b_{A_3}00+4 \tag{2.46}$$

The first two digits, a_{z_1} and a_{z_2} are related to the atomic number, while the digits b_{A_1}, b_{A_2} and b_{A_3} denote the mass number. Afterward, two zeros are added and an exponent +4 is at the end. There are three other data after the fission product identifier, which are separated by a blank space: the excited state, the value of the yield and finally its uncertainty. As an example, in Figure 2.7 is showed the yield information of the ^{133}Xe* and the ^{135}Xe fission products of ^{235}U. The information related to these isotopes is the following:

$$\begin{array}{cccc} \multicolumn{2}{c}{^{133}\mathrm{Xe}^{*}} & \multicolumn{2}{c}{^{135}\mathrm{Xe}} \\ 5.413300+4 & \text{ZAI identifier} & 5.413500+4 & \text{ZAI identifier} \\ 1.\ 000000+4 & \text{Excited state} & 0.\ 000000+4 & \text{Excited state} \\ 1.885870-5 & \text{Yield's value} & 7.851250-4 & \text{Yield's value} \\ 1.206950-5 & \text{Uncertainy} & 4.710750-5 & \text{Uncertainy} \end{array} \tag{2.47}$$

The extraction of these data requires to find the fission product identifier, and then search the following three values, with the possibility that this information can be contained in two different and consecutives lines. Finally, in order to use the cumulative yield for fission products with metastable states, it is necessary to carry out the summa of them. In other words:

$$\gamma_{\text{cumulative}}\left({}^{A}_{Z}X\right)=\sum_{i}\gamma_{\text{cumulative}}\left({}^{A}_{Z}X^{i}\right) \tag{2.48}$$

Where i is the metastable state of the fission product.

2.6 A numerical approach to the fission product yields.

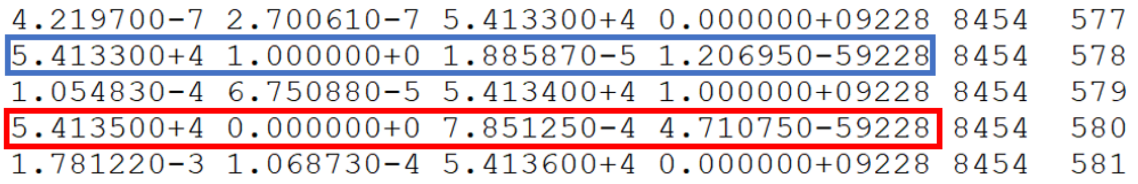
Based on the analysis of the last section, it is possible to summarize the yield assignation as follows: if an isotope is at the beginning of an isobaric chain (i.e. at the beginning of a column of Figure 2.36), then it will have a cumulative yield. Also, all the other elements in an isobaric chain will have an independent yield.

There are some important findings when the cumulative yield is interpreted as a factor that simulates the contributions related to the elements that were not considered. For example, we can suppose that in a complete network we have the following isobaric chain (which in this example will be draw in a horizontal form):

$$^{\searrow}\boxed{{}^{A}_{Z}X_1}\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+1}X_2}\rightarrow\cdots\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+i-1}X_i}\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+i}X_{i+1}}\rightarrow\cdots\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+n-1}X_n} \tag{2.49}$$

If only the isotopes that appear after X_i are considered, it is possible to reduce the linear chain as follows:

$$^{\searrow}\boxed{{}^{A}_{Z+i-1}X_i}\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+i}X_{i+1}}\rightarrow\cdots\rightarrow{}^{\searrow}\boxed{{}^{A}_{Z+n-1}X_n} \tag{2.50}$$



```
4.219700-7 2.700610-7 5.413300+4 0.000000+09228 8454  577
5.413300+4 1.000000+0 1.885870-5 1.206950-59228 8454  578
1.054830-4 6.750880-5 5.413400+4 1.000000+09228 8454  579
5.413500+4 0.000000+0 7.851250-4 4.710750-59228 8454  580
1.781220-3 1.068730-4 5.413600+4 0.000000+09228 8454  581
```

Figure 2. 7. Figure 2.7. Fission yield information of ^{235}U, for the ^{133}Xe* and the ^{135}Xe fission products. The information related to ^{133}Xe* are enclosed in the blue rectangle, and the corresponding to ^{135}Xe in the red one.

2.6.1 Solution to the complete scheme.

It is possible to simulate, in a numerical way, the scheme given in (2.49), in order to compare it with the scheme given in (2.50). Essentially, it is necessary to find the solution of both systems and compare them. After a process of linearization of the scheme given in (2.49), it is possible to find the following linear chains:

$$\begin{gathered}
\boxed{\begin{array}{c}\text{Heavy}\\\text{isotope}\end{array}} \xrightarrow{\text{fission}} X_1 \xrightarrow{\beta^-} X_2 \xrightarrow{\beta^-} \dots \xrightarrow{\beta^-} X_{i-1} \xrightarrow{\beta^-} X_i \\
\boxed{\begin{array}{c}\text{Heavy}\\\text{isotope}\end{array}} \xrightarrow{\text{fission}} X_2 \xrightarrow{\beta^-} X_3 \xrightarrow{\beta^-} \dots \xrightarrow{\beta^-} X_{i-1} \xrightarrow{\beta^-} X_{i-1} \\
\vdots \\
\boxed{\begin{array}{c}\text{Heavy}\\\text{isotope}\end{array}} \xrightarrow{\text{fission}} X_{i-1} \xrightarrow{\beta^-} X_i
\end{gathered} \qquad (2.51)$$

The heavy isotope will be denoted by H with an initial concentration equal to $H(0)$. Since the contribution due to the fission process is investigated, it will be assumed that all the other elements in (2.51) have an initial concentration equal to zero. Using the modified Bateman equation, it is possible to compute the contribution to X_i, for a given time t, due to the first linear chain:

$$X_i(t,X_1) = H(0) b^{\text{eff}}_{H,X_1} \lambda^{\text{eff}}_H \prod_{d=1}^{i-1} b^{\text{eff}}_{d,d+1} \lambda^{\text{eff}}_d \left[\sum_{k=0}^{i} e^{-\lambda_i^{\text{eff}} t} \prod_{\substack{j=0\\ j\neq i}}^{n} \frac{1}{\lambda_j^{\text{eff}} - \lambda_k^{\text{eff}}} \right]$$

With $\lambda_H^{\text{eff}} = \lambda_0^{\text{eff}}$ and where $b^{\text{eff}}_{d,d+1}$ denotes the fraction of atoms of the isotope X_{d+1} that are generated by the transformation of the atoms of the isotope X_d. Similarly, the λ_d^{eff} is the effective removal coefficient of X_d. For the general case, with $1 \le p \le i$:

$$X_i(t,X_p) = H(0) b^{\text{eff}}_{H,X_p} \lambda^{\text{eff}}_H \prod_{d=p}^{i-1} b^{\text{eff}}_{d,d+1} \lambda^{\text{eff}}_d \left[\sum_{k=1}^{i} e^{-\lambda_i^{\text{eff}} t} \prod_{\substack{j=1\\ j\neq i}}^{n} \frac{1}{\lambda_j^{\text{eff}} - \lambda_k^{\text{eff}}} \right] \qquad (2.52)$$

Finally, the concentration of $X_i(t)$ can be found through the following summa:

$$X_i(t) = \sum_{p=1}^{p=i} X_i(t, X_p) \tag{2.53}$$

It is convenient to express (2.53) as follows:

$$\begin{aligned} X_i(t) &= \sum_{p=1}^{p=i-1} X_i(t, X_p) + X_i(t, X_i) \\ &= \sum_{p=1}^{p=i-1} X_i(t, X_p) + H(0) b_{H,X_i}^{\text{eff}} \lambda_H^{\text{eff}} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right] \end{aligned} \tag{2.54}$$

The last equation will be called the solution to the complete scheme.

2.6.2 Comparison between the scheme solutions.

Through the cumulative yield, the solution to the reduced scheme is given by:

$$X_{i,\text{cumulative}}(t) = H(0) b_{H,X_i,\text{cumulative}}^{\text{eff}} \lambda_H^{\text{eff}} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right] \tag{2.55}$$

Assuming that the concentration given in (2.54) and (2.55) needs to be equal, it is possible to set:

$$\sum_{p=1}^{p=i-1} X_i(t, X_p) + H(0) b_{H,X_i}^{\text{eff}} \lambda_H^{\text{eff}} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right] = H(0) b_{H,X_i,\text{cumulative}}^{\text{eff}} \lambda_H^{\text{eff}} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right]$$

From which it is possible to write:

$$b_{H,X_i,\text{cumulative}}^{\text{eff}} = \frac{\sum_{p=1}^{p=i-1} X_i(t, X_p)}{H(0) \lambda_H^{\text{eff}} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right]} + b_{H,X_i}^{\text{eff}} \tag{2.56}$$

Using the definition given in (2.18), considering one group of energy:

$$b_{H,X_i,\text{cumulative}}^{\text{eff}} = \frac{\gamma_{\text{cumulative}} \phi \sigma_{f,H}}{\lambda_H^{\text{eff}}}, \qquad b_{H,i}^{\text{eff}} = \frac{\gamma_{\text{direct}} \phi \sigma_{f,H}}{\lambda_H^{\text{eff}}}$$

Replacing this in (2.56) it follows that:

$$\gamma_{\text{cumulative}} = \frac{\sum_{p=1}^{p=i-1} X_i(t, X_p)}{H(0) \phi \sigma_{f,H} \left[\frac{e^{-\lambda_X^{\text{eff}} t} - e^{-\lambda_i^{\text{eff}} t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}} \right]} + \gamma_{\text{direct}} \tag{2.57}$$

This last equation is very useful because it allows finding a relationship between the cumulative and the direct yield. Additionally, it represents a contribution of the present thesis, because at least to the best knowledge of the author, there is a lack of information about the numerical analysis of the yields and the relationship between them in terms of the time evolution.

2.6.3 Some applications of the equation (2.57).

Through equation (2.57) it is possible to express who is the function $C(t)$, which was described in equation (2.39):

$$C(t) = \frac{\sum_{p=1}^{p=i-1} X_i(t, X_p)}{H(0)\phi\sigma_{f,H}\left[\frac{e^{-\lambda_X^{\text{eff}}t} - e^{-\lambda_i^{\text{eff}}t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}}\right]} \tag{2.58}$$

This equation allows answering where the cumulative yield can be applied. Firstly, as it was mentioned before, the values of the cumulative yields that are contained in the ENDF/B-VII.1 library are constant in time. It is possible to explain this situation through the exponential functions inside the term $\sum_{p=1}^{p=i-1} X_i(t, X_p)$. Clearly the experimental cumulative yields assume that:

$$\frac{\sum_{p=1}^{p=i-1} X_i(t, X_p)}{\left[\frac{e^{-\lambda_X^{\text{eff}}t} - e^{-\lambda_i^{\text{eff}}t}}{\lambda_i^{\text{eff}} - \lambda_H^{\text{eff}}}\right]} \approx \text{constant} \tag{2.59}$$

The condition (2.58) is very useful to determine when a cumulative yield can be used, because through a graphical analysis it will be possible to determine, for different energies, an interval of time where the cumulative yield fulfilled it. The study of the mathematical behavior of (2.59) can have impact, because even when the modern and future burnup codes will not use cumulative yields, it is possible that such data continue being useful to other applications.

In fact, the cumulative yields are not used exclusively in burnup problems. They have a crucial role in heat fission calculations, in waste management, in estimation of gamma and delayed neutron emission, and even with nuclear tests (Britt et al., 2010), (Privas et. al., 2016). On the other hand, the paradigm's change about the use of only independent yield is not overnight, and it is possible that some codes (not necessary burnup codes) will continue to use cumulative yields. Therefore, a good propose to a future work consist on the detailed study of such equation.

2.6.4 Topology of the decay and transmutation network.

There is an important question related to the yields and the fission products: given a set of isotopes, does exist a minimal decay and transmutation network that contain them, and whose error's concentration (computed from it) are below certain value ϵ?

In other words, if there is a special interest in a set of isotopes, it could be possible to build a minimal decay and transmutation network who contain it (with the possibility of containing other more) and whose concentration value will be below to a given value ϵ? The importance behind this question is related to the possibility to compare two different burnup codes, as well as to the possibility to optimize the execution time involved in the solution of the Bateman equation. If the answer is positive, the following question will be: is this network unique?

In order to answer this question, it is necessary to study the structure of the decay and transmutation network, as well as to carry out a study of all the number of linear chains that contributes to the concentration of a given element. This last task can be made through combinatorics and analysis of all the different paths that lead to the isotope whose concentration is searching. In order to show that procedure, the following structure will be considered:

$$\begin{tikzcd}
{\searrow}\boxed{{}^{A}_{Z}X_i} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+1}_{Z}X_i} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+2}_{Z}X_i} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+3}_{Z}X_i} \arrow[r] \arrow[d] & {} \\
{\searrow}\boxed{{}^{A}_{Z+1}X_j} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+1}_{Z+1}X_j} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+2}_{Z+1}X_j} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+3}_{Z+1}X_j} \arrow[r] \arrow[d] & {} \\
{\searrow}\boxed{{}^{A}_{Z+2}X_k} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+1}_{Z+2}X_k} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+2}_{Z+2}X_k} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+3}_{Z+2}X_k} \arrow[r] \arrow[d] & {} \\
{\searrow}\boxed{{}^{A}_{Z+3}X_l} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+1}_{Z+3}X_l} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+2}_{Z+3}X_l} \arrow[r] \arrow[d] & {\searrow}\boxed{{}^{A+3}_{Z+3}X_l} \arrow[r] \arrow[d] & {} \\
{} & {} & {} & {} &
\end{tikzcd} \tag{2.60}$$

As an example, the number of all the linear chains who contribute to ${}^{A+3}_{Z+3}X_l$ will be computed. In order words, the isotope ${}^{A+3}_{Z+3}X_l$ will be the target, or the final isotope in the paths. In practical burnup problems, it is more probable to find (n,γ), β^- and β^+ (or EC) reactions than other reactions. Even when the neutron reaction $(n,2n)$ is important to several heavy nuclide's chains, it is possible to ignore it for the fission products. The same is true for the α decay. Therefore, each of the isotopes belonging to (2.60) can be interpreted as the end of a route or a path, in which it is only possible to move to the right or down. The first movement is related to the capture reaction and the second with the beta decay.

In order to compute the concentration of ${}^{A+3}_{Z+3}X_l$, the combinatoric analysis will begin considering only the contribution due to linear chains whose initial element is ${}^{A}_{Z}X_i$. It is possible to note that all these linear chains have 3 reactions capture and 3 beta decays, which can be corroborate in the following examples:

$$\begin{gathered}
{}^{A}_{Z}X_i \xrightarrow{n,\gamma} {}^{A+1}_{Z}X_i \xrightarrow{n,\gamma} {}^{A+2}_{Z}X_i \xrightarrow{n,\gamma} {}^{A+3}_{Z}X_i \xrightarrow{\beta^-} {}^{A+3}_{Z+1}X_j \xrightarrow{\beta^-} {}^{A+3}_{Z+2}X_k \xrightarrow{\beta^-} {}^{A+3}_{Z+3}X_l \\
{}^{A}_{Z}X_i \xrightarrow{\beta^-} {}^{A}_{Z+1}X_j \xrightarrow{\beta^-} {}^{A}_{Z+2}X_k \xrightarrow{\beta^-} {}^{A}_{Z+3}X_l \xrightarrow{n,\gamma} {}^{A+1}_{Z+1}X_l \xrightarrow{n,\gamma} {}^{A+2}_{Z+2}X_l \xrightarrow{n,\gamma} {}^{A+3}_{Z+3}X_l \\
{}^{A}_{Z}X_i \xrightarrow{\beta^-} {}^{A}_{Z+1}X_j \xrightarrow{n,\gamma} {}^{A+1}_{Z+1}X_j \xrightarrow{n,\gamma} {}^{A+2}_{Z+1}X_j \xrightarrow{n,\gamma} {}^{A+3}_{Z+1}X_j \xrightarrow{\beta^-} {}^{A+3}_{Z+2}X_k \xrightarrow{\beta^-} {}^{A+3}_{Z+3}X_l \\
{}^{A}_{Z}X_i \xrightarrow{n,\gamma} {}^{A+1}_{Z}X_i \xrightarrow{n,\gamma} {}^{A+2}_{Z}X_i \xrightarrow{\beta^-} {}^{A+2}_{Z+1}X_j \xrightarrow{\beta^-} {}^{A+2}_{Z+2}X_k \xrightarrow{n,\gamma} {}^{A+3}_{Z+2}X_k \xrightarrow{\beta^-} {}^{A+3}_{Z+3}X_l
\end{gathered} \tag{2.61}$$

The structure given in (2.61) can be expressed in terms of the involved reactions as:

$$\begin{array}{cccccc} \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{\beta^-} & \boxed{\beta^-} & \boxed{\beta^-} \\ \boxed{\beta^-} & \boxed{\beta^-} & \boxed{\beta^-} & \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{n,\gamma} \\ \boxed{\beta^-} & \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{\beta^-} & \boxed{\beta^-} \\ \boxed{n,\gamma} & \boxed{n,\gamma} & \boxed{\beta^-} & \boxed{\beta^-} & \boxed{n,\gamma} & \boxed{\beta^-} \end{array} \tag{2.62}$$

From (2.62) it is possible to note that each path consists of three β^- and three (n,γ) reactions. In fact, each route or path that begins with ${}^{A}_{Z}X_i$ and ends with ${}^{A+3}_{Z+3}X_l$, needs having three beta decay and three beta reactions, and clearly it is possible to find the number of linear chains that fulfill this condition through permutations with repetitions (Grimaldi, 2003). In other words, the number of linear chains is equal to the permutation of 3 elements with repetition in a total of six places:

$$\frac{6!}{3!\,3!} = 20 \tag{2.63}$$

To generalize this outcome, it is necessary to replace the mass number $A+3$ and the atomic number $Z+3$ for arbitraries $A+k$ and $Z+n$. Therefore, the number of linear chains who begin with ${}^{A}_{Z}X$ and ends with ${}^{A+k}_{Z+n}X$, with $1 \leq k, n$, there will be at most:

$$\frac{(k+n)!}{k!\,n!} \tag{2.64}$$

For the second step, it is necessary to solve the problem considering all the possible contribution, and not only those who begin with the isotope ${}^{A}_{Z}X_i$. In other words, the original problem was to find all the linear chains that ends with a given isotope ${}^{A+k}_{Z+n}X$ and, the above discussion only solves the question of what number of linear chains begin with a ${}^{A}_{Z}X$ isotope and ends with ${}^{A+k}_{Z+n}X$.

Therefore, it is necessary to carry out a second generalization, in which all the possible linear chains that ends with ${}^{A+k}_{Z+n}X$ will be considered. In order to carry out this last task it is necessary to observe that the decay and transmutation schemes, like the one shown in (2.60), have a rectangular shape[3]. In this rectangle, the size of the width is related to the mass number, and the height with the atomic number.

Due to this rectangular shape, it is possible to locate the isotope whose concentration is desired in the right lower corner of a given rectangle, while in the left upper corner will be the initial isotope of the largest path that ends with the desired isotope.

Now, from this initial "giant" rectangle, it is necessary to build the set of all the possible rectangles that end with the desired isotope, which can be built removing rows and columns. For example, in the scheme (2.65) is showed how a small rectangle is built, where the first column and the first row have been removed in (2.60).

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[3] It is possible that voids or empty spaces exist in the networks. However, the general shape can be considered as a rectangle.

$$\left[\begin{array}{cccccc}
{}^{\searrow}\boxed{{}^{A+1}_{Z+1}X_j} & \rightarrow & {}^{\searrow}\boxed{{}^{A+2}_{Z+1}X_j} & \rightarrow & {}^{\searrow}\boxed{{}^{A+3}_{Z+1}X_j} & \rightarrow \\
\downarrow & & \downarrow & & \downarrow & \\
{}^{\searrow}\boxed{{}^{A+1}_{Z+2}X_k} & \rightarrow & {}^{\searrow}\boxed{{}^{A+2}_{Z+2}X_k} & \rightarrow & {}^{\searrow}\boxed{{}^{A+3}_{Z+2}X_k} & \rightarrow \\
\downarrow & & \downarrow & & \downarrow & \\
{}^{\searrow}\boxed{{}^{A+1}_{Z+3}X_l} & \rightarrow & {}^{\searrow}\boxed{{}^{A+2}_{Z+3}X_l} & \rightarrow & {}^{\searrow}\boxed{{}^{A+3}_{Z+3}X_l} & \rightarrow \\
\downarrow & & \downarrow & & \downarrow &
\end{array}\right], \left[\begin{array}{cccc}
{}^{\searrow}\boxed{{}^{A+2}_{Z+2}X_k} & \rightarrow & {}^{\searrow}\boxed{{}^{A+3}_{Z+2}X_k} & \rightarrow \\
\downarrow & & \downarrow & \\
{}^{\searrow}\boxed{{}^{A+2}_{Z+3}X_l} & \rightarrow & {}^{\searrow}\boxed{{}^{A+3}_{Z+3}X_l} & \rightarrow \\
\downarrow & & \downarrow &
\end{array}\right] \tag{2.65}$$

For each of these small rectangles it is necessary to carry out the counting process that was described in the step 1, using equation (2.64). For a given "giant" rectangle, with width of a isotopes, and a high of b isotopes, the number of possible small rectangles will be $a \times b - 1$.

Therefore, in order to obtain the total number of linear chains that have as final element the isotope ${}^{A}_{Z}X$, it is necessary to perform the following sum:

$$\sum_{j=Z-a}^{Z}\sum_{i=A-b}^{A}\frac{(j+i)!}{k!\,n!}-1 \tag{2.66}$$

This expression is very useful, because through it an estimated time of computational calculation can be computed. In other words, if the mean time related to the solution of a linear chain of a path is known, then an approximate time to solve all these linear chains can be obtained through a multiplication of (2.66) by such mean time.

The counting approach to the decay and transmutation networks allows finding the total number of linear chains that contribute to a given solution. Nevertheless, the graph approach is more powerful because it provides information about the relationship of the elements belonging to these structures. In other words, this last approach allows answering the first question that was set at the beginning of the present section.

Under the graph approach, each isotope can represent a vertex or a node, and a reaction (decay process or capture) can be viewed as an edge (or link). Particularly, some decay and transmutation networks can be treated as tree structures, which is very useful in order to carry out the linearization process.

In her thesis dissertation, Maria Pusa (Pusa, 2013) provided an insightful analysis of the burnup matrix through a graph theory approach. Essentially, such analysis can be extended to decay and transmutation structures, in order to find the isotopes that are strongly connected, and in this way to permute the corresponding burnup matrix. In fact, even when a burnup code does not use the linear chain method, and instead it applies a matrix method like CRAM, it is necessary to perform a study of the decay and transmutation networks. In this case, such a study will allow building the burnup matrix, and as in the case of the Pusa's study, permuting it. Therefore, this study is not limited to the linear chain method.

Another interesting work related to the topology of the decay and transmutation network was developed by Stanisz and his colleagues (Stanisz et. al, 2019), who developed a trajectory period folding, who is very close to the TTA methodology that will be explained in the following Chapter.

At least to the best knowledge of the author of this thesis, the graph and topological approach to the decay and transmutation network is a fertile topic, where there are a few studies, and where there is the possibility to carry out important contributions. Particularly, this topic will be considered as part of future work.

2.6.5 Uncertainty related to the fission yields products.

An important issue in the development of nuclear reactor codes is the uncertainty analysis. Such analysis can be defined as a study of the propagation of the uncertainties related to nuclear data (Mills, 2014), and particularly through the different calculations inside the routines of a computational code. This analysis is necessary, especially with transport/diffusion couplings, and therefore it is convenient to make a brief discussion about such topic related to the fission product yields.

The result of a given calculation of a burnup code is influenced or has a sensitivity to at least three sources:

1) the effects related to the reactor physics calculations involved in the determination of the neutron flux, as long with the uncertainties related to the solution of the neutron transport equation (in the case of Monte Carlo codes)[4],

2) the effects related to the numerical methods and the computational round errors,

3) sensitivity due to nuclear data, as well as to the correlations between the uncertainties of it.

A common methodology used to compute the sensitivity terms consist in to repeating a series of calculations, varying the value of the nuclear data starting from the best estimate value, and to observe the spread of the results. Nevertheless, this procedure does not take into account the correlation neither the constraints between the uncertainties of the data. For the uncertainties related to cross sections, it is possible to carry out such variations with the Variance-Covariance Matrices, through which it is possible to consider the correlations between reactions, energies and isotopes (Leray et. al., 2017).

Unfortunately, in the case of the fission product yields, only the uncertainties are provided in the international evaluations, but not the correlations, and therefore it is not possible to apply the same procedure. Therefore, there is a recent effort in the development of methodologies that take into account correlations between fission yields considering conservation equations, for which is necessary the generation of covariance matrices (Pigni et. al., 2013).

There are different methodologies to generate these covariance matrices, for example through perturbation theory with the "Five Gaussians and Wahl's models" (Pigni et. al., 2013), through updating data with the Bayesian/General Least-Squares (GLS) method (Kawano and Chadwick, 2013), through the Monte Carlo perturbation using the GFE code (Schmidt et. al., 2016), and

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[4] Some authors also include the uncertainties related to the normalization factor (García-Herranz et. al, 2008).

with the Q-matrix approach for generating covariances for the direct fission yields through the cumulative fission yields (Mills, 2014).

From the last methodologies, the work developed by Mills has a relationship with the discussion that was carried out in Section 2.6.2. As it was showed in that section, it is possible to compute the cumulative yield through the direct yield and the solution of the isobaric chain given in equation (2.36). Since the cumulative yield's measurements have smaller uncertainties that the direct ones, it is possible to use them to generate covariance matrices. Mills set the following equation, which can be proved to be equal to the equation (2.57):

$$Y_i^c = \sum_j Y_j^i Q_{j,i} \tag{2.67}$$

Where Y represents the yield and the superscripts c and i denote the cumulative and independent (or direct) type, the subscripts j and i refers to the isotopes X_j who decays to the isotope X_i, and $Q_{j,i}$ is the fraction of such decays. If the variance equation is applied to (2.67) it follows:

$$var(Y_i^c) = var\left(\sum_j Y_j^i Q_{j,i}\right) \tag{2.68}$$

Through the formula for the variance of a sum, the equation (2.68) becomes:

$$var(Y_i^c) = \sum_j var\left(Y_j^i\right) Q_{j,i}^2 + 2\sum_j \sum_k Q_{j,i} Q_{k,i}\, cov\left(Y_j^i, Y_k^i\right) \tag{2.69}$$

As it was mentioned in Section 2.5.5, the ENDF/B-VII.1 library contains the values $var(Y_i^c)$, and $var\left(Y_j^i\right)$. The values $Q_{j,i}$ can be obtained through the solution of the isobaric chain discussed in Section 2.6.3. Therefore, in equation (2.69) the only values that are unknown are $cov\left(Y_j^i, Y_k^i\right)$, and, clearly, they can be obtained through such equation. The topic of uncertainties related to the fission product yields is relatively recent, and it also represent an important field to develop future contributions. Additionally, it will be necessary to study this field as a part of the work that is being developed in the AZTLAN Platform project.

There is an interesting fact about the values of the uncertainties $var(Y_i^c)$ and $var\left(Y_j^i\right)$ found in the ENDF/B-VII.1 library. In some cases, these variances are considerably large and can be comparable with the value of the fission yields, specially, for the direct or independent's type. Some examples of this behavior are listed in Table 2.7. Particularly, this is true for the direct or independent type.

2.6.6 Uncertainty related to the effective removal coefficients.

In terms of the effective removal coefficients, it is possible to carry out an uncertainty analysis based on the Taylor series expansion of the concentration function X. Such procedure is known as the linear sensitivity analysis and it is implemented in codes like ACAB or CASEMATE (García-Herranz et. al., 2008).

Table 2. 7. Examples of isotopes whose yields have a significant variance.

Isotope	Y^c	$var(Y^c)$	Y^i	$var(Y^i)$
^{95}Mo	6.5196x10^{-2}	4.85 x10^{-4}	4.952930x10^{-12}	3.1698x10^{-12}
^{131}Xe	3.1788x10^{-4}	8.700x10^{-5}	4.912820x10^{-9}	2.4062x10^{-9}
^{133}Xe	1.9654x10^{-3}	1.246x10^{-3}	2.558684x10^{-5}	1.1586x10^{-5}
^{87}Sr	2.4899x10^{-9}	1.589x10^{-9}	1.324457x10^{-8}	7.0650x10^{-9}
^{133}I	3.6556x10^{-3}	2.339x10^{-3}	1.654684x10^{-3}	7.7043x10^{-4}
^{133}Cd	4.5014x10^{-11}	2.873x10^{-11}	4.501480x10^{-11}	2.8809x10^{-11}

If it is considered that the concentration function X depends on the following set of variables $\mathcal{X} = \{x_1, x_2, \dots, x_n\}$, among which there are the λ_i^{eff} constants, it is possible to set that:

$$X_k = \overline{X_k} + \sum_i \frac{\partial X_k}{\partial x_i}(x_i - \overline{x_1}) + \cdots \tag{2.70}$$

Where $\overline{x_i}$ represents the best estimate value of the variable x_i, and $\overline{X_k}$ is the solution obtained with the values $\{\overline{x_1}, \overline{x_2}, \dots, \overline{x_n}\}$ (Chaitanya Tadepalli and Subhash, 2018). For a first order approach it follows that:

$$X_k - \overline{X_k} \cong \sum_i \frac{\partial X_k}{\partial x_i}(x_i - \overline{x_1}) \tag{2.71}$$

Applying the same procedure used in (2.68):

$$varX_k \cong \sum_i \sum_j \frac{\partial X_k}{\partial x_i}\frac{\partial X_k}{\partial x_j}\, cov(x_i - \overline{x_1}) \tag{2.72}$$

The term defined as $S_{ki} = (\partial X_k / \partial x_i) \cdot (x_i / X_k)$, is the sensitivity coefficient of the concentration function. In this case, the uncertainty and the sensitivity are related with the derivative of the modified Bateman equation with respect to the effective lambda coefficient. In a recent work developed by Chaitanya Tadepalli and Subhash (Chaitanya Tadepalli and Subhash, 2018), a set of simplified recursive relations of such derivatives has been developed.

As it will be showed in Chapter 5, there is an alternative way to build in a fast way such derivatives through a symbolical computational algorithm, which was developed in the present thesis. From the above discussion, it is clear that the modified Bateman equation has an important role in the uncertainty analysis, and therefore its study continues being important.

2.7 The artificial dependence on time in burnup problems.

In Section 2.3 the effective removed lambdas, λ_i^{eff}, and the effective branching ratios, $b_{i,i+1}^{\text{eff}}$, were considered constant in time. Nevertheless, for some isotopes who undergoes nuclear reactions, such parameters are not constant, because they depend on the flux and the microscopic cross section, which acquire an artificial dependence on time due to the burnup process.

2.7.1 The flux dependence on time.

It is possible to understand this phenomenon through the dependence of the flux on the material composition. As it is known, when the diffusion or the transport equation is solved, the material composition is an important part of such solution through the macroscopic cross sections:

$$\Sigma_r = X\sigma_r \tag{2.73}$$

Where X is the atomic density and σ_r is the microscopic cross section for the reaction r. The term given in (2.7) is very useful to compute the different ration rates. Then, it is possible to set the following relationship between the flux and the atomic density:

$$\phi(E) \equiv \phi(E, X) \tag{2.74}$$

As it was defined in Chapter 1, in a burnup problem the composition of the core change in time, due to the transformation of the isotopes and the different reactions that they undergo. Therefore, it is clear that the atomic density is a function of time. In other words, $X \equiv X(t)$. Considering this, it is clear that the relationship described in (2.74) can be written as:

$$\phi(E, X) \equiv \phi(E, t) \tag{2.74}$$

In the last equation the relationship between the flux and the time in a burnup problem has been showed.

2.7.2 An integral the cross section.

The dependence on time of the microscopic cross section, on the other hand, is artificial and it is possible to discuss it through its definition. As it is known, the microscopic cross section usually is described as a proportionally constant through the following equation (Lamarsh, 2001):

$$\begin{matrix}\text{Number of collision per} \\ \text{second (in entire target)}\end{matrix} = \sigma n v N \mathcal{A} T \tag{2.76}$$

Such definition arises from a theoretical experiment, where a beam containing n monoenergetic neutrons per cm^3 with a velocity v, impinges on a target of thickness T, area $\mathcal{A}$ and atom density N. If the equation (2.76) is divided by the volume of the target, given by $\mathcal{A}T$, it is possible to define the collision density F:

$$F = \sigma N n v = \Sigma n v \tag{2.77}$$

From the last equation comes the definition provided in (2.73). Even when this definition set that the microscopic cross section is constant, it does not explain the nature of such constancy.

Therefore, it is more convenient to develop a definition starting from the macroscopic cross section Σ and considering its relationship with length. As it is known, Σ is the expected number of neutron-nucleus collision per unit of length of path traveled (Meghreblian and Holmes, 1960). Essentially, it is a necessary a formulation from a point of view of the neutron instead of the theoretical experiment, who describes the properties of the materials through the space. In addition, it is convenient to include the speed of such neutron as a variable.

The function $h(r)$ will be defined as the probability per unit of time that a neutron who has a distance r from a nucleus, will suffer a collision with it. For a differential length ds, Σ can be computed as:

$$\Sigma = \int_0^\infty \frac{NdV \cdot h(r)dt}{ds} \tag{2.78}$$

In this case, the integral has been defined in terms of the time, considering a total of NdV nucleus in a differential volume dV. The next step consists of making a change of variable, considering that in a differential time dt, the neutron travels a distance given by:

$$dt = \frac{ds}{v} \tag{2.79}$$

Clearly the integration limits remain unchanged. On the other hand, since a reference frame centered on the neutron is used, then the differential of volume corresponds to a sphere $dV = 4\pi r^2 dr$, therefore (Meghreblian and Holmes, 1960):

$$\begin{aligned}\Sigma &= \int_0^\infty \frac{NdV \cdot h(r)dt}{ds} = \int_0^\infty \frac{N(4\pi r^2 dr) \cdot h(r)\frac{ds}{v}}{ds} \\ &= \int_0^\infty N(4\pi r^2 dr) \cdot \frac{h(r)}{v} = N \underbrace{\int_0^\infty \frac{4\pi r^2}{v} \cdot h(r)dr}_{\sigma}\end{aligned} \tag{2.80}$$

In the last equation the term $h(r)dr$ can be interpreted as a probability distribution, per unit of time, that a neutron who has a distance between r and $r + dr$ from a given nucleus, will suffer a collision with such nucleus. Therefore, the microscopic cross section can be interpreted as an integral of a weighted probability function, who only depends of the position r.

Strictly speaking, the dependence on time has been changed by a dependence on energy, which can be found in the term $1/v$ of the integral. In other words, the microscopic cross section only depends on the type of the nucleus and on the energy.

2.7.3 Artificial dependency on time of the microscopic cross section.

In the past section it was analyzed that the microscopic cross section does not depends on time, instead it is defined in terms of the isotope and the energy of the incident neutron. Nevertheless, this parameter is used in burnup calculations in two ways:

1) through the definition provided in (2.15), corresponding to the homogenized one group cross section:

$$\sigma_{i,m} = \frac{\int_0^\infty \sigma_{i,m}(E)\phi(E)dE}{\int_0^\infty \phi(E)dE} \tag{2.81}$$

2) through the multigroup energy discretization:

$$\sigma_{i,m}^{g} = \frac{\int_{E_i}^{E_{i+1}} \sigma_{i,m}(E)\phi(E)dE}{\int_{E_i}^{E_{i+1}} \phi(E)dE} \tag{2.82}$$

For a group g defined as the interval of energy between E_i and E_{i+1}. Both (2.81) and (2.82) use the flux as a weight function, and since such function depends on the time, then the microscopic cross section acquires an artificial dependency on time.

2.7.4 An approximated solution.

An important conclusion for the past sections is that, for effective removal coefficients who involve neutron-nucleus reactions, the following mass balance equation can be written as:

$$\frac{dX_{i+1}}{dt} = b_{i,i+1}^{\text{eff}}\lambda_i^{\text{eff}}X_i - \lambda_{i+1}^{\text{eff}}X_i \tag{2.83}$$

$$= b_{i,i+1}\left(\phi(\overline{X})\right)\lambda_i\left(\phi(\overline{X})\right)X_i(t) - \lambda_{i+1}\left(\phi(\overline{X})\right)X_i$$

Where $\overline{X}$ denotes the total composition of the core, which clearly depends of the function of X_i and X_{i+1}. In other words, the mass balance equation has the following structure:

$$\frac{dX_{i+1}}{dt} = G(X_i, t)X_i(t) - L(X_{i+1}, t)X_{i+1}(t) \tag{2.84}$$

Since the functions G and L depends on X_i and X_{i+1} respectively, the equation (2.84) becomes in a not linear differential equation, and therefore the system cannot be solved as it was made until now. A methodology that is used to overcome this difficulty consist of discretizing the time in a set of intervals, $\Delta t_i, \Delta t_{i+1}, \ldots,$. Then, for each interval of time, the neutron flux ϕ will be assumed not depending on time, i.e. the parameters $b_{i,i+1}^{\text{eff}}\lambda_i^{\text{eff}}$ and $\lambda_{i+1}^{\text{eff}}$ will be considered constant, and the modified Bateman equation can be used. Then, the neutron flux will be updated as long with the microscopical cross section for the next interval, repeating the process.

For example, beginning with the interval $\Delta t_i = t_{i+1} - t_i$, the system (2.83) will be solved, obtaining $\overline{X}(t_{i+1})$. Afterward, this material composition, $\overline{X}(t_{i+1})$ will be used in the solution of the neutron diffusion or in the neutron transport equation, considering it as constant, and obtaining the neutron flux $\phi(t_{i+1})$. This new flux will be used or updated, in turn, to compute the microscopical cross section $\sigma(t_{i+1})$ and to solve equation (2.83) for the following time interval $\Delta t_{i+1} = t_{i+2} - t_{i+1}$. Then, the new material composition $\overline{X}(t_{i+2})$ will be found, after of which the procedure is repeated. This approximated solution is equivalent to a coupling between a solver of the Bateman's equation with a solver of the neutron diffusion or the transport equation. Such coupling is described in Figure 2.8.

2.7.5 The predictor-corrector method.

It is possible to improve the methodology described in the past section through an averaging process called the "predictor-corrector method".

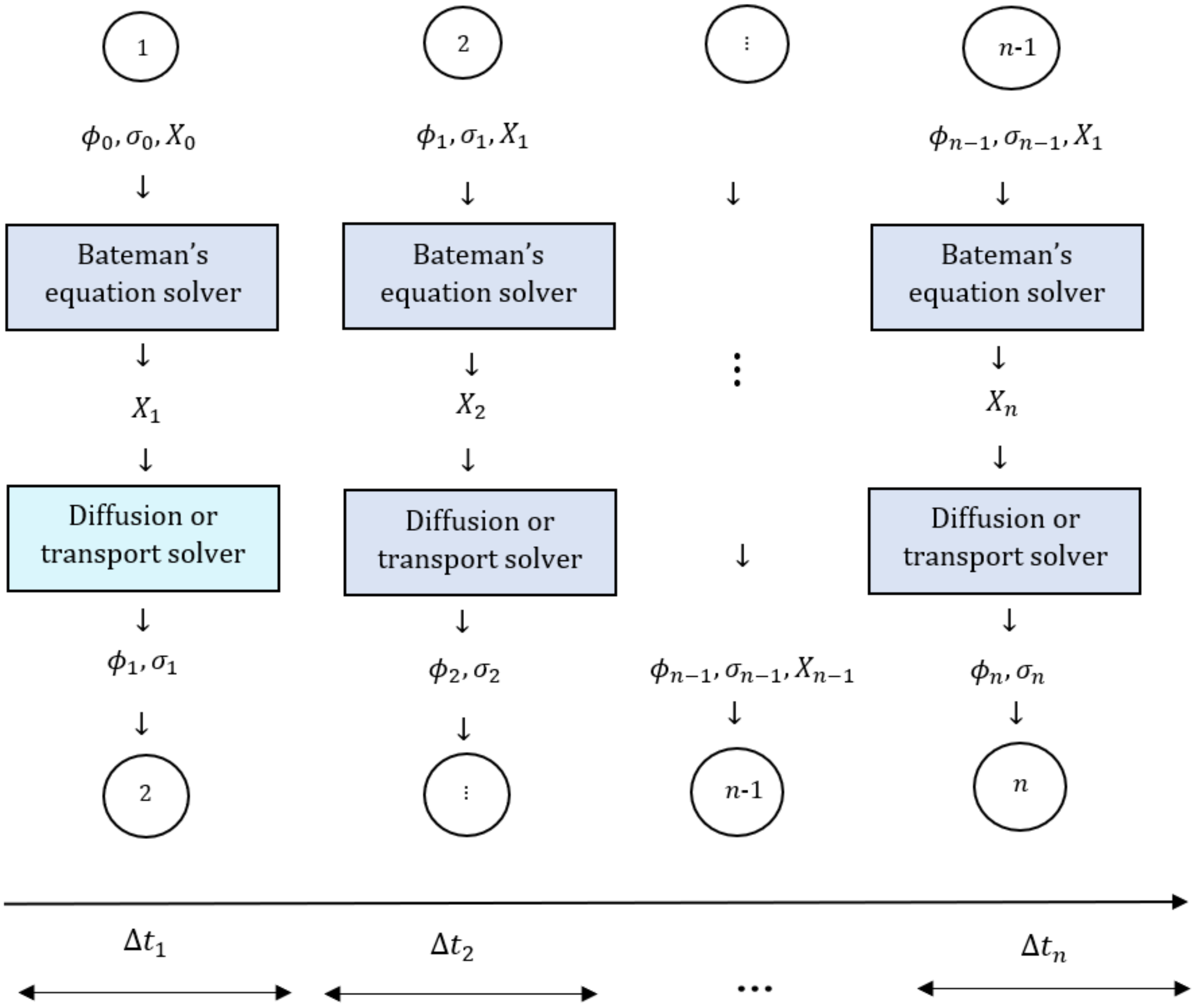

Figure 2. 8. Scheme of the discretization of time and coupling for a burnup process. The vertical relationships show the coupling between a Bateman's equation solver and a diffusion/transport solver. The horizontal arrows show the discretization in time

Such methodology can be explained using as example the first step of burnup. As it was mentioned in the last section, when the material composition $\overline{X}_1$ is computed during the first-time step, it is considered that the neutron flux ϕ_0 remains constant. This can be represented as:

$$B(X_0, \phi_0, \sigma_0, t_1) \rightarrow \overline{X_{1,p}} \tag{2.85}$$

Where B represents a burnup solver, which uses the input data given by X_0, ϕ_0, σ_0, for a time step t_1. The sub index p will refer to the concentration computed in a predictor step. After this, the neutron flux is calculated through a diffusion/transport solver, using the $\overline{X_{1,p}}$ computed previously. In other words:

$$(D/T)(\overline{X_{1,p}}) \rightarrow \phi_1, \sigma_1 \tag{2.86}$$

Where $(D/T\ )$ represents a Diffusion/Transport Solver. Once this new neutron flux and the microscopic cross section have been computed, it is necessary to repeat the calculation for the same time step, but with the updated neutron flux and microscopic cross section, i.e.:

$$B(X_0, \phi_1, \sigma_1, t_1) \rightarrow \overline{X_{1,c}} \tag{2.87}$$

Where the sub index c will refer to the corrector step. Finally, the concentration for the first-time step can be computed as:

$$\overline{X_1} = \frac{\overline{X_{1,p}} + \overline{X_{1,c}}}{2} \tag{2.88}$$

Then, this mean concentration will be used to compute the adjusted neutron flux $\overline{\phi_1}$ and the microscopic cross section $\overline{\sigma_1}$. In the scheme showed in Figure 2.9 is summarized this procedure. The discussion about the mathematical justification of the predictor-corrector method is related to the Euler constant extrapolation method, and more formally with serval methodologies to approximate nonlinear differential equations (Douglas and Jones, 1963), (Cash, 1983), (Voss and Khaliq, 1999). Nevertheless, such formal discussion is beyond the scope of the present thesis.

There are more advances methodologies of the predictor-corrector method, which are based in using extrapolated reaction rates for the predictor step as well as interpolated reactions rates for the corrector calculations. Such methodologies are used in the well-known Monte Carlo transport code SERPENT, and they have been studied mainly through the works carried out by Isotalo and Aarnio (Isotalo and Aarnio, 2011).

2.7.6 Sub-steps methodology.

It would be desirable to use small time steps for a burnup calculation, with the purpose to provide a better estimation of the material composition $\bar{X}$ as well as the neutron flux and the microscopic cross section. Unfortunately, as it is suggested (Knott and Yamamoto, 2010), the calculations of the neutron flux and the microscopic cross section dominate the execution time of a lattice physics computation.

Therefore, it is not convenient to use a large number of time steps, because the execution time will increase considerably. The methodology described in the last section allows to improve the approximation of considering the neutron flux and the microscopic cross section constant, but there is another methodology that allows to improve the limitation of the number of time steps that can be used in a burnup problem.

Such methodology consists of defining a second set of time steps of a given time steps, i.e. defining sub-steps. In order to explain the idea behind such methodology, two-stages of a given burnup problem will be considered, which are represented as:

$$\underset{\{X_0, \phi_0, \sigma_0\}}{\text{Initial conditions}} \quad \overset{\Delta t}{\rightarrow} \quad \underset{\{X_1, \phi_1, \sigma_1\}}{\text{First time-step}} \tag{2.89}$$

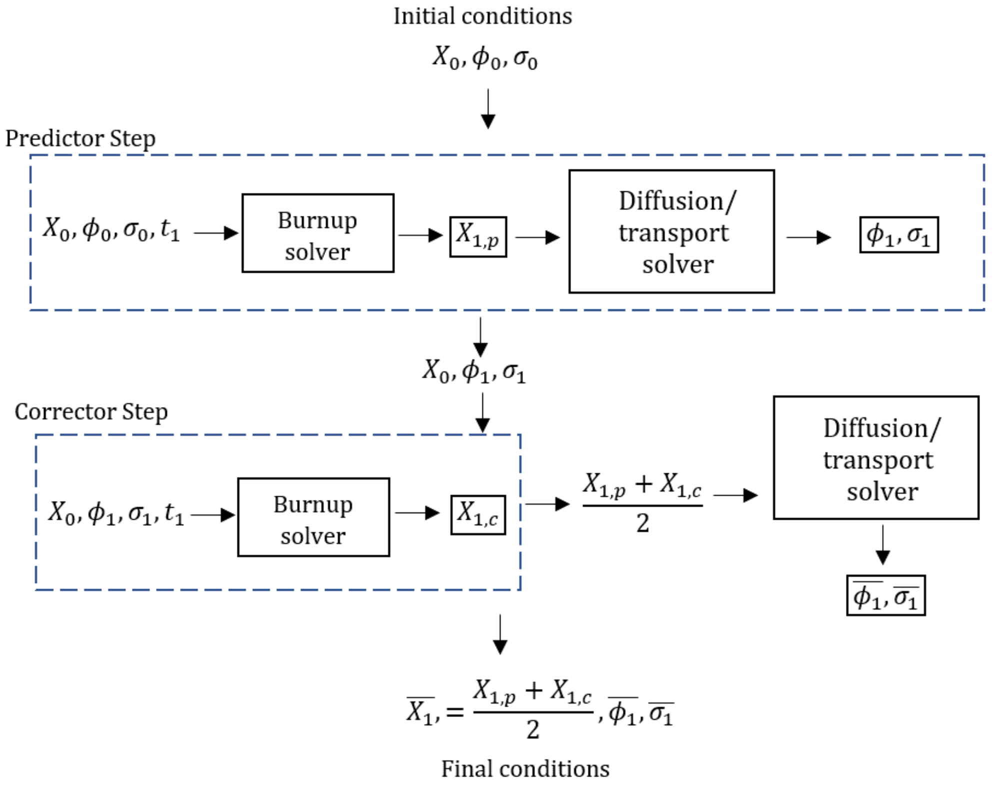

Figure 2. 9. Scheme of the predictor-corrector methodology.

As it was discussed before, during the time interval Δt, both ϕ_0 and σ_0 are considered constant. At the end of such interval in t_1, there is a suddenly change from ϕ_0, σ_0 to ϕ_1, σ_1 due to the update through the diffusion/transport code, using X_1 as an input. This change can be represented as a stair-step graph:

$$\phi(t)\sigma(t) = \begin{cases} \phi_0\sigma_0 & t \in [0, t_1) \\ \phi_1\sigma_1 & t = t_1 \end{cases} \qquad (2.90)$$

Clearly the equation (2.90) is not continuous in $t = t_1$. For some scenerios it is useful to produce a set of values $\phi(t)_{i_j}\sigma(t)_{i_j}$, $0 < i_1 < i_1 < \cdots < i_n < t_1$, through a process of interpolation between $\phi_0\sigma_0$ and $\phi_1\sigma_1$. It is worth nothing that such generation does not require using a diffusion/transport code. Therefore:

$$\phi(t)\sigma(t) = \begin{cases} \phi_0\sigma_0 & t \in [0, i_1) \\ \phi_{i_1}\sigma_{i_1} & t \in [i_1, i_2) \\ \phi_{i_2}\sigma_{i_2} & t \in [i_2, i_3) \\ \vdots & \vdots \\ \phi_{i_n}\sigma_{i_n} & t \in [i_n, t_1) \\ \phi_1\sigma_1 & t = t_1 \end{cases} \qquad (2.91)$$

Depending on the type of the interpolation process, this new stair-step graph can be closer to the actual behavior of the function $\phi(t)\sigma(t)$. Figure 2.10 shows an example of such procedure for a linear interpolation (Isotalo, 2013). One of the most important implications of (2.91) is related to the new size of the time step, and to the balance equation of certain fission products.

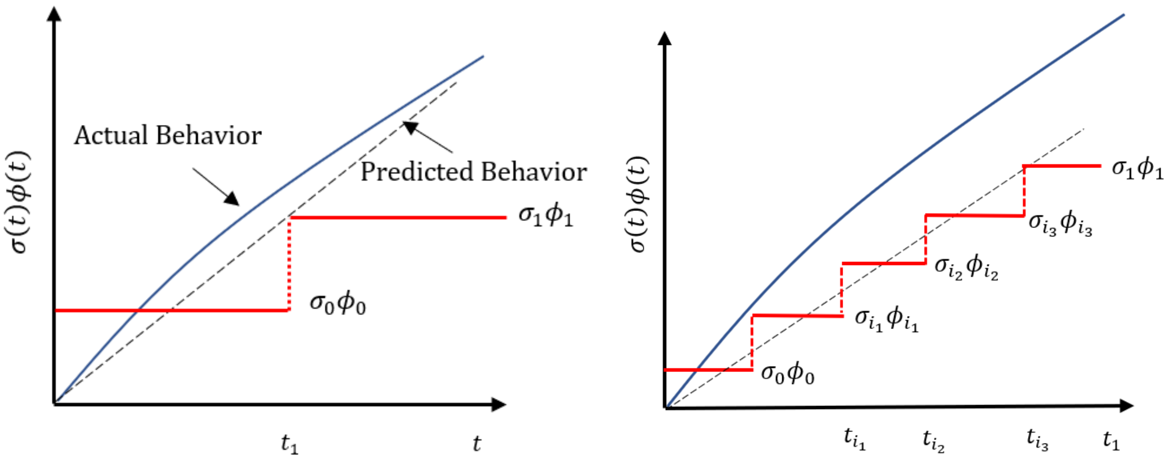

Figure 2. 10. Example of a sub-step methodology where an original time step has been divided in three sub steps. In the left part, a standard procedure of a predictor-corrector calculation is depicted.

Leppänen explained this last through the analysis of the fission product ^{131}I, who is a low-absorbing fission product with a continuously changing production rate (Leppänen, 2012). Essentially, this author shows that the mass balance equation for some isotopes can be written as:

$$\frac{dX_j}{dt} = P(t) - \lambda_j X_j \approx P - \lambda_j X_j \tag{2.92}$$

Where the production term, $P(t)$, who originally depends on time, can be approximated by a constant value over a time step for certain cases, where the main contribution to X_j comes from a heavy isotope whose concentration has a small variation on the given time step. In such scenario the solution of equation (2.92) is equal to:

$$X(t) = \frac{P}{\lambda_j}\left(1 - e^{-\lambda_j t}\right) \tag{2.93}$$

If the burnup time step is considerably longer than the half-life of the isotope, the concentration of the isotope will reach the most part of the equilibrium concentration at the end of such time step:

$$X \approx \frac{P}{\lambda_j} \tag{2.94}$$

The problem is that P depends considerably on the value $\sigma(t)\phi(t)$, which is treated as constant in the time-step interval. Therefore, for a procedure similar to the left part of Figure 2.10, this constant value $\sigma_0\phi_0$ differs considerably from the final value $\sigma_1\phi_1$, and therefore the production rate P is lower than the actual value and the concentration X is underestimated. With the sub-step methodology, the time step is lower and therefore the concentration from equation (2.93) can be approximated in a better way. It is worth mentioning that the use of the sub-step methodology does not automatically improve the results of all the fission products, as

Leppänen suggests (Leppänen, 2012), because as it was discussed the procedure depends on the comparison of the time-steps with the half-life of the isotopes, as well with the condition of equilibrium.

Additionally, Knott and Yamamoto (Knott and Yamamoto, 2010) analyzed the importance of the sub-step methods for reducing the error of temporal discretization, and for avoiding the underestimation of the burnup due to the power normalization.

2.8 Comparison between burnup codes.

In one of the stages of the development of the code Szilard, it was very important to have a tested burnup code in order to perform comparisons. As a first option it was used the Monte Carlo SERPENT Code, but when some issues related with the fission product appeared, it was used instead the Monte Carlo N-Particle X code (MCNPX) in its version 2.6.2 (Pelowitz, 2008).

The reason behind this change was related to some discrepancies between the concentration of some fission products between SERPENT and Szilard. Once several possible causes of such discrepancies were discarded, it was considered the possibility that the origin was in the decay and transmutation networks that were used. Unfortunately, the way in which the linear chains are formed and used by SERPENT are not documented (Leppänen, 2017), and therefore a it was impossible to carry out a correct comparison.

On the other hand, the specific isotopes that are included in the calculations performed by MCNPX are well documented. This code used three schemes named TIER, which uses different numbers of fission products. TIER 1 contains 12 fission products, TIER 2 considers 85 and TIER 3 uses 220. This last are listed in Table 2.8.

Using this information, a set of linear chains were built for Szilard, through which it was possible to make more adequate comparisons. During such process an important conclusion was obtained about the relationship between the neutron flux and the fission products.

2.8.1 The neutron flux and the decay and transmutation network.

A very interesting conclusion was obtained when the results of MCNPX were compared with the results of SERPENT, which shows that even when two codes have similar results for the behavior of neutron flux, this not implies that the concentration of the fission products will share the same behavior, even more it is possible that the percentual error between the concentrations will be considerable.

The data in which is based the following discussion is related to a comparison that was carried out simulating a unit cell in infinite medium, in a thermal neutron flux spectrum. Even when the configuration of this simulation will be described in Section 4, for the moment a general discussion of the results it is possible in order to exemplified the issues related with the comparison between codes. As it is possible to conclude from Figure 2.11, the neutron flux computed with both codes has the same behavior, and from the Figure 2.12 it is possible to conclude that the percentual error between the results is lower than 1.4%. In fact, the graph has its maximum value for the time equal to 12.5 days. In other words, the results for the neutron flux for both codes are very close.

Table 2. 8. List of isotopes that are considered in the TIER 3 in MCNPX.

^{69}Ga	^{71}Ga	^{70}Ge	^{72}Ge	^{73}Ge	^{74}Ge	^{76}Ge	^{74}As	^{75}As	^{74}Se
^{76}Se	^{77}Se	^{78}Se	^{79}Se	^{80}Se	^{82}Se	^{79}Br	^{81}Br	^{78}Kr	^{80}Kr
^{82}Kr	^{83}Kr	^{84}Kr	^{85}Kr	^{86}Kr	^{85}Rb	^{86}Rb	^{87}Rb	^{84}Sr	^{86}Sr
^{87}Sr	^{88}Sr	^{89}Sr	^{90}Sr	^{88}Y	^{89}Y	^{90}Y	^{91}Y	^{90}Zr	^{91}Zr
^{92}Zr	^{93}Zr	^{94}Zr	^{95}Zr	^{96}Zr	^{93}Nb	^{94}Nb	^{95}Nb	^{97}Nb	^{92}Mo
^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{99}Mo	^{100}Mo	^{99}Tc	^{96}Ru	^{98}Ru
^{99}Ru	^{100}Ru	^{101}Ru	^{102}Ru	^{103}Ru	^{104}Ru	^{105}Ru	^{106}Ru	^{103}Rh	^{105}Rh
^{102}Pd	^{104}Pd	^{105}Pd	^{106}Pd	^{107}Pd	^{108}Pd	^{110}Pd	^{107}Ag	^{109}Ag	^{111}Ag
^{106}Cd	^{108}Cd	^{110}Cd	^{111}Cd	^{112}Cd	^{113}Cd	^{114}Cd	^{116}Cd	^{113}In	^{115}In
^{112}Sn	^{113}Sn	^{114}Sn	^{115}Sn	^{116}Sn	^{117}Sn	^{118}Sn	^{119}Sn	^{120}Sn	^{122}Sn
^{123}Sn	^{124}Sn	^{125}Sn	^{126}Sn	^{121}Sb	^{123}Sb	^{124}Sb	^{125}Sb	^{126}Sb	^{120}Te
^{122}Te	^{123}Te	^{124}Te	^{125}Te	^{126}Te	^{128}Te	^{130}Te	^{132}Te	^{127}I	^{129}I
^{130}I	^{131}I	^{132}I	^{133}I	^{134}I	^{135}I	^{123}Xe	^{124}Xe	^{126}Xe	^{128}Xe
^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{133}Xe	^{134}Xe	^{135}Xe	^{136}Xe	^{133}Cs	^{134}Cs
^{135}Cs	^{136}Cs	^{137}Cs	^{130}Ba	^{132}Ba	^{133}Ba	^{134}Ba	^{135}Ba	^{136}Ba	^{137}Ba
^{138}Ba	^{140}Ba	^{138}La	^{139}La	^{140}La	^{136}Ce	^{138}Ce	^{139}Ce	^{140}Ce	^{141}Ce
^{142}Ce	^{143}Ce	^{144}Ce	^{141}Pr	^{142}Pr	^{143}Pr	^{145}Pr	^{142}Nd	^{143}Nd	^{144}Nd
^{145}Nd	^{146}Nd	^{147}Nd	^{148}Nd	^{150}Nd	^{147}Pm	^{148}Pm	^{149}Pm	^{151}Pm	^{144}Sm
^{147}Sm	^{148}Sm	^{149}Sm	^{150}Sm	^{151}Sm	^{152}Sm	^{153}Sm	^{154}Sm	^{151}Eu	^{152}Eu
^{153}Eu	^{154}Eu	^{155}Eu	^{156}Eu	^{157}Eu	^{152}Gd	^{153}Gd	^{154}Gd	^{155}Gd	^{156}Gd
^{157}Gd	^{158}Gd	^{160}Gd	^{159}Tb	^{160}Tb	^{156}Dy	^{158}Dy	^{160}Dy	^{161}Dy	^{162}Dy
^{163}Dy	^{164}Dy	^{165}Ho	^{162}Er	^{164}Er	^{166}Er	^{167}Er	^{168}Er	^{170}Er	^{169}Tm

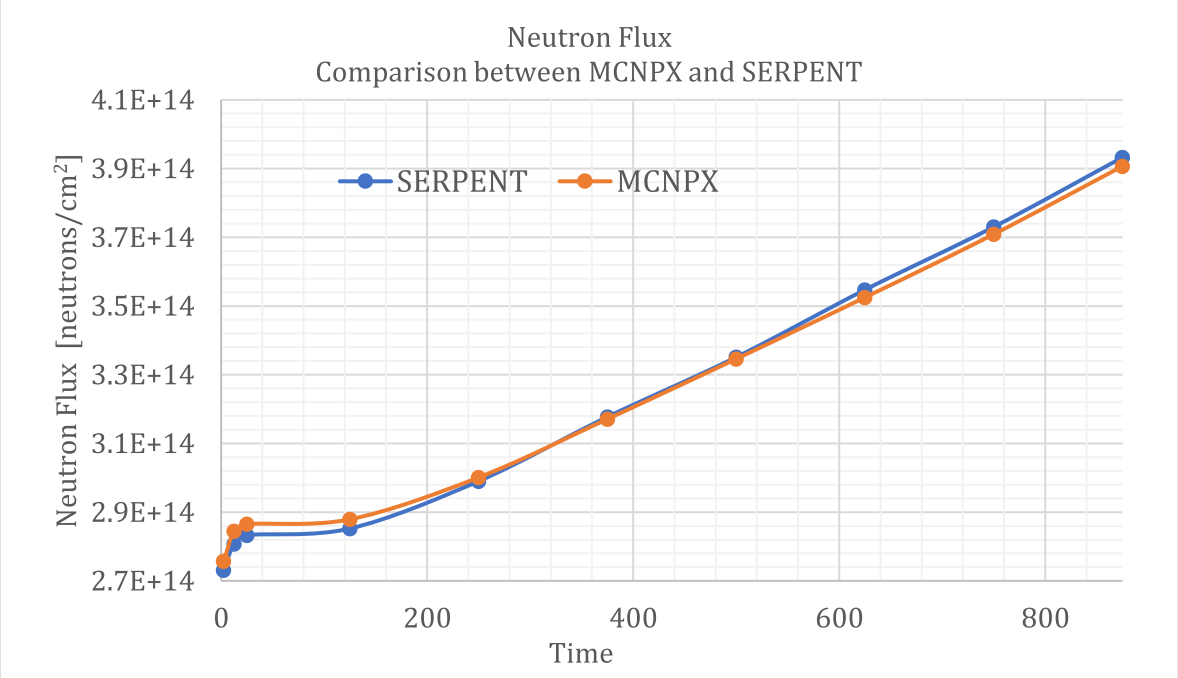

Figure 2. 11. Comparison between the neutron flux computed between MCNPX and SERPENT for the same simulation.

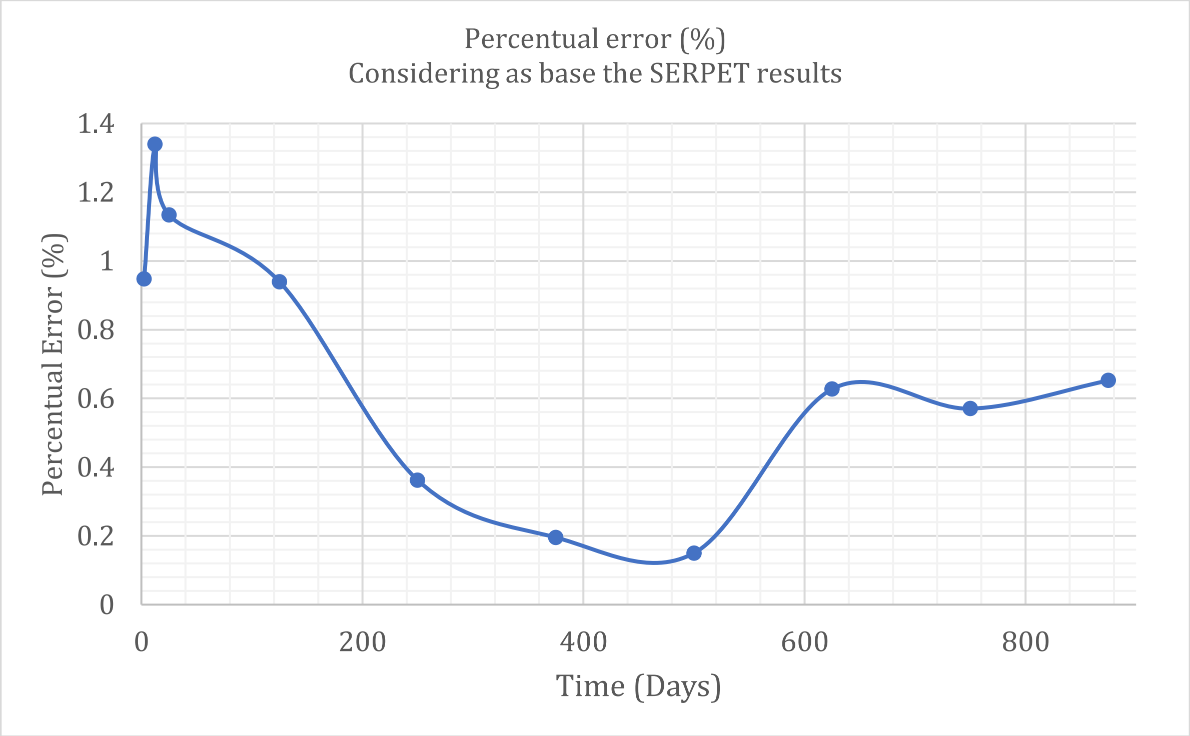

Figure 2. 12. Percentual error between the data showed in Figure 2.11 vs time.

2.8.2 Discrepancies between the concentration of some fission products.

As it was discussed in the past section, for the studied case there are not considerable differences between the neutron fluxes computed by MCNPX and SERPENT. Nevertheless, for some fission products the opposite is true, because there are marked discrepancies. Three examples are showed from Figure 2.13 to Figure 2.18, where the behavior between the concentrations are different. In total, at least 39 isotopes of the TIER 3 given in Table 2.8 showed similar discrepancies between their concentrations computed with MCNPX and SERPENT.

The author of this thesis concluded that the origin of the discrepancies can be related to the different decay and transmutation networks or to the distinct predictor-corrector method that are used by the codes. The first possibility is based in the analysis of several fission products, and it can be illustrated with the study of ^{109}Ag and the graphs given in Figure 2.15 and Figure 2.16. This isotope belongs to the following isobaric chain, which is formed by negative beta decay:

$$\overset{\searrow}{\boxed{^{109}\text{Tc}}} \xrightarrow[0.87\text{ s}]{\beta^-} \overset{\searrow}{\boxed{^{109}\text{Ru}}} \xrightarrow[34.5\text{ s}]{\beta^-} \overset{\searrow}{\boxed{^{109}\text{Rh}}} \xrightarrow[80\ s]{\beta^-} \overset{\searrow}{\boxed{^{109}\text{Pd}}} \xrightarrow[13.7012\text{ h}]{\beta^-} \boxed{^{109}\text{Ag}} \tag{2.95}$$

^{109}Ag also belongs to an isobaric chain given by electron capture decay:

$$\boxed{^{109}\text{Sb}} \xrightarrow[17\text{ s}]{\beta^+} \boxed{^{109}\text{Sn}} \xrightarrow[18\text{ min}]{\beta^+} \boxed{^{109}\text{In}} \xrightarrow[4.2\text{ h}]{\beta^+} \boxed{^{109}\text{Cd}} \xrightarrow[462.6\text{ d}]{\beta^+} \boxed{^{109}\text{Ag}} \tag{2.96}$$

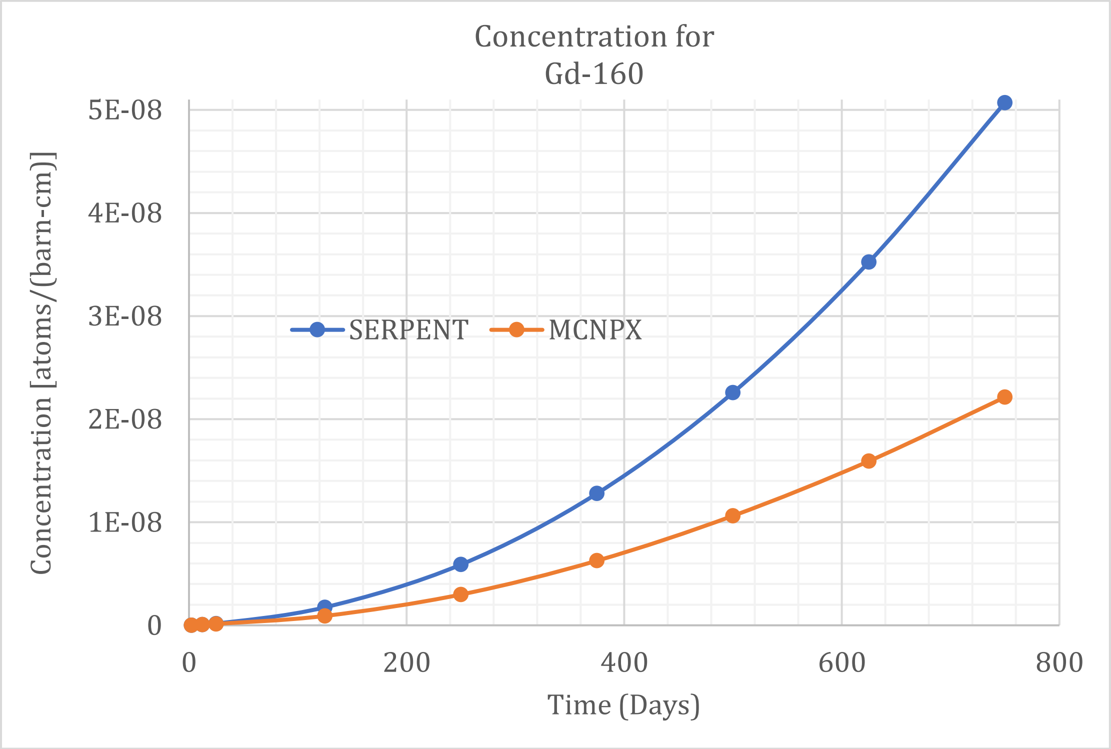

Figure 2. 13. Concentration as a function of time for ^{160}Gd computed with the code SERPENT, and the code MCNPX, for a case of study described in Chapter 4.

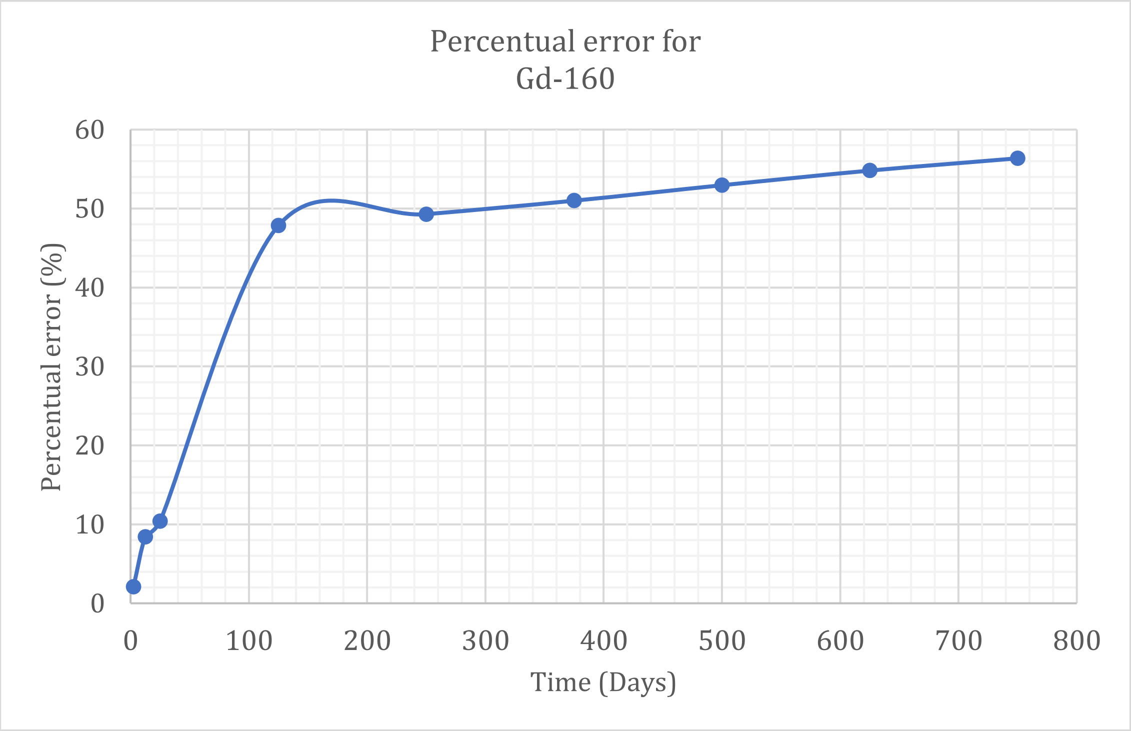

Figure 2. 14. Percentual error for the concentration showed in Figure 2.13.

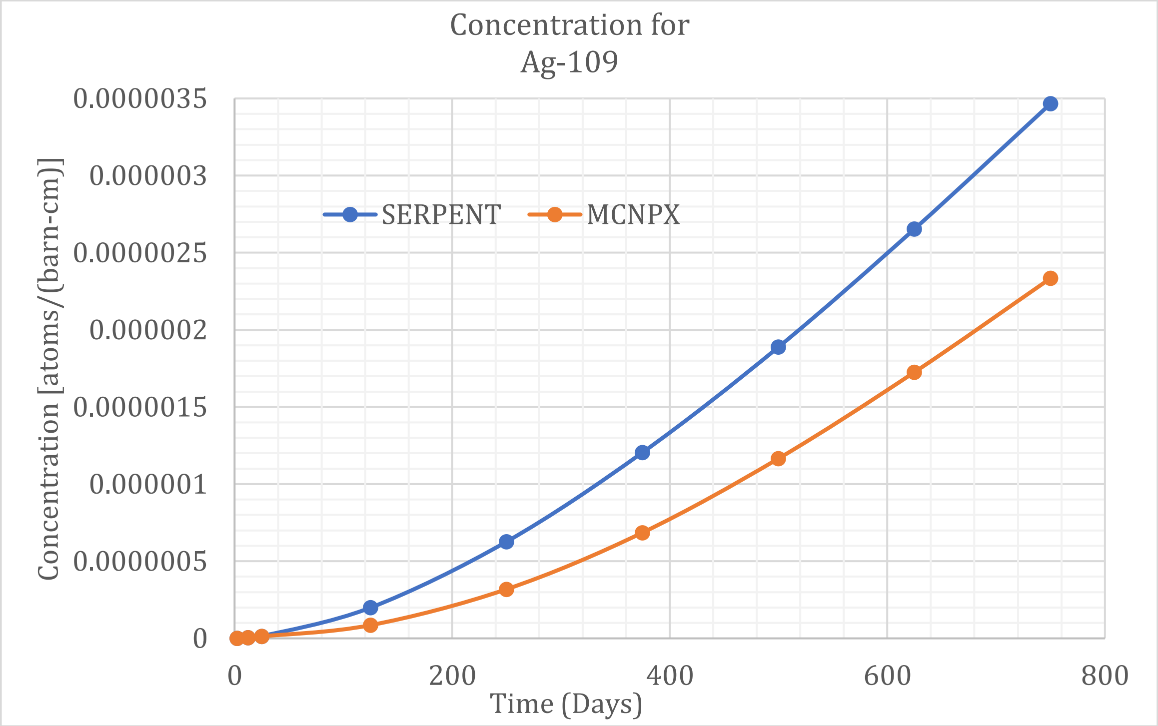

Figure 2. 15. Concentration as a function of time for ^{109}Ag computed with the code SERPENT, and the code MCNPX, for a case of study described in Chapter 4.

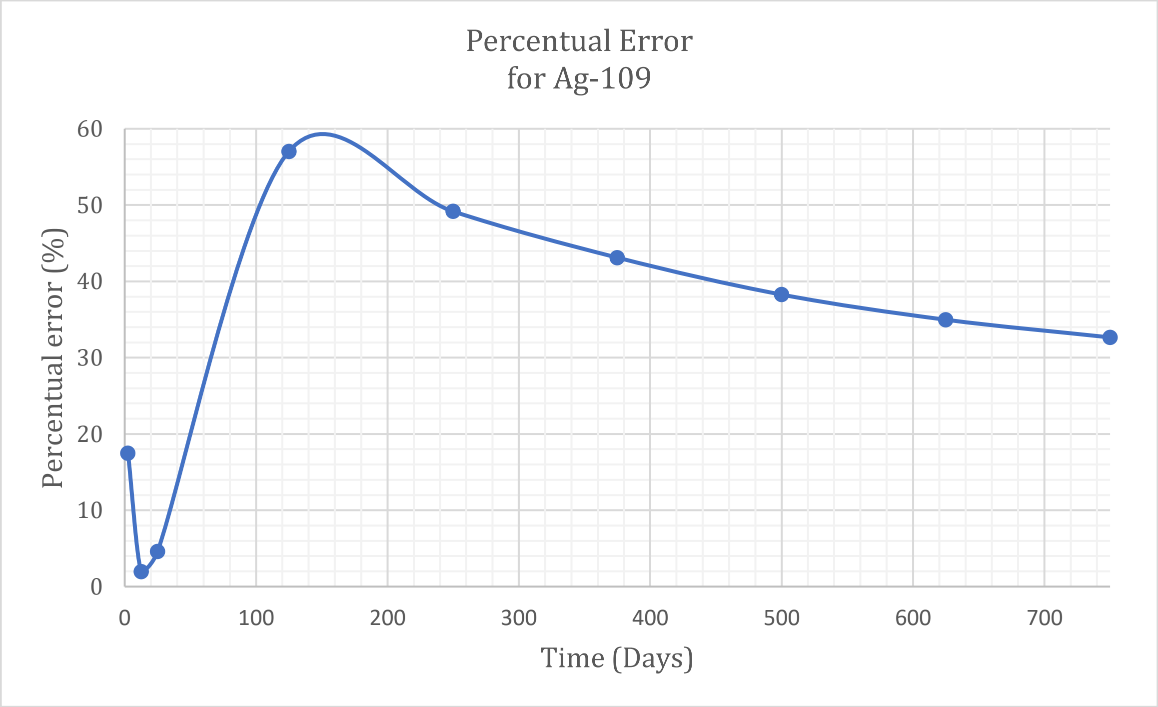

Figure 2. 16. Percentual error for the concentration showed in Figure 2.15.

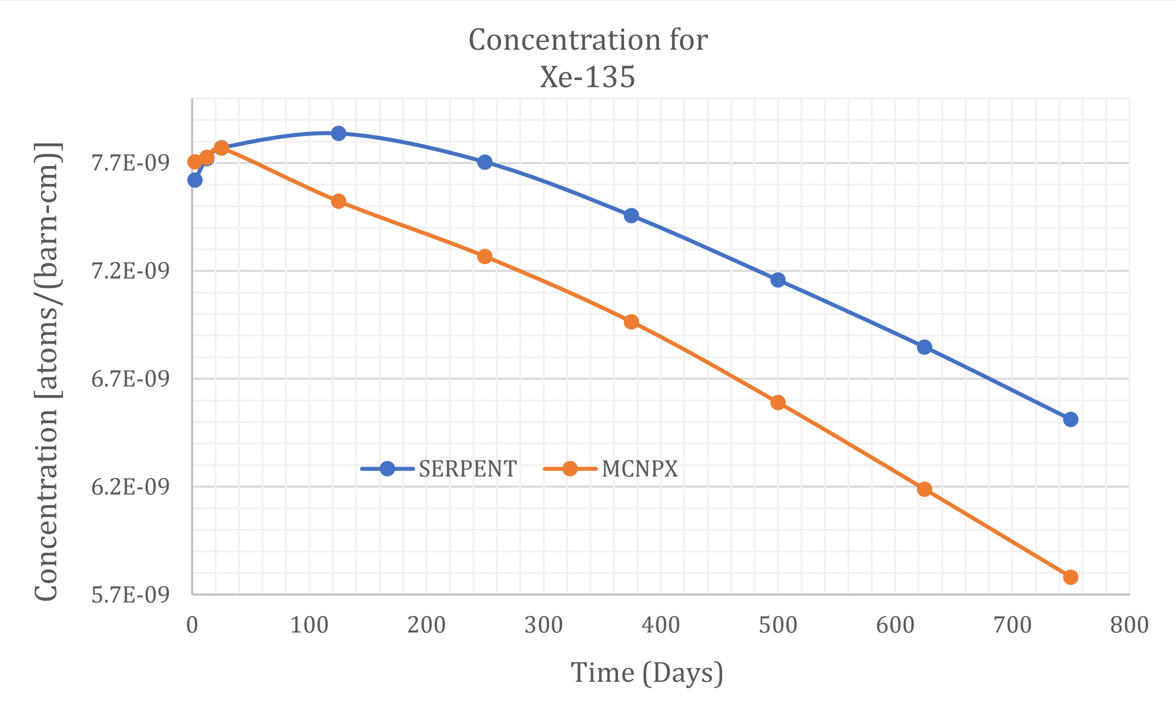

Figure 2. 17. Concentration as a function of time for ^{135}Xe computed with the code SERPENT, and the code MCNPX, for a case of study described in Chapter 4

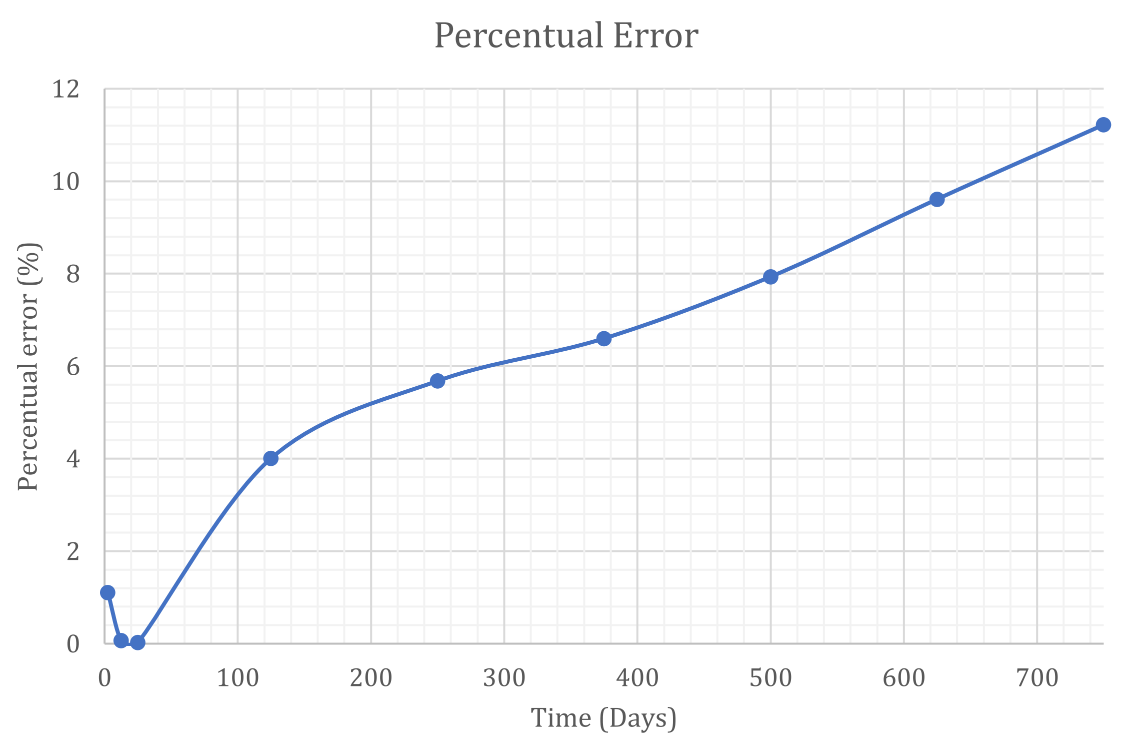

Figure 2. 18. Percentual error for the concentration showed in Figure 2.17.

From the scheme of (2.95) it is possible to conclude that the ^{109}Ag is not produced directly by the fission process. On the other hand, all the isotopes belonging to the isobaric chain in (2.96) are not produced by fission, or there are not enough data about them in the ENDF/B-VII.1 library, because they are not included as fission products of the ^{235}U and ^{239}Pu.

Therefore, the concentration only depends on the analysis of the scheme (2.95). The half-lives of the ancestors of ^{109}Ag in such scheme fulfill the condition of the first case given in Section 2.5.2., therefore it is adequate to use the cumulative yield for ^{109}Ag.

From Table 2.8 it is clear that MCNPX use this type of yield for ^{109}Ag, because there are not included the ancestors of this isotope in such table. Nevertheless, if the isotope ^{109}Pd (one of the ancestors) is not included, the concentration of ^{109}Ag will be underestimated, due to the omission of contributions originated by the following sequence radioactive captures:

$$\begin{tikzcd}
\overset{\searrow}{\boxed{^{104}\mathrm{Pd}}} \arrow[r] & \overset{\searrow}{\boxed{^{105}\mathrm{Pd}}} \arrow[r] & \cdots \arrow[r] & \overset{\searrow}{\boxed{^{108}\mathrm{Pd}}} \arrow[r] & \overset{\searrow}{\boxed{^{109}\mathrm{Pd}}} \arrow[d] \\
 & & & & \boxed{^{109}\mathrm{Ag}}
\end{tikzcd} \tag{2.97}$$

In other words, the isotope ^{109}Pd is a link between the contribution due to radioactive capture from ^{108}Pd, as well as to the set of linear chain given by the sequence $^{104}\mathrm{Pd} \rightarrow {}^{105}\mathrm{Pd} \rightarrow \cdots \rightarrow {}^{109}\mathrm{Pd}$ and those that have one of the isotopes of it. Thus, if ^{109}Pd is not included, then several contribution paths will be omitted, and therefore the concentration ^{109}Pd will be underestimated. It is possible to corroborate this underestimation from the graph Figure 2.15.

A more detailed study would require to analyze the value of the microscopic cross section of capture for ^{108}Pd , in order to determine if such reaction can be discarded. Additionally, it will be necessary to study the value of the cumulative (or direct) yield for the isotopes belonging to the sequence $^{104}\mathrm{Pd} \rightarrow {}^{105}\mathrm{Pd} \rightarrow \cdots \rightarrow {}^{109}\mathrm{Pd}$, to find their actual contribution to the concentration of ^{109}Ag. Clearly, all these tasks are complex, and they corroborate that the type assignation of a yield type is not straightforward. Nevertheless, the most important conclusion for this example, if that if two burnup codes have similar results for the neutron flux, this not means that the concentration of the fission products also will be similar.

Since the ^{109}Ag fission product is stable, the analysis of the differences between MCNPX and SERPENT was reduced to the decay and transmutation network. However, the analysis of other isotopes as ^{135}Xe is more complex, because the differences between the codes can be related also to the predictor-corrector and sub steps methodology. For such isotope, it is necessary to study the following isobaric chain:

$$\overset{\searrow}{\boxed{^{135}\mathrm{Sb}}} \xrightarrow[1.68\ \mathrm{s}]{\beta^-} \overset{\searrow}{\boxed{^{135}\mathrm{Te}}} \xrightarrow[19\ \mathrm{s}]{\beta^-} \overset{\searrow}{\boxed{^{135}\mathrm{I}}} \xrightarrow[6.57\ \mathrm{h}]{\beta^-} \overset{\searrow}{\boxed{^{135}\mathrm{Xe}}} \xrightarrow[9.14\ \mathrm{h}]{\beta^-} \tag{2.98}$$

According to the isotopes given in Table 2.8, the code MCNPX uses a cumulative yield for ^{135}I because the other two ancestors ^{135}Sb and ^{135}Te does not appear. Based on the half-lives of these ancestors, it is possible to conclude that a use of a cumulative yield for ^{135}I is adequate, as well as the assignation of a direct yield for ^{135}Xe. Unlike the analysis of the ^{109}Ag, in this case the differences between the results between the codes that is shown in Figure 2.18 cannot be related to the decay and transmutation network, because the most important contribution to this isotope are being considered in Table 2.8. Nevertheless, its short half-life suggests that an adequate treatment of this isotope can be accomplished through a sub step methodology.

The MCNPX's version that was used includes a predictor-corrector methodology (Bomboni et. al., 2010), (Fensin, 2008), but it does not use a sub step methodology as Serpent does (Isotalo, 2010). Therefore, the author of the present thesis considers that the differences that are showed in Figure 2.17 and Figure 2.18 can be explained according to these methodologies.

2.9 The constant lambda decay.

The findings describing in all the past sections show that there are several issues related to nuclear data required for a burnup code. Until now such issues have a relationship with the branching rations, the fission yields, and the microscopic cross sections. In terms of the decay lambda constant, there are some topics that were studied by the author of the present thesis that not necessary are fundamental to the development of a burnup code, but that represent interesting facts about the standard treatment of the decay constant in nuclear engineering.

The first of these facts is related with the common assumption of the constancy of the parameter λ in the radioactive decay law. What are the reasons behind this assumption? Are there theoretical foundations that guarantee this? What is the time-region where this can be assumed? The second fact is related with a more mathematical interpretation of the lambda constant through a probability approach. These two topics will be discussed in the following subsections.

2.9.1 A briefly quantum formulation of the decay process.

In Section 1.2.1 was described the discovery of the radioactive law by Rutherford, which was essentially empirical and was obtained through experimental means. The theory that was developed later about the decay process had a probabilistic interpretation. It considered that a radioactive atom can undergo decay with a certain probability, which is independent of past decaying process. Therefore, the variation of the number of such radioactive atoms, $X(t)$ will be represent by the following differential equation:

$$\frac{dX(t)}{dt} = \lambda X(t) \tag{2.99}$$

Whose solution is:

$$X(t) = X(0)e^{-\lambda t} \tag{2.100}$$

This last equation, nevertheless, does not explain the mechanism who produces the decay, and clearly has an experimental base. A more formal interpretation of this phenomena appeared with the developing of the quantum mechanics under the study of nuclear stability and unstable quantum systems. It is considered that the first formal treatment of this problem was carried out by Weisskopf and Wigner (Weisskopf and Wigner, 1930).

The quantum mechanics formulation of the problem consists of finding the probability that a quantum system with an initial state ψ measured at time $t = 0$, will have the same state measured at time t. In other words, the problem is finding the probability that the physical situation does not change in the interval $(0, t)$ (Fonda et. al., 1978). In order to find such probability, it is necessary to use the following functions:

$$\begin{gathered} A(t) = (\psi, \exp(-iHt)\,\psi) \\ P(t) = |A(t)|^2 \end{gathered} \tag{2.101}$$

Where H is the Hamiltonian, and i is the imaginary unit. If there is a set of identical systems, whose number can be represented by $X(t)$, the mathematical formulation for the problem discussed above can be given by:

$$X(t) = X(0)P(t) \tag{2.102}$$

Which is the quantum version of (2.100). The study of the function $P(t)$ allows to conclude that for larger and smaller times, the behavior of such function cannot be considered as exponential. For larger times, Krylov and Fock (Fonda et. al., 1978) found that $A(t)$ fulfills the following Riemann-Lebesgue lemma:

$$\lim_{t \to \infty} A(t) = 0 \tag{2.103}$$

Later Khalfin concluded that $A(t)$ cannot be purely exponential, using a theorem on Fourier transforms which requires that such function must to fulfill:

$$\int_{-\infty}^{+\infty} dt \frac{|\log\,|A(t)||}{1 + t^2} < +\infty \tag{2.104}$$

In order to guarantee the convergence of this integral for the upper limit, it is necessary that:

$$|\log|A(t)|| \overset{t \to \infty}{\sim} Bt^{2-p} \tag{2.105}$$

For a constant B and $p > 1$. Clearly, this implies that:

$$A(t) \overset{t \to \infty}{\sim} \exp(c_1 t^{2-p}) = \exp\,(c_1 t^q)$$

Since $p > 1$, $q = 2 - p < 1$. Using (2.103), it follows that $c_1 < 0$, and therefore:

$$A(t) \overset{t \to \infty}{\sim} \exp(-ct^q)\,, \quad q < 1, c > 0 \tag{2.106}$$

From (2.101) and (2.106) it is clear that:

$$P(t) = |A(t)|^2 \overset{t \to \infty}{\sim} \exp\,(-2ct^q) \tag{2.107}$$

The last equation shows that for a larger time, the behavior of (2.107) cannot be considered purely exponential due to the power q. For the case of very small values, the discussion is more complex, because it is related to the eigenstates of the operator H and with the study of the derivative of $P(t)$ at the origin (Fonda et. al., 1978). Essentially such analysis concludes that:

$$P(t) \underset{t\to 0^+}{>} \exp(-\lambda t) \tag{2.108}$$

Which means that the function $P(t)$ is greater than an exponential behavior for small times. Through equation (2.107) and (2.108) it is possible to conclude that there are derivations from the exponential decay law for very small times and very large times. In other words, in such cases it does not have sense to talk about a lambda constant decay.

2.9.2 Experimental considerations.

There have been experimental studies about the deviations of the exponential law. The earlier researches were carried by Rutherford in 1911, who studied the α decay in ^{222}Rn, which has a half-life of 3.82 days. Rutherford traced the decay curves to 27 half-lives, without finding important deviations (Rutherford, 1911). Later, in 1961 Winter carried out studies for the ^{56}Mn ($T_{1/2} = 2.58\ h$) for β^- decay until 34 half-lives, finding similar results (Winter, 1962). In 1972, Butt and Wilson continued the Rutherford's study of the isotope ^{222}Rn until 40 half-lives obtaining the same conclusions (Butt and Wilson, 1972).

In 1984 Gopych and his colleagues studied the β^- decay process of the ^{116m}In ($T_{1/2} = 54.3\ \mathrm{min}$) for 33 half-lives, without finding any derivations of the decay curve from exponentiality shape (Gopych et. al., 1984). In 1988 Norman and his colleagues carried out the first study for small times, considering the beta decay for the ^{60}Co and ^{56}Mn isotopes, for lower limits of $\leq 10^{-4}T_{1/2}$ and $0.3\,T_{1/2}$ respectively (Norman et. al., 1988), finding a behavior consistent with purely exponential behavior.

In 1988 Gopych and Zalyubovskii concluded that, in order to find notable effects for a nonexponential behavior considering the isotope ^{40}K, it will be necessary to extend the experimental analysis at least to $10^{-38}T_{1/2}$. This is a very important conclusion that was reconsidered by Norman and his colleagues, who performed tests with this isotope, confirming the validity of the decay law at least to $10^{-10}\ T_{1/2}$ (Norman et. al., 1995).

Dykhne and Tkalya proposed a new method for checking the exponentiality of the radioactive law in 1998, obtaining measurements for times longer than 50 $T_{1/2}$ (Dykhne and Tkalya, 1998). Afterwards in 2006, Rothe and his colleagues found the first experimental proof of the turnover in to the nonexperimental decay regime, concluding that the deviations of the power law are related to the cases where the width of the energy distribution is large compared to the released energy (Rothe et. al., 2006). Semkov reported in 2007 the following conditions for the deviations of the radioactive law, which are experimentally unviable (Semkov, 2007):

$$\begin{gathered} -\lambda t < 10^{-14} \\ \lambda t > 71 \end{gathered} \tag{2.109}$$

Based on the above discussion, it is possible to consider that the exponential radioactive law is valid for the times related to practical burnup problems. Finally, it is worth to mentioning that recently Pommé and his colleagues carried out an analysis about the claim that the neutrinos could affect the decay constants through the fifth-force interaction, concluding that such claim is false (Pommé, 2018).

2.9.3 A basic probabilistic study of the lambda constant.

Meghreblian and Holmes proposed an interesting study about the macroscopic cross section through probabilistic theory, which is known as the "as good as new" hypothesis (Meghreblian and Holmes, 1960). Essentially such hypothesis consists of considering that if a neutron does not undergo a collision or a reaction in the interval $[0, x_1]$, then the probability that it undergoes a reaction will be computed starting from x_1, as the neutron is as good as new.

It is possible to formulate a similar hypothesis for the radioactive decay and the lambda constant, considering that if an atom survives a time t without decay, then it is as good as new. From equation (1.3), it is possible to define the probability that an atom survives a time t without decay as:

$$p(t) = e^{-\lambda t} \tag{2.110}$$

In the last equation a frequency probability given by $n(t)/n(0)$ was used. The related probability that an atom will decay before the time t, measure from $t = 0$ is called the distribution function for the first decay and it is given by:

$$F(t) = 1 - p(t) = 1 - e^{-\lambda t} \tag{2.111}$$

From this point the frequency for the first decay before a time t can be obtained from (2.111) through derivation:

$$f(t) = \frac{d}{dt}F(t) = \lambda e^{-\lambda t} \tag{2.112}$$

Strictly speaking, the last three equations were deduced in an informal way, because they started from a frequency definition. Nevertheless, using the "as good as new" hypothesis it is possible to find such relationships in a more formal way, from a mathematical probabilistic theory. Firstly, it is necessary to define a probability that an atom decay before reaching the time t as:

$$P(\mathcal{T} \leq t) \equiv F(t) \tag{2.113}$$

Where $\mathcal{T}$ is a random variable. Using (2.113) the conditional probability of decay in the interval $(t, t + \tau]$ without decaying in the interval $(0, t]$ is given by:

$$P(t < \mathcal{T} \leq t + \tau | \mathcal{T} > t) \tag{2.114}$$

In this point the hypothesis as good as new needs to be used to find $F(t)$ and $f(t)$. The probability on (2.114), under such hypothesis, is equivalent to the probability that an atom will decay before a time τ. In other words:

$$P(t < \mathcal{T} \leq t + \tau | \mathcal{T} > t) = P(\mathcal{T} \leq \tau) \tag{2.115}$$

From the conditional probability, it is possible to set the following equation:

$$P(t < \mathcal{T} \leq t + \tau | \mathcal{T} > t) = \frac{P(t < \mathcal{T} \leq t + \tau, \mathcal{T} > t)}{P(\mathcal{T} > t)} \tag{2.116}$$

The probability $P(t < \mathcal{T} \leq t + \tau, \mathcal{T} > t)$ is the probability that the atom decays in the interval $(t, t + \tau)$, and that it decays after the time t. If such probability is isolated from (2.116), it follows:

$$P(\mathcal{T} > t) P(t < \mathcal{T} \leq t + \tau | \mathcal{T} > t) = P(t < \mathcal{T} \leq t + \tau, \mathcal{T} > t) \tag{2.117}$$

Replacing (2.115) in (2.117):

$$P(\mathcal{T} > t) P(\mathcal{T} \leq \tau) = P(t < \mathcal{T} \leq t + \tau, \mathcal{T} > t) \tag{2.118}$$

On the other hand, $P(t < \mathcal{T} \leq t + \tau, \mathcal{T} > t) = P(t < \mathcal{T} \leq t + \tau)$ due to the as good as new hypothesis. Therefore:

$$P(\mathcal{T} > t) P(\mathcal{T} \leq \tau) = P(t < \mathcal{T} \leq t + \tau) \tag{2.119}$$

Considering that:

$$P(\mathcal{T} > t) P(\mathcal{T} \leq \tau) = [1 - F(t)][F(\tau)]$$

And:

$$P(t < \mathcal{T} \leq t + \tau) = F(t + \tau) - F(t)$$

It is possible to rewrite the equation (2.119) as:

$$[1 - F(t)][F(\tau)] = F(t + \tau) - F(t) \tag{2.120}$$

Dividing (2.120) by τ:

$$[1 - F(t)] \frac{[F(\tau)]}{\tau} = \frac{F(t + \tau) - F(t)}{\tau} \tag{2.121}$$

Computing the limit $\tau \to 0$:

$$\lim_{\tau \to 0} [1 - F(t)] \frac{[F(\tau)]}{\tau} = [1 - F(t)] \lim_{\tau \to 0} \frac{[F(\tau)]}{\tau} = \lim_{\tau \to 0} \frac{F(t + \tau) - F(t)}{\tau} \tag{2.122}$$

It is assumed that the derivative of $F(t)$ exists, then it follows that:

$$[1 - F(t)] \lim_{\tau \to 0} \frac{[F(\tau)]}{\tau} = F'(t) \tag{2.123}$$

The last equation implies that the limit $\lim_{\tau \to 0} \frac{[F(\tau)]}{\tau}$ must to exists and it needs to be a constant, which will be defined as k:

$$[1 - F(t)] k = F'(t)$$

The solution of the last differential equation is given by:$-kt = \ln|1 - F(t)|$, where it is found that:

$$F(t) = 1 - e^{-kt} \tag{2.124}$$

The value of the constant can be found as:

$$\lim_{\tau \to 0} \frac{[F(\tau)]}{\tau} = k = F'(0) \tag{2.125}$$

If follows that $k = \lambda$. On the other hand, it is possible to define the probability distribution $f(t)$ as:

$$\frac{d}{dt} F(t) = f(t) = \lambda e^{-\lambda t} \tag{2.126}$$

Particularly $f(0) = \lambda$. Since $f(t)dt$ can be interpreted as the probability that an atom will decay in an interval given by $[t, t + dt]$, then $f(0) = \lambda dt$ can be interpreted as the probability that an atom will decay in an interval $[0, dt]$. In other words, it is possible to consider the lambda constant as a probability of decay per unit of time. This last interpretation of the lambda constant is very intuitive, but it has a strong relationship with the meaning of the limit in (2.125), which is a probability per unit of time computed around the time equal to zero.

2.10 Conclusions of the Chapter.

In the present chapter three parameters related to burnup problems were discussed: the branching ratio, the microscopic cross section and the fission product yield. Such parameters allow solving more complex structures, which are known as decay and transmutation networks. With small modifications in the Bateman equation, it is possible to include such parameters obtaining a more general solution.

Nevertheless, from the discussion carried in the present chapter, it is possible to conclude that the implementation of the branching ratio and the fission product yield is not a straightforward process, because these tasks are related to the study of the structure or topology of the decay and transmutation network. On the other hand, the implementation of the microscopic cross section is related to the study of the non-linearity of the mass balance equation, as well as to a discretization on time.

The use of the branching ratios requires applying a linearization process to the network, as well as a process of superposition. The first procedure breaks-down the original structure in a set of linear chains, and the second one consists of determining which contribution will be considered to the calculation of the total concentration. The use of the fission product yields requires a study of the isobaric chains, and an analysis of the half-lives of the isotopes that belong the them. Additionally, it is necessary to take into account the capture reaction's sequences that contributes to the final concentration of a given isotope.

Capture reactions, where the fission is included, involve the non-linearity of the differential mass balance equations. In order to overcome this situation, a time-discretization is necessary.

Nevertheless, there are some issues related to the most appropriate way to make such discretization, that originate the predictor corrector and the sub steps methodology.

Since there are several ways in which these parameters can be included in a burnup solver, the comparison between two different burnup codes regarding the concentration of the fission products is a very hard task. As it was possible to conclude in the present chapter, even when two codes have similar outcomes for the neutron flux, this does not imply that the fission product concentration will be equal. In fact, three examples were showed in the present chapter, where percentual error differences were considerable, being in one of them close to 60%.

Finally, it is possible to conclude that the study of the branching ratios, the fission product yield, and the microscopic cross section is necessary to develop a burnup code, and it is possible to make contribution to the solution of the Bateman equations through them.

Chapter 3. Contribution to the linearization process.

As it was described in the previous chapter, it is possible to break a decay and transmutation network in a set of linear chains, solving each one with the Bateman's solution or some of its variations, and finally apply a superposition process to take account of all valid contributions. The first part of this procedure is known as the linearization process, and jointly with the superposition, such methodology is called the "linear chain method".

The common way to build these linear chains is using a Depth-First-Search (DFS) algorithm, which consists in finding every possible path in a network, tracking the decay and transmutation reaction for a set of isotopes until one stable appears or there is no more information to continue. At this point, the algorithm moves backwards searching a branch or an untraveled path, and then the procedure is repeated.

In the present chapter, an alternative new algorithm for building linear chains is developed, which uses a special notation and reduces the problem of finding paths to the problem of ordering a sequence of characters (known as string in computer science). The proposed notation allows summarizing the transmutation network information in a single line, in the same way as Newick's three format does it. Unlike the DFS, the developed algorithm has not a backward routine, but it has a "fill" procedure instead. The last property decreases the computational time when the linear chains are building, and it can be useful with cyclic chains.

3.1 The linearization process.

In order to describe the linearization process, the transmutation and decay network illustrated in Figure 3.1 will be considered. The elements X_i, with $1 \leq i \leq 19$, represent the isotopes, and the factors $b_{i,j}$ are the branching ratios. For this network, the standard linearization process begins with the element X_1, since it has two branches, it is necessary to choose one, and "put on hold" the other. Suppose the isotope X_4 is chosen.

From this point, there are not branches in the following isotopes. Indeed, there is a "segment" or a linear chain with the elements X_4, X_9 and X_{12}. Finally, there are three cases for a final element: 1) it has not progenies, 2) there is not available information about it, 3) it will be defined as the final element. The last case is useful when it has been decided to "truncate" or "cut off" a chain.

Once a final element is reached, the set of isotopes traversing becomes a linear chain. In the example given in Figure 3.1 such linear chain will be:

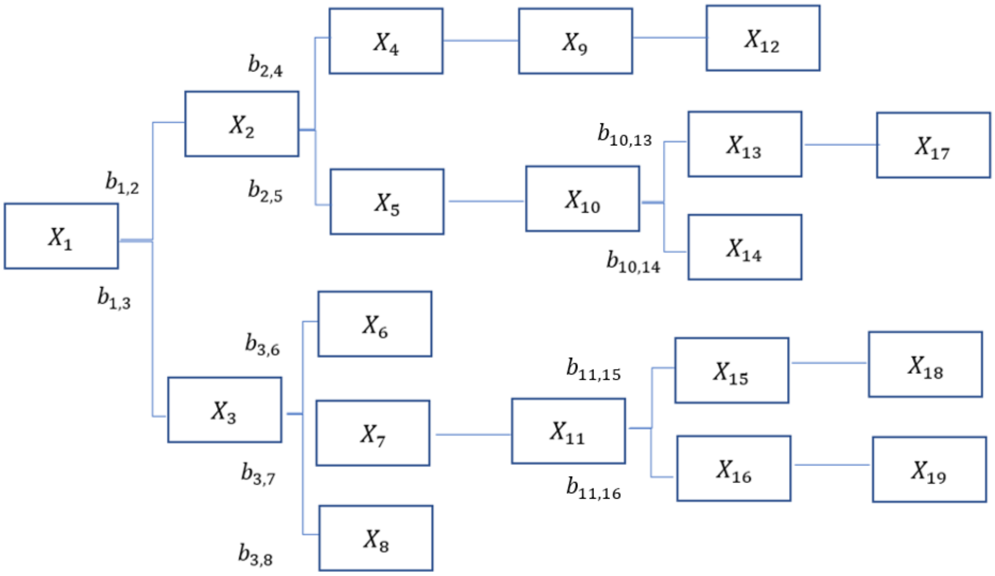

Figure 3. 1. Scheme of a transmutation and decay network.

$$X_1 \rightarrow X_2 \rightarrow X_4 \rightarrow X_9 \rightarrow X_{12} \tag{3.1}$$

After this, it is necessary to travel the linear chain obtained in the opposite direction until the first branch with an element "put on hold" is found. In the case of the example this isotope is: X_5. The branching element X_2, who originates X_5, is taken as a reference point, and it is searched in the last immediate linear chain, i.e. in (3.1). Once this isotope is located, it is necessary to take all the previous elements in that linear chain, which in the present case is only X_1. These elements will form the "base" or the "stem" of the following linear chain. This process is repeated until there are not elements "put on hold". Figure 3.2 shows the outcome of this process, i.e. the linearization of the network of the Figure 3.1.

If the isotopes are called "nodes", and the paths between elements are considered as "edges", it is possible to see the network as a "tree", and the procedure of searching can be considered as a method for traversing tree. In data structure's theory, there are two main algorithms used to traversing trees: the "Depth-First Search" (DFS) and the "Breadth-First Search" (BFS) (Thulasiraman et. al, 2016). The linearization method above discussed is a standard "Depth-First Search" algorithm. As its name suggests, this algorithm first finds the last element (the deepest) in a "steam" or "path", and then search in the unexplored nodes.

The DFS is the most direct way to find linear chains in a decay and transmutation network and it is also very intuitive. It is important to note that in the burnup problems, there are not networks "a priori". Instead, they are built through the information from nuclear data libraries as ENDF or JEFF, and their structure is determined by the special treatment of the code. In the next section it will be discussed how the DFS is used, and it will justify the importance of improving this process of linearization.

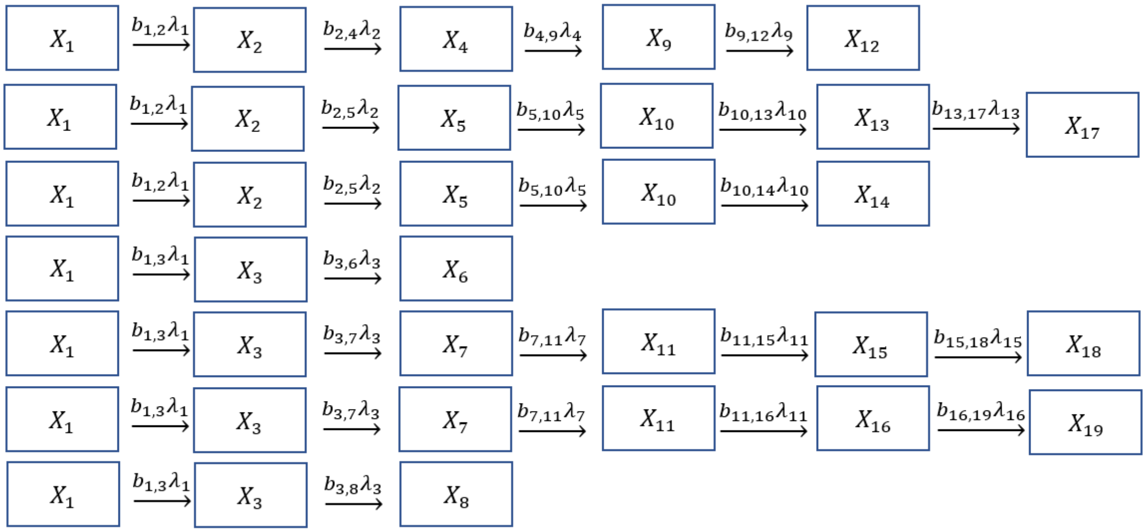

Figure 3. 2. Linearization of the network depicted in Figure 3.1.

3.2 The relevance of linearization.

One of the early references to the linearization process is the England's work about the "fission cross sections" (England, 1962a). In the Part I of his report, England shows how to break a section of the transmutation and decay network related with ^{134}Xe. In the appendices A and B of the same report, England explains how the independent linear chains can be solved using an analytical solution. Nevertheless, England's explanation is merely intuitive and does not offer an explicit methodology for the linearization.

Time after, the burnup code CINDER (England, 1962b) implemented England's procedure, and it is, probably, the first code using the linearization process. Even when the idea behind the linearization process was merely intuitive in the first stages of the development of burnup codes, a formal mathematical proof about linearization was offered by Raykin and Shlyakhter (Rayking and Shlyakhter, 1989). In their work, it is strictly proved that any transmutation network can be broken into independent depletion chains if the burnup equations are linear in concentrations.

After CINDER was developed, other codes who used the linear appeared, among which are ORIGEN (Bell, 1973), WIMS (Leszczynski et al. 2007), DCHAIN (Tasaka, 1976), HELIOS (Studsvik, 2008), BISON-C (Cetnar, 2000), MONTEBURNS (Poston and Trellue, 1999), PENBURN (Manalo, 2008), among others. Unfortunately, in their documentation there is not a detailed treatment about the linear chains building, and even with a fewer exceptions, the DFS algorithm is not mentioned.

This lack of information about the linearization method can be explained if it is considered the computation limitations, and the conservative focus that some codes had. Since the number of linear chains that are used in a code impacts considerably in the execution's time, it is very probable that the linearization process was carried out only once, using a probed scheme that were developed heuristically. In other words, the codes always work with the same set of linear chains that produce adequate results.

Besides, not all the linear chains are significant to compute the nuclides concentration, and they can be neglected. In fact, the main advantage of one of the most powerful linear chain methodologies, known as the Transmutation Trajectory Analysis (TTA), consists in reducing the set of linear chains, considering only those having a meaningful contribution (Cetnar, 2006).

The above reasons seem to justify the little attention that the linearization process have received, nevertheless a new paradigm has emerged recently in the development of nuclear core analysis codes, which consists essentially in carry out several computations "on the fly" during the global core simulation (Ivanov, 2008, Zhang, 2008). Unlike the common and traditional approach where, for example, pre-generated tables for a homogenized node were calculated for these parameters, this new paradigm involves a greater use of computational resources to achieve best estimates. These new procedures are known as "Next Generation Methods" (NGM).

Even when this new approach does not consider the linearization process in burnup calculations, it suggests using more detailed computations and more complex analysis. Therefore, it is possible to extend this paradigm in the transmutation and decay network's study, to include a great number of isotopes, and to track all the possible reactions with fewer approximations. In this way, it will be possible to gain flexibility respecting to the number of linear chains included, as well as its length.

For the above discussion, it is justified the development of alternative linearization's algorithms, that provides with longer linear chains obtained in a shorter time interval and tracking a greater number of elements in the network. Besides, this allows defining the set of linear chains according to different scenarios or problems, and thus avoiding the use of a fixed scheme.

It seems that the last fact is true specially in another branch of nuclear engineering, namely: "nuclear activation calculations", in which the Bateman's equations are also solved with a similar methodology. For example, in the recently developed ACTYS activation code (Chaitanya et al., 2017), the linear chains (also called "pathways") are automatically generated at each time step, adapting its procedure to several cases, and avoiding excessive time consumption.

3.3 The algorithm based in sorting strings elements.

One of the key points about the proposed algorithm is related with the "a priori" information of the isotopes, and their reactions that come from nuclear data libraries, such as ENDF or JEFF. As it was discussed in the past chapter, through these libraries it is possible to know if an isotope is stable, and who are their descendants or "daughters". This information can be managed in data structures like lists or dictionaries, and the DFS's queries are reduced to know if the isotope decays/transmutes and, if the answer is true, what are the products of these transformations.

As it will be discussed in the next section, both queries can be summarized in a single consult about who are the descendants, using two vectors and a special notation. Later, the whole structure of a transmutation and decay network can be condensed in one vector, and from it the linearization process is straightforward. Particularly, the procedure to build linear chains is equivalent to ordering a character's sequence, also called "string" in computer science.

3.3.1 Information's condensation, and indexes generation.

In order to describe the procedure of generation of indexes, the transmutation and decay network showed in the Figure 3.1 will be considered, as well as the empty vectors or lists V_1 and V_2. The first element in the network X_1, is adding to V_1, and a string "P0" is stored in V_2:

$$V_1 = [X_1] \quad ; \qquad V_2 = [\mathrm{P0}] \tag{3.2}$$

After this, it is necessary to define two iterative functions: "Add-indexes" and "Add-elements", which are showed in the Algorithm 3.1 and Algorithm 3.2, respectively. The first one builds the necessary notation to the process of linearization, and the second one stores this information in a special structure. Once the first iteration of "Add-elements" function is carried out, the structures of V_1 and V_2 are the following:

$$V_1 = [X_1, X_2, X_3]; \qquad V_2 = [P0, P0-1, P0-2] \tag{3.3}$$

Note that both vectors must have the same number of elements, and that there is a one-to-one correspondence (a bijective function) between them. Therefore, the information in the i-position in V_2, belongs to the i-element in V_1. The dash character ("-") in the items in V_2, allows identifying the relationship between the elements in V_1. For example, for X_3 the notation "$P0 - 2$" is used, which means that this element is the "second daughter" or the "second descendent" of "$P0$". In the present case, "$P0$" correspond to X_1. Then, for the second iteration of "Add-elements" function, V_1 and V_2 have the following structure:

$$V_1 = [X_1, X_2, X_3, X_4, X_5]; \qquad V_2 = [P0, P0\text{-}1, P0\text{-}2, P0\text{-}1\text{-}1, P0\text{-}1\text{-}2] \tag{3.4}$$

Again, it is possible to track a pathway or a linear chain in V_2 using the dash character. It is only needed to follow the ordering sequence of the strings. From (3.4) following pathways can be extracted:

$$\begin{gathered} P0 \to P0\text{-}1 \to \ P0\text{-}1\text{-}1 \\ P0 \to P0\text{-}1 \to P0\text{-}1\text{-}2 \end{gathered} \tag{3.5}$$

Essentially, the elements in (3.4) have been ordered, following the paths. This procedure of ordering the sequence of strings can be addressed with a more detail in Section 3.3.2. Through the correspondence with V_1, the following two linear chains can be built from the pathways given in (3.5):

$$\begin{gathered} X_1 \to X_2 \to \ X_4 \\ X_1 \to X_2 \to X_5 \end{gathered} \tag{3.6}$$

If (3.6) is compared with the Figure 3.1, it is possible to realize that the algorithm traverses the network one generation at time. In other words, it first finds the descendants for one element, store them, and then repeated this procedure for the following elements in the same generation.

Algorithm 3.1 Add-indexes

```
Input: Two integer numbers, $j$ and $k$, regarded as indexes
Output: A character string stored in $V_2$

STEP 1 Select the item in the $k$- position in $V_2$
STEP 2 Build the character string "$-j$"
STEP 3 Concatenate the character string built in STEP 2, with the item in the $k$-position in $V_2$ i.e.:

        $V_2[k] + " - j"$

STEP 4 Store $V_2[k] + " - j"$ in $V_2$:

        $V_2 \leftarrow V_2[k] + " - j"$
```

Algorithm 3.2 Add-elements

```
Input: The element in the $j$-position in $V_1$
Output: Add elements to $V_1$ and call the "Add-indexes" function

If $V_1[j]$ is different from "End":
    ■ STEP 1 Check the "descendants" for the element in the $j$-position in $V_1$
    ■ STEP 2 Count the number of "descendants", and call that value as "$n$"

    ■ If $n$=0:
        ■ Add "End" to $V_1$ :

                $V_1 \leftarrow "End"$

        ■ Call the Add-indexes function with input's values $j$ and $i$:

                Add-indexes $(j, i)$
    ■ If $n$ is different from 0:
        ■ For $i = 1, \dots, n$:
            ■ Add the $i$-descendent (from a total of $n$) of the $j$- position element in $V_1$:

                    $V_1 \leftarrow i$-descendent of $V_1[j]$

            ■ Call the Add-indexes function with input's values $j$ and $i$:

                    Add-indexes $(j, i)$
```

Therefore, the algorithm is not a DFS's type, because it does not search first in depth, instead it searches first in breadth. In computer sciences, this methodology is known as Breadth-First-Search (BFS) algorithm.

As it was mentioned earlier, the DFS method carries out two queries: 1) if the isotope decays/transmutes and 2) what are the products of these transformations. Nevertheless, if the information about the isotopes is treated before the queries, it is possible to reduce these consultations to only one. Therefore, if there is a "data-base" with the descendants of the isotopes, it is only necessary to consult if the isotope decays/transmutes, and then to select one of the descendants and "put on hold" the rest. The proposed algorithm makes the same consultation, but it does not "put on hold" elements, instead it builds the notation previously mentioned.

For the 10th iteration the structure of the vectors will be the following:

$$V_1 = \ [X_1, X_2, X_3, X_4, X_5, X_6, X_7, X_8, X_9, X_{10}, \mathrm{End}, X_{11}, \mathrm{End}, X_{12}, X_{13}, X_{14}];$$

$$V_2 =$$

[P0, P0-1, P0-2, P0-1-1, P0-1-2, P0-2-1, P0-2-2, P0-2-3, P0-1-1-1, P0-1-2-1, P0-2-1-1, P0-2-2-1, P0-2-3-1, P0-1-1-1-1, P0-1-2-1-1, P0-1-2-1-2]

3.3.2 Sorting based in the dash character

Two paths were found in (3.5) through a sorting of the elements of the vector V_2. It is possible to explain in a more formal way how this procedure it is carry out. An arbitrary item in X_k will be considered, which has the following element in V_2:

$$P0 - \cdots - n - m - j \tag{3.7}$$

In order to search the immediate ancestor and successor elements of X_k, it is just necessary to find in V_2 the next items:

$$P0 - \cdots - n - m, \qquad P0 - \cdots - n - m - j - 1 \tag{3.8}$$

It can be noted, that the dash character and the notation structure provide a useful symbolic identifier. Starting from (3.8), for the ancestor element it is necessary to "back up" one dash character, omitting the j element. For the successor, it is necessary to go forward one "dash" character, and search the "1" element that corresponds to the first successor.

Once these items have been found in V_2, it is possible to find the corresponding elements in V_1 through their positions, from the bijective relationship between V_1 and V_2. Nevertheless, this search method in its standard form can represent a high computational time cost, because the "back up" and "forward" steps in the element (3.7) are string operations. The first one is equivalent to a "split" process and the second one involves a concatenation procedure. Then, if this process is repeated for all the elements, it is possible that the computation time will be greater than the related with DFS algorithm, and therefore it would not exist an interest in this methodology.

Fortunately, there is an alternative form to carry out this search, that represents one of the main advantages for the present contribution, and whose computational time cost is minimal compared with the standard methodology previously mentioned. It turns out that, because of using "dashes", the notation structure has hierarchical characteristics that makes it perfect for sorting techniques, being unnecessary to use strings operations.

The best option for the present case is a sort method based in the lexicographic order, which (speaking in a non-rigorous way) compares ASCII characters codes, that are essentially numbers, and then orders the string elements based in its equivalent values. For example, the "A" character has an ASCII value code of "65", the "4" character has "54", and the dash character has "45". So, if these three elements are compared, from lowest to highest, the result would be: "-", "4" "A".

In computer science (Harzheim,2005) this kind of sorting uses a Cartesian product criterion, which is based in the comparison of two strings, taking element by element. It is possible to exemplify this with a simple case where the strings have two components or characters, i.e. a string with length 2. To build this kind of strings it is necessary to use two sets, A and B, each one having all the characters allowed in the strings structure. Now, the set of all strings of length 2, named S, can be defined as the cartesian product of A and B:

$$S = A \times B = \{(a, b) | a \in A, b \in B\} \tag{3.9}$$

Now for S, the lexicographic order is given as:

$$(a, b) \leq (a', b') \text{ if and only if } a < a' \text{ or } \ (a = a' \text{ and } \ b \leq b') \tag{3.10}$$

This definition can be generalized for strings of an arbitrary length, and it is very suitable for the notation element in (3.7) and (3.8). After the elements in V_2 are sort applying a lexicographic order, the result is as follows:

$$V_2^* = \\ [\mathrm{P0}, \mathrm{P0\text{-}1}, \mathrm{P0\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}1\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}2}, \mathrm{P0\text{-}1\text{-}2\text{-}1}, \mathrm{P0\text{-}1\text{-}2\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}2\text{-}1\text{-}2}, \\ \mathrm{P0\text{-}2}, \mathrm{P0\text{-}2\text{-}1}, \mathrm{P0\text{-}2\text{-}1\text{-}1}, \mathrm{P0\text{-}2\text{-}2}, \mathrm{P0\text{-}2\text{-}2\text{-}1}, \mathrm{P0\text{-}2\text{-}3}, \mathrm{P0\text{-}2\text{-}3\text{-}1}] \tag{3.11}$$

The asterisk superscript indicates that V_2 has been sorted. Finally, several programming languages have native functions for sorting sets, vectors or list whom elements are strings. Therefore, after the condensation process described in Section 3.3.1, it is only necessary to apply a simple instruction to sort the elements in V_2. The linearization process is not yet complete, but as result of the sorting it has been generated a structure in (3.11) with various features will be useful to break-down the network more easily.

3.3.3 Notation's properties and linearization process.

In order to discuss some properties of the notation the first five elements in (3.11) will be considered:

$$\mathrm{P0}, \mathrm{P0\text{-}1}, \mathrm{P0\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}1\text{-}1}, \mathrm{P0\text{-}1\text{-}1\text{-}1\text{-}1} \tag{3.12}$$

From the relationship with V_1, the equivalent elements are:

$$X_1, X_2, X_4, X_9, \mathrm{X}_{12} \tag{3.13}$$

The last sequence is precisely the first linear chain appearing in Figure 3.2. Now, the following four elements will be analyzed

$$\text{P0-1-2, P0-1-2-1, P0-1-2-1-1, P0-1-2-1-2} \tag{3.14}$$

Again, through the relationship between V_1 and V_2 it is possible to obtain the following linear chain:

$$X_5, X_{10}, X_{13}, X_{14} \tag{3.15}$$

In this case, there is not a complete linear chain. Nevertheless, if the first three elements are observed, it is possible to conclude that they belong to the second chain appearing in Figure 3.2. Even more, except[5] for X_{17}, these three elements build the last part of the second linear chain. The two missing items of this linear chain, X_1 and X_2, have already appeared in (3.13). Now, if the second and third linear chains in Figure 3.2 are compared, they are identical except for their last element: the second one ends with X_{13} and the third with X_{14}.

For the following sequence in V_2^*:

$$\text{P0-2, P0-2-1, P0-2-1-1} \tag{3.16}$$

With their corresponding items in V_1:

$$X_3, X_6, \text{End} \tag{3.17}$$

It is possible to conclude that this is a fragment of the fourth linear chain in Figure 3.2. Again, the first element missing, X_1, has already appeared in (3.13), and also in this case the string "End" appears.

From the above discussion it is possible to note that the sequences (3.12), (3.14) and (3.16) were extracted from (3.11), searching the elements where the dashes number in the items in V_2^* decrease. Also, in these extracted elements, the dashes number increases, or remains constant in its final part, as shown in (3.14) where P0-1-2-1-1 and P0-1-2-1-2 have the same number of dashes. Finally, the missing elements in (3.15) and (3.17) are items with a dashes number decreased by at least one unit, compared with the initial elements in (3.14) and (3.16).

The above properties can be summarized as follows:

1) In a linear chain, the dashes number in consecutive elements increases. Therefore, if there are two consecutive elements in V_2^*, with a growing dashes number, then these belong to the same linear chain; even more, they are ordered.

---

[5] This element does not appear, because we make only 10 iterations of the **Add-elements** function.

2) If k −consecutive elements in V_2^* have the same dashes number from the position j, then it is necessary to duplicate $k-1$ times the previous “segment” of linear chain until the position $j-1$.
3) If the dashes number decreases between two consecutive elements, j and $j+1$ in V_2^*, then the element j is the final of the current linear chain, and the $j+1$ item is where the following linear chain begins.

A formal proof of these properties is given in Appendix A. With the first and third properties, it can be concluded that when the dashes number decreases then a linear chain “fragment” has been generated. All the possible linear chain fragments of the example are listed in Table 3.1.

The second property represents an advantage over the DFS. It will be supposed that there are k consecutive elements in V_2 with the same dashes number. This correspond to the following linear chains structure (supposing that the repetition begins in the j position):

$$\begin{cases} X_0 \rightarrow X_1 \rightarrow \cdots \rightarrow X_{j-1} \rightarrow X_1 \\ X_0 \rightarrow X_1 \rightarrow \cdots \rightarrow X_{j-1} \rightarrow X_2 \\ \qquad\qquad\quad \cdots \\ X_0 - X_1 \rightarrow \cdots \rightarrow X_{j-1} \rightarrow X_k \rightarrow \end{cases} \tag{3.18}$$

The arrow in X_k means the possibility of having more elements from the position j, i.e. the possibility that this linear chain continues. The proposed notation allows knowing this kind of repetitive structure easily unlike to the DFS, which for the same scheme would performs k queries to know if the j element has more descendants, and therefore consuming more computational time. Additionally, the kind of structure in (3.18) is common if linear chains with a uniform length are generated.

The result of applying the second property to (3.15) is:

$$X_5 \rightarrow X_{10}, \rightarrow X_{13} \qquad ; \qquad X_5 \rightarrow X_{10} \rightarrow X_{14}$$

3.3.4 Breaking and Structuring Algorithm.

From V_2^*, the process of linearization can be divided in two parts: 1) A breaking-down routine, and 2) a Structuring routine. In the first part, the vector V_2^* is divided in linear chain fragments, while in the last part such fragments will be complete, filling the missing elements and duplicating (if applicable) the linear chain with the same dashes number, as in the case of (318). The algorithms related with these routines are showed in Algorithm 3.3 and Algorithm 3.4. bracket’s notation will be used to represent vector or lists, as well as the colon operator whose use is common in MATLAB o Python Programming.

Table 3. 1. Linear chain segments in V_2^*.

	Linear chain fragment in V_2^*	Equivalent element in V_1
1	[P0, P0-1, P0-1-1, P0-1-1-1, P0-1-1-1-1]	$[X_1, X_2, X_4, X_9, X_{12}]$
2	[P0-1-2, P0-1-2-1, P0-1-2-1-1, P0-1-2-1-2]	$[X_5, X_{10}, X_{13}, X_{14}]$
3	[P0-2, P0-2-1, P0-2-1-1]	$[X_3, X_6,$End$]$
4	[P0-2-2, P0-2-2-1]	$[X_7, X_{11}]$

Algorithm 3.3. Breaking-down routine

Input: A sorted vector/list V_2^*, whose ordering methodology is based in a lexicographic order
Output: A vector/list L, whose elements are all the linear chain fragments in V_2^*, and a set U, whose elements are vectors with the dash's numbers of the elements in L

```
■ $l$ = length of $V_2^*$ (i.e. the number of elements in $V_2^*$)
■ $L = [\,]$, i.e. $L$ is an empty vector.
■ $U = [\,]$, i.e. $U$ is an empty vector.
■ $c = l$
If $l > 2$:
    While $c \neq 0$:
        ■ $S_1 = [\ ]$ (an empty momentary vector)
        ■ $M_1 = [\,]$
        ■ $d = 0$
        ■ $i = 1 + c$
        While $d \geq 0$ and $i < l$:
            ■ $S_1 \leftarrow V_2^*[i]$
            ■ $M_1 \leftarrow$ dashes number of $V_2^*[i]$
            ■ $d$ = dashes number in $V_2^*[i+1]$ −dashes number in $V_2^*[i]$
            ■ $i = i + 1$
        If $i = l$:
            If dashes number in $V_2^*[i] \geq V_2^*[i-1]$:
                ■ $S_1 \leftarrow V_2^*[i]$
                ■ $L \leftarrow S_1$
                ■ $M_1 \leftarrow$ dashes number of $V_2^*[i]$
                ■ $U \leftarrow M_1$
                ■ $c = 0$
            Else:
                ■ $L \leftarrow S_1$
                ■ $L \leftarrow [V_2^*[i]]$
                ■ $U \leftarrow M_1$
                ■ $U \leftarrow$ [dashes number of $V_2^*[i]$]
                ■ $c = 0$
        Else:
            ■ $L \leftarrow S_1$
            ■ $U \leftarrow M_1$
            ■ $c = i - 1$
Else:
    If $l = 1$:
        ■ $L = V_2^*$
        ■ $U$ = [dashes number of $V_2^*[1]$]
    Else:
        "The vector $V_2^*$ is empty"
```

Algorithm 3.4. Structuring algorithm

Input: The vector/list L, which contains all the linear chain fragments of a vector V_2^*, and vector U, whose elements are vectors/lists that contain the dashes number information of the vector/list L
Output: The vector R, which contains all the linear chains of a network

```
■ $l$ = length of $L$, i.e. the number of elements in $L$
■ $m$ = length of $U$
■ $K = [\,]$
■ $K \leftarrow L[1]$
For $i = 2, \dots, l$:
    ■ $a = L[i][1]$
    For $j = 1, \dots, n$ (where $n$ is the length of $K$):
        If $a$ in $K[j]$:
            ■ $p$ = position of $a$ in $K[j]$
            ■ $T = K[j][1{:}p-1]$
            ■ $K \leftarrow T + L[i][1]$
For $i = 1, \dots, m$:
    ■ $S$ = elements that are repeated in $M[i]$
    ■ $O = [\,]$
    ■ $O \leftarrow K[i]$
    ■ $z = 0$
    ■ $g$ = length of $K[i]$ – length of $M[i]$
    For $j = 1, \dots, s$ (where $s$ is the length of $S$):
        ■ $c$= number of times that the element $S[j]$ appears in $M[i]$
        ■ $p$= first position where the element $S[j]$ appears in $M[i]$
        ■ $p_z = p + g - z$
        For $u = 1, \dots, c-1$:
            ■ $R \leftarrow O[-1][1{:}p_z - 1] + O[-1][p_z - 1 + u]$

        If $j \neq s$ (where $l$ is the length of $K[i]$):

            ■ $O \leftarrow O[-1][1{:}p_z - 1] + O[-1][p_z - 1 + c] + O[-1][p_z + c{:}]$
        Else:
            ■ $R \leftarrow O[-1][1{:}p_z - 1] + O[-1][p_z - 1 + c]$

        $z = z + c - 1$
```

3. 4. Additional notation properties and sorting methods.

3.4.1 Comparison with Newick's notation.

In addition to improve the running time, another contribution of the proposed algorithm is its notation, which can provide information of the transmutation and decay networks used in a specific problem. Besides, the algorithm's notation can be used to standardize the set of networks used between two different burnup codes, allowing the interchange of schemes.

For this task it is necessary to using a matrix column with two elements, which will be V_1 and V_2. Later, with the proposed algorithm the set of linear chains can be extracted from these vectors in a straightforward way.

In mathematics and computer science, there are several proposed notations for these types of structures, also called tree structures, being Newick's one of the most popular. This notation uses "comas" and "parentheses" to denote the edges of a tree and their structure. For example, for the scheme of Figure 3.1, the Newick's notation for the $X_1, X_2, \dots, X_8$ elements will be:

$$((X_4, X_5)X_2, (X_6, X_7, X_8)X_3\ )X_1 \tag{3.19}$$

With a right-to-left hierarchy, the parentheses contain the "direct descendants" for an isotope. So, the father or ancestor of a set of elements is on the right, outside of the parenthesis around these items. For the the following generation in the scheme of Figure 3.1, the corresponding Newick's notation will be:

$$\big(\big((X_9)X_4, (X_{10})X_5\big)X_2, (X_6, (X_{11})X_7, X_8)X_3\ \big)X_1 \tag{3.20}$$

And finally, for the complete network:

$$\big(\big(((X_{12})X_9)X_4, (((X_{17})X_{13}, X_{14})X_{10})X_5\big)X_2, (X_6, (((X_{18})X_{15}, (X_{19})X_{16})X_{11})X_7, X_8)X_3\ \big)X_1$$

Also, it is possible to include more information in the Newick's notation using the colon symbol. For example, for the first line it is possible to include the type of decay related with an isotope, or its branch ratio:

$$\big((b_{2,4}{:}\,X_4, b_{2,5}{:}\,X_5\big)b_{1,2}{:}\,X_2, \big(b_{3,8}{:}\,X_6, b_{3,7}{:}\,X_7, b_{3,8}{:}\,X_8\big)b_{1,3}{:}\,X_3\ \big)X_1$$

Clearly, the Newick's notation is a powerful tool to represent decay and transmutation networks. Nevertheless, there are several notable differences between the proposed algorithm's notation and the Newick's. Particularly, Newick's notation have the following advantages over the first one:

1) That notation can summarize all the information of a network in a single line or vector, using the colon symbol, while the proposed algorithm's notation necessary requires at least two vectors: one for the elements, and another for the dashes structure.
2) Read and interpret the Newicks's notation is a relative straightforward process, because it is possible to find an element and his ancestor, or his descendants, in a simpler way due the parentheses and comas. Even when this process can become complex with many elements, it is easier than read and interpret the proposed algorithm's notation, where it is necessary to use the relationship between the vector V_1 and V_2, and the position of the elements in each of these vectors.

On the other hand, its disadvantages the Newick's notation have some disadvantages over the proposed algorithm, which are:

1) Build linear chains starting from Newick's notation will require a search algorithm that deals with the parentheses and the right-to-left hierarchy. This process can be extremely difficult, because that notation generates nested structures, and it is necessary to identify the initial and final parentheses for a set of given elements.

2) The proposed algorithm's notation can provide information about the number of linear chains that will be generated in the linearization process. It is only necessary to build the vector V_2^*, i.e. the vector V_2 under a sorting process, and later apply an elementary counting process. On the other hand, obtain that information from Newick's does not seem to be a direct process, because the counting procedure necessarily requires build the linear chains.

3.4.2 Building the linear chains through the Newick's Notation.

The first advantage of the proposed algorithm over the Newick's notation will be discussed in more detail. For such analysis, it will be considered the structure given in (3.20). Such structure can be traversed from left to right, counting the number of left parentheses, "(", and searching for the first right parentheses, ")". In the present case, this element will be found after the isotope X_9, i.e.: $(((X_9)$.

The next item on the right of this parenthesis will be the "father", or "ancestor", of all the elements grouped in the most nested structure, i.e., the set or elements between the first right parentheses ")", and the last left parentheses, "(".

In our example, that element is X_4:

$$\left(\left((X_9 \qquad \overset{\substack{\text{First right parenthesis}\\ \downarrow}}{)} \qquad \underbrace{X_4}_{\text{Father}}\ , (X_{10})X_5\right) X_2, (X_6, (X_{11})X_7, X_8)X_3\right)$$

There may be more than one element in the parenthesis to the left of X_4, and therefore it is necessary to select one of them, and "put on hold" the rest, in the same way that we did it with the DFS's algorithm. In the present example, the only isotope is X_9, which will be after X_4:

$$X_4 \rightarrow X_9 \qquad (3.21)$$

Then, there are two cases for the next element to the right of X_4 in (3.20): 1) this element is a coma or 2) this element is a right parenthesis.

For the first case, a coma would imply that there are more elements belonging to the same "generation" or "depth" of X_4. Since it is necessary to find the "father" or "ancestor" of X_4 to build the linear chain, the other elements of the same generation will be ignoring, including their parenthesis. To do that, it is necessary to walk the element, while counting the left and right parenthesis that will be found, until the number of right parenthesis will be more than the left's type. This difference between the type of parenthesis would mean that the end of the parenthesis has been found, which contains all the elements belonging to the same generation. For the second case, where there is a right parenthesis after X_4, this would imply that there are not more elements in the same generation. Then, the next item will be to the linear chain.

In the present example there is a coma after X_4, and therefore it is necessary to walk the parenthesis to the right, until the isotope X_5 will be reached. Up to this point the counting is: one left parenthesis (to the left of X_{10}), and two of the right type. During the walk, it is necessary to "store" all the elements that are in the same generation of X_4, and put them on hold.

The next step is to take the element that has have found at the right of X_5, in other words X_2, and adding it at the beginning of the linear chain in (3.21):

$$X_2 \rightarrow X_4 \rightarrow X_9 \tag{3.22}$$

Then, this process is repeated, until there are not more elements "put on hold". The described procedure is like a DFS's algorithm, even when the linear chains are built starting from the end, instead of the beginning. Therefore, using the Newick's notation to build linear chains is equivalent to use a DFS's algorithm.

It was mentioned earlier in this section, that the proposed algorithm's notation can be used to standardize the set of networks used between two different burnup codes. Then, the objective of the detailed analysis that was carried out was to show that even when the Newick's notation has several advantages, the process of linearization starting from it cannot be straightforward, neither direct or fast.

3.4.3 The superposition process

In addition to the advantages described before, the proposed notation may be useful to determine which contribution will be considered in the process the superposition process that was discussed in Section 2.4.3. For example, the first two linear chains of Figure 3.2 will be considered:

$$\begin{gathered} X_1 \rightarrow X_2 \rightarrow X_4 \rightarrow X_9 \rightarrow X_{12} \\ X_1 \rightarrow X_2 \rightarrow X_5 \rightarrow X_{10} \rightarrow X_{13} \rightarrow X_{17} \end{gathered} \tag{3.23}$$

There are two common elements that appear in both lines, X_1 and X_2. The concentration of the isotopes of the two lines can be obtained using the Bateman equation. Nevertheless, when the second linear chain is solved, it is necessary to ignore these elements, otherwise their concentration will be overestimated. The reason for this, is not only that they are repeated, but rather that they have the same "position" in both (first and second place), and they are in the same order; therefore, the sequence represents the same physical phenomenon in the original network.

Now, if it is assumed that there is another linear chain with the following structure (that not appears in Figure 3.2):

$$X_1 \rightarrow X_2 \rightarrow X_3 \rightarrow \cdots \rightarrow X_i \rightarrow X_1 \rightarrow X_2 \rightarrow X_4 \rightarrow X_9 \rightarrow X_{12}$$

In this structure also appears the sequence $X_1 \rightarrow X_2$, with all the rest of elements belonging to the first linear chain in (3.23). Nonetheless, in this case these elements must not be ignored, because even when the items are repeated and they have the same order, they not are in the same position, and therefore this sequence represents a different physical phenomenon. Particularly in this case, the elements have a different "genealogy", because they have distinct "ancestors" in the network. In the first linear chain, X_1 does not have a "father" or an "ancestor", while in the last chain the isotope X_1 was the product or "daughter" of the isotope X_i.

Therefore, the procedure to decide which concentration will be taken into account, requires walking through the set of linear chains, identifying if an element appears for the first time in it, in which case their concentration will be considered. In another case, it is necessary to determine if the sequence of ancestors of the given isotope is also repeated, as well as its sequence of successors.

A detailed procedure of this task is described in the Helios code manual (Studsvik, 2008), using a set of special numbers or indices, also in the Kevin Manilo's master thesis (Manalo, 2008). In the last reference, the author developed an algorithm to "enumerate" the linear chains, using four types of ID values: a zero value for elements whose concentration must not be considered, a negative value when the contribution due to these elements must not be considered, a value of -99 of the last two combined scenarios, and finally, an integer and common values for elements whose concentration must be taken into account.

In any case, there are at least two methods to assign these values: 1) when the linear chains have been built, 2) during the process of linearization.

If set of linear chains already has been generated, the process of assigning values can consume a great amount of time, depending on the number of the elements in the set, because it requires the procedure described in Section 2.4.3. On the other hand, if the values are assigned while the linear chains are built, the process is faster because it is possible to use several properties of the network. For example, it is possible to know when the sequences share the same ancestors, because this often happens with the presence of the branches of the network.

Now, if it is desired to share the linear chains information between two burnup codes, these values should be included. Hence, the proposed algorithm's notation, through V_2^*, can be used to assign these values during the linearization process.

As it was mentioned before, the proposed notation allows to identify the branching points, the elements whose contribution must to be considered, and therefore is very suitable for the "enumerating procedure" of the superposition process, being other of its advantages.

3.4.4 Cyclic chains.

There is a class of networks whose structure contains a repeated pattern that under a linearization process leads to an infinite number of linear chains. For example, the following transmutation scheme can be considered as a cyclic chain:

$$ {}^{A}_{Z}X \xrightarrow{n,\gamma} {}^{A+1}_{Z}X \xrightarrow{n,2n} {}^{A}_{Z}X_1 \rightarrow \cdots \tag{3.24}$$

The first element reappears at the final part; therefore, the pattern is repeated from this point. This case can happen with several heavy elements as ^{235}U and ^{238}U. If this were a linear chain, its length would be infinite, and the problem would be solved by restricting the length of the depth of search. Unfortunately, there may be branches and then the problem becomes complex.

To illustrate this, suppose that in Figure 3.1 the element X_{14} is actually X_2, i.e. it reappears in that node. Note that the element X_2 generates two branches at the beginning of the network, which have X_4 and X_5 as their initial elements. Therefore, if this element reappears in the position of X_{14}, these branches will appear again and again, generating an infinite network.

This type of networks is called “cyclic chains” and have an impact in the Bateman’s equation, since its analytic solution presents problems if there are two elements with the same effective decay constant (that is the case if there are two repeated elements in a network’s pathway).

In Section 1.9 it was discussed a methodology to solve such structures through a general solution, which is based in the Laplace transform, and in Chapter 4 and Chapter 5 such topic will be addressed in a more detailed way. As previously mentioned, the linearization process is applied to a cyclic chain, the result will be an infinity number of linear chains. Nevertheless, if the number of generations considered is restricted (i.e. the depth of search), it is possible to address the problem in a satisfactory way, and the number of linear chains can be finite, even if this number is large. The error due to this approach is acceptable.

This procedure is used in the ALARA (Wilson 1999) activation code, and an earlier reference to it is in DCHAIN2 (Tasaka, 1980). Depending on the considered length, the number of linear chains can grow significantly, and therefore, using a standard DFS algorithm cannot be adequate. In the Section 3.5, the developed algorithm is compared with the DFS in several cases, among which the cyclic chain scenario is notorious. A more extensive discussion about the linear chains will be given in Chapter 4 and Chapter 5.

3.4.5 Computing the number of linear chains

In Section 3.4.1 it was stated that one disadvantage of the Newick’s notation is that there is not a straightforward method that allows knowing the number of linear chains that will be generated. This contrast with the proposed algorithm’s notation, from which this value can be obtained. In the present section, this subject will be treated in more detail.

Based in the properties listed in section 3.3.3, it is possible to count the number of linear chains that will be generated from the vector V_2^*. If the number of dashes in V_2^* is always growing, then the number of linear chains will be equal to one. Otherwise, when the number of dashes between two consecutive elements in V_2^* decreases, then there is a “breaking point”. The total number of “breaking points” in V_2^* will be represented by α.

As was mentioned in Section 3.3.2, the vector V_2^* can be divided into a set of “segments” of linear chains, whose cardinality is equal to α. For a given segment, denoted by i, the number of dashes is increasing or remains constant in certain sections. The element at the end of such segment i will have the maximum number of dashes for this segment, and will be denoted as m_i.

Then, it is necessary to define the function $n_i(k), 1 \leq k \leq m_i$, which will count the number of elements inside the segment i, that have k dashes. Therefore, the total number of linear chains that will be generated from the segment i will be equal to:

$$\sum_{k=1}^{m_i} (n_i(k) - 1) + 1 = \sum_{k=1}^{m_i} n_{i,k} - m_i + 1 \tag{3.25}$$

Finally, the total number of linear chains that will be generated from vector V_2^* will be:

$$\sum_{i=1}^{\alpha} \sum_{k=1}^{m_i} n_{i,k} - m_i + 1 \tag{3.24}$$

3.5 Comparison with DFS

3.5.1 The comparison scheme.

Strictly, a formal comparison between two algorithms must be carried out based on their time and spatial complexity. A theoretical study of this topic involves an analysis of the steps and the structure of the algorithms, using the big O notation (also called Bachmann-Landau notation).

Nonetheless, this kind of study is beyond the scope of the present thesis, and therefore an empirical approach will be used instead. The proposed comparative scheme can be divided in three parts:

1) A network of decay and transmutation of heavy isotopes, in which the fission process is presented, but the fission products do not appear explicitly.
2) A set of networks that have a fission product as their initial element, which in turn are the first elements of isobaric chains.
3) A set of networks that have a fission product as their initial element.

Starting from this scheme, it is possible to build the network for a burnup problem in two different ways, each one using the part 1), and choosing between the part 2) and the part 3). The fission process for a heavy isotope will be simulated supposing that there is a descendant called “Fission Product”, and that it is the final element in a linear chain. In other words, this “Fission Product” will be treated as a stable isotope.

Table I in the Appendix B illustrates this procedure for the case of ^{235}U, considering only the (n, γ), $(n, 2n)$ and the fission reactions. For this scheme, there are 53 linear chains with a length or depth of five elements, and the “Fission Product” isotope (abbreviated as “FP”) also has been added.

For the part 2), as it was described in the past chapter, there is a set of networks whose first element is a fission product, and that it is at the beginning of an isobaric chain. After the linearization process of these elements, the product will be a set A, of linear chains with the following structure:

$$A = \begin{cases} P_1 & X_{11} & \dots & X_{1n} \\ P_2 & X_{21} & \dots & X_{2n} \\ \dots & \dots & \dots & \dots \\ P_n & X_{n1} & \dots & X_{nn} \end{cases} \tag{3.25}$$

Where P_1, P_2,..., P_n are fission products, and the elements X_{ij} represent the descendent j for the fission product i. In (3.25) it has been assumed that linear chains have the same length, but this assumption is not necessary. Then, the set A must be inserted in the linear chains that were generated in the part 1) using the “Fission Product” as a link.

For example, for a linear chain with the following structure $X_1^{part\ 1} \rightarrow X_2^{part\ 1} \rightarrow \cdots \rightarrow X_{n-1}^{part\ 1} \rightarrow$ "Fission Product" (where the superscript indicates that it was generated in the part 1), it is necessary to replace the "Fission Product" element by the set A:

$$X_1^{part\ 1} \rightarrow X_2^{part\ 1} \rightarrow \cdots \rightarrow X_{n-1}^{part\ 1} \rightarrow A$$

Which in turn produces the set A' :

$$A^{'} = \left\{\begin{array}{llllllll} X_1^{part\ 1} & X_2^{part\ 1} & \dots & X_{n-1}^{part\ 1} & P_1 & X_{11} & \dots & X_{1n} \\ X_1^{part\ 1} & X_2^{part\ 1} & \dots & X_{n-1}^{part\ 1} & P_2 & X_{21} & \dots & X_{2n} \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ X_1^{part\ 1} & X_2^{part\ 1} & \dots & X_{n-1}^{part\ 2} & P_n & X_{n1} & \dots & X_{nn} \end{array}\right. \tag{3.26}$$

As it was described above, the linear chains can be built in two separated steps: first for the heavy isotopes, and then for the fission products. Up to this point it may seem enough to consider only the part 2), with the isobaric scheme. Unfortunately, there are some cases where using only this scheme can cause problems. For example, if the isobaric scheme that begins with ^{131}In is used, it will be necessary to use linear chains with a length of at least six elements to simulate the isotope ^{131}Xe. So, it is possible that it will be a special interest in an isotope whose position in an isobaric chain is distant, and therefore the length of linear chains should be large.

As it was explained the past chapter, there is a way to avoid this using the cumulative yield, and redefining the start of an isobaric chain. In the example of ^{131}In, an isobaric chain that begins with ^{131}I or ^{131}Te can be considered, using a cumulative yield for them, reducing the length of the linear chains. Additionally, if an algorithm to track a specific reaction path is used, the isobaric chain scheme can be inadequate because it is possible that isotope in which there is a special interest, will be different from an initial isobaric element. Therefore, for the part 3) it is possible to define a set of networks whose initial element is not the start of an isobaric linear chain.

Before continuing with the description of the comparison it is important to mention that the following tests were run on a 2.6 GHz-3.5 GHz[6] Intel i7, 6700 HQ, under a 64-bit Windows, using the Python 2.7 programming language.

3.5.2 Heavy isotopes comparison.

For this part, the isotopes ^{235}U and ^{238}U will be considered as the initial elements in the transmutation and decay networks. In Table II of the Appendix B, all the isotopes that appear in each network are listed, with the corresponding reactions that were considered. That information was extracted from the ENDF/B-VII.1 Library.

As it was mentioned before, the combination of the reaction $(n, 2n)$ and (n, γ) can lead to the cyclic chain scenario, and since they are included in the present case, the growth of the number

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[6] The processor's specifications have two frequencies: a basic of 2.6 GHz and a turbo maxim of 3.5 GHz; the last one was used in the present work.

of linear chains can be significant. The Figure 3.3 shows the results for ^{235}U, and the Table 3.2 contains the corresponding data. This graph, whose y-axis is in logarithm scale, considers the running time in seconds as a function of the depth or chain's length.

It is possible to observe that, in the first eight values in the graph, the DFS's time is greater than the corresponding time of the proposed algorithm. Even more, if the quotient between them is computed, using the DFS's time in the numerator (these values are in the fifth column in Table 3.2), there are values ranging from 6.5 to 1.35 for the first eight points. Since there is an interest in determine how many times, in average, the proposed algorithm is faster than the DFS, it is convenient to compute the geometric mean in the first eight points. This value is equal to 2.43, and therefore the proposed algorithm is 2.43 times faster than the DFS for the first eight values, corresponding to a depth from 4 to 11 elements.

Due to the proximity of the curves for the chain's length of 11, it is advisable to use the proposed algorithm up to the value of length 10. Nevertheless, if the α reaction is not considered, it is possible to extend the length above this value. Such omission can be considered in practical problems, because it involves the following linear chain:

$$^{235}\text{U} \rightarrow {}^{231}\text{Th} \rightarrow {}^{231}\text{Pa} \rightarrow \text{Fission Product}$$

Fortunately, the decay constant for the α reaction of ^{235}U is $7.038x10^{8}$ years, and therefore the contribution of fission products due to ^{231}Pa can be neglected. Then, for practical cases it is a very good approximation not to consider the α reaction. Nevertheless, in the present study such reaction was considered only to include all the main reactions related with ^{235}U.

It may seem that the use of the proposed algorithm becomes inconvenient and disadvantageous for greater values of chain's length. Nevertheless, it is necessary to analyze two key aspects before to conclude that. First one: until now it has been taken for granted that ordering elements is a fast task, but it was not mentioned that it depends on the number of items to order, their characteristics, and the sorting method used. Particularly, for the present example with a depth of 13 items, the vector V_2, described in Section 3.3.2, has 67823 elements, a considerable big number to be ordered. Additionally, the structure of its two last elements provides an idea of the complexity of the comparisons:

'P0-2-2-1-2-1-2-1-2-1-2-1-1-0', 'P0-2-2-1-2-1-2-1-2-1-2-1-1-1'

Then, the first key point to consider is that the proposed algorithm was programmed using a standard function, and therefore its running time can be improved with a more powerful and complex method of sorting. Currently, there are several sorting algorithms whose implementation often depends on the nature of the elements to order, such as the "Merge sort", the "Quick sort", the "Cocktail sort", among others (Knuth, 1997). These algorithms have different characteristics that not only involve distinct running times, but also have differences related with their stability, their convergence, etc.

Therefore, implementing a more powerful and complex sorting method may require a deeper study of the characteristics of the items to be ordered, which can be extremely complex. But this topic leads to the second key point. It is possible to observe in Table 3.2 and Figure 3.4, that for a chain's length of 11, there are 8544 linear chains. Now, within these linear chains there are nearly 3000 "Fission product" elements, where the set A from (3.25) must be inserted.

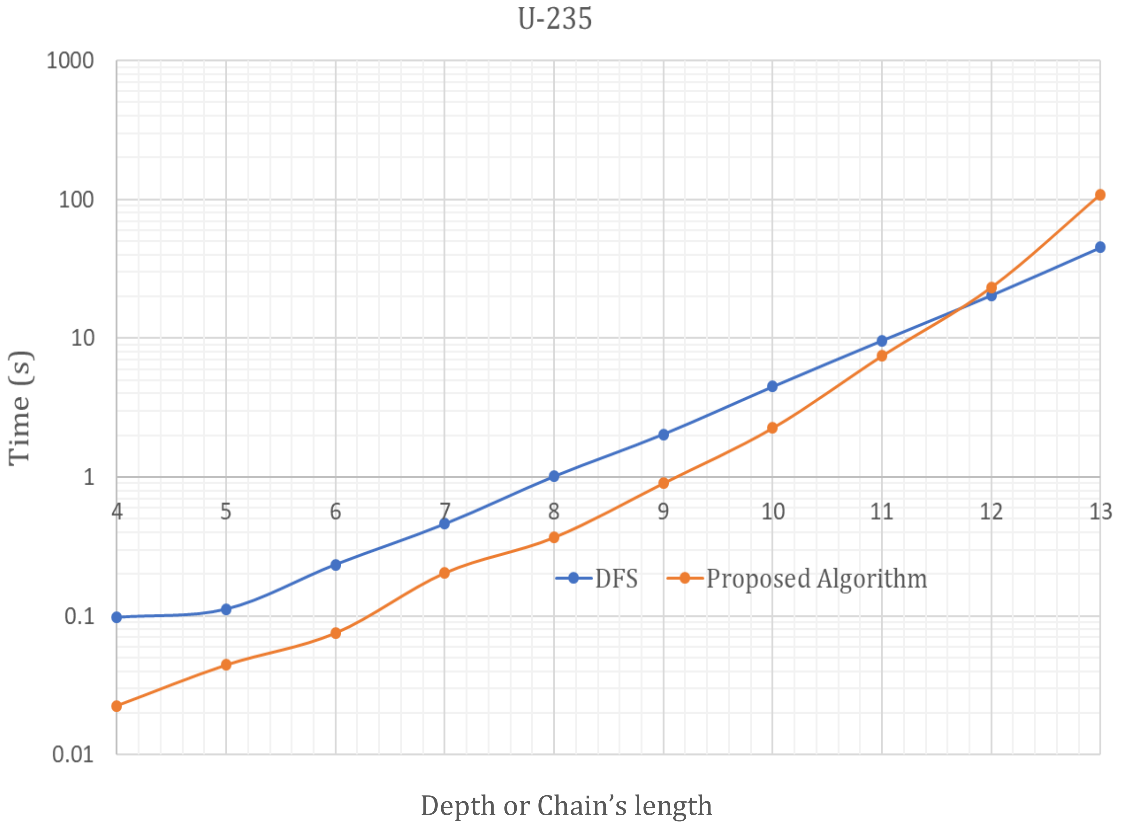

Figure 3. 3. Comparison of Running time vs the chain's length, between the DFS and the proposed algorithm for the ^{235}U.

Table 3. 2. Comparison for the running time between the DFS's and the proposed algorithm for^{235}U .

Length of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
4	51	0.0974	0.0225	4.3157
5	106	0.1118	0.0443	2.5216
6	207	0.2345	0.0752	3.1167
7	430	0.4596	0.2038	2.2547
8	874	1.0119	0.3668	2.7581
9	1855	2.0376	0.9011	2.2612
10	3981	4.4856	2.2515	1.9922
11	8544	9.6323	7.4464	1.2935
12	18698	20.4035	23.225	0.8785
13	40197	45.1705	108.64	0.41575

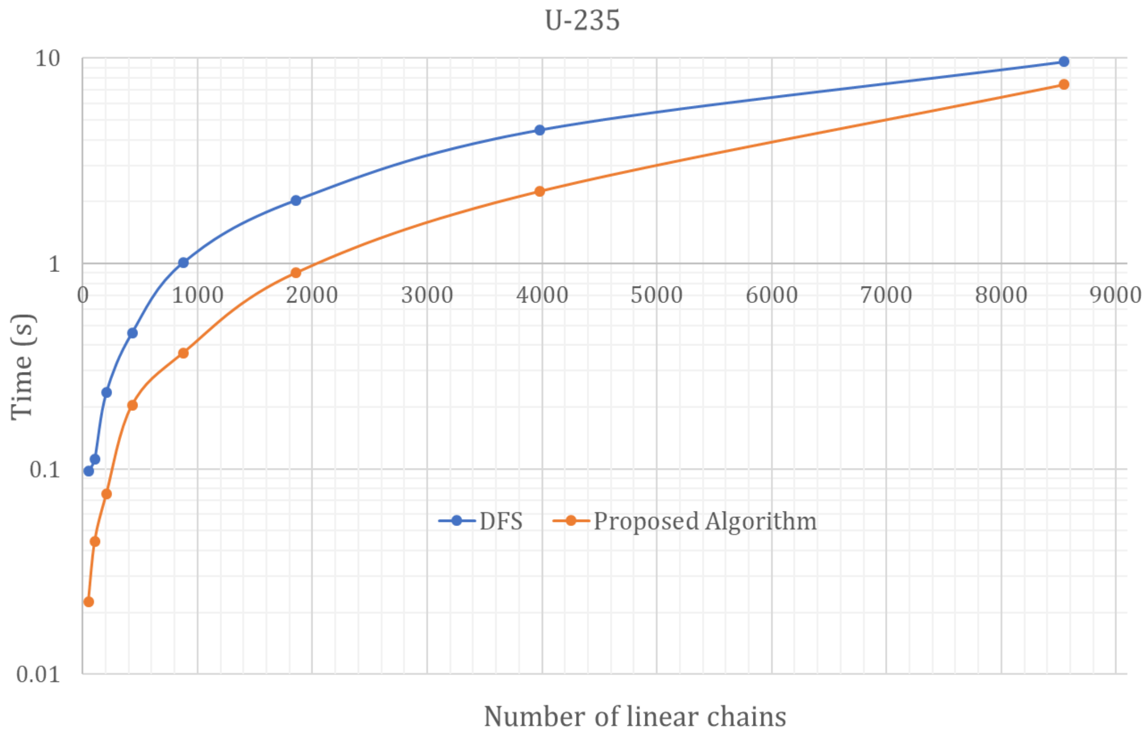

Figure 3. 4. Graph of the running time vs the number of linear chains for the first eight chain's lengths for ^{235}U.

Now, if it is assumed that the set A has 50 linear chains related to fission products (a low value, in fact), then the set A' will have 150,000 linear chains, a large number. Even more, the number of linear chains can be astronomical if it is noted that for a chain's length of 13, there are about 40 mil linear chains for the part 1), and that the set A often has at least 200 linear chains: i.e. the set A' will be at least one million of elements.

Therefore, the proposed algorithm is faster than the DFS in a region of practical applications, and if the user wants a more general case, the algorithm can be improved using a more complex and powerful sorting method. In Figures 3.5-3.6, similar results for ^{238}U were obtained with their corresponding values in the Table 3.3. For this isotope, the geometric means for the first 8 values of the comparison quotient is 2.816. Additionally, to corroborate the results, it was verified that the number of linear chains obtained from the DFS were the same as those obtained with the proposed algorithm. Finally, it is important to discuss an interesting fact related to the way in which the number of linear chains increase. For all the cases, when the depth or length is increased by one, the number of linear chains is increasing by a factor greater than 2, but less than 3. If an exponential function is used as an approximation to fit the dates, following equation is obtained for the ^{235}U case:

$$y = \quad 2.4805e^{0.741x} \tag{3.27}$$

Where y is the number of linear chains, and x represents the depth or chain's length. Figure 3.7 shows the graph of (3.27) compared with the results of ^{235}U.

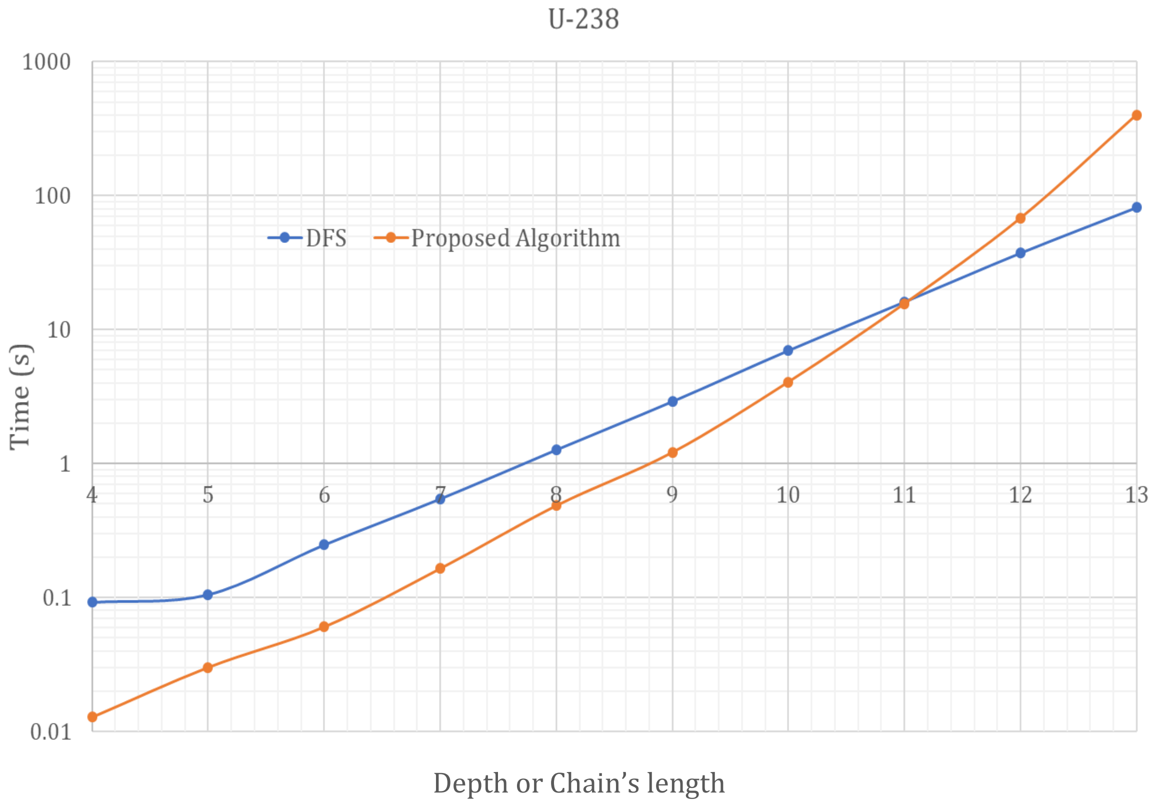

Figure 3. 5. Running time vs the chain's length, comparison between the DFS and the proposed algorithm for ^{238}U.

Table 3. 3. Comparison for the running time between the DFS's and the proposed algorithm for ^{238}U.

Length of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
4	31	0.0926	0.0127	7.2479
5	79	0.1056	0.0301	3.5078
6	199	0.2482	0.0603	4.1144
7	506	0.5472	0.1642	3.3319
8	1197	1.2724	0.4846	2.6256
9	2859	2.9198	1.2104	2.4121
10	6502	7.0203	4.0597	1.7292
11	15124	16.1745	15.5876	1.0376
12	33983	37.4541	67.7777	0.5526
13	78241	81.9111	399.6479	0.2049

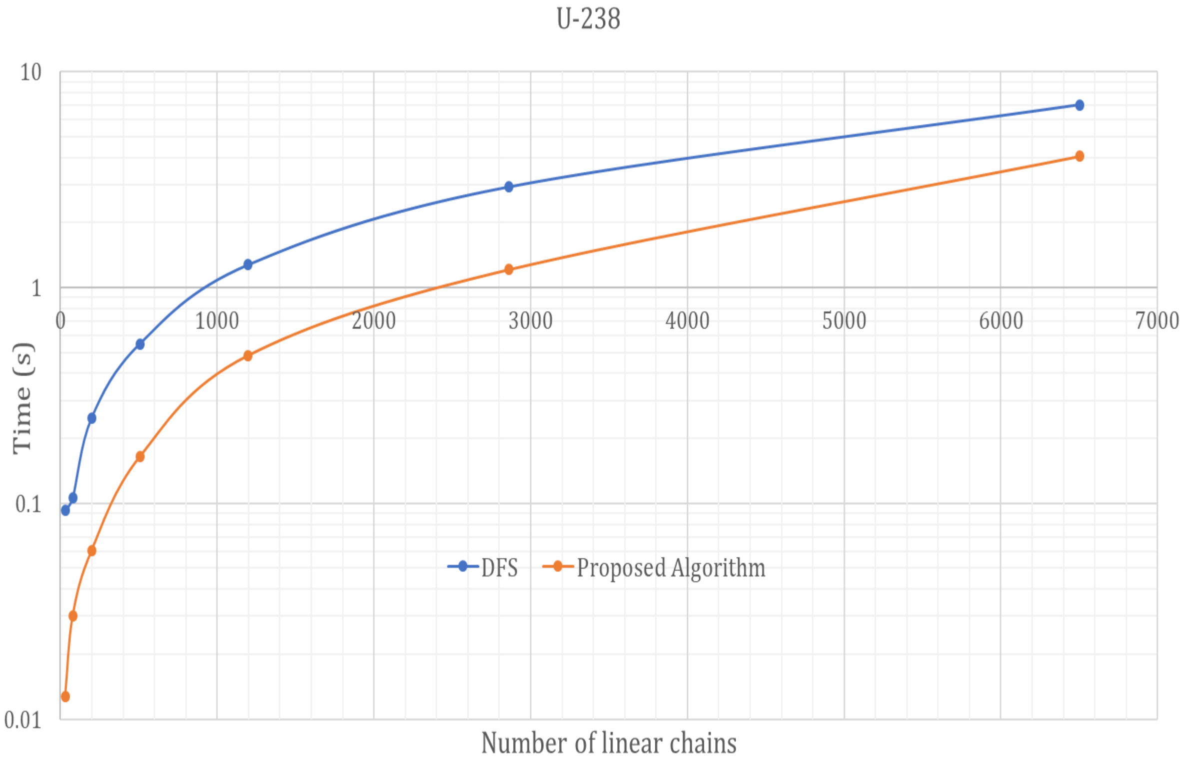

Figure 3. 6. Graph of the running time vs the number of linear chains for the first eight chain's lengths for ^{238}U.

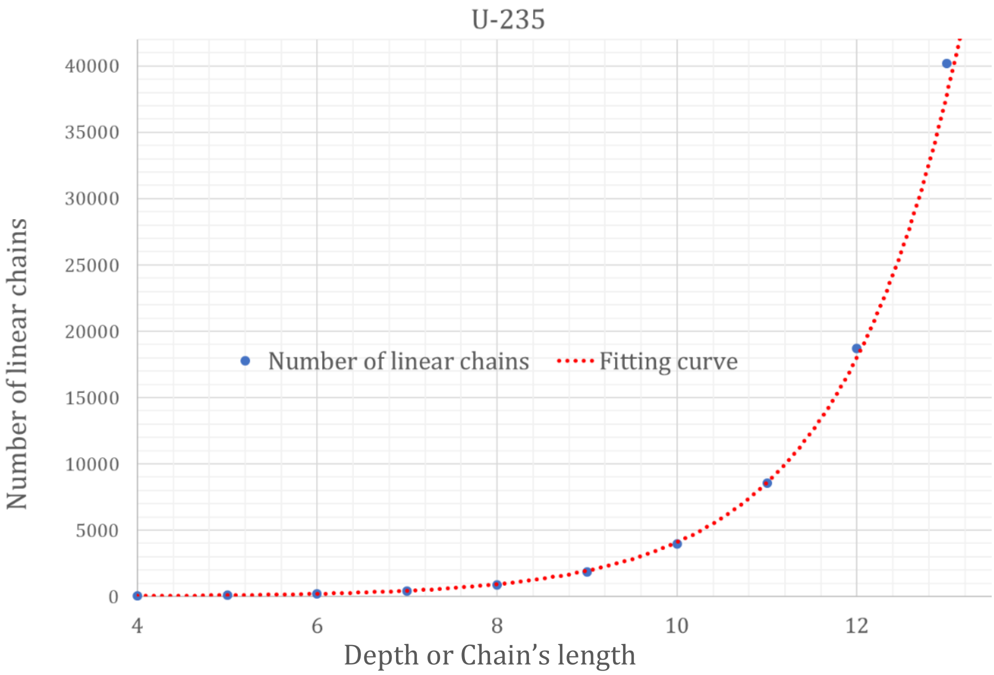

Figure 3. 7. Graph of the number of linear chains vs chain's length, where the fitting curve (computed using an exponential function) is showed

3.5.3 Fission products analysis.

For the present section the fission products ^{87}Se, ^{95}Rb, ^{131}Sb and ^{135}Sb will be considered. In each case, those isotopes will be the initial elements in a transmutation and decay network. In the Table III and Table IV of the Appendix B all the reactions considered for these isotopes, and all the elements that appear in the corresponding networks are listed. This information was mainly extracted from the ENDF/B-VII.1 library, and several schemes were sketched using the online data library in atom.kaeri.ie.kr.

Analysis for ^{87}Se.

According to atom.kaeri, the isotope ^{87}Se does not have possible "parent" nuclides, and it has the beta decay and the beta + neutron reactions. From this element it is possible to build the following isobaric chain:

$$^{87}\text{Se} \xrightarrow{\beta^-} {}^{87}\text{Br} \xrightarrow{\beta^-} {}^{87}\text{Kr} \xrightarrow{\beta^-} {}^{87}\text{Rb} \xrightarrow{\beta^-} {}^{87}\text{Sr} \tag{3.28}$$

The isotope ^{87}Sr is stable, and therefore it is the final element in the isobaric chain. The right arrow after it implies that this element has a transmutation reaction: (n, γ), showed in Table III. Then, ^{87}Se is a fission product at the beginning of an isobaric chain, and this case is related with the part 2) of the proposed comparative scheme of Section 3.5.1. Figure 3.8 shows the running times of the proposed algorithm and the DFS, versus the chain's length. Table 3.4 contains the corresponding data.

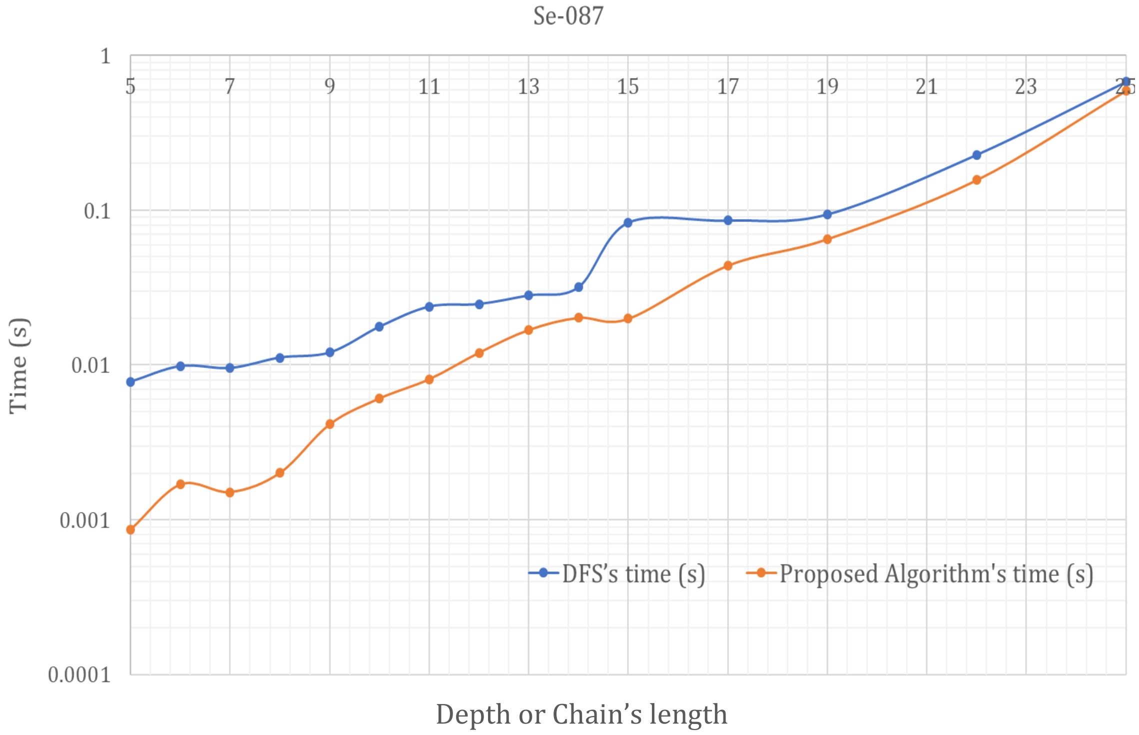

Figure 3. 8. Running time vs the chain's length, comparison between the DFS and the proposed algorithm for ^{87}Se.

Table 3. 4. Comparison for the running time between the DFS's and the proposed algorithm for ^{87}Se.

Length of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
5	2	0.0077	0.0008	8.9767
6	2	0.0098	0.0016	5.8116
7	4	0.0095	0.0015	6.3253
8	6	0.0111	0.0020	5.5479
9	10	0.0120	0.0041	2.9039
10	16	0.0176	0.0060	2.9041
11	16	0.0238	0.0081	2.9434
12	16	0.0247	0.0120	2.0620
13	32	0.0281	0.0168	1.6705
14	32	0.0318	0.0201	1.5793
15	48	0.0832	0.0199	4.1830
17	80	0.0858	0.0438	1.9562
19	80	0.0941	0.0650	1.4464
22	336	0.2285	0.1565	1.4603
25	976	0.6774	0.0008	1.1478

Unlike the heavy isotopes, for the fission products it is necessary to use a greater depth, because there are several connections between isobaric chains. For example, in the present case the isobaric chain in (3.28) is related with the corresponding chain of ^{88}Se. Such connection is showed in (3.29) through the ^{87}Rb and ^{87}Sr isotopes.

$$\begin{array}{ccccccccc} \boxed{^{87}\mathrm{Se}} & \xrightarrow{\beta^-} & \boxed{^{87}\mathrm{Br}} & \xrightarrow{\beta^-} & \boxed{^{87}\mathrm{Kr}} & \xrightarrow{\beta^-} & \boxed{^{87}\mathrm{Rb}} & \xrightarrow{\beta^-} & \boxed{^{87}\mathrm{Sr}} \\ & & & & & & \downarrow (n,\gamma) & & \downarrow (n,\gamma) \\ \boxed{^{88}\mathrm{Se}} & \xrightarrow{\beta^-} & \boxed{^{88}\mathrm{Br}} & \xrightarrow{\beta^-} & \boxed{^{88}\mathrm{Kr}} & \xrightarrow{\beta^-} & \boxed{^{88}\mathrm{Rb}} & \xrightarrow{\beta^-} & \boxed{^{88}\mathrm{Sr}} \\ & & & & & & \downarrow (n,\gamma) & & \downarrow (n,\gamma) \end{array} \tag{3.29}$$

It is important to mention that not all the reactions of transmutation were included for the isotopes. For example, the isotope ^{88}Sr has a cross section related with the reaction $(n, 2n)$, which was not included in the present analysis. The reason for this is that for a thermal reactor the value of its cross section is practically zero. Even when the isotope ^{87}Sr is stable, it is possible to build linear chains from ^{87}Se with a length of 25 elements through the connections between isobaric chains by the reactions (n, γ) and $(n, 2n)$. An example of this linear chain is the following:

$$
\begin{aligned}
&{}^{87}\mathrm{Se} \to {}^{87}\mathrm{Br} \to {}^{87}\mathrm{Kr} \to {}^{87}\mathrm{Rb} \to {}^{87}\mathrm{Sr} \to {}^{88}\mathrm{Sr} \to {}^{89}\mathrm{Sr} \to {}^{89}\mathrm{Y} \to {}^{90}\mathrm{Y} \to {}^{90}\mathrm{Zr} \to {}^{91}\mathrm{Zr} \to \\
&{}^{92}\mathrm{Zr} \to {}^{93}\mathrm{Zr} \to {}^{94}\mathrm{Zr} \to {}^{95}\mathrm{Zr} \to {}^{95}\mathrm{Nb} \to {}^{95}\mathrm{Mo} \to {}^{96}\mathrm{Mo} \to {}^{97}\mathrm{Mo} \to {}^{98}\mathrm{Mo} \to {}^{99}\mathrm{Mo} \to \\
&{}^{99}\mathrm{Tc} \to {}^{100}\mathrm{Tc} \to {}^{100}\mathrm{Mo} \to {}^{101}\mathrm{Mo} \to
\end{aligned}
\tag{3.30}
$$

As indicated in Figure 3.8, the proposed algorithm is faster than the DFS in all the range. The time difference is remarkable for the first points, and at least for the 5-8 lengths, the comparison quotient is greater than 5. Unlike the case of the heavy isotopes, where the number of linear chains is increasing with the depth (due mainly to the cyclic chains), in this case there is a constant value for several depths. This happens in linear chains where the elements have only one descendant, i.e. where there are not branches. In the present case, several isotopes in (3.30) have only one descendant. The results show that the proposed algorithm is the best option to build linear chains in the range of depth from 5 to 25. From the depth values of 5 to 12, the geometric mean of the comparison quotient is 4.1827. Then, on average, the proposed algorithm is 4.1827 times faster than the DFS for that range.

Analysis for ^{95}Rb.

Similar results for the isotope ^{95}Rb are showed in Figure 3.9, and their corresponding data is in Table 3.5. In this case, the proposed algorithm curve intersects and becomes larger than the DFS's for the last values. Nevertheless, as in the case of ^{87}Se, in the first depth values the difference between the algorithms is remarkable. In the range of depth values from 5 to 15, where the proposed algorithm seems be more advantageous, the geometric mean of the comparison quotient is 3.469.

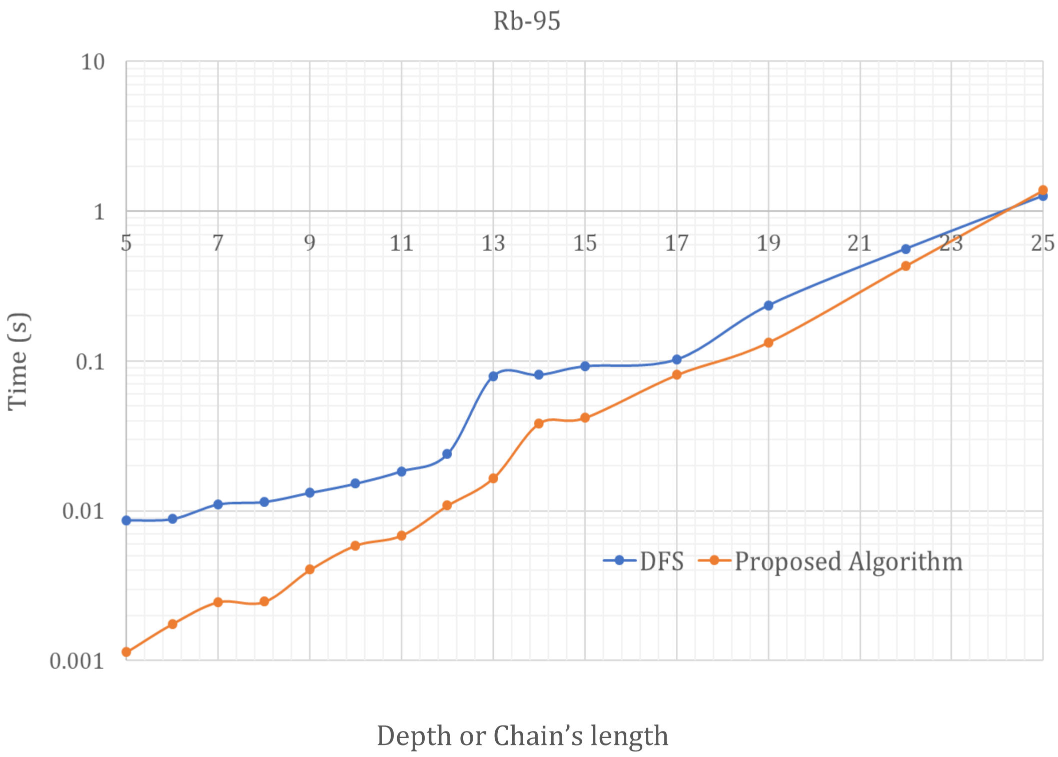

Figure 3. 9. Running time vs the chain's length, comparison between the DFS and the proposed algorithm for ^{95}Rb.

Table 3. 5. Comparison for the running time between the DFS's and the proposed algorithm for ^{95}Rb .

Lenght of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
5	3	0.0086	0.0011	7.5733
6	4	0.0088	0.0017	5.0078
7	4	0.0110	0.0024	4.4819
8	4	0.0114	0.0024	4.6354
9	4	0.0131	0.0040	3.2625
10	12	0.0151	0.0058	2.5838
11	20	0.0183	0.0068	2.6788
12	32	0.0239	0.0108	2.2090
13	40	0.0795	0.0165	4.8081
14	60	0.0807	0.0383	2.1049
15	68	0.0922	0.0418	2.2040
17	124	0.1025	0.0808	1.2692
19	80	0.0941	0.0650	1.4464
22	336	0.2285	0.1565	1.4603
25	976	0.6774	0.5901	1.1478

Analysis for^{131}Sb and ^{135}Sb

Figure 3.10 and Figure 3.11 show similar results for ^{131}Sb and ^{135}Sb, and tables 3.6 and 3.7 contain the corresponding data. The isobaric chains, whose first elements are these isotopes contain the elements ^{131}Xe and ^{135}Xe, respectively, which commonly are important to burnup problems.

Strictly speaking, the isobaric chain related with ^{131}Xe should start with the isotope ^{131}In instead of ^{131}Sb, nevertheless the information of some of the elements belonging to this structure is missing in the ENDF library. As in the past cases, the proposed algorithm is faster than the DFS for all the points in the interval. For the ^{131}Sb, there is a remarkable "gap" between the curves in the range of depth values from 9 to 13. In this interval the geometric mean of the comparison quotient is 3.5282.

There is an interesting fact related with the points where the number of linear chains remains constant. For example, for ^{131}Sb, in the range of depth values from 11 to 17, the number of linear chains is constant and it is equal to 33. For a depth of 11, the DFS's time is 0.109450531 seconds, while that for the next value of depth, 12, the DFS's time is 0.100898 seconds, clearly this last value is less than the first. This seems to make no sense, because the DFS's algorithm makes more queries in the last case. Something similar happens between the depth value of 12 and 13. After verifying that there were no errors in the DFS algorithm, the most plausible answer is related with the computer architecture, and with the function used to measure the elapsed time.

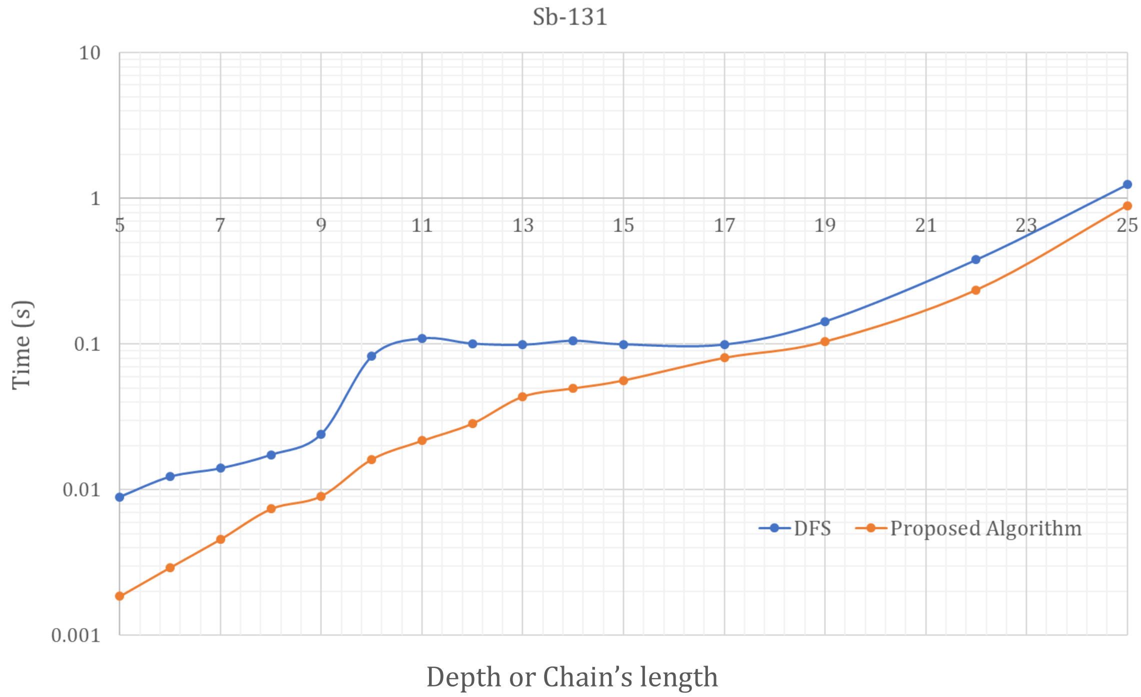

Figure 3. 10. Running time vs the chain's length, comparison between the DFS and the proposed algorithm for^{131}Sb.

Table 3. 6. Comparison for the running time between the DFS's and the proposed algorithm for ^{131}Sb .

Lenght of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
5	3	0.0089	0.0018	4.7996
6	6	0.0122	0.0029	4.1871
7	9	0.0140	0.0045	3.0634
8	12	0.0173	0.0073	2.3481
9	18	0.0241	0.0090	2.6615
10	24	0.0823	0.0161	5.0862
11	33	0.1094	0.0217	5.0401
12	33	0.1008	0.0285	3.5361
13	33	0.0989	0.0436	2.2661
14	33	0.1055	0.0497	2.12203
15	33	0.0997	0.0564	1.76705
17	33	0.0993	0.0806	1.23249
19	63	0.1430	0.1042	1.37164
22	243	0.3794	0.2353	1.61223
25	1023	1.2453	0.8949	1.39145

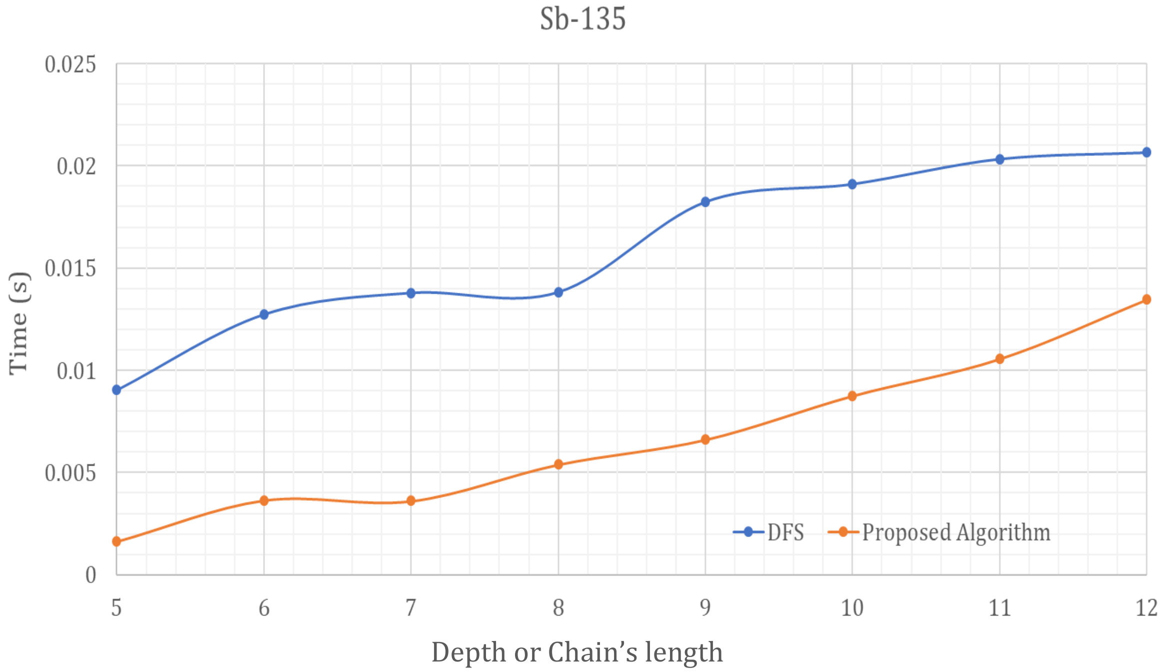

Figure 3. 11. Running time vs the chain's length, comparison between the DFS and the proposed algorithm for ^{135}Sb.

Table 3. 7. Comparison for the running time between the DFS's and the proposed algorithm for ^{135}Sb .

Length of linear chains	Number of linear chains generated	DFS's time (s)	Proposed Algorithm's time (s)	Comparison quotient
5	3	0.0090	0.0016	5.5235
6	4	0.0127	0.003647	3.4936
7	5	0.0137	0.003611	3.8192
8	6	0.0138	0.0053	2.5632
9	6	0.0182	0.0066	2.7597
10	6	0.0191	0.0087	2.1825
11	6	0.0203	0.0105	1.9253
12	6	0.0206	0.0134	1.5338
13	6	0.0265	0.0146	1.8133
14	12	0.0292	0.0162	1.7972
15	12	0.0762	0.0220	3.4565
17	36	0.0919	0.0301	3.0562
19	78	0.1138	0.0809	1.4071
22	300	0.4350	0.21234	2.0486
25	1260	1.5652	1.08811	1.4384

The difference in the running times is of the order of ms. As it was mentioned before, the processor that was used have a variable clock rate and it is possible that there are some variations in time, which were not significant for the other cases, but that for the present problem are comparable with the running time.

On the other hand, in Python programming language there are several options to measure the elapsed time (Martelli et. al, 2005), and when this type of test is carrying, it is important to keep the computer "as quiet" as possible, because the elapsed time can be affected by other process in the computer.

Even when the number of process were maintaining to a minimum when we the simulations were performed; it is possible that there were some tasks that affected our results. It is possible that this problem is only present in the DFS's execution, due the way in which it performs the queries, but it is necessary to carry out a detailed analysis on this subject, which is beyond the scope of the present work.

Additionally, the problem is presented with values of depth where the number of linear chains remains constant, and therefore, where few operations were carried out. This can be observed in the first running times where the number of linear chains is not constant, and the running time is increasing.

3.6 Conclusions of Chapter 3

In this chapter a new algorithm to build linear chains from a transmutation and decay network was developed. This algorithm uses a special notation based in the use of the dash symbol, and it carries out most of the linearization process through a sorting process, using a lexicographical ordering. The proposed algorithm was compared with the standard algorithm used for the linearization process: the depth first search. This analysis was carried out using the algorithms running times, versus the depth or chain's length or the linear chains.

For the comparison process an empirical numerical methodology was used, which is based on dividing the linearization process in two steps: 1) one for the heavy isotopes that present the fission process, 2) for the isotopes that are fission products. For the heavy isotopes step the elements ^{235}U and ^{238}U were considered.

For the ^{235}U case, the proposed algorithm was faster than the DFS's algorithm in the range of the chain's length values of 4 to 11. In that interval, on average, the proposed algorithm is 2.43 times faster than the DFS's. For chain's length values greater than 11, the proposed algorithm seems to be disadvantageous, but it was possible to conclude that this drawback can be solved if a more advanced and complex sorting algorithm is used. Additionally, the length's interval where the proposed algorithm is faster can be extended if the α reaction is ignored.A similar behavior is observed for ^{238}U, where the proposed algorithm is faster than the DFS's being on average 2.816 times faster than the DFS's, in the range of depth values of 4 to 11.

For the fission products step, the isotopes ^{87}Se, ^{95}Rb, ^{131}Sb and ^{135}Sb were considered, with a range of chain's length values of 5 to 25. In all the cases, the proposed algorithm was faster than the DFS's.

In addition to the running time, the proposed algorithm provides a useful notation, which would allow a standardization of the burnup schemes between two different codes. This notation has several properties, within which is it possible to know the number of linear chains before they are built.

From the above facts it is possible to obtain at least three general conclusions. Firstly, the linear chain method can be improved in terms of the execution time, in the part related to the generation of the linear chains. At least to the best know of the author, this topic has not been addressed in literature. Therefore, this is field where it is possible to carry out potential contributions. Since, the mass balance analysis where is determined what isotopes will be considered to compute the final concentration, depends on the linearization method, an improvement in this last is equivalent to an improvement in the mass balance analysis.

Secondly, using a notation to denote a decay and transmutation network allows to share schemes between burnup codes. Such schemes are more useful than only to specific the isotopes that are considered for a given burnup problem. As it was discussed in the past Chapter, in order to make proper comparison between two codes it is necessary to consider the same decay and transmutation network, and therefore the proposed notation will be useful.

Finally, an important conclusion was obtained from the analysis of the grow of the linear chains as function of their length. For the case of heavy isotopes, the number of linear chains grows in an exponential way, due to the presence of the reactions (n,γ) and $(n,2n)$, which jointly produce cyclic chains. In the case of the fission products, it is possible that the number of linear chains remains constant for certain intervals of length.

This last is an important conclusion, because the cyclic chains are present in the most of the burnup problems. Therefore, if the presence of such structures produces a huge amount of linear chains, which impacts on the execution's time, it is necessary to develop a deeper study of this topic, which will be addressed in the following chapter.

Chapter 4. Study of the cyclic chains.

Among the recent research topics related to the linear chain method, it is the development of general solutions, which overcome one of the most remarkable limitations of the Bateman's solution: the presence of repeated elements. As it was described in Chapter 1, one of the conditions in the original Bateman's formulation was the assumption that all the isotopes in a linear chain were different. Such condition guarantees that several subtractions in the denominator do not become zero, and therefore it allows that the solution is well defined.

Nevertheless, as it was showed in the past chapter, transmutation and decay networks known as cyclic chains can lead to the generation of linear chains with repeated elements, and therefore the Bateman´s solution fails. Also, it is possible that singularities appear when two different isotopes in a linear chain have the same removal coefficients, even when this last case is not common. Two approaches were developed in order to solve the difficulty of this singularities. The first one is to introduce small modifications in the repeated removal coefficients, which avoid the divisions by zero. The second approach is to develop more general solutions.

In the present chapter, the decay and transmutations schemes that contains loops or cycles are studied, beginning with their classification and identifying the simplest one. Afterward two approaches to solve them are proposed. Additionally, a comparison of these approaches is carried out, finding that they are easier to implement and provide several advantages over the standard linear chain method.

4.1 Definition of cyclic chains.

As it was discussed in the past chapter, there is a special type of networks where the linearization process fails. Such networks contain structures called "cyclic chains", which appear commonly in several burnup and activation problems.

It seems that the first author who analyzed this type of chains and proposed a procedure to deal with them was Tasaka (Tasaka, 1980), who defined it as "a loop chain where a nuclide transforms certain times and then transforms to the original nuclide". Based in Tasaka's definition, a cyclic chain can be represented in a network diagram through a closed loop between two or more nuclides. For example, in Figure 4.1 there is a cyclic chain given by the succession of isotopes $X_1 \rightarrow X_7 \rightarrow X_9 \rightarrow X_{10} \rightarrow X_{11} \rightarrow X_1$, which have been highlighted in yellow.

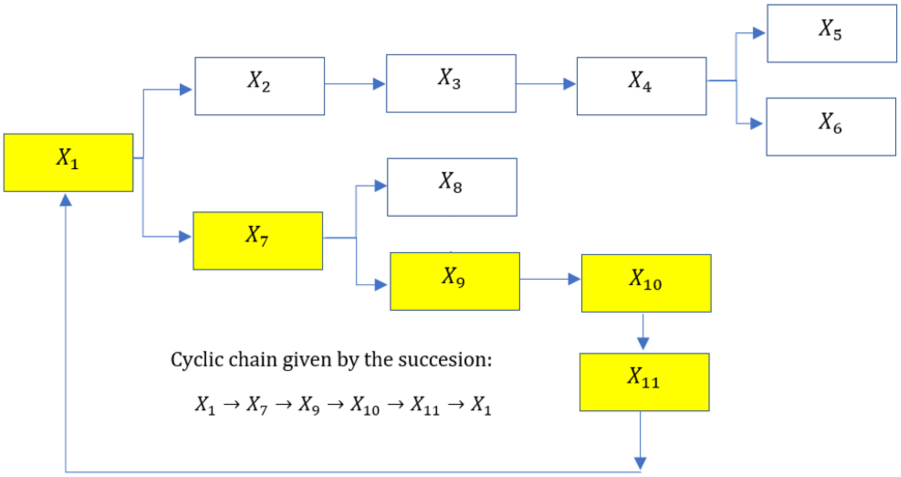

Figure 4. 1 Example of a decay and transmutation network which contains a cyclic chain.

It is not possible to use equations (1.24) (2.5) to solve this loop or cycle due to several reasons. Firstly, the balance equations of the cyclic chain cannot be modelled by a system like the given in (1.12) because the corresponding equation for X_1, in the loop of Figure 4.1, has a gain term, which does not appear in the original mass balance equation given in Chapter 1. In fact, in such chapter was considered that X_1 has not contributions. Therefore, the actual mass balance equation for X_1 based in Figure 4.1 is:

$$\frac{dX_1}{dt} = \underbrace{b_{11,1}\lambda_{11}X_{11}}_{\substack{\text{gain term that does} \\ \text{not appear in (2.5)}}} - \lambda_1 X_1 \tag{4.1}$$

Except for this term, all the other balance equations of the loop can be perfectly described by the equations of the form given in (2.5). Therefore, a first approach to find the concentration of the isotopes that are included in this loop is to assume that $b_{11,1}\lambda_{11}X_{11} \approx 0$, which is reduced to treat these isotopes as if they belong to a linear chain. This approach can be improved in turn, through the use of an artificial succession of isotopes given by

$$X_1 \to X_7 \to X_9 \to X_{10} \to X_{11} \to X_1 \to \cdots \tag{4.2}$$

In other words, the element X_1 has been added at the end of the linear chain in order to "simulate" the contribution $b_{11,1}\lambda_{11}X_{11}$ to X_1. This methodology, where a repeated element has been added to a linear chain is known as "artificial linear chain". Such name suggests that the structures approximate in an artificial way the contributions of the loops. There are notorious issues related to this process, which will be discussed in the following section.

4.2 Implications of simulating the loops as artificial linear chains.

The procedure of building artificial linear chains, adding the isotope X_1 at the end in (4.2), is equivalent to duplicate an infinite number of times all the elements that are originated from the decay and transmutation of this isotope.

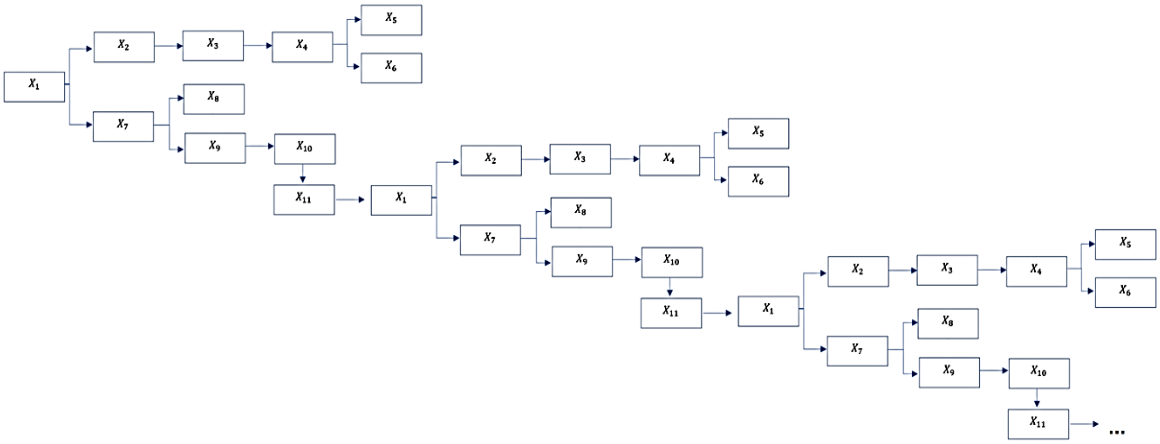

Figure 4. 2. Duplication of the decay and transmutation network of Figure 4.1. This scheme is the result of building an artificial linear chain.

Particularly, since this element is the beginning of the structure in the example showed in Figure 4.1, it implies that all the transmutation and decay network appearing after it, will be repeated as it is represented in Figure 4.2, where only three times of this process of duplication are showed. In other words, the extension of the linear chain in (4.2) will be infinite. Therefore, it is necessary to approximate the linear chain with a finite extension.

The first author that described this process of traveling a cyclic chain, was Wilson, who published a discussion about the duplication in 1999 (Wilson, 1999). In his work, Wilson compared this procedure to the process of converting a connected graph into a n-array tree, and called it as "tree straightening" procedure. It is possible to conclude that, under this procedure, the transmutation and decay network will have a finite number of isotopes that appear an infinite number of times, and it cannot be possible to traverse it in a full way. In other words, the use the linearization process cannot be applied in such structures, because such procedure will not have an end.

Wilson proposed a method to overcome, to some extension, this problem. Essentially it is possible to approximate the solution of the linear chain, limiting the process of duplication to an appropriate number of times, which means that the deep of searching in the linearization process will be limited. Nevertheless, even when this methodology, the number of generated linear chains grows in an exponential way for the cases of cyclic chain, as it was discussed in the past chapter. Therefore, it is important to study alternative ways to approach this problem, in order to provide a more adequate answer.

Before continue, it is necessary to note that the duplication methodology is an approximation due to two facts. Firstly, the mass balance equation does not correspond to the actual structure. And secondly, even when an artificial linear chain is built, their extension need being approximate. Another difficulty related to the structure given in (4.2) is the presence of repeated elements. As it was discussed in Section 1.9, when there are two or more elements with the same effective decay constant, the Bateman's solution cannot be applied, and it is necessary to use a more general solution, as the equation (1.149), or introduce small modifications in the effective lambda constant. These both topics will be discussed in the Chapter 5.

4.3 A first approach to the cyclic chains classification.

As for its complexity, the cyclic chains can be divided into two classes: the pure and the compound. The first one can be defined as a loop, where all the nuclides belong to the same cyclic chain. On the other hand, the compound type is the structure where at least one nuclide belongs to at least two different loops. Among the structures of the first type, there are two additional classes regarding the place where the loop is present: the “first position” and the “on-path position” class. Figure 4.3 shows some examples of this classification.

In such figure the curve arrows represent a sequence of isotopes, and the gray rectangle represent a particular isotope for which there are at least two interpretation. For example, in the first two diagrams of Figure 4.3 it is possible to observe that all, except for the isotope denoted by the gray rectangle, the other elements in the cyclic chain have only one daughter and one father. Therefore, in the pure’s type the gray rectangle represent the initial isotope, or the connector isotope. On the other hand, the gray rectangles in the compound type correspond to isotopes that belong to more than one cyclic chain.

Strictly speaking, the main objective of this study is to model the compound’s type, because in nuclear engineering it appears constantly, in special with heavy isotopes, as is showed in Figure 4.4. In such figure only the reactions that involve isotopes of uranium have been represented, b the fission reaction has been omitted as well as the alpha and beta decay, and all the network related to plutonium and neptunium isotopes.

In other words, even with the complexity of Figure 4.4, the full network is even more complex. A first approach to the treat these cyclic chains consists of finding the solution for the first two cases, the “first position” and the “on path position” and, starting from here, to develop a methodology that allows treating more general cases.

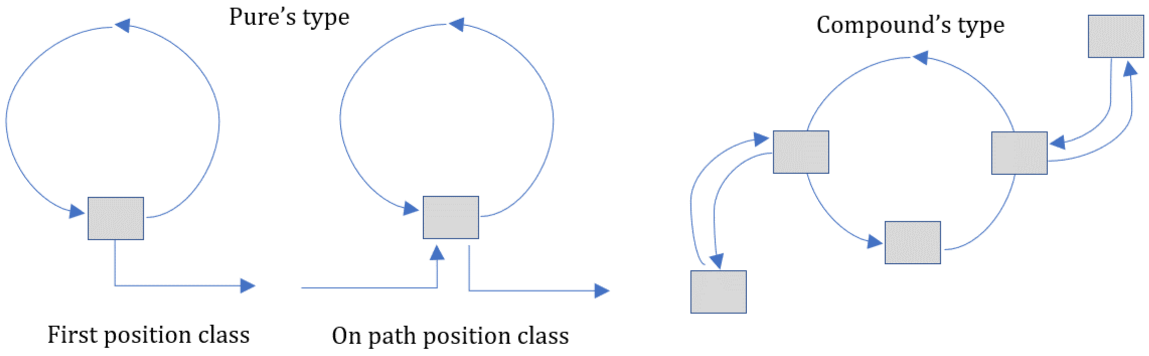

Figure 4. 3. Classification of the cyclic chains. The gray rectangles represent nuclides, and the blue straight arrows and curved arrows indicate succession of isotopes. The extreme left is a pure cyclic chain that appears at the beginning of a network. The next diagram shows a loop that appears on the path of a network. The right structure is a compound cyclic chain where some nuclides belong to more than one loop.

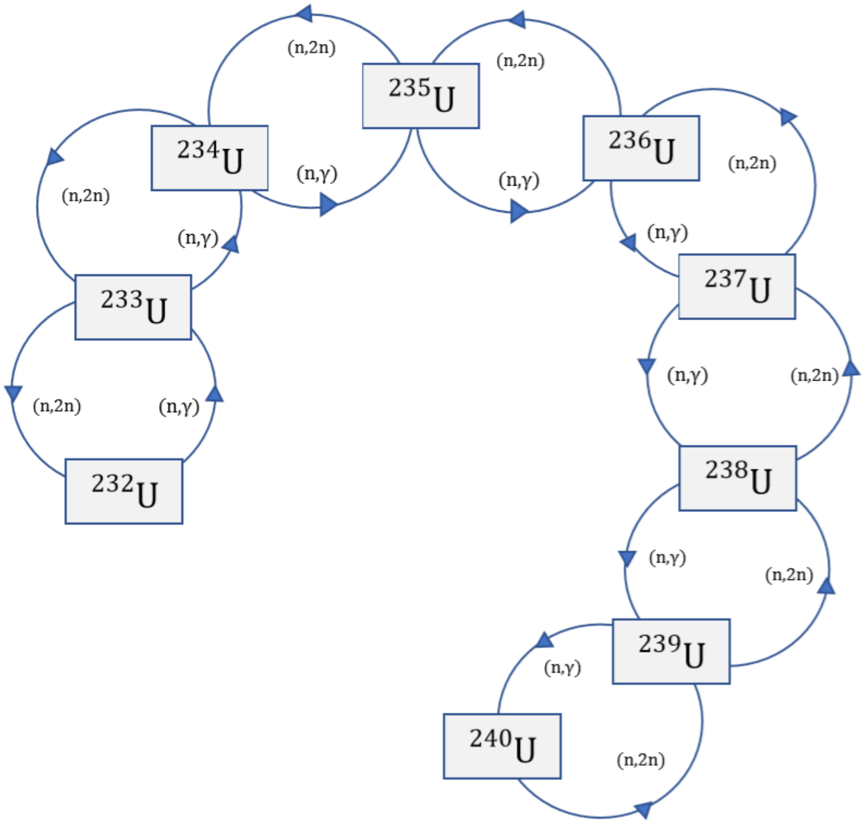

Figure 4. 4. An example of compound cyclic chain related to isotopes of uranium. Such structure is a simplification of the actual case, because the fission reaction, the beta and alpha decay, and the networks related to neptunium and plutonium isotopes have been omitted.

4.4 Analysis for the pure type of first position class.

A pure loop will be denoted as the succession of elements $X_1 \to X_2 \to \cdots \to X_n \to \boxed{X_1}$, where X_1 is the initial isotope. Such element has been boxed at the end to the chain to denote that, starting from this element, the linear chain is repeated. The balance equation to this cyclic chain can be set using the same initial conditions that were used in (1.14)[7]:

$$\begin{cases} \dfrac{dX_1}{dt} = & b_{n,1}\lambda_n X_n - \lambda_1 X_1 \\ \dfrac{dX_2}{dt} = & b_{1,2}\lambda_1 X_1 - \lambda_2 X_2 \\ \quad\vdots & \vdots \\ \dfrac{dX_n}{dt} = & b_{n-1,n}\lambda_{n-1}X_{n-1} - \lambda_n X_n \end{cases} \xrightarrow{\mathcal{L}\{\}} \begin{cases} s\tilde{x}_1 - X_1(0) & = & b_{n,1}\lambda_n\tilde{x}_n - \lambda_1\tilde{x}_1 \\ s\tilde{x}_2 & = & b_{1,2}\lambda_1\tilde{x}_1 - \lambda_2\tilde{x}_2 \\ \vdots & \vdots & \vdots \\ s\tilde{x}_n & = & b_{n-1,n}\lambda_{n-1}\tilde{x}_{n-1} - \lambda_n\tilde{x}_n \end{cases}$$

If the transform terms are isolated it follows that:

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[7] By simplicity, the effective removal coefficient and branch ratios will be denoted as λ and b, respectively

$$\rightarrow \begin{cases} \tilde{x}_1 & = & \dfrac{X_1(0) + b_{n,1}\lambda_n\tilde{x}_n}{s+\lambda_1} \\ \tilde{x}_2 & = & \dfrac{b_{1,2}\lambda_1\tilde{x}_1}{s+\lambda_2} \\ \vdots & \vdots & \vdots \\ \tilde{x}_n & = & \dfrac{b_{n-1,n}\lambda_{n-1}\tilde{x}_{n-1}}{s+\lambda_n} \end{cases} \tag{4.3}$$

After multiple substitutions it is possible to obtain an equation for $\tilde{x}_n$:

$$\tilde{x}_n = \left[\frac{\alpha(n-1)X_1(0)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n) - \alpha(n)}\right], \tag{4.4}$$

Where:

$$\alpha(n) = \prod_{j=1}^{n} b_{j,j+1}\lambda_j \tag{4.5}$$

Equations (4.4) is significative if it is compared with the equivalent expression in (1.15). If presence of the branch ratios is omitted in this last expression, the equation for $\tilde{x}_n$ is identical to (4.4), except for the term $\alpha(n)$ that appears in the denominator. Denoting the solution that corresponds to the linear and the cyclic chain as X_n^L and X_n respectively, it is possible to show that:

$$X_n \rightarrow X_n^L \text{ when } \alpha(n) \approx 0 \tag{4.6}$$

Now, considering the definition of $\alpha(n)$, it is possible to conclude that the situation given in (4.6) is very realistic, because the order of magnitude of the product of the effective removal coefficients is very small. Then, this justify the approximation of the cyclic chain with the linear chain case.

4.4.1 Solution for the case $k = n$

The exact solution of the equation (4.4) can be found if it is assumed that:

$$\prod_{j=1}^{n} b_{j,j+1}\lambda_j = \alpha(n) < |(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)| \tag{4.7}$$

Since s is a complex variable in the Laplace transform space, the implications of this inequality are not straightforward, nevertheless, for the moment it will suppose it. Equation (4.4) can be written as follows:

$$\tilde{x}_n = \frac{\alpha(n-1)X_1(0)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)}\left[\frac{1}{1 - \dfrac{\alpha(n)}{(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n)}}\right] \tag{4.8}$$

Considering (4.7), it is possible to use the following series expansion to the right side of the equation (4.8):

$$\frac{1}{1-z}=\sum_{j=0}^{\infty} z^j, \quad |z|<1, \tag{4.9}$$

Therefore:

$$\left[\frac{1}{1-\dfrac{\alpha(n)}{(s+\lambda_1)(s+\lambda_2)\ldots(s+\lambda_n)}}\right]=\sum_{j=0}^{\infty}\left[\frac{1}{(s+\lambda_1)(s+\lambda_2)\ldots(s+\lambda_n)}\right]^j(\alpha(n))^j \tag{4.10}$$

Through (4.10) it is possible to rewrite (4.8) as:

$$\tilde{x}_n=\frac{X_1(0)}{\lambda_n b_{n,1}}\sum_{j=0}^{\infty}\left[\frac{1}{(s+\lambda_1)(s+\lambda_2)\ldots(s+\lambda_n)}\right]^{j+1}(\alpha(n))^{j+1} \tag{4.11}$$

The element inside the brackets in (4.11) can be rewritten using (1.18) and (1.19):

$$=\frac{X_1(0)}{\lambda_n b_{n,1}}\sum_{j=0}^{\infty}\left[\sum_{i=1}^{n}\frac{a_i}{s+\lambda_i}\right]^{j+1}(\alpha(n))^{j+1} \tag{4.12}$$

If it is supposed that the inverse Laplace function of $\tilde{x}_n$ exists, then it is possible to write:

$$X_n=\mathcal{L}^{-1}\{\tilde{x}_n\}=\frac{X_1(0)}{\lambda_n b_{n,1}}\mathcal{L}^{-1}\left\{\sum_{j=0}^{\infty}\left[\sum_{i=1}^{n}\frac{a_i}{s+\lambda_i}\right]^{j+1}(\alpha(n))^{j+1}\right\}$$

In this last step it is necessary to make another supposition, considering that the series in curly brackets is absolutely convergent. Therefore, the inverse Laplace operator can be introduced into the summation:

$$X_n=\frac{X_1(0)}{\lambda_n b_{n,1}}\sum_{j=0}^{\infty}\left[\mathcal{L}^{-1}\left\{\sum_{i=1}^{n}\frac{a_i}{s+\lambda_i}\right\}\right]^{j+1}(\alpha(n))^{j+1} \tag{4.13}$$

From (1.24) it follows that:

$$\mathcal{L}^{-1}\left\{\sum_{i=1}^{n}\frac{a_i}{s+\lambda_i}\right\}=\frac{X_n^L}{\alpha(n-1)X_1(0)} \tag{4.14}$$

Using the last step, it is possible to write (4.13) as:

$$X_n=\frac{X_1(0)}{\lambda_n b_{n,1}}\sum_{j=0}^{\infty}\left[\frac{b_{n,1}\lambda_n X_n^L}{X_1(0)}\right]^{j+1} \tag{4.15}$$

Where X_n^L is the solution of the term n in a linear chain. Since that:

$$\frac{b_{n,1}\lambda_n X_n^L}{X_1(0)} < 1$$

It follows that:

$$X_n = \frac{X_1(0)}{\lambda_n b_{n,1}} \frac{X_n^L b_{n,1}\lambda_n}{X_1(0)} \sum_{j=0}^{\infty} \left[\frac{b_{n,1}\lambda_n X_n^L}{X_1(0)}\right]^j = X_n^L \frac{1}{1 - \frac{b_{n,1}\lambda_n X_n^L}{X_1(0)}}$$

Therefore:

$$X_n = X_n^L \frac{X_1(0)}{X_1(0) - b_{n,1}\lambda_n X_n^L} \tag{4.16}$$

4.4.2 Analysis of the Assumptions of the pure cyclic solution.

Three assumptions were made in the past section:

1) The inequality in (4.7) is true.
2) The inverse Laplace transform in (4.12) exists.
3) The series in (4.13) is absolutely convergent.

Even when the study of these three assumptions is beyond the scope of this thesis, it is possible to discuss, in general terms, the theory behind them.

As Clayton and his colleagues pointed out (Clayton et. al., 1961), through the theory of functions, it is known that very small values of t correspond to very large values of s, and vice versa. Then, equation (4.7) represents a condition for both s and t. Nevertheless, it is necessary to be very careful with this kind of sentences regarding the values of s, because it is a complex variable, and because there is not a total order in $\mathbb{C}$, it is necessary to study this condition in a deepest way.

Regarding the second assumption, it is possible to proof that the inverse Laplace exists using the integral Fourier-Mellin formula (Brown and Churchill, 2004) which was described in (1.25). Nevertheless, this proof is far to be easy. Even more, in most practical cases in engineering, the existence of the inverse Laplace transform is taken for granted, and it is searching for a correct representation of it in tables or using the convolution theorem.

The third condition about the absolute convergence of the series is related again with the study of the variable s. In order to analyze the proposed solution in a formal way it is necessary to investigate about these topics, but they will be part of a future work. Finally, equation (4.16) is very useful, because it allows comparing the solution of a cyclic chain, and the solution of a linear chain, in fact if $b_{n,1}\lambda_n \approx 0$, then

$$X_1(0) - b_{n,1}\lambda_n X_n^L \approx X_1(0) \quad \text{and} \quad X_n \approx X_n^L \tag{4.17}$$

The condition $b_{n,1}\lambda_n \approx 0$ is less restrictive that condition (4.6). If this term is very small, then the contribution of the isotope X_n to the isotope X_1 is neglected.

4.4.3 Lack of symmetry and convolution approach.

A remarkable disadvantage of the system in (4.3) is the lack of symmetry between the equations of the terms corresponding to the isotope k, with $2 \leq k \leq n-1$, and the isotope n. Due to this lack of symmetry, equation (4.17) cannot be extended to the other isotopes of another index k. In other words:

$$X_k \neq X_k^L \frac{X_1(0)}{X_1(0) - b_{k,k+1}\lambda_k X_k^L} \tag{4.18}$$

In this sense the index n has a special meaning, because it denotes the element in the position before the term that has been considered as the first. All the other elements, between this isotope and the first one, are denoted by the k index. As a first approach, it is possible to use (4.16) to compute the solution for the isotope X_{n-1}. According to the system in (4.3):

$$\tilde{x}_{n-1} = \frac{\tilde{x}_n(s+\lambda_n)}{b_{n-1,n}\lambda_{n-1}} = \frac{1}{b_{n-1,n}\lambda_{n-1}}(s\tilde{x}_n + \lambda_n\tilde{x}_n) \tag{4.19}$$

From this equation it is possible to compute $\mathcal{L}^{-1}\{s\tilde{x}_n\}$. Considering:

$$D(s) = s, \qquad F(s) = \tilde{x}_n$$

And:

$$d(t) = \mathcal{L}^{-1}\{D(s)\} = \delta'(t), \quad f(t) = \mathcal{L}^{-1}\big(F(s)\big) = X_n$$

Where δ is the Dirac delta function. Through the convolution theorem (Zill, 2009):

$$\mathcal{L}^{-1}\{s\tilde{x}_n\} = \int_0^t \delta'(t-u)X_n(u)du = X_n'(t) = \frac{dX_n}{dt} \tag{4.20}$$

Where in the last part it has been used a property of δ. Using (4.16) and (4.20) it is possible to obtain:

$$X_{n-1} = \mathcal{L}^{-1}\{\tilde{x}_{n-1}\} = \frac{1}{b_{n-1,n}\lambda_{n-1}}(\mathcal{L}^{-1}\{s\tilde{x}_n\} + \lambda_n\mathcal{L}^{-1}\{\tilde{x}_n\})$$

$$X_{n-1} = \frac{1}{b_{n-1,n}\lambda_{n-1}}\left(\frac{dX_n}{dt} + \lambda_n X_n\right) \tag{4.21}$$

It is possible to note that the last equation is equivalent to (1.31), which was deduced through the backward method that was described in Section 1.6. In fact, if the convolution theorem is applied again to (4.22), the result will be equal to the backward solution for $n-2$. Therefore, the backward Bateman solution given in (1.55) can be used to compute the concentration of the other isotopes in the structure (4.3). Unfortunately, such equation involves high-order derivatives of X_n, which need to be computed from (4.16). Such task is hard and it is necessary to develop a general formula for them, which will be carried out in the following section.

4.4.4 Formula for the derivatives.

When the backward formulation is used, it requires to compute the following high order derivatives:

$$\frac{d^i X_n}{dt^i}, \quad 1 \leq i \leq n \tag{4.22}$$

It is convenient to rewrite equation (4.16) for X_n as follows:

$$X_n = \frac{X_1(0)}{b_{n,1}\lambda_n} \frac{b_{n,1}\lambda_n X_n^L}{X_1(0) - b_{n,1}\lambda_n X_n^L} \tag{4.23}$$

Considering the following functions:

$$U(t) = \ b_{n,1}\lambda_n X_n^L(t), \ \ V(t) = \frac{U(t)}{a - U(t)} \tag{4.24}$$

The problem to find a formula for the derivatives of (4.22) is equivalent to find a formula for the derivatives of $V(t)$ and for the derivatives of $U(t)$, defined in (4.24). It is possible to compute the first four derivatives of $V(t)$ in a straightforward way:

$$\begin{gathered} V' = \frac{aU'}{(a-U)^2}, \quad V'' = \frac{aU''}{(a-U)^2} + \frac{2a(U')}{(a-U)^3}, \\ V^{(3)} = \frac{aU^{(3)}}{(a-U)^2} + \frac{6aU''U'}{(a-U)^3} + \frac{6a(U')^3}{(a-U)^4} \end{gathered} \tag{4.25}$$

$$V^{(4)} = \frac{aU^{(4)}}{(a-U)^2} + \frac{8aU^{(3)}U'}{(a-U)^3} + \frac{6a(U'')^2}{(a-U)^3} + \frac{36U''(U')^2}{(a-U)^4} + \frac{24(U')^4}{(a-U)^5} \tag{4.26}$$

At first sight, it seems difficult to find a general formula because the coefficients and the number of terms does not exhibit a clear relationship. Nevertheless, after careful observation, it was found a pattern related to the derivatives of U. In fact, it is possible to observe a tendency between the sum of the orders of the derivatives of U and the order of the derivative of V.

For example, for the case of $V^{(3)}$, there are three terms. In the first one a derivative $U^{(3)}$ appears, in the second term, the functions U' and U'' are present, which correspond to the orders of 1 and 2 respectively. If such orders are added, the outcome is 3. In the last term, the term that appears is $(U')^3$, which is equivalent to have three derivatives of order one, and whose sum is again 3. As it can be observed, the sum of the orders in the derivatives in each term is 3. It is possible to corroborate a similar pattern for $V^{(4)}$:

$$\overbrace{V^{(4)}}^{\text{order }=4} = \frac{\overbrace{aU^{(4)}}^{\text{order 4}}}{(a-U)^2} + \frac{\overbrace{8aU^{(3)}U'}^{\text{order 3 +order 1= order 4}}}{(a-U)^3} + \frac{\overbrace{6a(U'')^2}^{\text{order 2 + order 2 =order 4}}}{(a-U)^3} \tag{4.27}$$

$$\frac{\overbrace{36U''(U')^2}^{\text{order 2 + order 1 + order 1= order 4}}}{(a-U)^4} + \frac{\overbrace{24(U')^4}^{\text{order 1 + order 1 + order 1 + order 1= order 4}}}{(a-U)^5}$$

As it can be noted in (4.27), if the orders of derivatives are added in each term, the result is 4. Even when the derivatives of U are multiplying, their orders can be analyzed as a sum. Such tendency is related to the integer's partitions, which are all the possible combinations in which an integer n can be represented as a sum of positive integers. In the last two cases, the partitions of 3 are: 3, 2+1, and 1+1+1, whereas the partitions of 4 are: 4, 3+1, 2+2, 2+1+1 and 1+1+1+1.

Strictly, there is not a closed-form equation to obtain all the partitions of a given integer, nevertheless, there is software like Wolfram Alpha that can provide them in a very fast way. Following the previous reasoning, it can be inferred some properties for $V^{(5)}$. Firstly, it is possible to expect that this function will have as many terms as elements in the partitions of 5, and the distributions of derivatives of U in each term will correspond to the configurations of these partitions. Therefore:

$$\begin{aligned}\overbrace{V^{(5)}}^{\text{order =5}} &= \frac{\overbrace{c_1 aU^{(5)}}^{\text{order 5}}}{(a-U)^{k_1}} + \frac{\overbrace{c_2 aU^{(4)}U'}^{\text{order 4+order 1=order 5}}}{(a-U)^{k_2}} + \frac{\overbrace{c_3 aU^{(3)}U''}^{\text{order 3+order 2= order 5}}}{(a-U)^{k_3}} \\ &+ \frac{\overbrace{c_4 aU^{(3)}(U')^2}^{\text{order 3 + order 1 + order 1=order 5}}}{(a-U)^{k_4}} + \frac{\overbrace{c_5 a(U'')^2U'}^{\text{order 2 + order 2 + order 1= order 5}}}{(a-U)^{k_5}} \\ &+ \frac{\overbrace{c_6 aU''(U')^3}^{\text{order 2 + order 1 + order 1+order 1=order 5}}}{(a-U)^{k_6}} + \frac{\overbrace{c_7 a(U')^5}^{\text{order 1+order 1 + order 1 + order 1+order 1=order 5}}}{(a-U)^{k_7}}\end{aligned} \tag{4.28}$$

4.4.5 Sequences related to partitions.

The following step is to find the coefficients $c_1, \dots, c_7$ and $k_1, \dots, k_7$ in equation (4.28), for this task it is possible to assume that they have a relationship with the partitions. Before continuing it is worth mentioning that the partitions can be ordered in decreasing order. Therefore, the terms in (4.28) must be written in such way, because the coefficients will be dependent on the position. In other words, the index 1,2, … ,7 denote the position of the term.

After a search of sequences related to the partitions, it was found that the succession of numbers denoted by A049019 in the OEIS's code[8], predicts the coefficients $c_1, \dots, c_7$. Henceforth, such sequence will be represented as $p_{19}(z)$, whose first 18 terms are the following:

$$1, 1, 2, 1, 6, 6, 1, 8, 6, 36, 24, 1, 10, 20, 60, 90, 240, 120,$$

---

[8] On-Line Encyclopedia of Integer Sequences. https://oeis.org/

$$1, 12, 30, 20, 90, 360, 90, 480, 1080, 1800, 720, \dots \tag{4.29}$$

In order to use this sequence, it is only necessary to fragment it using the number 1 as a division point or delimiter:

$$\begin{gathered}\boxed{1}, \boxed{1, 2}, \boxed{1, 6, 6}, \boxed{1, 8, 6, 36, 24}, \boxed{1, 10, 20, 60, 90, 240, 120}, \\ \boxed{1, 12, 30, 20, 90, 360, 90, 480, 1080, 1800, 720}, \dots\end{gathered} \tag{4.30}$$

Then, as it can be observed, each resulted block corresponds to the set of coefficients of a given derivative of V. To locate the correct block, it is necessary to order them in an ascending form, then the position of the block will be equal to the order of V. In the present example, the derivative $V^{(5)}$ is being computed, and therefore it is necessary to locate the fifth block that was generated by the fragmentation process. Thus, the set $\{c_1, c_2, \dots, c_7\}$ corresponds to the set $\{1, 10, 20, 60, 90, 240, 120\}$ belonging to (4.30).

In most programming languages is possible to carry out the fragmentation of the sequence $p_{19}(z)$. It is only necessary to use a “split” function of strings, using as a character separator (or delimiter) the number 1. A procedure of fragmentation related to the format used in Python and MATLAB is described in Algorithm 4.1. Nevertheless, for higher values of the order of the derivative of V, the procedure of fragmentation has one disadvantage: the time. This issue is related to the number of elements of the sequence $p_{19}(z)$ that need to be considered with the purpose of carrying out the fragmentations, which becomes a huge number.

In fact, such number needs to be big enough to guarantee that the “block” containing the set of coefficients will be included, but not so big that the fragmentation procedure consumes a lot of computational time. An alternative method can overcome this difficulty. This consists in using two other sequences related to the partitions of integers. The first one is the sequence A000041, whose elements are the number of partitions of the integers beginning with zero, which will denote it as $p_{41}(z)$. The first fifteen numbers of $p_{41}(z)$ are the following:

$$1, 1, 2, 3, 5, 7, 11, 15, 22, 30, 42, 56, 77, 101, 135, \dots \tag{4.31}$$

In the present case, the value of z will correspond to the value of the order of the derivative of V. Therefore, it is possible to verify that $p_{41}(5) = 7$, which are the number of terms that are in (4.28). The second sequence has the OEIS’s code of A026905 and has as first elements:

$$1, 3, 6, 11, 18, 29, 44, 66, 96, 138, 194, 271, 372, 507, \dots \tag{4.32}$$

Such sequence will be denoted as p_{05}. As it can be noted, the elements in (4.32) are the partial sums of the numbers in (4.31), starting from $z = 1$. In other words:

$$p_{05}(z) = \sum_{k=1}^{z} p_{41}(k) \tag{4.33}$$

It is not necessary to carry out a fragmentation procedure when we use $p_{05}(z)$ and $p_{41}(z)$, because these sequences allow us to find the exact segment of the sequence $p_{19}(z)$ where we will find the coefficients of $V^{(k)}$.

Algorithm 4.1 Fragmentation and search procedure

```
Input: Order of the derivative of $V$
Output: Coefficients of the derivative of $V$

STEP 1: Select a subsequence of A049019 that contains its first element, and whose value is $n$. Store it as a string variable $x$:

    $A049019[:n] \rightarrow x$

STEP 2: Split the string variable $x$ using the number 1 as a separator (or delimiter), and store the generated list in the variable $y$:

    $split(x, 1) \rightarrow y$
IF (length $(y)$ ≥order of derivative):

    IF the split function of STEP 2 removed the separator:

        ■ For $i = 1, 2, \dots,$ length of $y$:
            ■ Add the element 1 at the beginning of the stored string in $y[i]$

    ■ Find the element in $y$ whose position is equal to the order of the derivative of $V$, and store it in the variable $z$:

        $y[\text{order of the derivative of V}] \rightarrow z$
ELSE:

    ■ Choose a larger value of $n$ and repeat the algorithm.
```

In fact, it is possible to corroborate that:

$$H = \{c_j | c_j \text{ is a coefficient of } V^k\} = \{p_{19}(q) | p_{05}(k) - p_{10}(k+1) \leq q \leq p_{05}(k)\} \qquad (4.34)$$

4.4.6 Exponent of the denominators.

The coefficients $k_1, \dots, k_7$ in (4.28) also have a relationship with the partitions. It is possible to determine their value considering the number of elements that are involved in a particular partition, which will be denoted by the function n_p. For the case of $V^{(5)}$ it follows that:

$$\begin{gathered} n_p(5) = 1, \qquad n_p(4+1) = n_p(3+2) = 2, \\ n_p(3+1+1) = n_p(2+2+1) = 3 \\ n_p(2+1+1+1) = 4, \qquad n_p(1+1+1+1+1) = 5 \end{gathered} \qquad (4.35)$$

And therefore:

$$k_l = n_p\left(P_n^l\right) + 1 \tag{4.36}$$

Where P_n^l is one element of the partition of the integer n, which is in the position l. Using (4.34) and (4.36) it is possible to complete the missing elements in (4.28):

$$\begin{gathered}V^{(5)} = \frac{aU^{(5)}}{(a-U)^2} + \frac{10aU^{(4)}U'}{(a-U)^3} + \frac{20aU^{(3)}U''}{(a-U)^3} + \frac{60aU^{(3)}(U')^2}{(a-U)^4} \\ \frac{90a(U'')^2U'}{(a-U)^4} + \frac{240a(U'')(U')^3}{(a-U)^5} + \frac{120a(U')^5}{(a-U)^6}\end{gathered} \tag{4.37}$$

Once a method to determine the distributions and orders of U has been developed, t is possible to propose a procedure to build the derivatives of V. Such procedure is described in Algorithm 4.2. It is worth mentioning that the equation is built in a symbolical sense, but the numerical evaluation is not carried out. The following step consist of computing the high-order derivatives of U.

4.4.7 Formula for the derivatives of U.

The derivatives of U can be computed using the balance equation of X_n^L. For the case of the first derivative:

$$\begin{aligned}U' = \frac{d}{dt}\left(b_{n,1}\lambda_n X_n^L(t)\right) &= b_{n,1}\lambda_n\frac{dX_n^L}{dt} = b_{n,1}\lambda_n\left(b_{n-1,n}\lambda_{n-1}X_{n-1}^L - \lambda_n X_n^L\right) \\ &= \psi(n-1,n)X_{n-1}^L - \lambda_n U\end{aligned} \tag{4.38}$$

Where:

$$\psi(k,m) = \prod_{j=k}^{m} b_{k,k+1}\lambda_k\,, \qquad 1 \le k \le m \tag{4.39}$$

For the second and third derivative it follows that:

$$U'' = \psi(n-2,n)X_{n-2}^L - \psi(n-1,n)\lambda_{n-1}X_{n-1}^L - \lambda_n U$$

$$\begin{gathered}U^{(3)} = \psi(n-3,n)X_{n-3}^L - \psi(n-2,n)(\lambda_{n-1} + \lambda_{n-2})X_{n-2}^L \\ +\psi(n-1,n)\lambda_{n-1}^2 X_{n-1}^L - \lambda_n U'\end{gathered}$$

And finally, the general formula for the derivatives of U is given by:

$$\begin{gathered}U^{(k)} = \psi(n-k,n)X_{n-k}^L - \psi(n-k+1,n)\left(\sum_{j=1}^{k}\lambda_{n-j}\right)X_{n-k+1}^L \\ +\psi(n-k+1,n)C_s(A_k,2)X_{n-k+2}^L - \psi(n-k+2,n)C_s(A_{k-1},3)X_{n-k+3}^K \\ + \cdots + (-1)^{k+1}\psi(n-1,n)C_s(A_1,k-1)X_{n-1}^L - \lambda_n U^{(k-1)}\end{gathered} \tag{4.40}$$

Algorithm 4.2 Equation for the derivative of order i of the function V

```
Input: $i \in \mathbb{N}$
Output: Equation for the derivative of order $i$ of the function $V$

STEP 1: Compute all the partitions of $i$, and list them in a descending order.
STEP 2: Compute the set of coefficients of $V^{(i)}$ using the Algorithm 4.1 or the Equation (4.34).
STEP 3: For each partition:
    i)   Determine how many numbers belong to the partition and call this quantity as $n_0$.
    ii)  For each number $s$ that belongs to the partition:
         a) Write the derivative of $U$, whose order is equal to $s$.
         b) Store the term built in a).
    iii) Multiply all the derivatives that were built in (ii).
    iv)  Compute the value $n_p$ using $n_0$.
    v)   Write the division of the product in (iii) by the expression $(a-U)^{n_p+1}$.
    vi)  Store the final term in the set $T$.
STEP 4: Write an equation given by the sum of all the terms contained in the set $T$.
STEP 5: Multiply each term of the equation built in (iii) by the corresponding coefficient computed in STEP 2.
```

Where A_k denoted the set whose elements are $\{\lambda_{n-1}, \lambda_{n-2}, \dots, \lambda_{n-k}\}$, and the function $C(A_k, r)$, with $1 \leq r \leq card(A)$, is the sum of all combinations with repetitions of r elements that belong to A_k. For example, $C(A_3, 3)$ is given by:

$$\lambda_{n-1}^3 + \lambda_{n-2}^3 + \lambda_{n-3}^3 + \lambda_{n-1}^2\lambda_{n-2} + \lambda_{n-1}^2\lambda_{n-3} + \lambda_{n-2}^2\lambda_{n-1} + \lambda_{n-2}^2\lambda_{n-3}$$

$$\lambda_{n-3}^2\lambda_{n-1} + \lambda_{n-3}^2\lambda_{n-2} + \lambda_{n-1}\lambda_{n-2}\lambda_{n-3}$$

4.5 Root-based solution.

4.5.1 Polynomial in Laplace transform space and roots.

An alternative solution of (4.3) can be obtained from the algebraic equation theory, which can be useful to carry out a comparison with equation (4.16). This solution arises from the roots of the following polynomial:

$$(s+\lambda_1)(s+\lambda_2)\dots(s+\lambda_n) - \alpha(n) \tag{4.41}$$

Under the assumption that all the roots are real, it is possible to rewrite the last polynomial as:

$$\prod_{j=1}^{n}(s-\beta_j), \quad \beta_j \text{ is a root of (4.41)} \tag{4.42}$$

In order to use symmetry in the analytic solution, the following variables will be defined:

$$\beta_j^* = -\beta_j, \ \ 1 \leq j \leq n \tag{4.43}$$

It is possible to obtain the analytical solution based on the roots of the isotope n, using a similar procedure like that described in (1.15)-(1.23):

$$X_n^A(t) = X_1(0) \prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.44}$$

As it can be observed, this last equation has a similar shape that equation (2.23), which is very useful to develop comparisons. As well as equation (4.16), equation (4.44) cannot be applied to other isotopes in the cyclic chain due to the lack of symmetry of the system. Nevertheless, through the backward equation of (1.31), it follows that:

$$X_{n-1}^A = \frac{1}{b_{n-1,n}^{\text{eff}} \lambda_{n-1}^{\text{eff}}} \left(\frac{dX_n^A}{dt} + \lambda_n^{\text{eff}} X_n^A \right) \tag{4.45}$$

$$= \left(X_1(0) \prod_{k=1}^{n-2} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{(\lambda_n^{\text{eff}} - \beta_i^*)}{(\beta_j^* - \beta_i^*)} \right)$$

Applying the same procedure for the general case k:

$$X_{n-k}^A = X_1(0) \prod_{m=1}^{n-k-1} b_{m,m+1}^{\text{eff}} \lambda_m^{\text{eff}} \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{u=n-k+1}^{n} (\lambda_u^{\text{eff}} - \beta_i^*) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.46}$$

4.5.2 Symmetry of the solution based on roots.

Using the described in Section 1.8.3, the following two functions will be defined:

$$F(B) = F(\beta_1^*, \beta_2^*, \dots, \beta_n^*) = \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.47}$$

$$M_k(B) = M_k(\beta_1^*, \beta_2^*, \dots, \beta_n^*) = \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{u=n-k+1}^{n} (\lambda_u^{\text{eff}} - \beta_i^*) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.48}$$

Where $B = \{\beta_i^* | \, 1 \leq i \leq n\}$. It is possible to show that $F(\beta_1^*, \beta_2^*, \dots, \beta_n^*)$ and $M_k(\beta_1^*, \beta_2^*, \dots, \beta_n^*)$ are symmetric in all their arguments related to the β^* variables. In other words:

$$F\left(\beta_1^*, \dots, \beta_i^*, \dots, \beta_j^*, \dots, \beta_n^*\right) = F\left(\beta_1^*, \dots, \beta_j^*, \dots, \beta_i^*, \dots, \beta_n^*\right) \quad 1 \le i, j \le n \tag{4.49}$$

And:

$$M_k\left(\beta_1^*, \dots, \beta_i^*, \dots, \beta_j^*, \dots, \beta_n^*\right) = M_k\left(\beta_1^*, \dots, \beta_j^*, \dots, \beta_i^*, \dots, \beta_n^*\right) \quad 1 \le i, j \le n \tag{4.50}$$

The last property implies that the position of the variables β^* does not matter, in other words, it is possible to make an arbitrary enumeration of the elements in B, and the outcome of the functions is the same. Under the previous argument the following equality is valid:

$$M_k(\beta_1^*, \beta_2^*, \dots, \beta_n^*) = M_k\left(\beta^*_{\varphi_1^B}, \beta^*_{\varphi_2^B}, \dots, \beta^*_{\varphi_n^B}\right) \tag{4.51}$$

Where the elements φ_k^B, $1 \le k \le n$, are the index of the positions of an arbitrary permutation of the elements of B. This is very useful in terms of computational time when some elements in B are removed, because several computational decisions steps can be omitted. For example, if the set $B_l = B - \{\beta_l^*\}$ is considered, it follows that:

$$M_k(B_l) = M_k(\beta_1^*, \dots, \beta_{l-1}^*, \beta_{l+1}^*, \dots, \beta_n^*) \tag{4.52}$$

$$= \sum_{\substack{i=1 \\ i \neq l}}^{n} e^{-\beta_i^* t} \prod_{u=n-k+1}^{n} (\lambda_u^{\text{eff}} - \beta_i^*) \prod_{\substack{j=1 \\ j \neq i \\ j \neq l}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.53}$$

As it can be noted, in the last equation there is a step of decision that verifies that $i \neq l$. Nevertheless, such step can be avoided through a new enumeration, for which:

$$M_k(\beta_1^*, \dots, \beta_{l-1}^*, \beta_{l+1}^*, \dots, \beta_n^*) = M_k\left(\beta^*_{\varphi_1}, \beta^*_{\varphi_2}, \dots, \beta^*_{\varphi_{n-1}}\right) \tag{4.54}$$

$$= \sum_{i=1}^{n-1} e^{-\beta^*_{\varphi_i} t} \prod_{u=n-k+1}^{n} (\lambda_u^{\text{eff}} - \beta^*_{\varphi_i}) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{\left(\beta^*_{\varphi_j} - \beta^*_{\varphi_i}\right)} \tag{4.55}$$

With the elements $\varphi_k^{B_l}$, $1 \le k \le n-1$, denoting the position of an arbitrary permutation of the elements of B_l. In equation (4.55) the step decision has been removed. While the depletion function F only depends on the arguments given by the variables β^*, the function M_k also depends on the variable k, that is related with the following set:

$$\Lambda_k = \{\lambda_{n-k+1}, \lambda_{n-k+2}, \dots, \lambda_n\} \tag{4.56}$$

Since this set is present in the function M_k in equation (4.50), through the product in the middle part of its expression, it is possible to verify that the function is also symmetrical in the argument k. Even more, if an arbitrary enumeration is used for the variables β^*, this can be inherited in a straightforward way by the set Λ_k, because both sets share the same subrange of index, given by $n-k+1, n-k+2, \dots, n$. Therefore, equation (4.55) can be written as:

$$\sum_{i=1}^{n-1} e^{-\beta^*_{\varphi_i} t} \prod_{u=n-k+1}^{n-1} (\lambda^{\text{eff}}_{\varphi_u} - \beta^*_{\varphi_i}) \prod_{\substack{j=1 \\ j\neq i}}^{n-1} \frac{1}{\left(\beta^*_{\varphi_j} - \beta^*_{\varphi_i}\right)} \tag{4.57}$$

4.5.3 Simplifications of the solution based on roots.

In certain cases, some variables β^* will be very closed to some removal coefficient λ^{eff}, which allows simplifying equation (4.46) using the symmetry discussed in the past section. For example, if $\lambda^{\text{eff}}_n \approx \beta^*_n$, then $\lambda^{\text{eff}}_n - \beta^*_n \approx 0$ and (4.48) is reduced to:

$$\sum_{i=1}^{n-1} e^{-\beta^*_i t} \prod_{u=n-k+1}^{n-1} (\lambda^{\text{eff}}_u - \beta^*_i) \prod_{\substack{j=1 \\ j\neq i}}^{n-1} \frac{1}{(\beta^*_j - \beta^*_i)}$$

In other words:

$$M_k(\beta^*_1, \dots, \beta^*_{n-1}, \beta^*_n) = M_k(\beta^*_1, \dots, \beta^*_{n-1}) \tag{4.58}$$

For the particular case where $k=1$, the last equation is reduced to:

$$M_1(\beta^*_1, \dots, \beta^*_{n-1}, \beta^*_n) = F(\beta^*_1, \dots, \beta^*_{n-1})$$

For the general case, where several variables β^* are approximately equal to the corresponding effective removal coefficient λ^{eff}, it is necessary to consider the following sets:

$$S = \{\beta^*_j \,|\, \beta^*_j \approx \lambda^{\text{eff}}_j, 1 \le j \le k\}, \qquad P = \{j | \beta^*_j \in S\} \tag{4.59}$$

Then:

$$M_k(\beta^*_1, \dots, \beta^*_{n-1}, \beta^*_n) \approx \sum_{\substack{i=1 \\ i\notin P}}^{n} e^{-\beta^*_i t} \prod_{\substack{u=n-k+1 \\ u\notin P}}^{n} (\lambda^{\text{eff}}_u - \beta^*_i) \prod_{\substack{j=1 \\ j\neq i \\ j\notin P}}^{n} \frac{1}{(\beta^*_j - \beta^*_i)} \tag{4.60}$$

Using the enumeration described in the previous section, the step decision given by $i \notin P$ and $u \notin P$ can be avoided, through the following equation:

$$M_k(\beta^*_1, \dots, \beta^*_{n-1}, \beta^*_n) \approx \sum_{i=1}^{n-p} e^{-\beta^*_{\varphi_i} t} \prod_{u=n-k+1}^{n-p} (\lambda^{\text{eff}}_{\varphi_u} - \beta^*_{\varphi_i}) \prod_{\substack{j=1 \\ j\neq i}}^{n-p} \frac{1}{\left(\beta^*_{\varphi_j} - \beta^*_{\varphi_i}\right)} \tag{4.61}$$

$$= M_k\left(\beta^*_{\varphi_1}, \beta^*_{\varphi_2}, \dots, \beta^*_{\varphi_{n-p}}\right) \tag{4.62}$$

Where p is the cardinality of the set P. Finally, if the set S is equal to the set Λ_k, then it is possible to use the following approximation:

$$M_k(\beta_1^*, \dots, \beta_{n-1}^*, \beta_n^*) = F(\beta_1^*, \beta_2^*, \dots, \beta_{n-k}^*)$$

This analysis can help to reduce the computational time involved when the analytic solution based on the roots is used.

4.6 Comparison schemes.

The main objective in this section is to compare the set of equations developed above with the modified Bateman solution given for a linear chain. It is worth mentioning that equations (4.16) and (4.46) are not necessary equivalent, because the first one was developed using certain assumptions related to the branching ratios and the effective removal coefficients. In such sense, equation (4.46) will be considered as the reference solution, because there were not approximations or assumptions involved in its development. Strictly speaking, this equation is not totally exact, because there is an error involved in the process of finding the roots. Nevertheless, since this searching is related to a polynomial function, it is possible to consider this error as insignificant.

Two examples related to cyclic chains will be analyzed. Such structures will consist of heavy isotopes, because these isotopes are commonly found in reactor nuclear problems. The first one consists in a pure cyclic chain composed by two isotopes, ^{235}U and ^{236}U, and the second is given by the sequence of isotopes $^{235}\mathrm{U} \rightarrow {}^{236}\mathrm{U} \rightarrow {}^{237}\mathrm{U} \rightarrow {}^{237}\mathrm{Np} \rightarrow {}^{238}\mathrm{Np} \rightarrow {}^{238}\mathrm{Pu} \rightarrow {}^{239}\mathrm{Pu} \rightarrow \boxed{{}^{235}\mathrm{U}}$. These schemes will correspond to the case $n = 2$ and the case $n = 7$, respectively, which are shown in Figure 4.5. For the case of two isotopes, a similar procedure to the one used by M. Blaauw has been used (Blaauw, 1993). For the other scheme, a sequence of isotopes reported by R. Dreher was used (Dreher, 2012). The models were fed with data that was obtained from the simulation of a unit cell in infinite medium, in a thermal neutron spectrum. Calculations were done with the well-known Monte Carlo code SERPENT (Leppänen et al., 2015). The description and main features of the unit cell are shown Figure 4.6 and Tables 4.1-4.3. For the decay constants the ENDF/VII.1 Library was used. The set of data used in the comparison is presented in Table 4.2, and the numerical operations were carried in the software Mathematica 11.3.0, using precision 30 in the case $n = 2$ and precision 40 in the case $n = 7$.

4.6.1 Case $n = 2$.

Superposition process.

From Table 4.1 it is known that both ^{235}U and ^{236}U have an initial concentration different from zero. In the previous developments it was always assumed that only one isotope has a concentration different from zero, which was the first element in the pure cyclic chain system given in (4.3) and (4.4). Therefore, in order to analyze a process of superposition, this structure will be solved in two steps. In the first one, the condition where only ^{235}U has a concentration different from zero will be used. Thereby, $\lambda_1 = \lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}$, $\lambda_2 = \lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}$, $b_{1,2} = b^{\mathrm{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}$, $b_{2,1} = b^{\mathrm{eff}}_{^{236}\mathrm{U},^{235}\mathrm{U}}$. In the second step of the superposition it will be considered that $X_{^{235}\mathrm{U}}(0) = 0$, and $X_{^{236}\mathrm{U}}(0) \neq 0$, and thus $\lambda_1 = \lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}$, $\lambda_2 = \lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}$, $b_{1,2} = b^{\mathrm{eff}}_{^{236}\mathrm{U},^{235}\mathrm{U}}$, $b_{2,1} = b^{\mathrm{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}$.

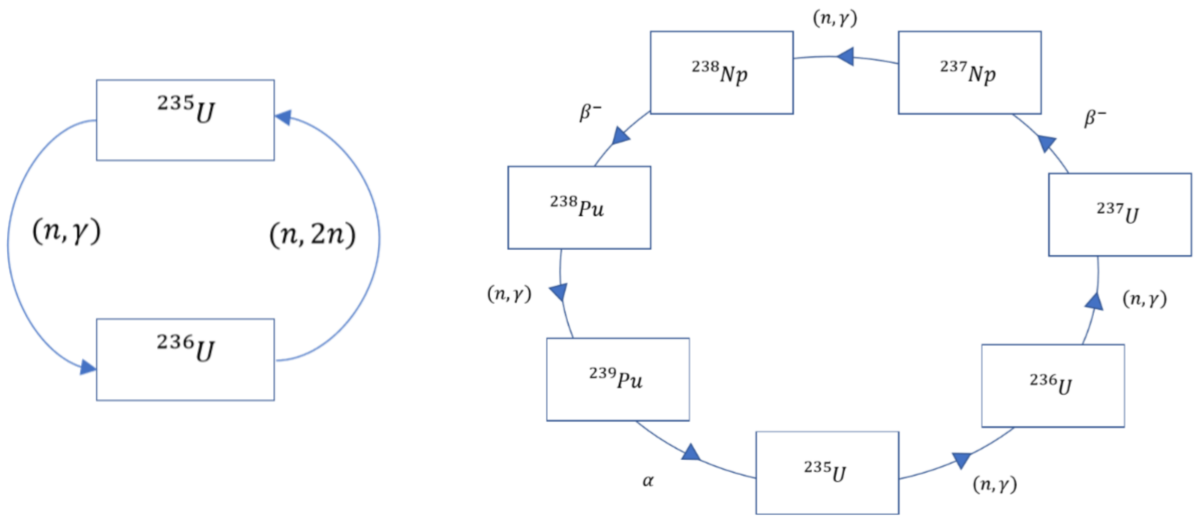

Figure 4. 5. Left side: case $n = 2$. Pure cyclic chain given by the isotopes ^{235}U and ^{236}U, through the reactions (n,γ) and $(n,2n)$. Right side: case n=7. Pure cyclic chain given by the sequence of isotopes that begins with ^{235}U and ends with ^{239}Pu.

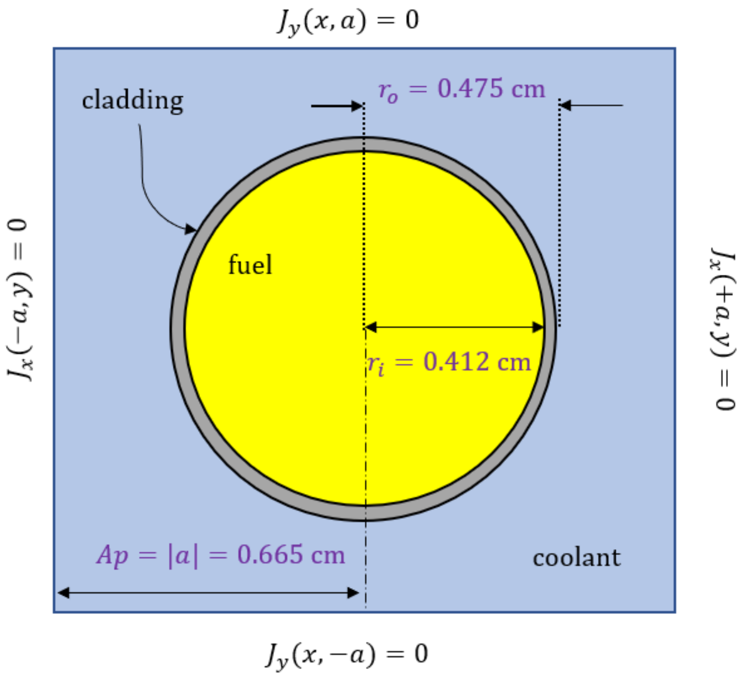

Figure 4. 6. Geometry of the unit cell in an infinite medium used in the comparison scheme. In such scheme the symbols J_x and J_y denote the magnitude of the neutron current density vector $\vec{J}$. This example was used in Chapter 2 to explain the comparison between the code MCNPX and SERPENT.

Table 4. 1. Main features of the unit cell used in the comparison scheme.

Parameter		Isotope	Initial concentration atoms/(b $\cdot$ cm)
Inner radius (cm)	0.412	^{234}U	$6.15138x10^{-6}$
Outer radius (cm)	0.475	^{235}U	$6.89185x10^{-4}$
Square's apothem (cm)	0.665	^{236}U	$3.16249x10^{-6}$
Coolant	Water	^{238}U	$2.17092x10^{-2}$
Cladding	Zircalloy		
Neutron Flux $(\frac{\text{neutrons}}{\text{cm}^2\cdot\text{s}})$	$2.72x10^{14}$		
Volume m^3	$5.33267x10^{-1}$		

Table 4. 2. Neutronic parameters of the isotopes belonging to the unit cell used in the comparison scheme.

Parameter	^{235}U	^{236}U	^{237}U	^{237}Np
$\sigma_f\ [b]$	$5.51954x10^{1}$	$3.0372x10^{-1}$	1.73591	$5.05637x10^{-1}$
$\sigma_{(n,\gamma)}\ [b]$	$1.17859x10^{1}$	8.65019	$6.29918x10^{1}$	$3.75076x10^{1}$
$\sigma_{(n,2n)}\ [b]$	$3.52596x10^{-3}$	$2.18561x10^{-3}$	$1.12769x10^{-2}$	$9.28667x10^{-4}$
$\sigma_f\cdot\phi\ [s^{-1}]$	$1.503275x10^{-8}$	$8.27197x10^{-11}$	$4.72784x10^{-10}$	$1.37712x10^{-10}$
$\sigma_{(n,\gamma)}\cdot\phi\ [s^{-1}]$	$3.20995026x10^{-9}$	$2.35592x10^{-9}$	$1.71561x10^{-8}$	$1.02154x10^{-8}$
$\sigma_{(n,2n)}\cdot\phi\ [s^{-1}]$	$9.60313x10^{-13}$	$5.95262x10^{-13}$	$3.07132x10^{-12}$	$2.52927x10^{-13}$
$\lambda\ [s^{-1}]$	$3.12298x10^{-17}$	$9.38495x10^{-16}$	$1.18852x10^{-6}$	$1.02516x10^{-14}$
λ^{eff}	$1.82437x10^{-8}$	$2.43924x10^{-9}$	$1.20615x10^{-6}$	$1.03533x10^{-8}$
$b_{1,2}^{\text{eff}}$	$1.75949x10^{-1}$	$9.65843x10^{-1}$	$9.85381x10^{-1}$	$9.86673x10^{-1}$
$b_{2,1}^{\text{eff*}}$		$2.4403x10^{-4}$		

Table 4. 3. Neutronic parameters of the isotopes belonging to the unit cell used in the comparison scheme.

Parameter	^{238}Np	^{238}Pu	^{239}Pu
$\sigma_f\ [b]$	$1.94989x10^{2}$	2.88213	$1.41846x10^{2}$
$\sigma_{(n,\gamma)}\ [b]$	$1.97274x10^{1}$	$4.11423x10^{1}$	$7.96234x10^{1}$
$\sigma_{(n,2n)}\ [b]$	$4.46679x10^{-3}$	$1.08646x10^{-3}$	$1.39825x10^{-3}$
$\sigma_f\cdot\phi\ [s^{-1}]$	$5.31063x10^{-8}$	$7.84963x10^{-10}$	$3.86325x10^{-8}$
$\sigma_{(n,\gamma)}\cdot\phi\ [s^{-1}]$	$5.37286x10^{-9}$	$1.12053x10^{-8}$	$2.16858x10^{-8}$
$\sigma_{(n,2n)}\cdot\phi\ [s^{-1}]$	$1.21655x10^{-12}$	$2.95903x10^{-13}$	$3.80821x10^{-13}$
$\lambda\ [s^{-1}]$	$3.78958x10^{-6}$	$2.50622x10^{-10}$	$9.11636x10^{-13}$
λ^{eff}	$3.84806x10^{-6}$	$1.22412x10^{-8}$	$6.03196x10^{-8}$
$b_{1,2}^{\text{eff}}$	$9.84802x10^{-1}$	$9.15377x10^{-1}$	$1.51134x10^{-5}$

In other words, the index position has been switched in the equations. In fact, this process is very similar to the one described by Tasaka in 1980 (Tasaka, 1980), where the loops were "broken" in two parts or linear chains, which later were solved using the standard Bateman equation, and finally their concentration were superposed.

4.6.2 Analysis of the roots.

For the case $n = 2$ equation (4.41) has two roots, which are given by:

$$\beta_{1,2} = \mp\sqrt{\left(\frac{\lambda_1^{\text{eff}} + \lambda_2^{\text{eff}}}{2}\right)^2 - \lambda_1^{\text{eff}}\lambda_2^{\text{eff}} + b_{1,2}^{\text{eff}}\lambda_1^{\text{eff}}b_{2,1}^{\text{eff}}\lambda_2^{\text{eff}}} - \left(\frac{\lambda_1^{\text{eff}} + \lambda_2^{\text{eff}}}{2}\right) \tag{4.63}$$

As it can be observed, equation (4.63) does not depend on the position index of the elements λ_i^{eff} and $b_{i,j}^{\text{eff}}$. In other words, the roots are the same regardless what the first isotope is in the pure cyclic chain. Therefore, both steps of the superposition will have the same roots[9].Once the data of Table 3.1 is replaced, the following values for the roots are obtained:

$$\beta_1 = -1.824378x10^{-8}s^{-1}, \quad \beta_2 = -2.439118x10^{-9}\ s^{-1} \tag{4.64}$$

With their respective values of β^*:

$$\beta_1^* = 1.824378x10^{-8}s^{-1}, \qquad \beta_2^* = 2.439118x10^{-9}\ s^{-1} \tag{4.65}$$

Using equation (4.46) for the root-based solution for case $n = 2$, it follows that:

$$X_1^A = X_1(0)\left(\frac{(\lambda_2^{\text{eff}} - \beta_1^*)}{\beta_2^* - \beta_1^*}e^{-\beta_1^* t} + \frac{(\lambda_2^{\text{eff}} - \beta_2^*)}{\beta_1^* - \beta_2^*}e^{-\beta_2^* t}\right) \tag{4.66}$$

And using equation (4.44):

$$X_2^A = \frac{b_{1,2}^{\text{eff}}\lambda_1^{\text{eff}}X_1(0)}{\beta_2^* - \beta_1^*}\left(e^{-\beta_1^* t} - e^{-\beta_2^* t}\right) \tag{4.67}$$

These two equations will be used in the two superposition steps. As it can be noted, the values of (4.65) are very similar to the removal coefficients $\lambda_{235_{\text{U}}}^{\text{eff}}$ and $\lambda_{236_{\text{U}}}^{\text{eff}}$. Particularly, the following condition is valid $\beta_1^* \approx \lambda_{235_{\text{U}}}^{\text{eff}}$ and $\beta_2^* \approx \lambda_{236_{\text{U}}}^{\text{eff}}$. These values can be compared using the percentual error given by:

$$\text{E}_{235_{\text{U}}} = 100\%\left(\frac{|\beta_1^* - \lambda_{235_{\text{U}}}^{\text{eff}}|}{\lambda_{235_{\text{U}}}^{\text{eff}}}\right) = 6.6269x10^{-4}\% \tag{4.68}$$

And, in a similar way:

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[9] The values of the roots are the same, but the sub-index of β^* changes. Therefore, β_1^* in the first superposition step has the same value of β_2^* in the second superposition step.

$$\mathrm{E}_{^{236}\mathrm{U}} = \;\; 4.9566x10^{-3}\% \tag{4.69}$$

The errors are so small, that the approximation $\beta^* \approx \lambda^{\text{eff}}$ can be considered and the analysis that we discussed in Section 4.5.3:

$$X_1^A \approx X_1^L \;\text{ and }\; X_2^A \approx X_2^L \tag{4.70}$$

In other words, in this case the modified Bateman solution for a linear chain is a very good approximation to solve a cyclic chain. The analysis of this approximation is very different that the one given by $b_{n,1}\lambda_n \approx 0$ and (4.6). In this case, the approximation only depends on the effective removal coefficients and the roots of the polynomial in (4.41), the branch ratios have not been considered. Additionally, this approximation does not consider a comparison of how small is a parameter, or how it is close to zero, instead it depends on a comparison with another parameter.

Another implication of the small errors is related to the localization of the roots of the polynomial in (4.41). As a first approach for other cases, the search of the roots can be performed in a neighborhood around the effective removal coefficients, which will reduce the time of the algorithm. Finally, the third implication allows approximating the solution of the system in the case where it will be appropriated. For example, the percentual errors in (4.68) and (4.69) have different orders of magnitude, therefore, the Bateman's solution of a linear chain can be used as an approximation of $X^A_{^{235}\mathrm{U}}$, but on the other hand, the root-based solution can be used to compute the concentration of ^{236}U.

4.6.3 First step of the superposition

Using equation (2.23), the modified Bateman equation for a linear chain will be equal to:

$$X^L_{^{235}\mathrm{U}} = X_{^{235}\mathrm{U}}(0)e^{-\lambda^{\text{eff}}_{^{235}\mathrm{U}}\cdot t}\,\frac{\text{atoms}}{\text{barn-cm}} = 6.89185\text{x}10^{-4}\cdot e^{-(1.82437\text{x}10^{-8})t}\,\frac{\text{atoms}}{\text{barn-cm}} \tag{4.71}$$

And:

$$X^L_{^{236}\mathrm{U}} = b^{\text{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}\lambda^{\text{eff}}_{^{235}\mathrm{U}}X_{^{235}\mathrm{U}}(0)\frac{\left(e^{-\lambda^{\text{eff}}_{^{235}\mathrm{U}}\cdot t} - e^{-\lambda^{\text{eff}}_{^{236}\mathrm{U}}\cdot t}\right)}{\lambda^{\text{eff}}_{^{236}\mathrm{U}} - \lambda^{\text{eff}}_{^{235}\mathrm{U}}}\,\frac{\text{atoms}}{\text{barn-cm}} = \tag{4.72}$$

$$= 1.39976\text{x}10^{-4}\left(e^{-(2.43924\text{x}10^{-9})t} - e^{-(1.82437\text{x}10^{-8})t}\right)\frac{\text{atoms}}{\text{barn-cm}} \tag{4.73}$$

For the case of the root-based solution the equation (4.66) will be valuated:

$$X^{A}_{^{235}\mathrm{U}} = 6.89179\text{x}10^{-4} e^{-(1.824378\text{x}10^{-8}\ s^{-1})t} + 5.27199\text{x}10^{-8} e^{-(2.449118\text{x}10^{-9}\ s^{-1})t}\,\frac{\text{atoms}}{\text{barn-cm}} \tag{4.74}$$

And for equation (4.67):

$$X^{A}_{^{236}\mathrm{U}} = 1.39974\text{x}10^{-4}\left(e^{-(2.449118\text{x}10^{-9}\ s^{-1})t} - e^{-(1.824378\text{x}10^{-8}\ s^{-1})t}\right)\frac{\text{atoms}}{\text{barn-cm}} \tag{4.75}$$

Finally, for the proposed equation given in (4.15):

$$X_{^{236}\mathrm{U}} = X_{^{235}\mathrm{U}}(0)\,\frac{X^{L}_{^{236}\mathrm{U}}\,\frac{\text{atoms}}{\text{barn-cm}}}{X_{^{235}\mathrm{U}}(0) - b^{\text{eff}}_{^{236}\mathrm{U},^{235}\mathrm{U}}\lambda^{\text{eff}}_{^{236}\mathrm{U}}X^{L}_{^{236}\mathrm{U}}} \tag{4.76}$$

Once the values are replaced, $X_{^{236}\mathrm{U}}$ will be equal to:

$$\frac{6.89185\text{x}10^{-4}\cdot 1.39976\text{x}10^{-4}\left(e^{-(2.43924\text{x}10^{-9})t} - e^{-(1.82437\text{x}10^{-8})t}\right)\frac{\text{atoms}}{\text{barn-cm}}}{6.89185\text{x}10^{-4} - 5.9526208\text{x}10^{-13}\left(1.39976x10^{-4}\left(e^{-(2.43924\text{x}10^{-9})t} - e^{-(1.82437\text{x}10^{-8})t}\right)\right)} \tag{4.77}$$

And for the case of $X_{^{235}\mathrm{U}}$:

$$X_{^{235}\mathrm{U}} = \frac{1}{b^{\text{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}\lambda^{\text{eff}}_{^{235}\mathrm{U}}}\left(\frac{dX_{^{236}\mathrm{U}}}{dt} + \lambda^{\text{eff}}_{^{236}\mathrm{U}}X_{^{236}\mathrm{U}}\right) \tag{4.78}$$

With:

$$\frac{dX_{^{236}\mathrm{U}}}{dt} = X_1(0)^2\ \frac{b^{\text{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}\lambda^{\text{eff}}_{^{235}\mathrm{U}}X^{L}_{^{235}\mathrm{U}} - \lambda^{\text{eff}}_{^{236}\mathrm{U}}X^{L}_{^{236}\mathrm{U}}}{\left(X_1(0) - b_{n,1}\lambda_n X^{L}_{n}\right)^2} \tag{4.79}$$

Before continuing with this first step of superposition, the denominator in equation (4.77) will be analyzed. Firstly, the function $e^{-(2.43924\text{x}10^{-9})t} - e^{-(1.82437\text{x}10^{-8})t}$ has a maximum value given by 0.63503, which is reached when $t = 1473.544$ days. Therefore:

$$5.9526208\text{x}10^{-13}\left(1.39976x10^{-4}\left(e^{-(2.43924\text{x}10^{-9})t} - e^{-(1.82437\text{x}10^{-8})t}\right)\right) \leq 8.33224x10^{-17} \tag{4.80}$$

Then, the denominator is, for practical purposes, equal to $6.89185\text{x}10^{-4}$, which means that equation (4.76) is essentially equal to $X^{L}_{^{236}\mathrm{U}}$. It was expected, because the condition given in (4.6) is valid. Since equations (4.78) and (4.79) are obtained through the backward method described in equation (1.31), it is possible to verify that $X_{^{235}\mathrm{U}}$ also tends to $X^{L}_{^{235}\mathrm{U}}$.Therefore, the analysis in this first step of the superposition is reduced to compare the root-based equation with the modified Bateman equation for a linear chain. Since the data from a unit cell is using with conditions that are very similar to the ones finding in a thermal reactor case, it is possible to conclude that the linear chain model can be a good approximation to model this pure cyclic chain.

Figure 4.7 and Figure 4.8 contain the results for the concentration of $^{235}\mathrm{U}$ and $^{236}\mathrm{U}$ for the first superposition step. In such graphs only the root-based solution appears, because the percentual error between this and the modified Bateman equation is so small that the two curves overlap. Figure 4.9 contains the percentual error between them, which was computed using the root-based solution as the reference. An interval of time between 1 hour (0.0417 days, approximately) and 1000 days was used, where it was assumed that the data in Table 4.2 remains constant.

It is important to consider such assumption, because the roots of equation (4.41) depend on the set of values of λ_i^{eff} and $b_{i,j}^{\mathrm{eff}}$ which in turn depends on the neutron flux and the microscopic cross sections. Strictly speaking, it is necessary to divide the calculations in a set of time steps, in which the effective removal coefficients must to be computed as well as the effective branch ratios, and therefore, the roots of equation (4.41) will not be the same. Nevertheless, the percentual errors given by (4.68) and (4.69) remain with the same order of magnitude, as it can be noted in Table 4.4, where each pair of roots has been computed in each burnup step, using the data obtained from SERPENT.[10] Based on Table 4.4, it is possible to justify the assumption that the data can be considered constant in time.

From this analysis it is possible to conclude that, for this first step of superposition, the modified Bateman equation for a linear chain is a very good approximation for the pure cyclic chain given by $^{235}\mathrm{U} \rightarrow \boxed{^{236}\mathrm{U}}$. In all the cases, the percentual error is less than 0.01%.

4.6.4 Analysis of the error.

As it can be observed in Figure 4.9, the error's curves have the same shape. In both cases, the minimum value is found in the first-time steps, then it undergoes for a great increment of almost seven order of magnitude at 100 days. From this point, the error increases slowly, and in a time lapse of 900 days, it grows by almost two orders of magnitude.

The error of the U-235 concentration is always greater than the error related to U-236. This can seem contradictory based in the errors (4.68) and (4.69). In other words, since $\beta^*_{^{235}\mathrm{U}}$ is closer to $\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}$, in comparison with the closeness of $\beta^*_{^{236}\mathrm{U}}$ to $\lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}$, it would be expected that the percentual concentration error of $^{235}\mathrm{U}$ was lower than the error related to $^{236}\mathrm{U}$.

This can be explained because the modified Bateman equation for a linear chain is using, and therefore is assuming that, instead of a cyclic chain, there is a linear succession of isotopes given $^{235}\mathrm{U} \rightarrow {}^{236}\mathrm{U}$. Therefore, the contribution to the concentration of $^{235}\mathrm{U}$ given by the reaction $^{236}\mathrm{U} \xrightarrow{n,2n} {}^{235}\mathrm{U}$ is ignoring and undervaluing the concentration of this isotope.

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[10] Such data is not shown in the present thesis. Tables 4.2 and 4.3, contain the values of the parameters only for the first burnup step.

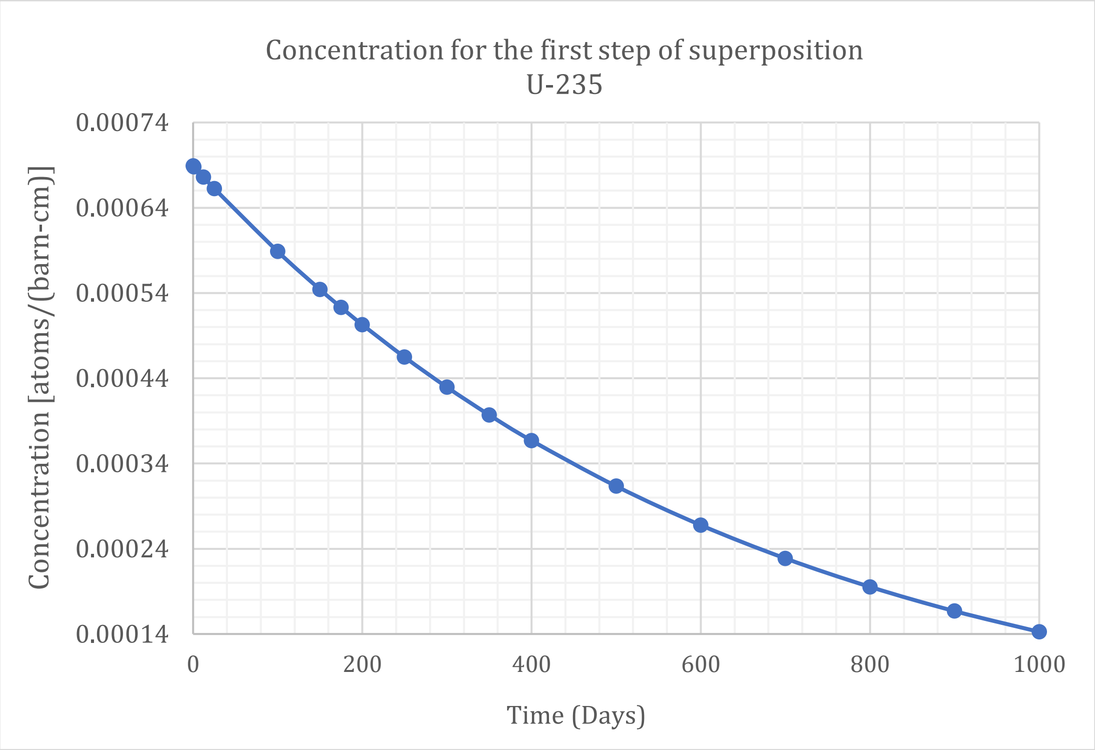

Figure 4. 7. Concentration of the isotope ^{235}U as a function of time, computed using the root-based solution for the first step of superposition. The modified Bateman equation was omitted in the graph, because both curves overlap.

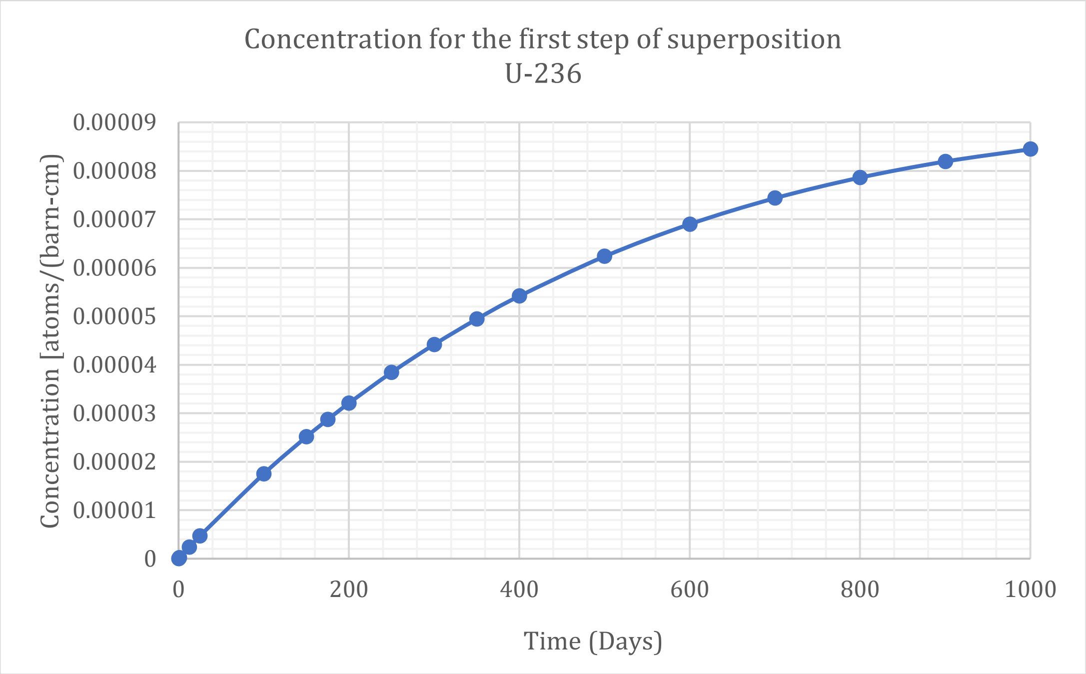

Figure 4. 8. Concentration of the isotope ^{236}U as a function of time, computed using the root-based solution for the first step of superposition. As in the case of Figure 4.7, the modified Bateman equation has been omitted.

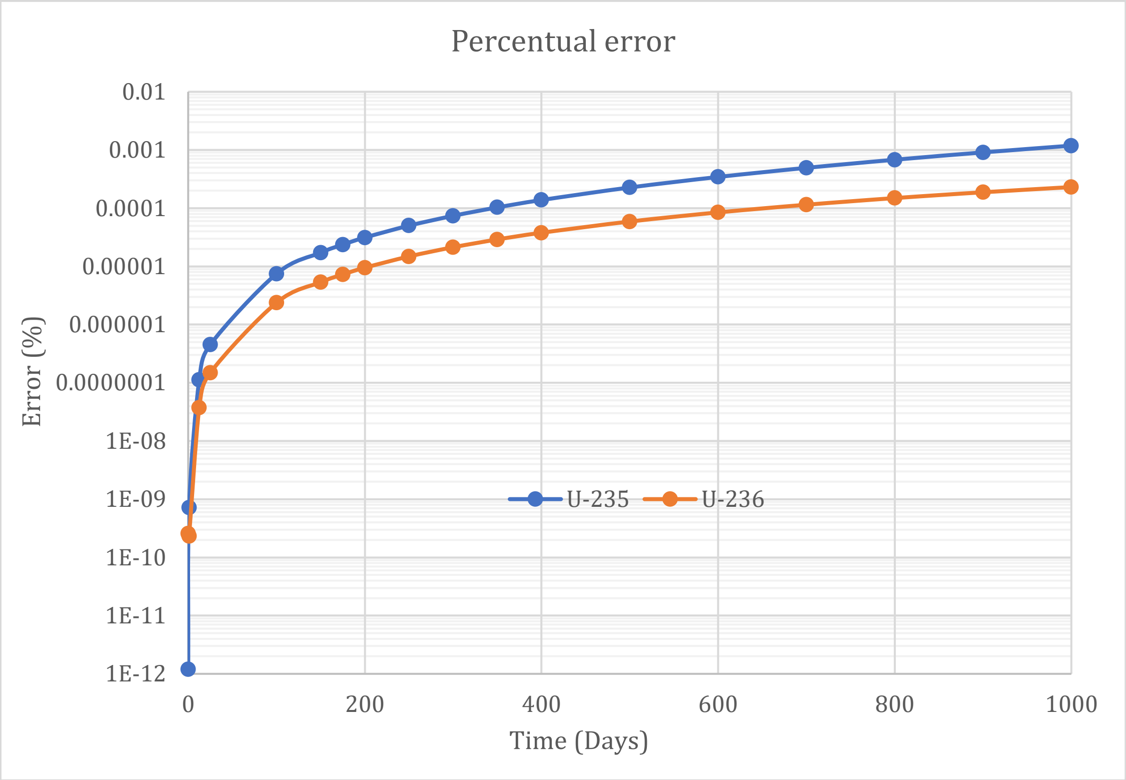

Figure 4. 9. Percentual error between the Root-based and the Modified Bateman solution for ^{235}U and ^{236}U, for the first step of superposition.

Table 4. 4. Comparison of the roots of equation (4.63) with the effective removal coefficients considering the variation on neutron flux.

Time (days)	$\beta^*_{^{235}\mathrm{U}}$	$\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}$	$\mathrm{E}_{^{235}\mathrm{U}}$	$\beta^*_{^{236}\mathrm{U}}$	$\lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}$	$\mathrm{E}_{^{236}\mathrm{U}}$
12.5	1.824378E-08	1.824366E-08	6.63E-04	2.439119E-09	2.439239E-09	4.96E-03
25	1.827399E-08	1.827386E-08	7.20E-04	2.563307E-09	2.563439E-09	5.14E-03
125	1.827096E-08	1.827083E-08	7.29E-04	2.583379E-09	2.583512E-09	5.15E-03
250	1.821497E-08	1.821483E-08	7.31E-04	2.536778E-09	2.536911E-09	5.25E-03
375	1.831622E-08	1.831607E-08	8.12E-04	2.527604E-09	2.527753E-09	5.89E-03
500	1.896815E-08	1.896799E-08	8.47E-04	2.519625E-09	2.519785E-09	6.38E-03
625	1.989494E-08	1.989477E-08	8.77E-04	2.549947E-09	2.550121E-09	6.84E-03
750	2.101352E-08	2.101333E-08	9.01E-04	2.613155E-09	2.613344E-09	7.25E-03
875	2.230737E-08	2.230717E-08	8.98E-04	2.713452E-09	2.713652E-09	7.38E-03
1000	2.368117E-08	2.368095E-08	9.08E-04	2.763975E-09	2.764190E-09	7.78E-03

Since ^{236}U is produced from ^{235}U, if the concentration of the second one is underestimated, then the concentration of the first one will also be underestimated. Finally, the difference of errors is explained because a small fraction of the reactions of ^{235}U generates ^{236}U.

4.6.5 Second step of the superposition

For the second step of superposition, it is possible to use some properties of the symmetry of the equations discussed in Section 4.5.2 with the purpose of reducing the amount of calculations to be done. Firstly, it is necessary to rewrite the modified Bateman equation for a linear chain, as:

$$X_n^L = X_1(0) \prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} F\left(\lambda_1^{\text{eff}}, \lambda_2^{\text{eff}}, \dots, \lambda_n^{\text{eff}}\right) \tag{4.81}$$

Where F was defined in equation (1.135). As it was discussed, one of the properties of the function F sets that $F\left(\lambda_1^{\text{eff}}, \lambda_2^{\text{eff}}, \dots, \lambda_n^{\text{eff}}\right) = F\left(\lambda_n^{\text{eff}}, \lambda_{n-1}^{\text{eff}}, \dots, \lambda_1^{\text{eff}}\right)$. In other words, the final isotope in the following two linear chains has the same function F:

$$\boxed{X_1 \to X_2 \to \cdots \to X_n}, \quad \boxed{X_n \to X_{n-1} \to \cdots \to X_1} \tag{4.82}$$

This property will be useful in the superposition case that is studied in this section, because when the linear chain is traveled in the opposite direction, it is possible to use the same function F for the second isotope in both linear chains. Using the following notation:

$$X_{i,j}^L, \qquad X_{i,j}(0) \tag{4.83}$$

In the last expressions, the index i denotes the isotope, whereas index j denotes the superposition step. With this notation, it is possible to show that for the linear chains in (4.82) the following equation is valid:

$$\frac{X_{n,1}^L \lambda_n^{\text{eff}}}{X_{1,1}(0)} \left(\prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}}\right)^{-1} = \frac{X_{1,2}^L \lambda_1^{\text{eff}}}{X_{n,2}(0)} \left(\prod_{k=1}^{n-1} b_{n-k+1,n-k}^{\text{eff}}\right)^{-1}$$

Thus:

$$X_{1,2}^L = X_{n,1}^L \frac{X_{n,2}(0) \lambda_n^{\text{eff}}}{X_{1,1}(0) \lambda_1^{\text{eff}}} \left(\prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}}\right)^{-1} \prod_{u=1}^{n-1} b_{n-u+1,n-u}^{\text{eff}} \tag{4.84}$$

In other words, the solution of the first isotope for the second superposition step was obtained in terms of the solution of the n isotope for the first superposition step. Therefore, for the calculations of $X_{^{235}\text{U}}^L$ for the second step of superposition, it is possible to use the solution of $X_{^{236}\text{U}}^L$ that was found in the first step. Replacing the values in (4.84) it follows that:

$$X^{L}_{^{235}\mathrm{U},2} = X^{L}_{^{236}\mathrm{U},1}\,\frac{X_{^{236}\mathrm{U},2}(0)}{X_{^{235}\mathrm{U},1}(0)}\,\frac{b^{\mathrm{eff}}_{^{236}\mathrm{U},^{235}\mathrm{U}}\lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}}{b^{\mathrm{eff}}_{^{235}\mathrm{U},^{236}\mathrm{U}}\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}} = 8.50948\mathrm{x}10^{-7}X^{L}_{^{236}\mathrm{U},1} \tag{4.85}$$

Under a similar reasoning, it is possible to extend the conclusions for the root-based equation:

$$X^{R}_{^{235}\mathrm{U},2} = 8.50948\mathrm{x}10^{-7}X^{R}_{^{236}\mathrm{U},1} \tag{4.86}$$

It is possible to note that both equations, (4.85) and (4.86) are being multiplied by the same constant factor $8.50948\mathrm{x}10^{-7}$, then it is possible to conclude that the error, between the two functions for the concentration of $^{235}\mathrm{U}$ in the second step of superposition, will be the same error for the first superposition step of the $^{236}\mathrm{U}$ concentration. Figure 4.10 shows the graph of the $^{235}\mathrm{U}$ concentration for the second superposition step. Again, the modified Bateman equation has been omitted in such graph, and it only shows the root-based solution, because both curves overlap. It can be verified that the curve has the same shape that Figure 4.8, but it is multiplied by a factor $8.50948\mathrm{x}10^{-7}$. As it can be observed, through equation (4.84) it is possible to save computational time, using the values that were found for the first superposition step. For the concentration of the isotope $^{236}\mathrm{U}$ in this second superposition step, the backward equation can be used:

$$X^{L}_{^{236}\mathrm{U},2} = \frac{1}{b_{^{236}\mathrm{U},^{235}\mathrm{U}}\lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}}\left(\frac{d}{dt}X^{L}_{^{235}\mathrm{U},2} + \lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}X^{L}_{^{235}\mathrm{U},2}\right) \tag{4.87}$$

Replacing equation (4.85) in equation (4.87), it follows:

$$X^{L}_{^{236}\mathrm{U},2} = 8.50948\mathrm{x}10^{-7}\,\frac{b_{^{235}\mathrm{U},^{236}\mathrm{U}}\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}}{b_{^{236}\mathrm{U},^{235}\mathrm{U}}\lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}}\left[\frac{1}{\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}b_{^{235}\mathrm{U},^{236}\mathrm{U}}}\left(\frac{d}{dt}X^{L}_{^{236}\mathrm{U},1} + \lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}X^{L}_{^{236}\mathrm{U},1}\right)\right]$$

Considering the following relationship:

$$\frac{\frac{d}{dt}X^{L}_{^{236}\mathrm{U},1} + \lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}X^{L}_{^{236}\mathrm{U},1}}{\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}b_{^{235}\mathrm{U},^{236}\mathrm{U}}} = X^{L}_{^{235}\mathrm{U},1} + X^{L}_{^{236}\mathrm{U},1}\,\frac{\left(\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}} - \lambda^{\mathrm{eff}}_{^{236}\mathrm{U}}\right)}{\lambda^{\mathrm{eff}}_{^{235}\mathrm{U}}b_{^{235}\mathrm{U},^{236}\mathrm{U}}}$$

Using the last equation, the expression (4.87) is rewritten as:

$$X^{L}_{^{236}\mathrm{U},2} = 4.588738\mathrm{x}10^{-3}X^{L}_{^{235}\mathrm{U},1} + 2.25929861\mathrm{x}10^{-2}X^{L}_{^{236}\mathrm{U},1} \tag{4.88}$$

Similarly

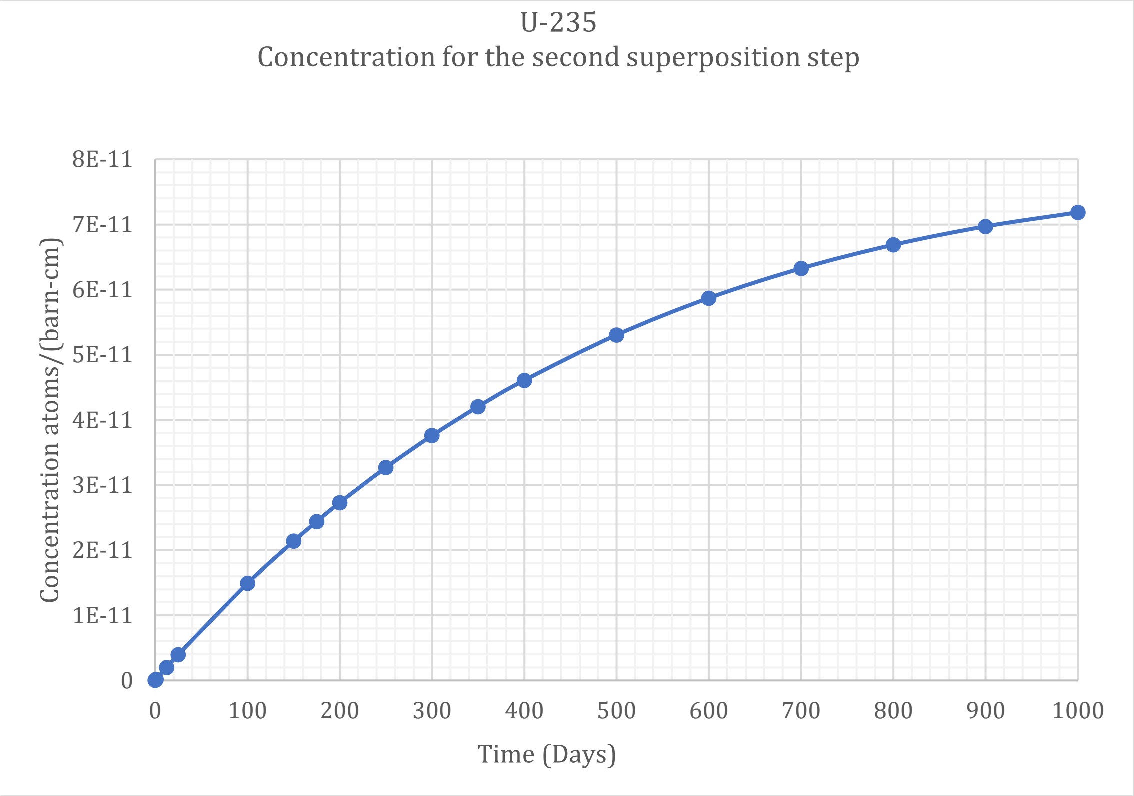

Figure 4. 10. Graph of the concentration of the isotope ^{235}U for the second superposition step. This graph has the same shape that the graph in Figure 4.8, but their values are multiplied by the factor $8.50948\mathrm{x}10^{-7}$.

$$X^{R}_{^{236}\mathrm{U},2} = 4.588738\mathrm{x}10^{-3} X^{R}_{^{235}\mathrm{U},1} + 2.25929861\mathrm{x}10^{-2} X^{R}_{^{236}\mathrm{U},1} \tag{4.89}$$

As it can be observed, it is possible to find the concentration of the isotope ^{236}U for the second step of superposition, in terms of the solution of the isotopes ^{235}U and ^{236}U found in the first step of superposition. In Figure 4.11 the graph of the concentration for the ^{236}U for the second step of superposition is shown. In this case, the graph has not the same shape that the one in Figure 4.7, because there is not symmetry between equations (4.88) and (4.89), and equations (4.71) and (4.74). The error between the root-based equation and the modified Bateman equation for this second step of superposition is shown in Figure 4.12. As in the first superposition step, it was not necessary to include equation (4.16) in the comparison, because it is equivalent to the modified Bateman equation. In fact, using a similar procedure that the one used in (4.77), the following denominator will be analyzed:

$$3.16249\mathrm{x}10^{-6} - 3.20995\mathrm{x}10^{-9}(1.19110\mathrm{x}10^{-10}\left(e^{-(2.43924\mathrm{x}10^{-9})t} - e^{-(1.82437\mathrm{x}10^{-8})t}\right) \tag{4.90}$$

The second term in this last expression fulfills the following inequaility:

$$3.20995\mathrm{x}10^{-9}(1.19110\mathrm{x}10^{-10}\left(e^{-(2.43924\mathrm{x}10^{-9})t} - e^{-(1.82437\mathrm{x}10^{-8})t}\right) \leq 2.42795\mathrm{x}10^{-19}$$

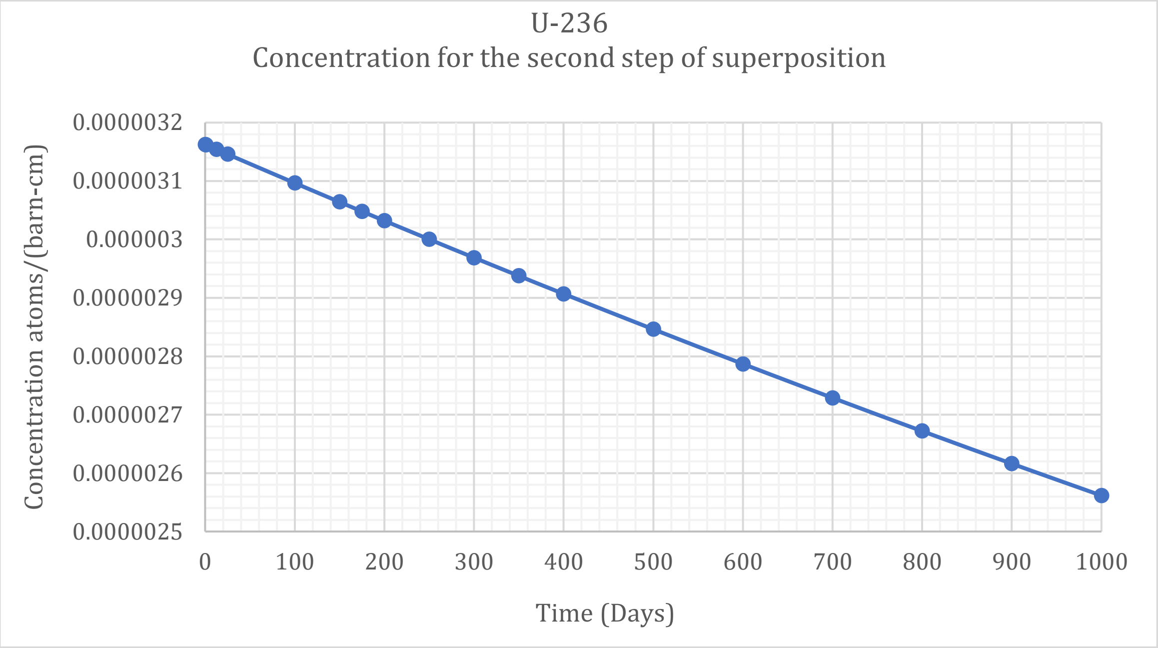

Figure 4. 11. Graph of the concentration of the isotope ^{236}U for the second step of superposition, computed with the root-based equation.

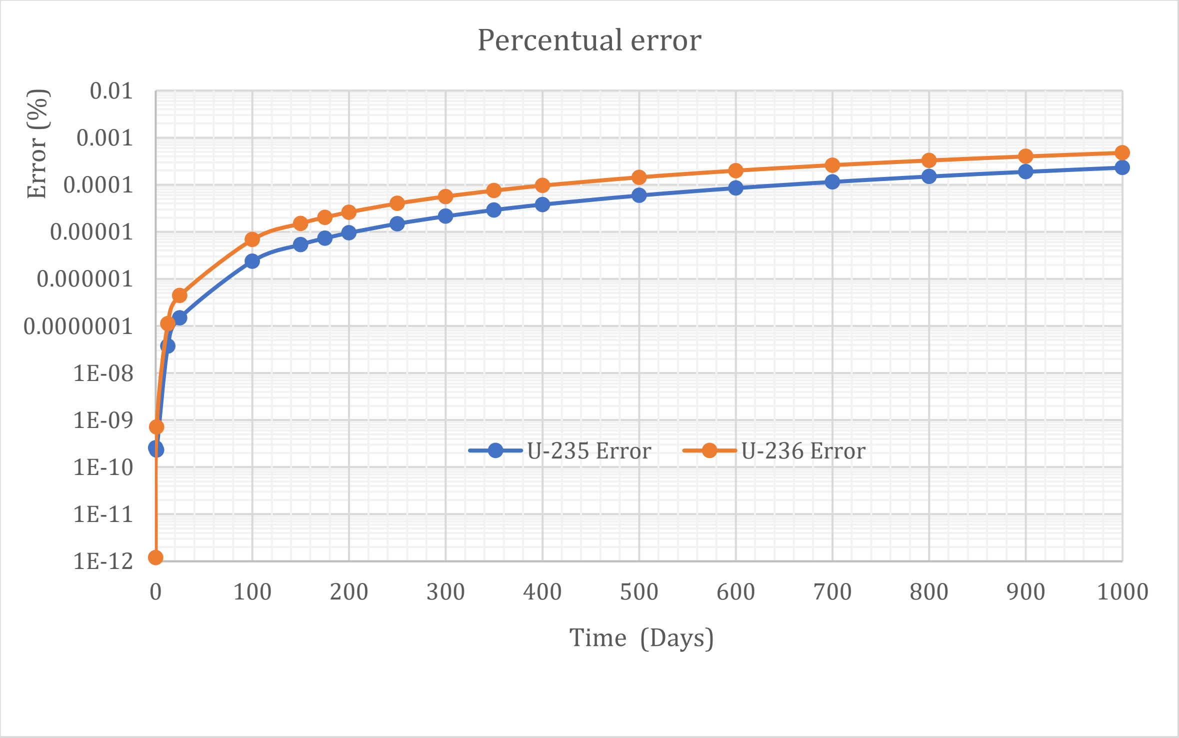

Figure 4. 12. Percentual error between the Root-based and the Modified Bateman solution for ^{235}U and ^{236}U, for the second superposition step.

Therefore, in terms of computational precision, equation (4.90) is equal to $3.16249\text{x}10^{-6}$, and therefore, equation (4.16) is reduced again to the modified Bateman equation.

4.6.6 Total concentration.

Finally, it is necessary to consider the two superposition steps in order to find the total concentration for the isotopes. Then, it follows that:

$$X^{R}_{^{235}\mathrm{U}} = X^{R}_{^{235}\mathrm{U},1} + X^{R}_{^{235}\mathrm{U},2} \quad \text{and} \quad X^{R}_{^{236}\mathrm{U}} = X^{R}_{^{236}\mathrm{U},1} + X^{R}_{^{236}\mathrm{U},2}$$

There are identical equations for $X^{L}_{^{235}\mathrm{U}}$ and $X^{L}_{^{236}\mathrm{U}}$. Practically, the contribution of the second superposition step to ^{235}U is insignificant in comparison with the value of the concentration of the first step. Therefore, the graph of the total concentration of this isotope is, essentially, the same that is shown in Figure 4.7. By the contrary, the total concentration of ^{236}U depends on the two superposition steps. In Figure 4.13 two contributions and the final concentration is showed. Again, the modified Bateman equation has been omitted in the graph, because its curve overlaps with the corresponding to the root-based equation.

Since the values of the microscopic cross section and the neutron flux that were used are very similar to the ones that are found in standard thermal nuclear reactor problems, these conclusions can be extended to other scenarios where the cyclic chain of ^{235}U and ^{236}U appears.

4.7 Final considerations of the superposition.

Equation (4.84) can be extended to a pure cyclic chain of length n where more than one isotope has an initial concentration different from zero. This generalization will be very useful, because it is possible to find relationships that save computing time. In order to discuss this generalization, the following pure cyclic chain will be considered:

$${}_{1}X^{1}_{1} \rightarrow {}_{2}X^{1}_{2} \rightarrow \cdots \rightarrow \boxed{{}_{n}X^{1}_{n}} \tag{4.91}$$

In the last equation three indexes have been used, the right lower one is used to distinguish the position of the isotopes in the structure under a numeration made from left to right. The left lower index is used to denote the isotope and the upper index denotes the superposition step. The following initial conditions will be used:

$${}_{i}X(t=0) \neq 0, 1 \leq i \leq n \tag{4.92}$$

In order to solve system (4.91) with the last initial conditions, it is necessary to break-down the procedure in n steps of superposition. In each of these steps, it will be considered that only one of the isotopes has a concentration different from zero, while the rest will be assumed with an initial concentration equal to zero. Therefore, in each superposition step, it will be necessary to define a new enumeration that will start with the isotope whose initial concentration is assumed different from zero.

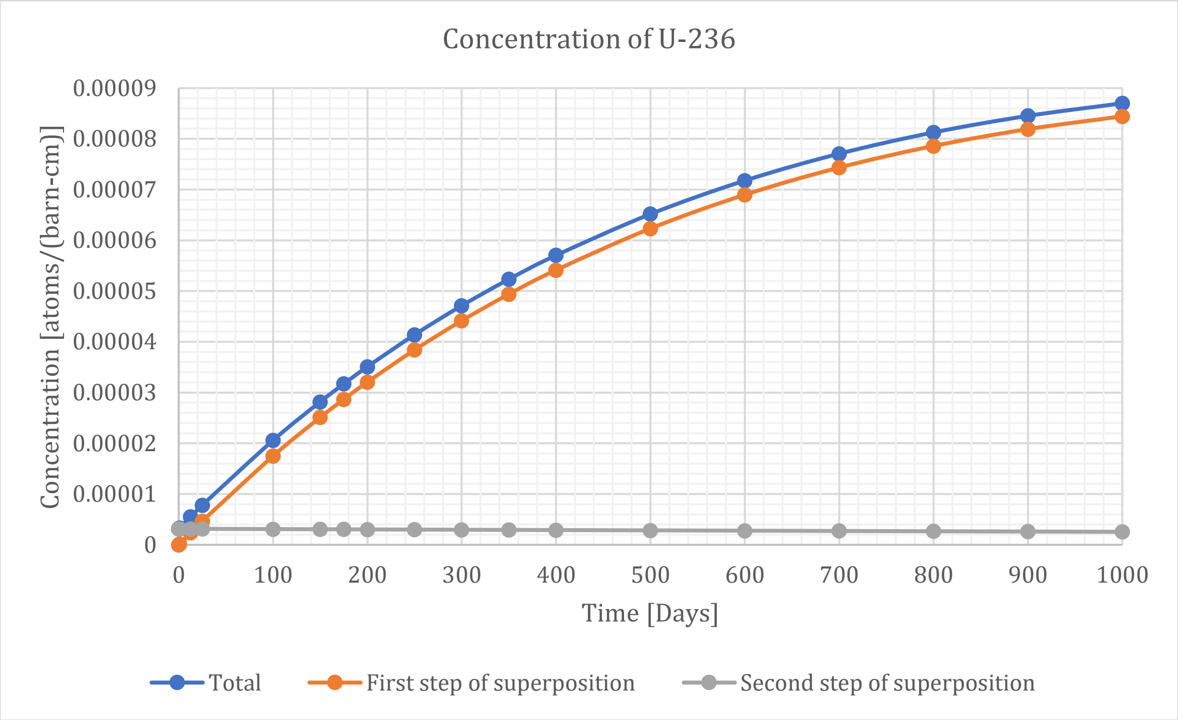

Figure 4. 13. Concentration of the isotope ^{236}U for the two superposition steps and for the total case.

Beginning with the first superposition step; it will be considered that only ${}_1X$ has an initial concentration different from zero. Then, the roots of the polynomial (4.41) corresponding to the system of (4.91) are computed, which will be denoted as ${}_1\beta_1^{\text{eff}},\ {}_2\beta_2^{\text{eff}}, \dots,\ {}_n\beta_n^{\text{eff}}$. With these roots, and assuming that only ${}_1X\ (0) \neq 0$ it is possible to set the root-based solution for the first superposition step for ${}_nX$ as:

$$
{}_nX^1 = \frac{{}_1X^1(0)}{{}_nb^{\text{eff}}_{n,1}\ {}_n\lambda^{\text{eff}}_n} \prod_{k=1}^{n} {}_kb^{\text{eff}}_{k,k+1}\ {}_k\lambda^{\text{eff}}_k\, F\left({}_1\beta_1^*,\ {}_2\beta_2^*, \dots,\ {}_n\beta_n^*\right) \qquad (4.93)
$$

In this case the right indexes in ${}_nX^1,\ {}_1X^1(0)$ and ${}_n\lambda^{\text{eff}}$ have been omitted, because their index position is the same that the index related to the isotope. For the second step, it is necessary to assume that only ${}_nX\ (0)$ is different from zero, and defining the new numeration of the isotopes belonging to the sequence (4.91):

$$
{}_nX_1^2 \to {}_1X_2^2 \to {}_2X_3^2 \to {}_3X_4^2 \to \cdots \to \boxed{{}_{n-1}X_n^2} \qquad (4.94)
$$

It can be noted that in this second case the isotope ${}_nX$ is considered as the “first” isotope in the cyclic chain. Besides this, there are no other differences in the properties of the cyclic chain defined in (4.91). It is possible to remove other indexes in equation (4.93). Firstly, since the roots of the polynomial in (4.41) do not depend on the initial concentration, neither on the numeration (or position) of the isotopes, it is possible to conclude that the roots of the system are the same that the roots of the system (4.94).

Besides, it is known that the function F is symmetrical in all its arguments, therefore the right index in the roots β^* can be ignored. Additionally, the left index in the product of the elements ${}_k b^{\text{eff}}_{k,k+1}\,{}_k\lambda^{\text{eff}}_k$ can be removed because in such multiplication all these parameters of the isotopes are involved. Then, when the solution of another isotope is built, this same product will be included, being this left index unnecessary. Considering the last arguments, the equation for the isotope ${}_{n-1}X^2$ of the system in (4.94) is:

$$ {}_{n-1}X_n^2 = \frac{{}_nX_1^2(0)}{{}_{n-1}b^{\text{eff}}_{n,1}\,{}_{n-1}\lambda^{\text{eff}}_n} \prod_{k=1}^{n} b^{\text{eff}}_{k,k+1}\lambda^{\text{eff}}_k\, F(\beta_1^*, \beta_2^*, \dots, \beta_n^*) \tag{4.95} $$

Using equation (4.93), it is possible to rewrite (4.95) as:

$$ {}_{n-1}X_n^2 = \frac{{}_nX_1^2(0)}{{}_1X^1(0)} \frac{{}_nb^{\text{eff}}_{n,1}\,{}_n\lambda^{\text{eff}}}{{}_{n-1}b^{\text{eff}}_{n,1}\,{}_{n-1}\lambda^{\text{eff}}_n}\,{}_nX^1 $$

Therefore, it is possible to compute the solution of the isotope ${}_{n-1}X$ in the second superposition step, using the solution ${}_nX^1$ found in the first superposition step. Following a similar reasoning, for the superposition step j with numeration given by:

$$ {}_{n-j+2}X_1^j \to {}_{n-j+3}X_2^j \to \cdots \to {}_nX_{j-1}^j \to {}_1X_j^j \to {}_2X_{j+1}^j \to \dots \to \boxed{{}_{n-j+1}X_n^j} \tag{4.96} $$

The following equation is valid:

$$ {}_{n-j+1}X_n^j = \mu^{j,1}_{n-j+1,n}\,{}_nX^1 \tag{4.97} $$

With:

$$ \mu^{j,1}_{n-j+1,n} = \frac{{}_nb^{\text{eff}}_{n,1}\,{}_n\lambda^{\text{eff}}\,{}_{n-j+2}X_1^j(0)}{{}_{n-j+1}b^{\text{eff}}_{n,1}\,{}_{n-j+1}\lambda^{\text{eff}}_n\,{}_1X^1(0)} \tag{4.98} $$

As it can be observed, using equation (4.98) it is possible to find the solution of the isotope with the last index n in each step of superposition, using the solution of the last isotope in the first step of superposition. Since the equations are proportional, it is only necessary to compute the solution ${}_nX^1$ for a set of time steps, and then multiply it by the factor in (4.98) in order to obtain the solution of the other superposition steps.

Now, is necessary to find relationships for the other isotopes in the first superposition step. This can be done through the backward method. Using equation (1.131) to equation (4.97), it follows that:

$$ {}_{n-j}X_{n-1}^j = \frac{\mu^{j,1}_{n-j+1,n}}{{}_{n-j}b^{\text{eff}}_{n-1,n}\,{}_{n-j}\lambda^{\text{eff}}_{n-1}} \left[\frac{d\,{}_nX^1}{dt} + {}_{n-j+1}\lambda^{\text{eff}}_n\,{}_nX^1\right] $$

Which can be rewritten as:

$$ {}_{n-j}X^{j}_{n-1} = \omega_j(0)\left[\varpi(0)\,{}_{n-1}X^{1} + \Delta_{n-j+1,n}\;{}_{n}X^{1}\right] \tag{4.99} $$

Where:

$$ \omega_j(u) = \mu^{j,1}_{n-j+1,n}\prod_{h=0}^{u}\frac{1}{{}^{j}b^{\mathrm{eff}}_{n-h-1,n-h}\quad {}^{j}\lambda^{\mathrm{eff}}_{n-h}},\ \varpi_j(u) = \prod_{h=0}^{u}\ {}^{1}b^{\mathrm{eff}}_{n-h-1,n-h}\quad {}^{1}\lambda^{\mathrm{eff}}_{n-h} $$

Where u is an integer, the upper left index in the branch ratios and effective removal coefficients denotes the superposition step and $\Delta_{x,y} = {}_{x}\lambda^{\mathrm{eff}} - {}_{y}\lambda^{\mathrm{eff}}$. Through equation (4.99), it is possible to find the concentration of the isotope in the position $n-1$ that belongs to the superposition step j. As it can be observed, such concentration is given as a linear combination of the concentration of the isotopes ${}_{n-1}X^{1}$ and ${}_{n}X^{1}$. Following a recursive procedure, it is possible to obtain an expression for the general case k:

$$ \begin{aligned} {}_{n-j+1-k}X^{j}_{n-k} = \omega_j(k-1)[\varpi(k-1)\,{}_{n-k}X^{1} \\ + \varpi(k-2)\left(\sum_{r=0}^{k-1}\Delta_{n-j+1-r,n-r}\right){}_{n-k+1}X^{1} \\ + \sum_{r=3}^{k}\varpi(k-r)\varrho_{\Delta}(r,k,j)\,{}_{n-1+r}X^{1} + \prod_{d=0}^{k-1}\Delta_{n-j+1-d,n}\;{}_{n}X^{1} \end{aligned} \tag{4.100} $$

Where $\varrho_{\Delta}(r,k)$ are coefficients related to the differences given by $\Delta_{x,y}$, which can be computed in a recursive way:

$$ \varrho_{\Delta}(r,k,j) = \begin{cases} \varrho_{\Delta}(r-1,k-1,j)\Delta_{n-j+2-k,n-(r-2)} + \varrho_{\Delta}(r,k-1,j), & 3 \le r \le k \\ \varrho_{\Delta}(r-1,k-1,j)\Delta_{n-j+2-k,n-(r-2)} + \prod_{d=0}^{k-1}\Delta_{n-j+1-d,n} & r > k \end{cases} $$

And, as part of the definition the following two equalities are defined:

$$ \varrho_{\Delta}(2,2,j) = \Delta_{n-j,n-1} + \Delta_{n-j+1,n}, \qquad \varrho(3,2) = \Delta_{n-j+1,n}\Delta_{n-j,n} $$

As equation (4.100) suggests, it is possible to find the solution of all the isotopes of other superposition steps, as a lineal combination of the solution of the isotopes for the first superposition step. Therefore, it is only necessary to compute the constants ω, ϖ, $\Delta_{x,y}$, and $\varrho_{\Delta}(r,k,j)$.

In general terms, the calculation of these constants is a less demanding task than computing the exponential functions and the product of inverse subtractions related with the terms ${}_{i}X^{1}$. Since basic arithmetical operations are less complex and less computational time consuming that the exponential function, it is expected that the calculations will be reduced when equation (4.100) is used.

It is worth mentioning that these last asseverations require a detailed algorithm analysis, and the use of the Big O notation, nevertheless these topics are beyond the scope of the present thesis, and these will be matter of a future research. Algorithm 4.3 summarizes the generalization of the superposition process and the use of equation (4.100).

Algorithm 4.3. Superposition process

Input: A pure cyclic chain of isotopes and their initial concentrations.

Output: The concentration for the isotopes of a pure cyclic chain for a set of time steps.

STEP 1: Build a set I that contains all the isotopes that belongs to the cyclic chain:

$$I \coloneqq \{i |\ i \text{ is a isotope of the pure cyclic chain}\}$$

STEP 2: Identify the isotopes of I that have an initial concentration different from zero and with them build the set I_0:

$$I_0 \coloneqq \{i \in I |\ {}_{i}X(0) \neq 0\}$$

STEP 3: Select an arbitrary isotope of I_0, which we will call i_0.
STEP 4: Starting from i_0, build the cyclic chain through the following sequence of isotopes:

$$i_0 \to i_{0+1} \to \cdots \to i_{0+card(I)-1}$$

Where:

$$\{i_0, i_{0+1}, \dots, i_{0+card(I)-1}\} = I$$

STEP 5: For the structure set in **STEP 4**, use equations (4.44), (4.46) to compute the concentration's function of the isotopes of the following sequence:

$${}_{i_0}X_1^1 \to {}_{i_{0+1}}X_2^1 \to \cdots \to {}_{i_0+card(I)-1}X_{card(I)}^1,$$

With:

$${}_{i_0}X_1^1(0) \neq 0, {}_{i_{0+h}}X_{1+h}^1(0) = 0, 1 \leq h \leq card(I) - 1$$

STEP 6. For j=1, 2, ..., card(I):

- Select the element i_{0+j}
- If $i_{0+j} \in I_0$:
  - Store the index j in the set K and build the sequence:
    $i_{0+j} \to i_{0+j+1} \to \cdots \to i_{0+card(I)-1} \to \cdots \to i_{0+j-1}$
  - Solve the following structure:
    ${}_{i_{0+j}}X_1^j \to {}_{i_{0+1}}X_2^1 \to \cdots \to {}_{i_0+card(I)-1}X_{card(I)}^1$
    Using (1.31) and **STEP 5** and the conditions:
    ${}_{i_0+j}X_1^j(0) \neq 0, {}_{i_{0+j+h}}X_{1+j+h}^j(0) = 0, 1 \leq h \leq card(I) - 1$
  - Store the computed concentrations

STEP 7. For u=0, 1, 2, ..., card(I)-1:

$${}_{i_{0+u}}X = {}_{i_{0+u}}X^1 + \sum_{k \in \mathrm{K}} {}_{i_{0+u}}X^k$$

4.8 case n=7.

4.8.1 Roots.

This case is related to the right side of Figure 4.5. Again, it is necessary to compute the roots of the following polynomial:

$$\left(s+\lambda^{\text{eff}}_{^{235}\text{U}}\right)\left(s+\lambda^{\text{eff}}_{^{236}\text{U}}\right)\left(s+\lambda^{\text{eff}}_{^{237}\text{U}}\right)\dots\left(s+\lambda^{\text{eff}}_{^{239}\text{Pu}}\right)-\alpha(7) \tag{4.101}$$

The roots of the last expression were found using the software Mathematica 11.3.0, which are shown in Table 4.5, with their respective values of β^{*}, as well as the percentual error given by their comparison with the effective removal coefficients. As in the case of $n=2$, it can be noted that the negative of the roots of the polynomial are very close to the effective removal coefficients. Particularly, the lowest percentual error is related to the isotopes ^{238}Np and ^{239}Pu.

Particularly, the lowest percentual error is related to the isotopes ^{238}Np and ^{239}Pu.

4.8.2 Results for ^{239}Pu

Using equation (4.44), the root-based equation for ^{239}Pu is equal to:

$$X^{A}_{^{239}\text{Pu}}(t)=X^{A}_{7}(t)=X_1(0)\prod_{k=1}^{6}b^{\text{eff}}_{k,k+1}\lambda^{\text{eff}}_{k}\sum_{i=1}^{7}e^{-\beta^{*}_{i}\,t}\prod_{\substack{j=1\\ j\neq i}}^{7}\frac{1}{\left(\beta^{*}_{j}-\beta^{*}_{i}\right)} \tag{4.102}$$

For the case of the modified Bateman equation, it follows:

$$X^{L}_{^{239}\text{Pu}}(t)=X^{L}_{7}(t)=X_1(0)\prod_{k=1}^{6}b^{\text{eff}}_{k,k+1}\lambda^{\text{eff}}_{k}\sum_{i=1}^{7}e^{-\lambda^{\text{eff}}_{i}\,t}\prod_{\substack{j=1\\ j\neq i}}^{7}\frac{1}{\left(\lambda^{\text{eff}}_{j}-\lambda^{\text{eff}}_{i}\right)} \tag{4.103}$$

The concentration of ^{239}Pu was obtained using the data in Tables 4.1-4.3 for a for a range of 5 days to 1000 days. Figure 4.14 shows both graphs, which are overlapped as in the case $n=2$.

Isotope	β	β^{*}	λ^{eff}	E(%)
^{235}U	-1.82437 x10^{-8}	1.82437 x10^{-8}	1.82436x10^{-8}	2.15839 x10^{-4}
^{236}U	-2.439230 x10^{-9}	2.439230 x10^{-9}	2.43923947 x10^{-9}	3.88253 x10^{-4}
^{237}U	-1.20616x10^{-6}	1.20616x10^{-6}	1.2061559x10^{-6}	3.34371x10^{-4}
^{237}Np	-1.035350x10^{-8}	1.035350x10^{-8}	1.035336x10^{-8}	1.322642x10^{-3}
^{238}Np	-3.848060x10^{-6}	3.848060x10^{-6}	3.848058x10^{-6}	4.115806x10^{-5}
^{238}Pu	-1.22411x10^{-8}	1.22411x10^{-8}	1.224119 x10^{-8}	7.927498 x10^{-4}
^{239}Pu	-6.03196 x10^{-8}	6.03196 x10^{-8}	6.03196184 x10^{-8}	3.04933 x10^{-5}

Table 4. 5. Roots and their comparison with the effective removal coefficients for the structure of the right side of Figure 4.5.

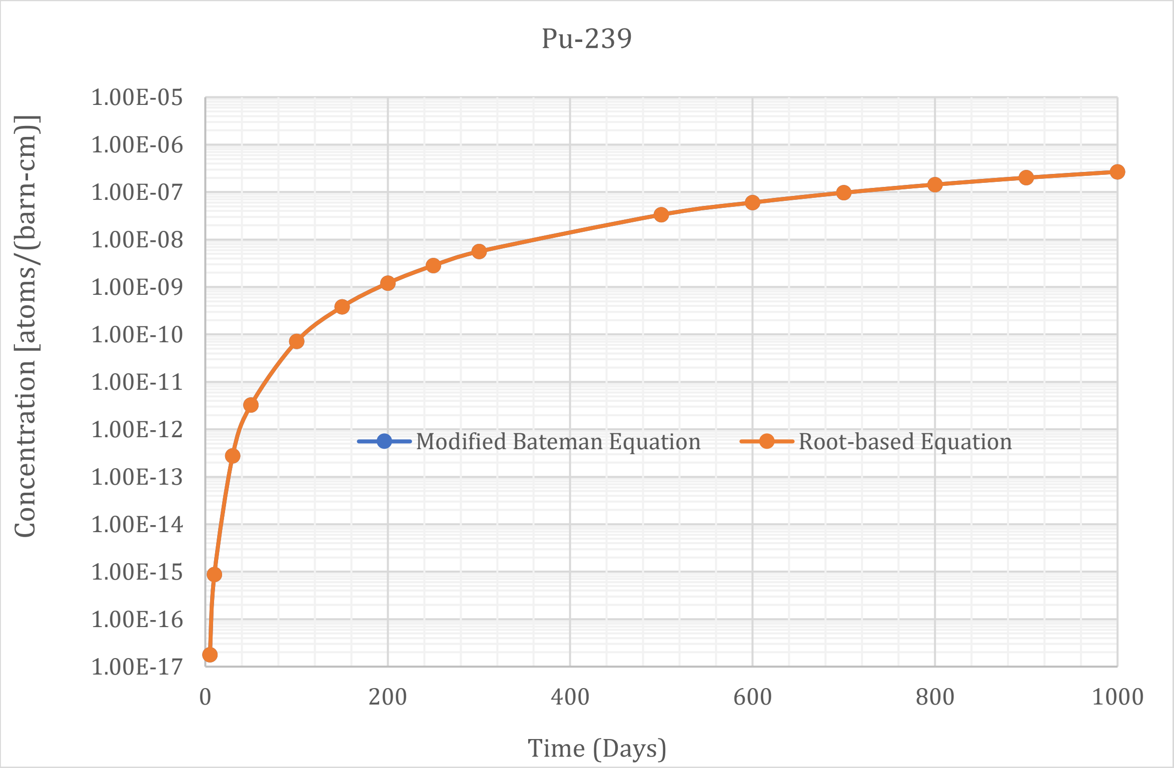

Figure 4. 14. Concentration for ^{239}Pu using the Modified Bateman Equation, and the Root Based Equation. The two curves overlap.

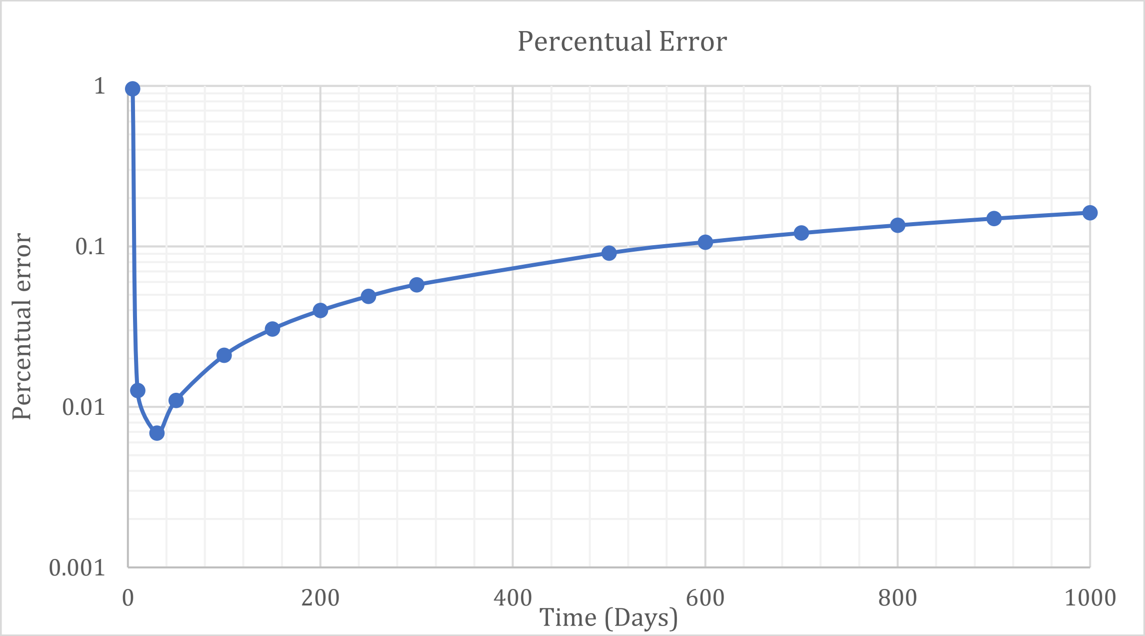

Figure 4. 15. Percentual error between the Modified Bateman Equation and the Root-based equation for the concentration of ^{239}Pu.

For this case a precision of 50 digits has been used, because several of the isotopes of the cyclic chain have a very small effective removal coefficients, which can produce that the equations yield negative values of concentration if the computational precision is not increased. This situation will be discussed in more detail in Section 4.8.3. For the percentual error that is shown in Figure 4.15, it is possible to note a similar behavior that the one related to Figure 4.9 and Figure 4.12. Except for the first point, that has a percentual error close to 1%, the rest of data shows an increasing error that starts in an order of 10^{-3}, and then it increases slowly to an error of the order of 10^{-1}.

In comparison with the case $n = 2$, where the error for the two superposition steps remained below of 0.01%, in this case, the percentual error remains below 1%. The main source of this error is related to the product of the inverse of the subtractions $\beta_j^* - \beta_i^*$ in equation (4.102), and with the corresponding term related to $\lambda_j^{\text{eff}} - \lambda_i^{\text{eff}}$ in equation (4.103).

Even when the percentual error between these parameters, β^* and λ^{eff}, were of the same order (or even of a lower order) of the corresponding percentual error of case $n = 2$, in this case there are several terms involved in the mentioned product, which was not the case with the cyclic chain given by ${}^{235}\text{U} \rightarrow \boxed{{}^{236}\text{U}}$.

In fact, in this last cyclic chain it is only necessary to compute the inverse of the differences: $\lambda_{^{236}\text{U}}^{\text{eff}} - \lambda_{^{235}\text{U}}^{\text{eff}}$, and $\beta_{^{236}\text{U}}^* - \beta_{^{235}\text{U}}^*$. Therefore, when more differences of terms that are close are involved, the error can increase. Nevertheless, this error is still acceptable. With respect to the first two points, at 5 and 10 days, it is possible to omit them due to lack of precision. As it will be shown in the following section, when small time steps are considered, it is necessary to increases the precision to values beyond 50 digits, in order to compute the error in a right way. The data suggest that this cyclic chain can be approximated using the modified Bateman equation for a linear chain.

4.8.3 Precision issues and negative values.

A recurrent issue with the Bateman equations is related to precision and the round-off difficulties when very small removal coefficients are used. These topics were identified since the beginning of the development of the linear chain method in the 50's and 60's. The first author that addressed this topic in a very detailed work was D. R. Vondy (Vondy, 1962), who carried out a complete analysis about the way in which the modified Bateman solution can be implemented in digital machine calculations, particularly in the IBM-7090 computer on Fortran programming language.

His work was based in the Lietzke and Claiborne (Lietzke and Claiborne, 1960), and Breslauer and Karricker (Breslauer and Karricker, 1960) articles. However, it seems that the precision and the round-off difficulties become the main disadvantage of the linear chain method, which made it the least popular among the burnup and activations codes, for at least two decades. Based on a description found in Tobias's work in 1980 (Tobias, 1980), where he suggested that this method was plagued with errors of this type, it is possible to conclude that the linear chain method was not popular in the early stages of the development of burnup codes.

Time		Precision					
Time	Time (s)	15	30	40	50	100	150
1 min	60	-3.42E-09	-2.86E-13	-4.06E-17	5.04E-20	3.97E-34	1.73E-40
2 min	120	-3.42E-09	-1.13E-13	-2.13E-16	1.66E-20	4.16E-34	1.11E-38
1 hr	3600	-3.44E-09	-1.87E-13	-1.13E-16	4.80E-20	8.07E-30	8.07E-30
6 hrs	21600	-7.20E-09	-1.99E-13	-1.60E-16	1.66E-19	3.71E-25	3.71E-25
12 hrs	43200	-7.33E-09	-2.05E-13	-5.75E-17	6.84E-20	2.34E-23	2.34E-23
1 day	86400	-7.61E-09	-2.36E-13	-3.39E-17	1.98E-19	1.45E-21	1.45E-21
5 days	432000	-2.50E-09	-1.88E-13	-2.17E-16	1.79E-17	1.79E-17	1.79E-17
10 days	864000	-8.78E-09	-2.86E-13	7.27E-16	8.79E-16	8.79E-16	8.79E-16
30 days	2592000	-4.15E-09	9.84E-14	2.78E-13	2.79E-13	2.79E-13	2.79E-13
50 days	4320000	-5.82E-09	2.92E-12	3.24E-12	3.24E-12	3.24E-12	3.24E-12
100 days	8640000	-2.67E-09	7.11E-11	7.12E-11	7.12E-11	7.12E-11	7.12E-11
150 days	12960000	-2.51E-09	3.84E-10	3.84E-10	3.84E-10	3.84E-10	3.84E-10
200 days	17280000	-2.69E-09	1.21E-09	1.21E-09	1.21E-09	1.21E-09	1.21E-09
250 days	21600000	-3.32E-10	2.84E-09	2.84E-09	2.84E-09	2.84E-09	2.84E-09
300 days	25920000	3.49E-10	5.60E-09	5.60E-09	5.60E-09	5.60E-09	5.60E-09

Table 4. 6. Concentration for ^{239}Pu computed with the Modified Bateman equation, for several precision values.

Certainly, there were authors that dealt with this problem in the past decades, included Tobias himself (Tobias, 1978), who developed new ways to write the Bateman equations in order to avoid these difficulties. Nowadays, some authors like Chaitanya Tadepalli and his colleagues (Chaitanya Tadepalli, 2016) continue to analyze this topic, and proposed mathematical tools to deal with precision problems. In the present case, in order to perform a right comparison, it is necessary to deal with this problem increasing the precision and selecting an adequate range of time. Using the Python language, version 3.5, and the multi-precision library *bigfloat*, an analysis of equation (4.102) and (4.103) is carried out varying the precision. Table 4.6 presents the results for the modified Bateman equation, for a range of time from 60 seconds to 1000 days. Table 4.7 shows the results for the root-based equation with the same conditions.[11]

Table 4. 7. Concentration for ^{239}Pu computed with the Modified Bateman equation, for several precision values.

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[11] Only two precision digits has been included in both tables, because the purpose of this analysis is to show negative values and the consistency of the data.

Time		Precision					
Time	Time (s)	15	30	40	50	100	150
1 min	60	1.00E-08	3.19E-13	-1.06E-17	-3.97E-20	-7.06E-35	1.73E-40
2 min	120	1.00E-08	3.61E-13	-8.05E-18	-7.95E-20	1.53E-34	1.11E-38
1 hr	3600	1.00E-08	2.22E-13	-6.78E-18	-7.95E-20	8.07E-30	8.07E-30
6 hrs	21600	9.56E-09	3.75E-13	-1.06E-17	-1.32E-19	3.71E-25	3.71E-25
12 hrs	43200	5.46E-09	3.89E-13	-8.05E-18	-1.32E-19	2.34E-23	2.34E-23
1 day	86400	8.19E-09	4.44E-13	-7.21E-18	-1.46E-19	1.45E-21	1.45E-21
5 days	432000	5.01E-09	4.72E-13	9.32E-18	1.77E-17	1.79E-17	1.79E-17
10 days	864000	7.28E-09	4.72E-13	8.71E-16	8.79E-16	8.79E-16	8.79E-16
30 days	2592000	9.56E-09	6.81E-13	2.79E-13	2.79E-13	2.79E-13	2.79E-13
50 days	4320000	5.01E-09	3.64E-12	3.24E-12	3.24E-12	3.24E-12	3.24E-12
100 days	8640000	7.74E-09	7.16E-11	7.12E-11	7.12E-11	7.12E-11	7.12E-11
150 days	12960000	1.14E-08	3.84E-10	3.84E-10	3.84E-10	3.84E-10	3.84E-10
200 days	17280000	9.10E-09	1.21E-09	1.21E-09	1.21E-09	1.21E-09	1.21E-09
250 days	21600000	6.83E-09	2.84E-09	2.84E-09	2.84E-09	2.84E-09	2.84E-09
300 days	25920000	1.00E-08	5.60E-09	5.60E-09	5.60E-09	5.60E-09	5.60E-09

All the results where the concentration has a negative value have been highlighted. As it can be observed, when 15 digits of precision are used, there are problems in both equations. In the case of the modified Bateman equation this precision is not enough to model the time steps that are lower of 300 days, and it produces negative results. In the other case, with the root-based equation (Table 4.7) there are not negative values, but instead there is not consistency in the results if they are compared with the values of the sixth column, which is considered as the correct data for this case.

Using precision 30, which was the precision that was used in the case $n = 2$, difficulties still arise. In the case of the modified Bateman equation, there are negative values for time steps less or equal to 10 days. For the root-based equation, there are inconsistent values, and particularly a result for a time step of 5 days with a percentual error given by:

$$E_{\%} = 100\% \cdot \frac{|4.72\text{x}10^{-13} - 1.79\text{x}10^{-17}|}{1.79\text{x}10^{-17}} = 2636771.508\% \qquad (4.104)$$

For the precision of 50 digits, the results are consistent for a range of time of 5 days onwards, and this was the reason that this precision was chosen to make a comparison of ^{239}Pu in the past section. Nevertheless, this precision is not enough to model the concentration value for values of less of 5 days. As it was mentioned before, the first two points in the graph of Figure 4.15 can be omitted, because they have their origin in a lack of precision. This can be verified in Table 4.6 and Table 4.7. Observing the sixth column of both tables, it is possible to conclude that the value of the concentration for a time of 5 days is close to $1.79x10^{-17}$. Since the sixth column uses the greatest precision value, such value is considered as the right one.

Nevertheless, in the fourth column in Table 4.7, which corresponds to the root-based equation, there is a value of the concentration for the same day, but with a precision of 50 is $1.77x10^{-17}$. Therefore, there is a difference with the same equation using two different values of precision, which is approximately equal to:

$$E_{\%} \approx 100\% \cdot \frac{\left|1.77483\text{x}10^{-17} - 1.79070\text{x}10^{-17}\right|}{1.79070\text{x}10^{-17}} = 0.8862\% \qquad (4.105)$$

This error is close to the first value of the graph of Figure 4.15, which is approximately equal to 0.9602%. If Table 4.6 is analyzed, which shows the data computed with the modified Bateman equation, it is possible to conclude that the modified Bateman equation gives a better result for the same time step and the precision of 50 digits. Nevertheless, if the percentual error between the results of the Modified Bateman Equation and the Root-based Equation is computed, using precision 100 instead of 50, it follows that for a time of 5 days this error approximately equal to:

$$E_{\%} = 100\% \cdot \frac{\left|1.79070\text{x}10^{-17} - 1.79187\text{x}10^{-17}\right|}{1.79070\text{x}10^{-17}} = 0.0012\% \qquad (4.106)$$

Which is a value close to the correct percentual error. Similarly, for the point of 10 days, there is a correct value of 0.0024%, instead of 0.012%. In practical cases these concentration values are very small, and usually the time steps related to them are of order of days. In fact, only a reduced number of isotopes are studied with small time steps, as it is the case of Xenon, whose study usually consider time steps of 50 hours onward (Itagaki et. al., 1993). This analysis was carried out in order to explain the apparent discrepancy in the first two points of Figure 4.1, and the need to increase the precision. Finally, it is possible to conclude that the percentual error in this cyclic chain is strongly influenced by the precision used, as it can be observed in Figure 4.16.

4.8.4 Results for ^{238}Pu

For the isotope ^{238}Pu, whose position in the cyclic chain is equal to $n-1=6$, it is possible to use the root-based equation (4.45):

$$X^{A}_{^{238}\text{Pu}} = X^{A}_{6} = \left(X_{^{235}\text{U}}(0) \prod_{k=1}^{4} b^{\text{eff}}_{k,k+1} \lambda^{\text{eff}}_{k} \sum_{i=1}^{7} e^{-\beta_i^{*}\, t} \prod_{\substack{j=1 \\ j\neq i}}^{7} \frac{\left(\lambda^{\text{eff}}_{^{239}\text{Pu}} - \beta_i^{*}\right)}{\left(\beta_j^{*} - \beta_i^{*}\right)} \right) \qquad (4.107)$$

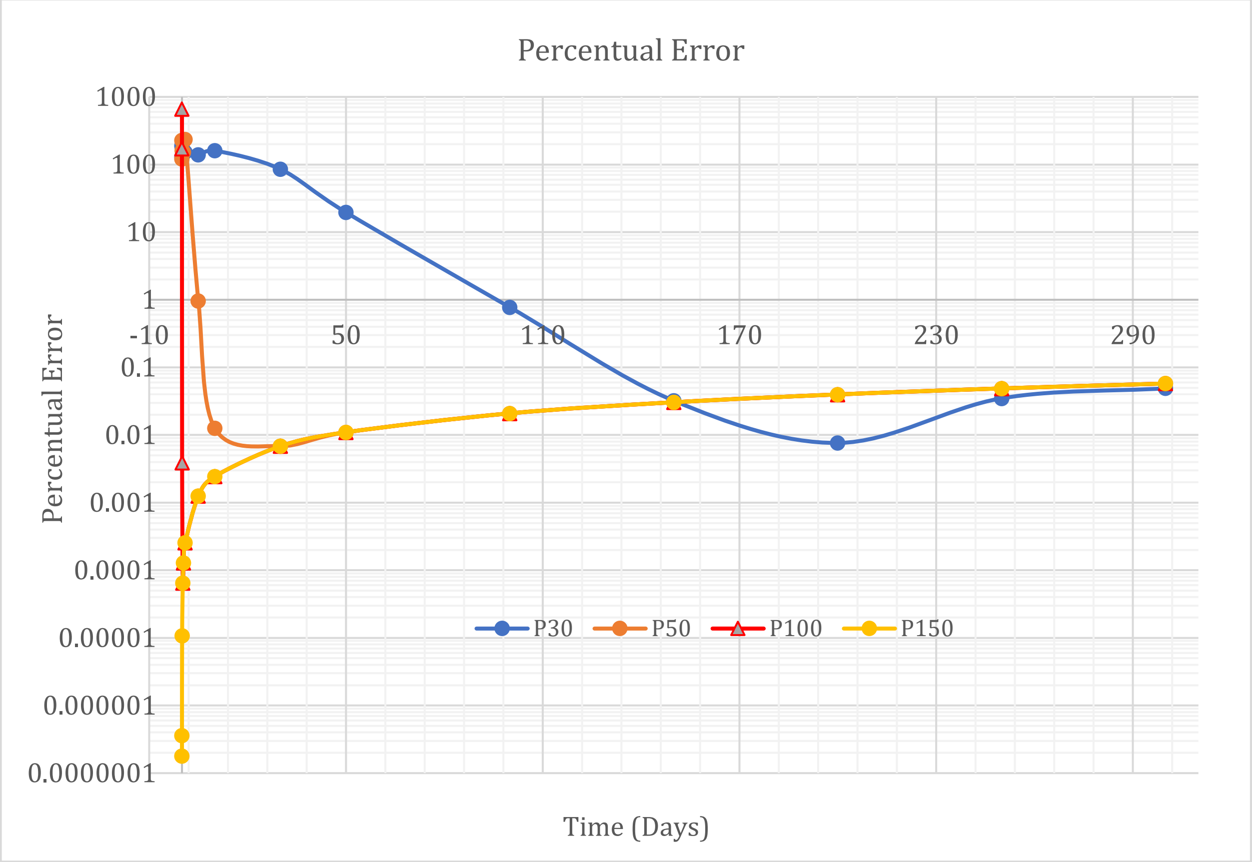

Figure 4. 16. Percentual error between the Modified Bateman Equation and the Root-Based Equation, for the isotope ^{239}Pu, as a function of time and with different values of precision.

The modified Bateman equation for this isotope is given by:

$$X^{L}_{^{238}\mathrm{Pu}} = X^{L}_{6} = X_{^{235}\mathrm{U}}(0) \prod_{k=1}^{5} b^{\mathrm{eff}}_{k,k+1} \lambda^{\mathrm{eff}}_{k} \sum_{i=1}^{6} e^{-\lambda^{\mathrm{eff}}_{i} t} \prod_{\substack{j=1 \\ j \neq i}}^{6} \frac{1}{\left(\lambda^{\mathrm{eff}}_{j} - \lambda^{\mathrm{eff}}_{i}\right)} \qquad (4.108)$$

With the same data and precision that was used for the isotope ^{239}Pu, the ^{238}Pu concentration was computed using equations (4.107) and (4.108). Figure 4.17 contains the graph for both equations, and Figure 4.18 contains the percentual error between these equations. Again, the curves overlap, even when there is a small difference in the first time-steps. This difference can be explained by the lack of precision as in the case of ^{239}Pu. In Figure 4.18 a percentual error curve with a greater precision is showed, which confirms this. The percentual error is less than 1% for all the time steps. Therefore, the results suggest that the linear chain can be a very good approximation to solve this particular cyclic chain. It was found that the results for the concentration of the isotopes ^{238}Np, ^{237}Np, ^{237}U and ^{236}U are very similar to the results of ^{239}Pu and ^{238}Pu. In fact, for all these isotopes the graphs overlap, and the error has the same behavior. Therefore, it was considered convenient to discuss only the ^{235}U of the rest of isotopes of the cyclic chain, because the forward method, discussed in Section 1.6, can be used to solve it.

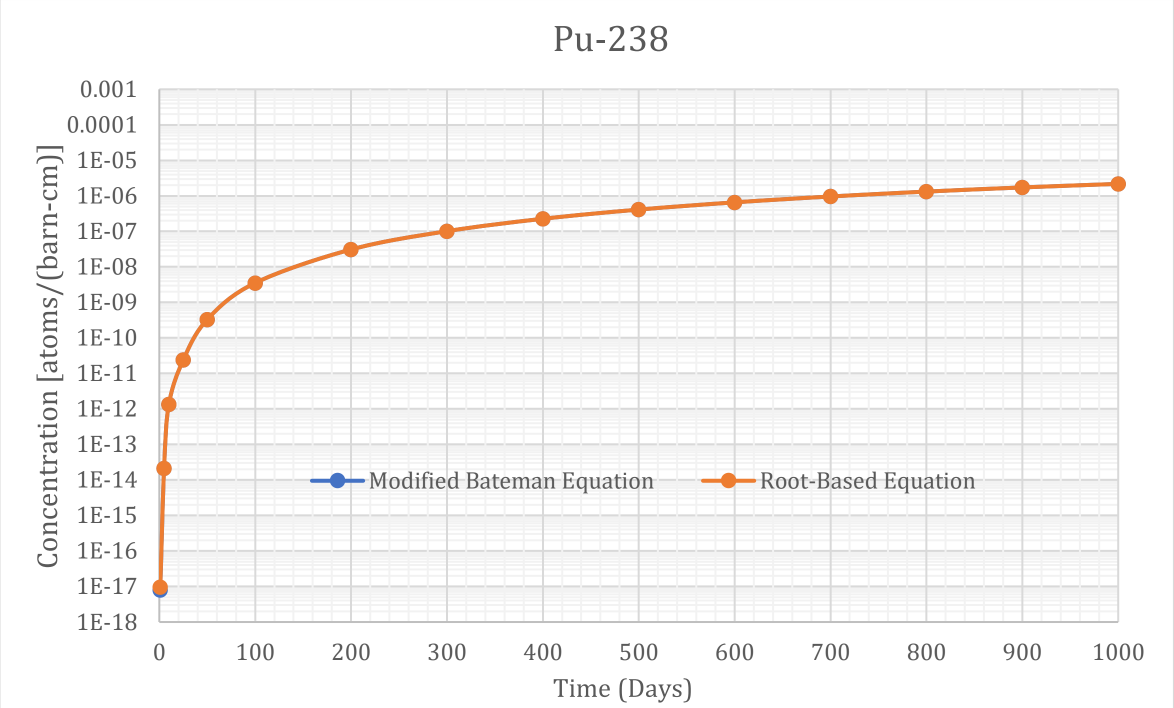

Figure 4. 17. Concentration for ^{238}Pu compute with the Modified Bateman Equation, and the Root Based Equation. The two curves overlap.

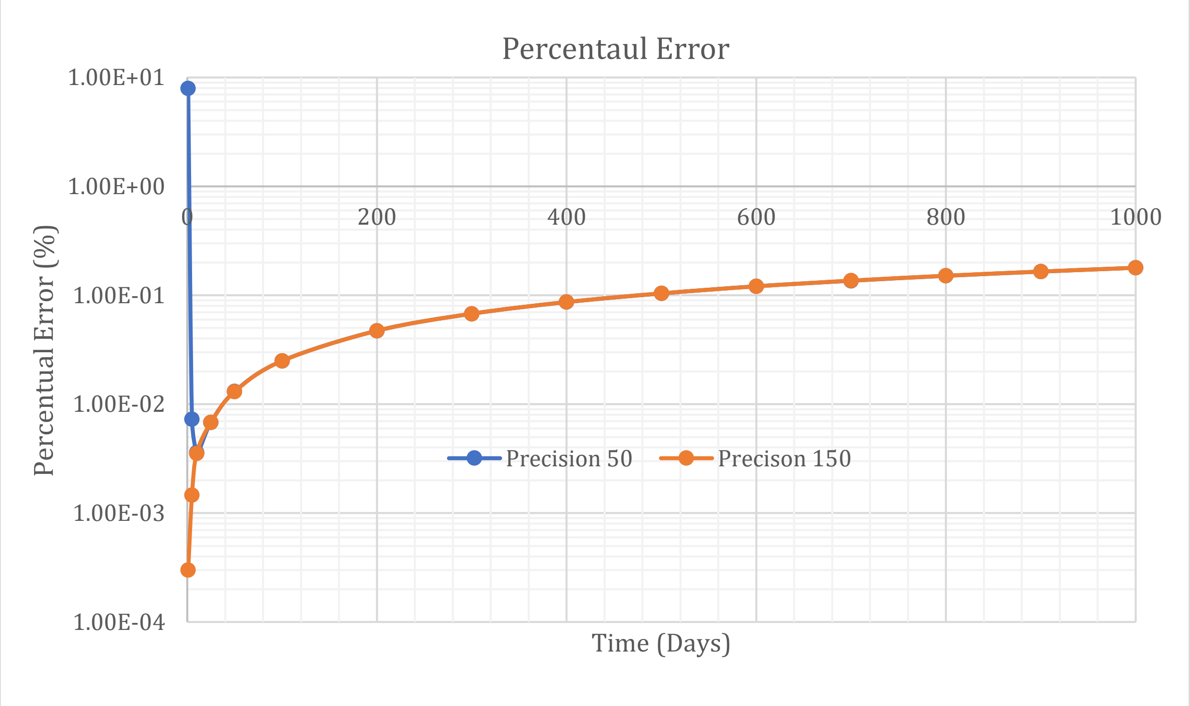

Figure 4. 18. Percentual error between the Modified Bateman Equation and the Root-Based Equation, for the isotope ^{238}Pu, as a function of time, and with two different values of precision.

4.8.5 The forward method for the root-based Equation.

Using the forward method given in (1.56) it is possible to find X_1^A in a cyclic chain, knowing the solution for X_n^A. Starting from equation (4.44) it follows that:

$$X_1^A = e^{-\lambda_1^{\text{eff}} t}\left(b_{n,1}^{\text{eff}} \lambda_n^{\text{eff}} \int_0^t X_n^A(t') e^{\lambda_1^{\text{eff}} t'}\, dt' + X_1^A(0) \right) \tag{4.109}$$

Firstly, the following term is computed:

$$\int_0^t X_n^A(t') e^{\lambda_1^{\text{eff}} t'} = \int_0^t X_1(0) \prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{n} e^{-\beta_i^* t'} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} e^{\lambda_1^{\text{eff}} t'} dt' \tag{4.110}$$

$$= X_1(0) \prod_{k=1}^{n-1} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \left(\sum_{i=1}^{n} \frac{e^{-(\beta_i^* - \lambda_1^{\text{eff}})t} - 1}{\lambda_1^{\text{eff}} - \beta_i^*} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \right) \tag{4.111}$$

If the subtraction is expanded:

$$\sum_{i=1}^{n} \frac{e^{-(\beta_i^* - \lambda_1^{\text{eff}})t} - 1}{\lambda_1^{\text{eff}} - \beta_i^*} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} = \sum_{i=1}^{n} \left(\frac{e^{-(\beta_i^* - \lambda_1^{\text{eff}})t}}{\lambda_1^{\text{eff}} - \beta_i^*} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} - \frac{1}{\beta_i^* - \lambda_1^{\text{eff}}} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \right)$$

Considering the definition where $\beta_{n+1}^* = \lambda_1^{\text{eff}}$, the term $\lambda_1^{\text{eff}} - \beta_i^*$ can be included in the product under the index j:

$$= \sum_{i=1}^{n} e^{-(\beta_i^* - \lambda_1^{\text{eff}})t} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{(\beta_j^* - \beta_i^*)} - \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.112}$$

Using the Reduction Theorem proved in Section 1.8.1:

$$-\sum_{i=1}^{n} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{\beta_j^* - \beta_i^*} = \prod_{j=1}^{n} \frac{1}{\beta_j^* - \beta_{n+1}^*} \tag{4.113}$$

Therefore:

$$\begin{aligned} &= \sum_{i=1}^{n} \frac{e^{-(\beta_i^* - \lambda_1^{\text{eff}})t} - 1}{\lambda_1^{\text{eff}} - \beta_i^*} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \\ &= \sum_{i=1}^{n} e^{-(\beta_i^* - \lambda_1^{\text{eff}})t} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{(\beta_j^* - \beta_i^*)} + \prod_{j=1}^{n} \frac{1}{\beta_j^* - \beta_{n+1}^*} \end{aligned} \tag{4.114}$$

Using the last expressions, the term $e^{-\lambda_1^{\text{eff}}t} b_{n,1}^{\text{eff}} \lambda_n^{\text{eff}} \int_0^t X_n^A(t') e^{\lambda_1^{\text{eff}}t'}\, dt'$ is equal to:

$$= X_1(0) \prod_{k=1}^{n} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \left(\sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{(\beta_j^* - \beta_i^*)} + \prod_{j=1}^{n} \frac{1}{\beta_j^* - \beta_{n+1}^*} e^{-\lambda_1^{\text{eff}} t} \right) \tag{4.115}$$

The last term can be written as:

$$= \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{\beta_j^* - \beta_i^*} e^{-\beta_i^* t}, \qquad i = n + 1 \tag{4.116}$$

Therefore, the equation (4.113) is equal to:

$$X_1(0) \prod_{k=1}^{n} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{n+1} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{\beta_j^* - \beta_i^*} \tag{4.117}$$

Thus, equation (4.109) can be rewritten as:

$$X_1^A = X_1(0) e^{-\lambda_1^{\text{eff}} t} + X_1(0) \prod_{k=1}^{n} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{n+1} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{n+1} \frac{1}{\beta_j^* - \beta_i^*} \tag{4.118}$$

This last equation is very useful, because it allows us to distinguish the solution related to the linear chain case, which correspond to the first term, from the contribution due to the cyclic chain.

4.8.6 Concentration of $^{235}\mathrm{U}$

Even when it is possible to use equation (4.44) to find $X_{^{235}\mathrm{U}}^A$, the forward method and equation (4.118) will be used instead, because this expression allows comparing the root-based equation and the modified Bateman equation in a simpler way. Using such equation, it follows that:

$$X_{^{235}\mathrm{U}}^A = X_{^{235}\mathrm{U}}^A(0) e^{-\lambda_{^{235}\mathrm{U}}^{\text{eff}} t} + X_{^{235}\mathrm{U}}^A(0) \prod_{k=1}^{7} b_{k,k+1}^{\text{eff}} \lambda_k^{\text{eff}} \sum_{i=1}^{8} e^{-\beta_i^* t} \prod_{\substack{j=1 \\ j \neq i}}^{8} \frac{1}{\beta_j^* - \beta_i^*} \tag{4.119}$$

On the other hand, the modified Bateman equation for this isotope is given by:

$$X_{^{235}\mathrm{U}}^L = X_{^{235}\mathrm{U}}^A(0) e^{-\lambda_{^{235}\mathrm{U}}^{\text{eff}} t} \tag{4.120}$$

Using this last equation, equation (4.119) can be rewritten as:

$$X_{^{235}\text{U}}^{A} = X_{^{235}\text{U}}^{L} + X_{^{235}\text{U}}^{A}(0)\prod_{k=1}^{7} b_{k,k+1}^{\text{eff}}\lambda_{k}^{\text{eff}}\sum_{i=1}^{8} e^{-\beta_i^* t}\prod_{\substack{j=1\\ j\neq i}}^{8}\frac{1}{\beta_j^* - \beta_i^*} \tag{4.121}$$

From the last equation it is possible to identify the contribution that is associated with the structure of the cyclic chain, and particularly with the reaction $^{239}\text{Pu} \overset{\alpha}{\rightarrow} {}^{235}\text{U}$. Figure 4.19 shows the graph of equations (4.120) and (4.121). As in the past cases, the curves overlap. The percentual error is shown in Figure 4.20. Again, a precision 50 has been used to compute this error. The contribution due to the cyclic chain is practically neglected. In fact, for a range of $1\text{ Day} \leq t \leq 1000\text{ Days}$ the following inequality is obtained

$$\begin{aligned} 1.64547\text{x}10^{-29}\left[\frac{\text{atoms}}{\text{b}\cdot\text{cm}}\right] \leq X_{^{235}\text{U}}^{A}(0)\prod_{k=1}^{7} b_{k,k+1}^{\text{eff}}\lambda_{k}^{\text{eff}}\sum_{i=1}^{8} e^{-\beta_i^* t}\prod_{\substack{j=1\\ j\neq i}}^{8}\frac{1}{\beta_j^* - \beta_i^*} \\ \leq 4.0733\text{x}10^{-12}\left[\frac{\text{atoms}}{\text{b}\cdot\text{cm}}\right] \end{aligned} \tag{4.122}$$

As it is known, this contribution is due to the alpha decay of ^{239}Pu to ^{235}U, which involves a half-life of 24110 years. From the results it can be concluded that the cyclic chain of the right side of Figure 4.5 can be modelled correctly with the modified Bateman equation.

4.9 Final considerations of the forward method.

The integral method described in Section 1.7 and Section 4.6.5 can be used to extend the model of pure cyclic chains to more general and complex structures. For the following discussion, the scheme that is shown in Figure 4.21 will be used. It has as a base a pure cyclic chain, denoted by the sequence $X_1 \rightarrow X_2 \rightarrow \cdots \boxed{X_n}$. From several of the isotopes that belong to this base structure, there are "branches" that emerge, forming linear chains from them, as the sequences given by $Y_1 \rightarrow Y_2 \rightarrow \cdots$ and $Z_1 \rightarrow Z_2 \rightarrow \cdots$.

Additionally, there is a linear chain whose final element, W_n, has a contribution to the cyclic chain. Using the forward method, the structures that emerge as branches from the cyclic chain can be solved. For the moment, the linear chain whose final element is W_n will be ignored and only the cyclic chain and the two branches given by the sequences $Y_1 \rightarrow Y_2 \rightarrow \cdots$ and $Z_1 \rightarrow Z_2 \rightarrow \cdots$ will be considered. Based on Figure 4.21, it is possible to set the balance equation for the following isotopes:

$$\frac{d}{dt}X_j = b_{j-1}^{\text{eff}}\lambda_{j-1}^{\text{eff}}X_{j-1} - \lambda_j^{\text{eff}}X_j, \quad \frac{d}{dt}X_k = b_{k-1}^{\text{eff}}\lambda_{k-1}^{\text{eff}}X_{k-1} - \lambda_k^{\text{eff}}X_k \tag{4.123}$$

And:

$$\frac{d}{dt}Z_1 = b_{k,Z_1}^{\text{eff}}\lambda_k^{\text{eff}}X_{j-1} - \lambda_{Z_1}^{\text{eff}}Z_1, \quad \frac{d}{dt}Y_1 = b_{j,Y_1}^{\text{eff}}\lambda_j^{\text{eff}}X_j - \lambda_{Y_1}^{\text{eff}}Y_1 \tag{4.124}$$

It can be observed that the first two equations only depend of the elements inside the pure cyclic chain. In fact, their root-based equations are set using expression (4.46):

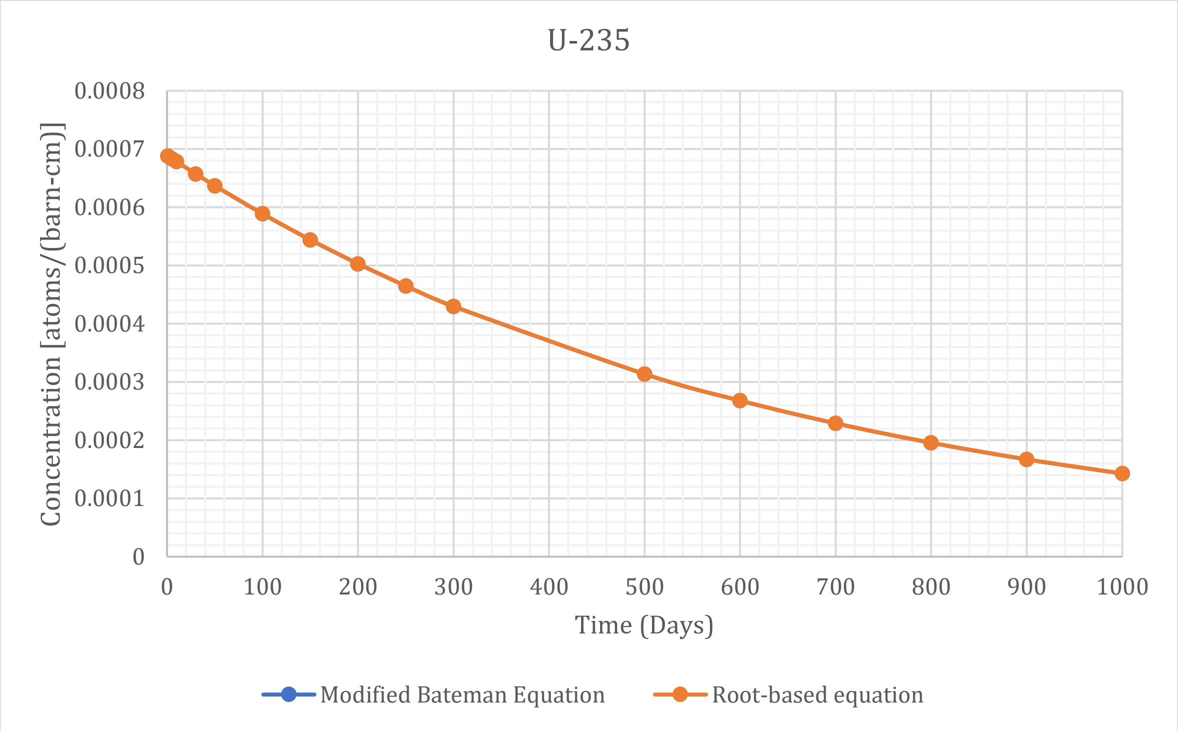

Figure 4. 19. Concentration for ^{235}U compute with the Modified Bateman Equation, and the Root Based Equation. The two curves overlap.

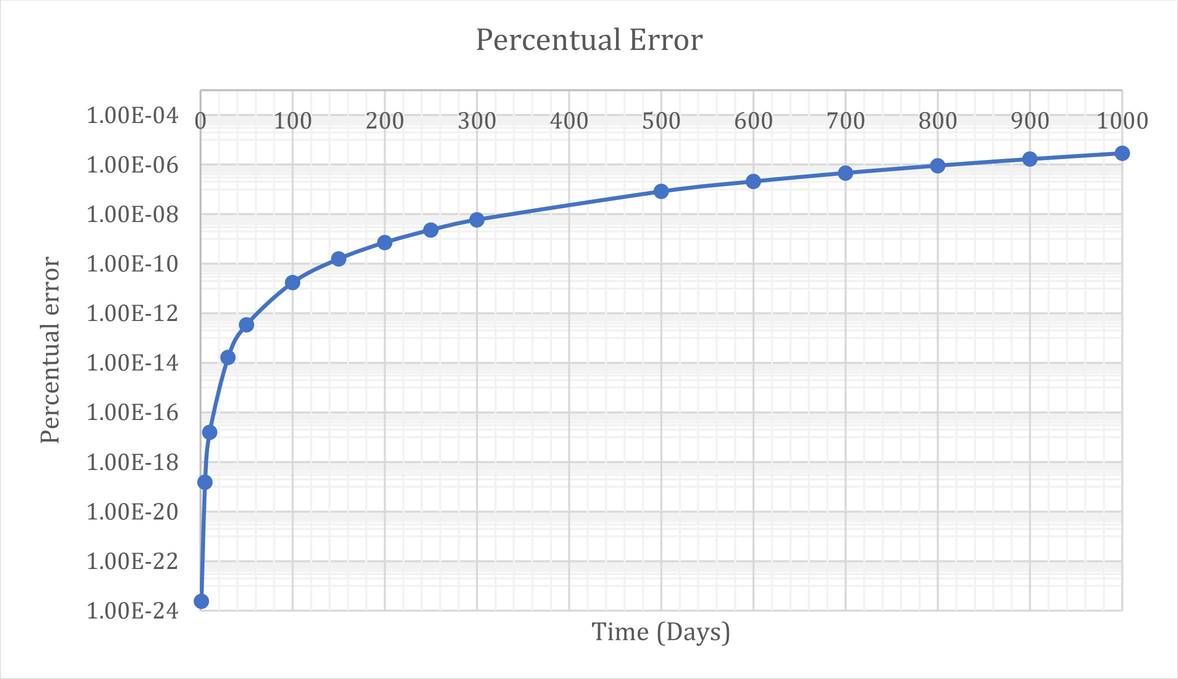

Figure 4. 20. Percentual error between the Modified Bateman Equation and the Root-Based Equation, for the isotope ^{235}U.

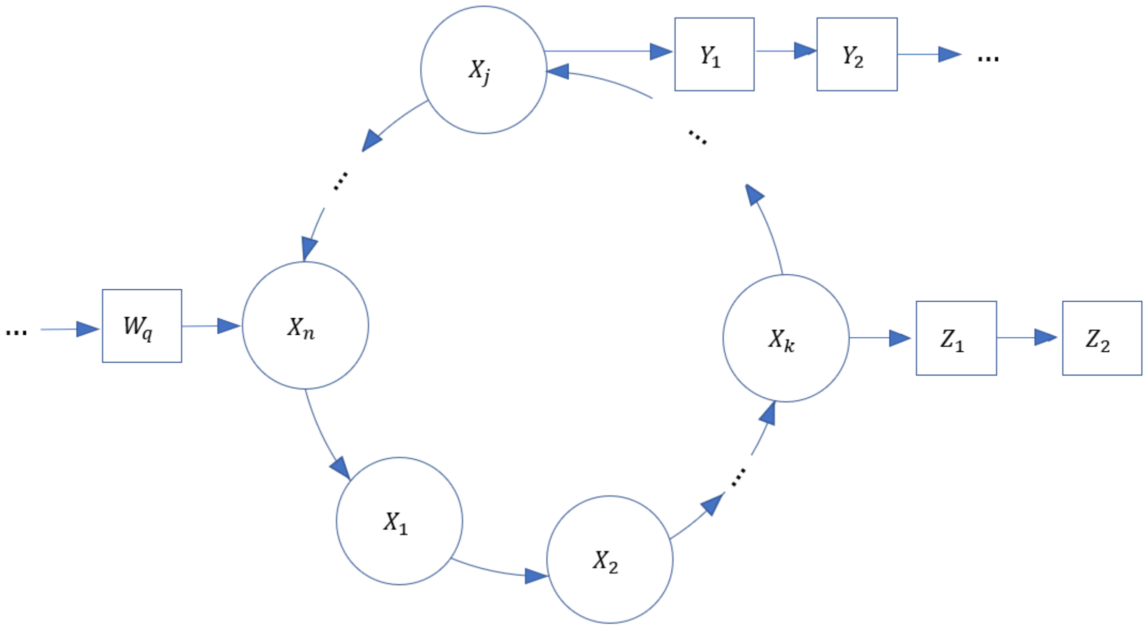

Figure 4. 21. Scheme of cyclic chain where some of its elements have branches, from which emerge linear chains. In addition, there is a linear chain that has a contribution to the cyclic chain through the isotope W_q.

$$X^A_{n-u} = X_1(0)\prod_{m=1}^{n-u-1} b^{\mathrm{eff}}_{m,m+1}\lambda^{\mathrm{eff}}_m \sum_{i=1}^{n} e^{-\beta_i^* t} \prod_{l=n-u+1}^{n} (\lambda_l^{\mathrm{eff}} - \beta_i^*) \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{(\beta_j^* - \beta_i^*)} \tag{4.125}$$

It is only necessary to replace the index u, by the indexes j and k. In other words, with branches that emerge and not enter to a pure cyclic chain, the solution of them is the same that has been discussed in the present thesis, and it is not necessary to make modifications. The reason of it lies inside the terms of lost in the balance equation. In the present example, the elements $\lambda_k^{\mathrm{eff}} X_k$ and $\lambda_j^{\mathrm{eff}} X_j$ represent all the losses that the isotopes undergo, and in these terms are included the transformations that have as a result the production of the isotopes $Z_1$and Y_1. In other words, it follows that:

$$\lambda_k^{\mathrm{eff}} = b^{\mathrm{eff}}_{k,k+1}\lambda_k^{\mathrm{eff}} + b^{\mathrm{eff}}_{k,Z_1}\lambda_k^{\mathrm{eff}} + \cdots, \quad \lambda_j^{\mathrm{eff}} = b^{\mathrm{eff}}_{j,j+1}\lambda_j^{\mathrm{eff}} + b^{\mathrm{eff}}_{j,Y_1}\lambda_j^{\mathrm{eff}} + \cdots$$

Therefore, when the pure cyclic chain is solved, it is considered these reactions in the system. Then, even when there are branches emerging from each isotope of a pure cyclic chain, the solution of this structure can be obtained ignoring all these elements. Furthermore, the balance equations can be solved using the forward method and equation (4.125). The variable v will be used to denote Z_1 or Y_1, and the expression $n-u$ to denote the position of the isotope in the cyclic chain:

$$X_v^A = e^{-\lambda_v^{\mathrm{eff}} t}\left(b^{\mathrm{eff}}_{n-u,v}\lambda^{\mathrm{eff}}_{n-u}\int_0^t X^A_{n-u}(t')e^{\lambda_v^{\mathrm{eff}} t'}\,dt' + X_v(0)\right) \tag{4.126}$$

To solve the last equation it is necessary to compute the following term $e^{-\lambda_v^{\text{eff}}t}\int_0^t X_{n-u}^A(t')e^{\lambda_v^{\text{eff}}t'}\,dt'$

$$e^{-\lambda_v^{\text{eff}}t}\int_0^t X_1(0)\prod_{m=1}^{n-u-1} b_{m,m+1}^{\text{eff}}\lambda_m^{\text{eff}}\sum_{i=1}^{n} e^{-(\beta_i^*-\lambda_v^{\text{eff}})t'} \times \prod_{l=n-u+1}^{n}(\lambda_l^{\text{eff}}-\beta_i^*)\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{dt'}{(\beta_j^*-\beta_i^*)}dt' \tag{4.127}$$

$$= X_1(0)\prod_{m=1}^{n-u-1} b_{m,m+1}^{\text{eff}}\lambda_m^{\text{eff}}\sum_{i=1}^{n}\frac{e^{-\beta_i^* t}-e^{-\lambda_v^{\text{eff}}t}}{\lambda_v^{\text{eff}}-\beta_i^*}\prod_{l=n-u+1}^{n}(\lambda_l^{\text{eff}}-\beta_i^*)\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{(\beta_j^*-\beta_i^*)} \tag{4.128}$$

Replacing (4.128) in (4.126), the concentration for the isotope v is:

$$X_v^A = X_1(0)b_{n-u,v}^{\text{eff}}\lambda_{n-u}^{\text{eff}}\prod_{m=1}^{n-u-1} b_{m,m+1}^{\text{eff}}\lambda_m^{\text{eff}} \left(\sum_{i=1}^{n}\frac{e^{-\beta_i^* t}-e^{-\lambda_v^{\text{eff}}t}}{\lambda_v^{\text{eff}}-\beta_i^*}\prod_{l=n-k+1}^{n}(\lambda_l^{\text{eff}}-\beta_i^*)\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{(\beta_j^*-\beta_i^*)}\right)+X_v(0)e^{-\lambda_v^{\text{eff}}t} \tag{4.129}$$

The last equation can be used to compute the concentration of the isotopes Y_1 and Z_1. It is only necessary to replace the variable v, and the variable u. The next step is to find the solution for the concentration of all the elements in the linear chains that emerge from the cyclic chain, and not only in their first term. Therefore, it is necessary to find a more general solution. In Appendix A it is shown that the general expression for the linear chain $Z_1, Z_2, \dots, Z_p$is given by:

$$X_{Z_p}^A = X_1(0)b_{k,Z_1}^{\text{eff}}\lambda_k^{\text{eff}}\prod_{h=1}^{n-2} b_{Z_h,Z_{h+1}}^{\text{eff}}\lambda_{Z_h}^{\text{eff}}\prod_{m=1}^{k-1} b_{m,m+1}^{\text{eff}}\lambda_m^{\text{eff}} \left(\sum_{i=1}^{n}F(\beta_i, Z_1, \dots Z_p)\prod_{l=k+1}^{n}(\lambda_l^{\text{eff}}-\beta_i^*)\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{(\beta_j^*-\beta_i^*)}\right) \tag{4.130}$$

A similar expression can be obtained for the linear chain $Y_1, Y_2, \dots, Y_n$. In fact, in the last equation it can be identified the index k, that is related to the isotope where the linear chain emerges, the index i that corresponds to the roots of the cyclic chain, and the index p that represents the position in the linear chain.

4.9.1 Linear chains that enter and the convolution theorem.

Unlike the linear chains that emerge from a cyclic chain, the linear chains that enter to the structure cannot be solved in a straightforward way. The main difficulty is that the equation (4.46) cannot be used as the base equation to perform the integration, as in the previous case, because its equation's system is modified. For example, for the structure in the Figure 4.21, the balance equations are the following:

$$\frac{d}{dt}X_n = b^{\text{eff}}_{W_q,X_n}\lambda^{\text{eff}}_{W_q}W_q + b^{\text{eff}}_{n-1,n}\lambda^{\text{eff}}_{n-1}X_{n-1} - \lambda^{\text{eff}}_{n}X_n, \tag{4.131}$$

$$\frac{d}{dt}W_q = b^{\text{eff}}_{W_{n-1},W_n}\lambda^{\text{eff}}_{W_{n-1}}W_{q-1} - \lambda^{\text{eff}}_{W_q}X_{W_q} \tag{4.132}$$

Equation (4.141) modifies the last equation of the system described in (4.3). For such equation it follows:

$$\tilde{x}_n = \frac{b_{n-1,n}\lambda_{n-1}\tilde{x}_{n-1}}{s+\lambda_n} + \frac{b^{\text{eff}}_{W_q,X_n}\lambda^{\text{eff}}_{W_q}\widetilde{w_q}}{s+\lambda_n} \quad , \quad \widetilde{w_q} = \mathcal{L}\{W_q\} \tag{4.133}$$

After successive substitutions in such system, and considering the same initial conditions that were used until now, it follows that:

$$\tilde{x}_n = \left[\frac{\alpha(n-1)X_1(0)}{\left(s+\lambda^{\text{eff}}_1\right)\left(s+\lambda^{\text{eff}}_2\right)\dots\left(s+\lambda^{\text{eff}}_n\right)-\alpha(n)}\right]\left[1+\frac{\Omega(n-1)b^{\text{eff}}_{W_q,X_n}\lambda^{\text{eff}}_{W_q}\widetilde{w}_q}{\alpha(n-1)X_1(0)}\right] \tag{4.134}$$

Where the function $\alpha(n)$ was defined in equation (4.5) and:

$$\Omega(n-1) = (s+\ \lambda_1)(s+\lambda_2)\dots(s+\lambda_{n-1}) \tag{4.135}$$

It is possible to find the inverse of (4.134) through the convolution theorem, which it is necessary to use two times. First, it is known that the inverse of the expression inside the first pair of brackets is equal to (4.44). Then, for the second pair of brackets, the inverse Laplace transform of 1 is equal to the Dirac Delta Function, and it is only necessary to find the following term:

$$\mathcal{L}^{-1}\left\{\frac{\Omega(n-1)b^{\text{eff}}_{W_q,X_n}\lambda^{\text{eff}}_{W_q}\widetilde{w_q}}{\alpha(n-1)X_1(0)}\right\} = \frac{b^{\text{eff}}_{W_q,X_n}\lambda^{\text{eff}}_{W_q}}{\alpha(n-1)X_1(0)}\mathcal{L}^{-1}\left\{\Omega(n-1)\widetilde{w_q}\right\} \tag{4.136}$$

4.9.2 First step of convolution.

Since W_q is an isotope in a linear chain, its solution is given by the modified Bateman equation. Then, the following step is to find the inverse Laplace transform of $\mathcal{L}^{-1}\{\Omega(n-1)\}$. Through the polynomial equation theory (Uspensky, 1963) it is known that (4.135) can be expressed as:

$$\Omega(n-1) = s^n + a_{n-1}s^{n-1} + \dots + a_1 = \sum_{j=0}^{n}\mathcal{J}(j)s^j \tag{4.137}$$

Where:

$$\mathcal{J}(0) = a_1 = \lambda_1^{\text{eff}}\lambda_2^{\text{eff}} \dots \lambda_{n-1}^{\text{eff}}, \qquad \mathcal{J}(n) = 1,\ \mathcal{J}(n-1) = a_{n-1} = \sum_{i=1}^{n} \lambda_i^{\text{eff}}$$

$$\mathcal{J}(n-k) = a_{n-k} = \sum_{h_1=h_k}^{n} \sum_{h_2=h_k-1}^{h_1-1} \dots \sum_{h_{k-1}=h_k-k}^{n-k-2} \sum_{h_k=k+1}^{n-k-1} \lambda_{h_1}\lambda_{h_2} \dots \lambda_{h_k} \tag{4.138}$$

The last expression can also be obtained using the analysis carried out in Section 1.6.2. It can be shown from the theory of the Dirac delta relationships that:

$$\mathcal{L}^{-1}\{s^n + a_{n-1}s^{n-1} + \dots + a_1\} = \delta^{(n)}(t) + a_{n-1}\delta^{(n-1)}(t) + \dots + a_1\delta(t) \tag{4.139}$$

Where the coefficients a_i, $1 \le i \le n$ are computed with (4.138). From (4.137) and (4.139) and using the convolution theorem it follows that:

$$\mathcal{L}^{-1}\{\Omega(n-1)\widetilde{w_q}\} = \int_0^t \sum_{j=0}^{n} \mathcal{J}(j)\, \delta^j(t-u) W_n(u) du$$

The integral can be computed using the following relationship (Wheeler, 1997):

$$\int f(y)\delta^{(n)}(y-x)dy = (-1)^n f^{(n)}(x)$$

Then

$$\mathcal{L}^{-1}\left\{\frac{\Omega(n-1) b_{W_q,X_n}^{\text{eff}} \lambda_{W_q}^{\text{eff}} \widetilde{w_q}}{\alpha(n-1)X_1(0)}\right\} = \frac{b_{W_q,X_n}^{\text{eff}} \lambda_{W_q}^{\text{eff}}}{\alpha(n-1)X_1(0)} \sum_{j=0}^{n} \mathcal{J}(j) \left[(-1)^j \frac{d^n W_q(u)}{dt^n}\right]_0^t \tag{4.140}$$

4.9.3 Second step of convolution.

Originally, the inverse Laplace transform of (4.134) was searched, and in the last section only one part of the procedure was carried out. Then, in order to solve the complete expression, it is necessary to solve the following integral, which comes from the convolution theorem:

$$X_n = \int_0^t \mathcal{L}^{-1}\left\{\frac{\Omega(n-1) b_{W_q,X_n}^{\text{eff}} \lambda_{W_q}^{\text{eff}} \widetilde{w_q}}{\alpha(n-1)X_1(0)}\right\}_{t-u} X_n^A(u)\, du \tag{4.141}$$

Where the subscript $t-u$ implies that the expression (4.140) was evaluated in $t-u$. As it can be observed, finding a solution of (1.141) is a very complex task. Even when the Laplace transform and the convolution theorem are very useful tools, it is necessary to carry out a more detailed study and a new relationship in this last case, where linear chains enter in a pure cyclic chain.

This topic will be matter of a future work, but the main objective in this section was to show that the forward method will be an essential part of the generalization of the solution of cyclic chain.

4.9.4 Conclusions of the Chapter 4.

In the present chapter the cyclic chains were studied from a new perspective, using an alternative mathematical model based in the mass balance equation, instead of using the standard linear chain approximation. Firstly, it was proposed a classification of such structures based in their complexity. When all the isotopes of a cyclic chains only belong to a single structure, then it was named a pure cyclic chain. In the other hand, if at least one isotope belongs to more than one loop, then it is a compound cyclic chain.

Though the present analysis, it was possible to compare the system of equations for a linear chain and the system related to the pure's type. Such comparison shows that both Laplace transforms have a very similar structure, and that they only differ in one term. This difference in turns, strongly depends on how small is a product of effective branching ratios and effective removal coefficients.

Therefore, as a first conclusion, it was proved mathematically that the linear chain method can approximate the solution of pure cyclic chains. This fact was already known, but it can be obtained from a numerical standpoint. Even more, in the present chapter it was identified on what this approximation depends on. Once the system of pure cyclic chain was compared, two solution were proposed to it. One of them was developed under some assumptions, and it was written in terms of a linear chain. The other solution was obtained through the calculation of the roots of a polynomial in the Laplace transform space.

As a second conclusion it was noted that the system of a pure cyclic chain, as well as the two proposed solution were not symmetrical. Therefore, in order to extend the application of these to other isotopes, it was necessary to use the forward and backward methods, which were developed in Chapter 1. Even when the resultant equations involve the computations of high order derivatives, it was possible to overcome this problem through the development of several expressions, some of them related with the topic of integer partitions of number theory.

Therefore, it was possible to conclude in mathematical terms, that the solutions for the pure cyclic chain can be expressed in terms of arithmetical and exponential expressions, in the same way that the standard Bateman solution for a linear chain. This fact is very important, because it implies that these solutions can be incorporated into a computational algorithm in a similar way that the standard Bateman solution. Additionally, through the depletion functions, it was possible to add some properties of symmetry to certain parts of the developed solutions.

For the numerical part, using typical data of a thermal nuclear reactor, it was possible to conclude that the standard Bateman equation is a very good approximation to the cyclic chains. Of the two developed solutions mentioned before, the power series one was practically equivalent to the standard modified Bateman equation in the studied scenarios. On the other hand, the results obtained with the root-based solution have a very small percentual error, being less than 0.1% in all the cases. Using the forward method, it was possible to write the root-based solution in terms of the modified Bateman equation. This is an important finding because it allows comparing both equations and to obtain a formula for the error involved when a cyclic chain is approximate using the linear chain method. Through this fact, it was possible to conclude that for the case $n = 7$, that was analyzed in the present work, the contribution to the concentration of ^{235}U due to the cyclic chain was neglected. Another important conclusion was that the root-based solution can be extended to other superposition steps through a set of equations and recursive formulas. Through a detailed analysis it was showed that it is only necessary to obtain the solution for the isotopes of a pure cyclic chain for the first superposition step, and then using them to obtain the solution for the rest of steps. This finding was used in the present numerical comparison to save computational time.

The possibility to extend the solution of the pure cyclic chain model was studied. It was showed that a structure that consists of a pure cyclic chain, from which linear chains emerge, can be successfully solved using the forward method. On the other hand, structures in which linear chains enter involve a more complex solutions related to the convolution theorem and several Dirac delta relationships. These last findings open the possibility to extend the analysis to more complex cyclic chains, and therefore to obtain a general alternative method to the linear chain approximation.

Finally, in general terms it was proved that the root-based solution and the power series solution are alternative methods to deal with cyclic chains, and, at least for thermal reactor scenarios are equivalent to the utilization of the modified Bateman equation. For the above discussion, it is possible to conclude that the pure cyclic chains can be solved through other alternative methods, which have similar properties to the standard Bateman solution.

Chapter 5. General solutions and the theory of increments.

As it was discussed in the previous chapters, one of the disadvantages of the Bateman's solution is that it cannot be used to solve linear chains with repeated elements, or where two different isotopes have the same effective lambda constant. There are two approaches that can be used in such cases: introducing small modifications to the effective lambda constants or using more general solutions. For the first methodology it is necessary to select an adequate modification, as an increment in the removal effective constants. On the other hand, the implementation of the last methodology has some issues related to the computational time, due to the huge amount of calculations involved in the analytical formulas.

In the present chapter these two topics are discussed and the contributions related to them are described. Firstly, an algebraic computational approach is proposed to solve some of the difficulties of the use of general equations, through an algorithm that builds the analytical general solution in a fast way. Secondly, a proposed related to the theory of increments is introduced. This chapter contains topics belonging to future researches.

5.1 Singularities of the Bateman equations.

As it was mentioned in past chapters, the singularities in the Bateman equation appear when there are least two elements in a linear chain with the same value for the lambda coefficients. That situation only happens in in two cases:

1) When there are repeated elements in the linear chain.

2) When two different elements have the same lambda coefficient.

As it was discussed in Chapter 3 and Chapter 4, for the first case it is possible to find repeated elements in a linear chain, if the (n, γ) and $(n, 2n)$ reactions occur in any order in a consecutive way. With one reaction a neutron is lost, and with the other a neutron is gained. Therefore, the mass balance is the same after the two reactions:

$$\begin{gathered} \ldots \to {}^{A}_{Z}X \xrightarrow{(n,\gamma)} {}^{A+1}_{Z}X \xrightarrow{(n,2n)} {}^{A}_{Z}X \to \cdots \\ \ldots \to {}^{A}_{Z}X \xrightarrow{(n,2n)} {}^{A-1}_{Z}X \xrightarrow{(n,\gamma)} {}^{A}_{Z}X \to \cdots \end{gathered} \tag{5.1}$$

Those reactions are very common among the heavy isotopes as it is shown in the following example:

$$^{235}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{236}\mathrm{U} \xrightarrow{(n,2n)} {}^{235}\mathrm{U} \xrightarrow{(n,2n)} {}^{234}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{235}\mathrm{U} \tag{5.2}$$

Additionally, as it was discussed in the past chapter, this type of linear chains present difficulties in the linearization process, because it generates branches that will appear again and again, generating an infinite network, as Figure 4.2 shows. In the linearization process it is necessary to move along the network (a process that is known as "traversing a tree" in computational science), and then if the length of the network is infinity, the process will generate an infinity number of linear chains, which in turn will have an infinity length.

It is interesting to analyze the infinity meaning in the standard Bateman solution:

$$X_n = X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^{n} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{\lambda_j - \lambda_i} e^{-\lambda_i t} \tag{5.3}$$

Since the last equation tends to infinity when λ_j tends to λ_i, at a first glance it may seem that the concentration for a given isotope will be infinity. Actually, this kind of infinity means that the number of "contributions" to the concentration will be infinite. Even more, when the concentration of an isotope is followed through a pathway or a route with an infinite length, the value of the contribution each time that the isotope appears decreases, and in fact it tends to zero. Therefore, even when the number of contributions tends to infinity, the value of them tends to zero, and thus the limit must exist.

The second scenario where the singularities appear is when two different elements have the same lambda coefficient. It is more difficult to find examples of this cases, and in fact it was thought, at first, that this situation was impossible. Nevertheless, Dreher (Dreher, 2013) provides two interesting examples about this situation:

$$\begin{gathered} ^{86}\mathrm{Ga} \xrightarrow{\beta^-(T_{1/2} > 300\ ns)} {}^{86}\mathrm{Ge} \xrightarrow{\beta^-(T_{1/2} > 300\ ns)} \dots \\ ^{60}\mathrm{As} \xrightarrow{p(T_{1/2} = 1\ ns)} {}^{59}\mathrm{Ge} \xrightarrow{2p(T_{1/2} = 1\ ns)} \dots \end{gathered} \tag{5.4}$$

In the case of ^{86}Ga and ^{86}Ge, the estimation of the half-life is given as a lower limit, instead of a specific value. On the other hand, for ^{60}As and ^{59}Ge, their half-lives were estimated by theoretical considerations. As it can be noted, in the Dreher's examples the elements have the same lambda coefficient due to the state of the library that was used. In this case, Dreher uses the Karlsruhe Nuclides Chart (Sóti et. al., 2019). Then, it is possible that in the future these values will be known in more detail, and therefore the lambda coefficient will be different. Even more, unlike the cyclic chain's case, where it is possible to give an interpretation or mean of the infinity, in this case it is more difficult to elaborate a plausible explanation.

In brief, the author of the present thesis considered that the second case, where the singularities appear, is related to the information of the data libraries, and not with a physical phenomenon.

Therefore, this case will be considered as an “artificial case”, that will depend on the state of the information about the isotopes and their properties.

5.2 Small modifications in lambda coefficients.

A first approach to overcome the issues related to the singularities, consists in introducing small modification in the set of repeated coefficient lambdas. Essentially, if there are m elements with the same lambda coefficient, it is necessary to add a “small” increment, Δ_c, as follows:

$$\lambda + k\Delta_c, \quad \text{where} \quad k = 0, \dots, m-1 \tag{5.5}$$

According to Isotalo (Isotalo, 2013), these modifications can be kept below the level of the uncertainty in initial data, due the effective decay constants are usually known to a few digits. The main difficult of introducing small modifications is associated with the correct choose of Δ_c. Cetnar (Cetnar, 2006) suggests that the accuracy of the numerical results depends on this task. Particularly, this last author used this procedure in the initial version of the TTA and the MCB codes.

Since in the early steps of the Szilard’s development not enough references were found about how to determine the value of Δ_c, a variation of this procedure was used. Taking advantage of the programming language Python, digit 5 was added at the final of the repeated lambda values in a recursive way. For example, it will be assumed that there are three lambda values equal to $2.342x10^{-2}\ s^{-1}$. In Szilard, this value is transformed to a string variable:

$$\lambda = \ 0.002342\,, \quad \text{with } \lambda \text{ as a string variable}$$

Then, using a string concatenate operation, the digit 5 is added at the end of the string character lambda:

$$\lambda_1 = \lambda + \text{"5"}{=}0.0023425 \ \text{ with } \lambda_1 \text{ as a string variable}$$

For the other repeated λ_2:

$$\lambda_2 = \lambda_1 + 5 = 0.00234255$$

In general, this is a recursive function, defined by:

$$\lambda_n = \lambda_{n-1} + \text{"5"} \tag{5.6}$$

This approximation brings adequate results for the heavy isotopes in Szilard, in comparison with a SERPENT’s output. Nevertheless, as it will be discussed later, this process is merely intuitive, and it is necessary to propose a more formal methodology to introduce the modifications. Such task can be carried out using the concept of limit, which can provide a more adequate answer about how to compute the value of Δ_c.

Finally, the procedure of introducing small variations continues being used in several codes, even in the recent ones. For example, in the code DEPTH (She et. al., 2013), ACTYS (Chaytania et al., 2017), TTA (Cetnar, 2006), among others. Additionally, this procedure is also used in recent studies about the first order decay networks (Sun et. al., 2012).

5.3 The Bateman solution with the increments.

It is important to rewrite the Bateman solution for the case where there are repeated elements. Such equation will be useful to discuss one of the most popular general solutions to the Bateman equations, as well as a methodology to find the increment Δ_c discussed before. This task is not straightforward, because it involves sums and products under the sigma and the Capital Pi notation (Σ and $\prod$). In the following developments, the standard Bateman solution without branching's and yields will be considered, in order facilitate the explanation. Equation (5.3) can be written as:

$$X_n(t) = \frac{X_1(0)}{\lambda_n}\sum_{i=1}^{n} \lambda_i \alpha_i\, e^{-\lambda_i t}, \qquad \text{where} \quad \alpha_i = \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{\lambda_j}{(\lambda_j - \lambda_i)} \tag{5.7}$$

It is a straightforward task to show that (5.3) and (5.7) are equivalents, in fact the main difference between the equations is the inclusion or reduction of the product of lambdas in the alpha coefficients. As a first case, a linear chain with n different isotopes, each of them appearing two times, will be considered. Afterwards, using this first approach, the general case will be deduced.

5.3.1 Case $m = 2$, generalization of the summa.

For a given linear chain with n different isotopes, there are a repeated element of each of them. It is possible to distinguish their lambda constant using an increment that depends of the isotope i, in other words:

$$\lambda_i,\ \ \lambda_i + \Delta_i,\ \ \text{with} \ \ 1 \leq i \leq n \tag{5.8}$$

For this case it is possible to generalize the Bateman equation in two steps. One of them related to the summation, and the other one related to the alpha coefficient in (5.7). In the first case it follows that:

$$\sum_{i=1}^{n} \lambda_i \alpha_i\, e^{-\lambda_i t} \rightarrow \sum_{i=1}^{n} \left(\lambda_i\, e^{-\lambda_i t} + (\lambda_i + \Delta_i) e^{-(\lambda_i + \Delta_i)t}\right) \alpha_i^{m\Delta_i} \tag{5.9}$$

In equation (5.9) the symmetry properties of the Bateman equation have been used in order to write the terms in this way. This step will be justified later. Furthermore, $\alpha_i^{m\Delta_i}$ is a generalization of the product in (5.7), where instead of having $(n-1)$ products, there are $2n-1$, because the lambdas with the increments have been included. An analysis of this product will be carried out later, but for the moment it is possible to represent such product using this variable. In (5.9) the repeated elements have been grouped under the same summation index n. It is possible to simplify (5.9) as:

$$\sum_{i=1}^{n} \left(\lambda_i\, e^{-\lambda_i t} + (\lambda_i + \Delta_i) e^{-(\lambda_i + \Delta_i)t}\right) \alpha_i^{m\Delta_i} \rightarrow \sum_{i=1}^{n} \sum_{m=0}^{1} (\lambda_i + m\Delta_i) \alpha_i^{m\Delta_i} e^{-(\lambda_i + m\Delta_i)t} \tag{5.10}$$

The last expression is the preliminary Bateman equation for the case where all the isotopes appears two times. Starting from the equation (5.10) it is possible to generalize the case to the scenario where the isotopes appeared an arbitrary number of times, instead of only two occasions. In order to make such generalization it is only necessary to modify the super index in the summation, considered the numbers m_i and μ_i. The first one is equal to the number of times that the isotope X_i appears. The second one is a useful coefficient that represents the lambda coefficient that will be modified, i.e. $\mu_i = m_i - 1$. Then, it follows:

$$\sum_{i=1}^{n}\sum_{m=0}^{1}(\lambda_i + m\Delta_i)\alpha_i^{m\Delta_i}e^{-(\lambda_i+m\Delta_i)t} \rightarrow \sum_{i=1}^{n}\sum_{m=0}^{\mu_i}(\lambda_i + m\Delta_i)\alpha_i^{m\Delta_i}e^{-(\lambda_i+m\Delta_i)t} \tag{5.11}$$

5.3.2 Generalization of the product, $\alpha_i^{m\Delta_i}$.

The second step in the generalization consists in finding who is $\alpha_i^{m\Delta_i}$, which is the real challenge for this procedure. For the moment the whole summation will be ignored. The standard form of the alpha coefficient without repeated elements is given by:

$$\alpha_i = \prod_{\substack{j=1\\ j\neq i}}^{n}\frac{\lambda_j}{(\lambda_j - \lambda_i)} = \frac{\lambda_1}{(\lambda_1 - \lambda_i)}\frac{\lambda_2}{(\lambda_2 - \lambda_i)}\cdots\frac{\lambda_{i-1}}{(\lambda_{i-1} - \lambda_i)}\frac{\lambda_{i+1}}{(\lambda_{i+1} - \lambda_i)}\cdots\frac{\lambda_n}{(\lambda_n - \lambda_i)} \tag{5.12}$$

As a first approach, the case where only the X_i element has one repeated element will be considered. In this scenario there are $n + 1$ isotopes, and the repeated one can be represented using the increment Δ_i, as $\lambda_i + \Delta_i$. Introducing this term in the product given in (5.12) it follows that:

$$\frac{\lambda_1}{(\lambda_1 - \lambda_i)}\cdots\frac{\lambda_{i-1}}{(\lambda_{i-1} - \lambda_i)}\boxed{\frac{\lambda_i + \Delta_i}{(\lambda_i + \Delta_i - \lambda_i)}}\frac{\lambda_{i+1}}{(\lambda_{i+1} - \lambda_i)}\cdots\frac{\lambda_n}{(\lambda_n - \lambda_i)} \tag{5.13}$$

If it is assumed that the isotope X_i appears m_i times, instead of two times, the last expression can be generalized. In this case, it is necessary to modify μ_i times the λ_i:

$$\underbrace{\lambda_i, \lambda_i + \Delta_1, \lambda_i + \Delta_2, \dots, \lambda_i + \Delta_{\mu_i}}_{m_i \text{ elements}}$$

Then the product in (5.14) is written as follows:

$$\frac{\lambda_1}{(\lambda_1 - \lambda_i)}\cdots\frac{\lambda_{i-1}}{(\lambda_{i-1} - \lambda_i)}\boxed{\frac{\lambda_i + \Delta_i}{(\lambda_i + \Delta_i - \lambda_i)}\frac{\lambda_i + 2\Delta_i}{(\lambda_i + 2\Delta_i - \lambda_i)}\cdots\frac{\lambda_i + \mu_i\Delta_i}{(\lambda_i + \mu_i\Delta_i - \lambda_i)}}\frac{\lambda_{i+1}}{(\lambda_{i+1} - \lambda_i)}\cdots\frac{\lambda_n}{(\lambda_n - \lambda_i)}$$

The elements of the last product can be sorted in the following way:

$$\frac{\lambda_1}{(\lambda_1 - \lambda_i)}\cdots\frac{\lambda_{i-1}}{(\lambda_{i-1} - \lambda_i)}\frac{\lambda_{i+1}}{(\lambda_{i+1} - \lambda_i)}\cdots\frac{\lambda_n}{(\lambda_n - \lambda_i)}\boxed{\frac{\lambda_i + \Delta_i}{(\lambda_i + \Delta_i - \lambda_i)}\frac{\lambda_i + 2\Delta_i}{(\lambda_i + 2\Delta_i - \lambda_i)}\cdots\frac{\lambda_i + \mu_i\Delta_i}{(\lambda_i + \mu_i\Delta_i - \lambda_i)}}$$

And using the product notation, this can be rewritten as:

$$\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{\lambda_j}{(\lambda_j-\lambda_i)}\prod_{k=1}^{\mu_i}\frac{\lambda_i+k\Delta_i}{(\lambda_i+k\Delta_i-\lambda_i)} \tag{5.14}$$

This expression was developed for the case where only the isotope X_i has μ_i repeated elements. The following generalization consists of considering the case where all the isotopes have repeated elements, for which it is only necessary to add the similar products of the right side of the equation (5.12). In other words:

$$\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{\lambda_j}{(\lambda_j-\lambda_i)}\prod_{k=1}^{\mu_1}\frac{\lambda_1+k\Delta_1}{(\lambda_1+k\Delta_1-\lambda_i)}\prod_{k=1}^{\mu_2}\frac{\lambda_2+k\Delta_2}{(\lambda_2+k\Delta_2-\lambda_i)}\prod_{k=1}^{\mu_3}\frac{\lambda_3+k\Delta_3}{(\lambda_3+k\Delta_3-\lambda_i)}\cdots \tag{5.15}$$

$$\cdots\prod_{k=1}^{\mu_{i-1}}\frac{\lambda_{i-1}+k\Delta_{i-1}}{(\lambda_{i-1}+k\Delta_{i-1}-\lambda_i)}\prod_{k=1}^{\mu_i}\frac{\lambda_i+k\Delta_i}{(\lambda_i+k\Delta_i-\lambda_i)}\prod_{k=1}^{\mu_{i+1}}\frac{\lambda_{i+1}+k\Delta_{i+1}}{(\lambda_{i+1}+k\Delta_{i+1}-\lambda_i)}\cdots\prod_{k=1}^{\mu_n}\frac{\lambda_n+k\Delta_n}{(\lambda_n+k\Delta_n-\lambda_i)}$$

If the first term (from left to right) is omitted in the last expression, it is possible to observe that there are n similar products with modified lambdas. The formulas are identical, except for the index of the considered element, and since all of them are multiplying, it is possible to summarize all with a product notation. In other words:

$$\prod_{l=1}^{n}\prod_{k=1}^{\mu_l}\frac{\lambda_l+k\Delta_l}{(\lambda_l+k\Delta_l-\lambda_i)}=\prod_{k=1}^{\mu_1}\frac{\lambda_1+k\Delta_1}{(\lambda_1+k\Delta_1-\lambda_i)}\prod_{k=1}^{\mu_2}\frac{\lambda_2+k\Delta_2}{(\lambda_2+k\Delta_2-\lambda_i)}\prod_{k=1}^{\mu_3}\frac{\lambda_3+k\Delta_3}{(\lambda_3+k\Delta_3-\lambda_i)}\cdots$$

$$\cdots\prod_{k=1}^{\mu_{i-1}}\frac{\lambda_{i-1}+k\Delta_{i-1}}{(\lambda_{i-1}+k\Delta_{i-1}-\lambda_i)}\prod_{k=1}^{\mu_i}\frac{\lambda_i+k\Delta_i}{(\lambda_i+k\Delta_i-\lambda_i)}\prod_{k=1}^{\mu_{i+1}}\frac{\lambda_{i+1}+k\Delta_{i+1}}{(\lambda_{i+1}+k\Delta_{i+1}-\lambda_i)}\cdots\prod_{k=1}^{\mu_n}\frac{\lambda_n+k\Delta_n}{(\lambda_n+k\Delta_n-\lambda_i)}$$

Particularly it is convenient to separate the double product in following way:

$$\prod_{l=1}^{n}\prod_{k=1}^{\mu_l}\frac{\lambda_l+k\Delta_l}{(\lambda_l+k\Delta_l-\lambda_i)}=\prod_{\substack{l=1\\ l\neq i}}^{n}\prod_{k=1}^{\mu_l}\frac{\lambda_l+k\Delta_l}{(\lambda_l+k\Delta_l-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{\lambda_g+k\Delta_g}{\left(\lambda_g+k\Delta_g-\lambda_i\right)} \tag{5.16}$$

If (5.16) is replaced in (5.15), it follows that:

$$\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{\lambda_j}{(\lambda_j-\lambda_i)}\prod_{\substack{l=1\\ l\neq i}}^{n}\prod_{k=1}^{\mu_l}\frac{\lambda_l+k\Delta_l}{(\lambda_l+k\Delta_l-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{\lambda_g+k\Delta_g}{\left(\lambda_g+k\Delta_g-\lambda_i\right)} \tag{5.17}$$

Two products inside (5.17) have the same product notation under the index n, and it is possible to reduce it if the subscript l is changed for j. Therefore, the expression for the product in the case where all the isotopes have repeated elements is equal to:

$$\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{\lambda_j}{(\lambda_j-\lambda_i)}\prod_{\substack{l=1\\ l\neq i}}^{n}\prod_{k=1}^{\mu_l}\frac{\lambda_l+k\Delta_l}{(\lambda_l+k\Delta_l-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{\lambda_g+k\Delta_g}{(\lambda_g+k\Delta_g-\lambda_i)} \tag{5.18}$$

Considering that $\lambda_j = \lambda_j + k\Delta_j$, when $k = 0$, it is possible to modify the product under k index, in such way that it starts from zero, and therefore the first two products can be summarized, in other words:

$$= \prod_{\substack{j=1\\ j\neq i}}^{n}\prod_{k=0}^{\mu_l}\frac{\lambda_j+k\Delta_j}{(\lambda_j+k\Delta_j-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{\lambda_g+k\Delta_g}{(\lambda_g+k\Delta_g-\lambda_i)} \tag{5.19}$$

In order to finalize the generalization of the $\alpha_i^{m\Delta_i}$ factor, a modification is needed in equation (5.19). If the original factor that appears in (5.7) is analyzed, it is possible to note that the lambda λ_i that appears in the denominator's subtraction is associated with the isotope X_i. Nevertheless, following the reasoning used in (5.9), where the changed $\lambda_i \rightarrow \lambda_i + m\Delta_i$ was made, the final step of generalization is given as:

$$\alpha_i^{m\Delta_i} = \prod_{\substack{j=1\\ j\neq i}}^{n}\prod_{k=0}^{\mu_l}\frac{\lambda_j+k\Delta_j}{(\lambda_j+k\Delta_j-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{\lambda_g+k\Delta_g}{(\lambda_g+k\Delta_g-(\lambda_i+m\Delta_i))} \tag{5.20}$$

Using (5.11) and (5.20) it is possible to write the Bateman general equation with modified lambda coefficients:

$$\sum_{i=1}^{n}\sum_{m=0}^{\mu_i}(\lambda_i+m\Delta_i)\prod_{\substack{j=1\\ j\neq i}}^{n}\prod_{k=0}^{\mu_l}\frac{\lambda_j+k\Delta_j}{(\lambda_j+k\Delta_j-\lambda_i)}\prod_{g=1}^{\mu_i}\frac{(\lambda_g+k\Delta_g)e^{-(\lambda_i+m\Delta_i)t}}{(\lambda_g+k\Delta_g-(\lambda_i+m\Delta_i))} \tag{5.21}$$

There are two remarkable facts in the last equation. The first one is related to the two different indexes under the increments, and the second one is the complexity of the equation, which is very different from the original Bateman equation given in (5.7). This formula will be very useful in several of the following sections.

5.4 Development of General solutions.

5.4.1 Review of general solutions.

As it was discussed before, the alternative approach to overcome the difficult of the repeated isotopes is to develop a more general solution of the mass balance system. In fact, in Chapter 1 in Section 1.9, it was showed that this procedure is a straightforward task using the Laplace Inversion Method. It is probable that Donald Clayton and his colleagues were the first who proposed in 1961, a general solution using this methodology. They studied the process of neutron capture, and they approached the case where there is more than one repeated element in successive transformations (Clayton et. al., 1961).

Nevertheless, at least to the best knowledge of the author of the present thesis, there are about 15 different works that developed solutions for the Bateman's equation with repeated elements, or that have relationships with the modeling of successive transformations with the same characteristics. These works do not necessary use the Laplace Inversion Method, and they belong to a wide range of disciplines, from astrophysics to transport theory, including nuclear engineering.

This fact can be explained if it is considered the enormous amount of processes that can be modelled using the balance equations given in (1.1). Nevertheless, due to the wide range of disciplines that deal with this problem, some of which are not necessarily related with each other, some of the proposed solutions were practically ignored by other authors, at least in nuclear engineering. This is a very interesting fact, because some authors developed their own solutions practically ignoring the previous works, and it seems that some of them did not know about the publications of authors from other disciplines. Nevertheless, this situation did not prevent that their solutions were obtained in many ingenious and original ways.

Before Clayton's work, there were other attempts to model the case of successive transformations with one repeated element, however these works cannot be considered as "general solutions" due to two reasons:

1) In some cases, the authors of these works only discussed specific situations were only one isotope was repeated, and they proposed particular equations for it, but they did not delve into the topic.
2) In other cases, the authors developed a methodology to treat cyclic chains, but not to obtain a specific equation.

In fact, the early attempt to describe this phenomenon belongs to Meyer and Schweilder. In 1927, these authors solved this difficulty through a recursive integral formula (Meyer and Schweilder, 1927), but they did not propose a general solution. Afterwards, in 1949, Rubinson considered the formula of Meyer and Schweilder when he developed the Bateman´s equation for the case where there is neutron flux (Rubinson, 1949), but he only showed the case where only one element is repeated.

Nearly two decades later, the developers of the depletion code ORIGEN, (Bell, 1973) obtain the solution to the elemental case where two isotopes have the same removal coefficients, and even when the procedure used to solve it is not mentioned, it is very possible that it consisted in to compute a limit. Then, in 1961, Clayton and his colleagues published the case with more than one repeated element in successive transformations originated by neutron captures. Even more, their work contains a discussion about the mathematical tools related to the inverse Laplace theorem, which motivated the discussion given in Section 1.5.

In 1978, Newman followed the research's line of Clayton, and published the exact solution of the s-process or slow neutron capture process (Newman, 1978). In his work, Newman developed a very similar expression to (1.149) using the Laplace method and the residue theorem. The only difference between the equations is the presence of branching coefficients, and the fact that the removal coefficients are only the capture cross sections in his work. It is possible to consider the Newman's work as the most early complete solution, because he considered the possibility where some elements were repeated, and others were not. This is a remarkable difference with the Claytont's work where only the case where all the elements were repeated was considered.

Therefore, from a general perspective it is possible to say that the solution to this problem was solved in a full way in 1978. Nevertheless, it is very interesting that in the nuclear engineering field the only author that cited the work of Newman was Vudakin in 1994 (Vudakin, 1994). In 1980, Tasaka (Tasaka, 1980) proposed a methodology to simulate the cyclic chains for the burnup code DCHAIN 2, that essentially consisted in transforming the loop in two linear chains and travel them in opposite directions.

It was until 1981 that a general solution was proposed in the field of nuclear engineering. Miles (Miles, 1981) developed a method that used recursive integral factors in order to remove singularities, obtaining a term very similar to (1.148). Eight years later, Raykin and Shlyakhter carried out an analysis about the cyclic chains, which are called in their work as feedback-loops (Raykin and Shlyakhter, 1989). As it was discussed in Chapter 1, the authors proposed a method related to their theory of transition probabilities (that can be interpreted as a more sophisticated integral method) that used depletion functions and the "back of the envelope" method developed by Lasche (Lasche, 1983).

In 1993, M. Blaauw published a work that included an equation with the same shape of (1.149), and he gave similar explicit expressions for the coefficients c_{ij} (Blaauw, 1993). His work is very interesting because he did not show the procedure by which he obtained the solution, instead he only gave it and used mathematical induction to show that it was correct. In 1994, Vudakin (Vudakin, 1994) published a methodology to find the general solution. The procedure followed by this author consisted in using an expansion in series related to the depletion functions described in Section 1.8.3. In 1996, Pommé (Pommé, 1996) and his colleagues published a work where they proposed activation and decay formulas that deal with the problem of equal removal coefficients. One year later, Mirzadeh and Walsh proposed a solution based in the recursive integral formula given by Rubinson (Mirzadeh and Walsh, 1997). Their solution is very interesting because it considers the combinations of removal coefficients that are equal.

In 1999, Wilson developed the activation code ALARA: Analytic and Laplacian Adaptive Radioactivity Analysis (Wilson, 1999). His work contained a full description of the Laplace Inverse Method described in Section 1.9. In the same year, Jovan Popovic' developed a solution related with metabolism kinetics (Popovic', 1999). In fact, in pharmacokinetics there are several works that used inverse Laplace methods and the Heaviside's expansion theorem to solve this kind of problems. In this work, Popovic proposed a solution where a compartment is repeated n times, nevertheless he did not generalize his solution to the case where more than one element is repeated.

In 2006, Cetnar published what can be considered as the most popular general solution in the nuclear engineering's field (Cetnar, 2006). Cetnar developed his solution using a limit procedure, and his work is also remarkable because it contains a description of the TTA method. In fact, the Bateman solution using increments that was described in the past section was based in his work. It is a remarkable fact that the Cetnar's solution has a very similar shape of the equation given by Pommé and his colleagues.

After Cetnar, M. Slodička and Balážová published two works that deal with the problem in the field of transport and first-order kinetics phenomena. Even when their works solved more complex and general processes, their solution to the case where two first-order reactions rates are repeated (that are equivalent to our effective removal coefficients) can be applied to generalize the Bateman's equation. Their first work was published in 2008 (Slodička and Balážová, 2008) and the second in 2010 (Slodička and Balážová, 2010), both are very similar regarding the topic of a solution with repeated elements, and the main difference between them lies in the procedure used to develop the method for the decomposition of multiple solute transport equations. Finally, in 2012, Raymond Dreher published other solution using a procedure of limit (Dreher, 2012). His method is very ingenious, because he translates the Bateman´s equation, with increments, to the formal definition of derivative. In fact, this solution depends on computing derivatives, as in the case of the Inverse Laplace Transform method. This review is summarized in Table 5.1. As it can be seen, several solutions have been developed in a wide range of different disciplines. Nevertheless, as it was discussed in the past Chapter, in all the cases these solutions only modelled the approximation of a cyclic chain using a linear chain.

5.4.2 Classification of the solutions.

From this section onwards, a general solution will be defined as a solution of the system:

$$\frac{dX_i(t)}{dt} = \lambda_{i-1}X_{i-1} - \lambda_i X_i \tag{5.22}$$

With the initial conditions:

$$X_i(t=0) = X_i(0) = 0 \text{ with } 2 \le i \le n,\ \ X_1(0) \neq 0 \tag{5.23}$$

Who considers the possibility to have repeated elements in a linear chain. Such solutions can be divided in four types, according to the mathematical tools that were used to obtained them:

1) Solution by Laplace Inverse Method.
2) Solution by integral formulation.
3) Solution by limit.
4) Solution by power series.

The first two methodologies were discussed in Section 1.9 and Section 1.7, respectively. The third methodology, based in a mathematical limit, is very useful to develop a theory of increments, and it is used in the most recent solution that was published by Dreher (Dreher, 2013). The limit solution will be studied in a more detailed way in the following sections.

The solution based in power series, as its name suggest, is based in to expand the exponential function that originally appears in the Bateman equation as power series, and to analyze the case when two or more lambda constants are equal. The study of the power series requires a more detailed analysis of the depletion functions described in Section 1.8.3, and therefore it is beyond the scope of the present thesis. Nevertheless, it is possible to say that the power series solution is strongly related to the work developed by Newman (1978).

It is worth mentioning that it is not a straightforward task to proof that the general solutions described before are equivalent. In other words, some of the solutions are expressed in a particular way, that is not obvious to conclude that are the same. For example, the solution that was developed by Cetnar is expressed in nested sums whereas that the expression that was found by Dreher is expressed in terms of multiple derivatives.

Year	Authors	Method	Year	Authors	Method
1927	Meyer and Schweildler	Integral	1994	Vukadin	Power series
1949	Rubinson	Integral	1996	Pommé et al.	Integral
1961	Clayton	Laplace	1997	Mirzadeh and Walsh	Integral
1973	Bell	Limit	1999	Wilson	Integral
1978	Newman	Laplace	1999	Popovic'	Laplace
1980	Tasaka	Integral	2006	Cetnar	Limit
1981	Miles	Integral	2008	Slodička and Balážová	Limit
1989	Raykin and Shlyakhter	Integral	2013	Dreher	Limit
1993	Blaauw	Laplace			

In the following section two general solutions that are based in the limit procedure will be described in general terms. A more formal discussion about this procedure will be discussed later, when the theory of increments will be explained.

Table 5. 1. Chronology of general solutions that were published in literature.

5.5 Main steps of the Cetnar's solution.

The Cetnar's solution consists of computing the following limit:

$$\lim_{\Delta_i,\,\Delta_j\to 0}\sum_{i=1}^{n}\sum_{m=0}^{\mu_i}(\lambda_i+m\Delta_i)\prod_{\substack{j=1\\ j\neq i}}^{n}\prod_{l=0}^{\mu_j}\frac{\lambda_j+l\Delta_j}{(\lambda_j+l\Delta_j\ -\lambda_i-m\Delta_i)}\prod_{\substack{k=1\\ k\neq m}}^{\mu_i}\frac{\lambda_i+k\Delta_i}{((k-m)\Delta i)}e^{-(\lambda_i+m\Delta_i)t}$$

The last expression will be analyzed in three parts as follows:

$$\lim_{\Delta_i,\,\Delta_j\to 0}\sum_{i=1}^{n}\sum_{m=0}^{\mu_i}\underbrace{(\lambda_i+m\Delta_i)}_{\text{First term}}\prod_{\substack{j=1\\ j\neq i}}^{n}\prod_{l=0}^{\mu_j}\underbrace{\left(\frac{\lambda_j+l\Delta_j}{(\lambda_j+l\Delta_j\ -\lambda_i-m\Delta_i)}\right)}_{\text{Second term}} \tag{5.24}$$

$$\times \prod_{\substack{k=1\\k\neq m}}^{\mu_i} \underbrace{\left(\frac{\lambda_j + l\Delta_j}{(\lambda_j + l\Delta_j \ - \lambda_i - m\Delta_i)}\right) e^{-(\lambda_i+m\Delta_i)t}}_{\text{Third term}}$$

Computing the limits in an independent way, beginning with the first and third part, it follows that:

$$\underbrace{\left(\lim_{\Delta_i\to 0}(\lambda_i+m\Delta_i)\right)}_{\text{First term}} \times \underbrace{\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\lambda_i + k\Delta_i\right)\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\frac{1}{\Delta_i}\right)\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\frac{1}{(k-m)}e^{-(\lambda_i+m\Delta_i)t}\right)}_{\text{Third term}} \tag{5.25}$$

Considering that $\lim\limits_{\Delta_i\to 0}(\lambda_i+m\Delta_i) = \lambda_i$ and:

$$\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\lambda_i + k\Delta_i = \lambda_i^{\mu_i-1}, \quad \lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\frac{1}{(k-m)} = (-1)^{m-1}\frac{1}{\mu_i!}\binom{\mu_i}{m} \tag{5.26}$$

It is possible to write (5.25) as:

$$\frac{\lambda_i^{\mu_i}e^{-\lambda_i}}{\mu_i!}\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\frac{1}{\Delta_i}\right)\lim_{\Delta_i\to 0}(-1)^{m-1}\binom{\mu_i}{m}e^{-(m\Delta_i)t} \tag{5.27}$$

In this point it is necessary to develop the exponential function as a power series:

$$e^{-(m\Delta_i)t} = \sum_{z=0}^{\infty}\frac{(-m\Delta_i t)^z}{z!} \tag{5.28}$$

Using (5.27) and (5.28) it is possible to rewrite the original limit that appears at the beginning of this section as:

$$\begin{aligned}&\sum_{i=1}^{n}\sum_{m=0}^{\mu_i}\lim_{\Delta_i,\Delta_j\to 0}\prod_{\substack{j=1\\j\neq i}}^{n}\prod_{l=0}^{\mu_j}\frac{\lambda_j + l\Delta_j}{(\lambda_j + l\Delta_j \ - \lambda_i - m\Delta_i)}\\ &\times\frac{\lambda_i^{\mu_i}e^{-\lambda_i}}{\mu_i!}\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\k\neq m}}^{\mu_i}\frac{1}{\Delta_i}\right)\lim_{\Delta_i\to 0}(-1)^{m-1}\binom{\mu_i}{m}\sum_{z=0}^{\infty}\frac{(-m\Delta_i t)^z}{z!}\end{aligned} \tag{5.29}$$

5.5.1 Limit of the second term.

Introducing the limit when $\Delta_j \to 0$ inside the second term of (5.24), it can be written as:

$$\lim_{\Delta_i \to 0} \prod_{\substack{j=1 \\ j \neq i}}^{n} \prod_{l=0}^{\mu_j} \lim_{\Delta_j \to 0} \frac{\lambda_j + l\Delta_j}{(\lambda_j + l\Delta_j \quad - \lambda_i - m\Delta_i)} = \lim_{\Delta_i \to 0} \prod_{\substack{j=1 \\ j \neq i}}^{n} \prod_{l=0}^{\mu_j} \frac{\lambda_j}{(\lambda_j \quad - \lambda_i - m\Delta_i)} \tag{5.30}$$

Since the term inside the product does not depend on l, the expression is reduced to:

$$\lim_{\Delta_i \to 0} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{(\lambda_j)^{\mu_j+1}}{(\lambda_j - \lambda_i - m\Delta_i)^{\mu_j+1}} \tag{5.31}$$

Considering that:

$$\frac{1}{(\lambda_j - \lambda_i - m\Delta_i)^{\mu_j+1}} = \frac{1}{\left(1 - \dfrac{m\Delta_i}{\lambda_j - \lambda_i}\right)^{\mu_j+1} \ (\lambda_j - \lambda_i)^{\mu_j+1}} \tag{5.32}$$

The expression (5.31) can be written as:

$$\lim_{\Delta_i \to 0} \prod_{\substack{j=1 \\ j \neq i}}^{n} \left(\frac{\lambda_j}{\lambda_j - \lambda_i}\right)^{\mu_j+1} (-1)^{\mu_j+1} \left(\frac{1}{\dfrac{m\Delta_i}{\lambda_j - \lambda_i} - 1}\right)^{\mu_j+1} \tag{5.33}$$

In this point it is necessary to make a power series expansion of the second term that contains m. For such work the following relationships are needed:

$$\frac{1}{(x - a)} = \frac{1}{a} \sum_{\omega=0}^{\infty} \left(\frac{x}{a}\right)^{\omega}, \qquad \left(\sum_{\omega=0}^{\infty} x^{\omega}\right)^{k+1} = \sum_{\omega=0}^{\infty} \binom{n+k}{k} x^{n} \tag{5.34}$$

Considering that $x = m\Delta_i/(\lambda_j - \lambda_i)$, it follows that:

$$\left(\frac{1}{\dfrac{m\Delta_i}{\lambda_j - \lambda_i} - 1}\right)^{\mu_j+1} = \sum_{\omega=0}^{\infty} \binom{\omega + \mu_j + 1}{\mu_j + 1} \left(\frac{m\Delta_i}{\lambda_j - \lambda_i}\right)^{\omega} \tag{5.35}$$

Using (5.33) and (5.35) it is possible to rewrite the second term of the original limit in (5.24) as:

$$\lim_{\Delta_i \to 0} \prod_{\substack{j=1 \\ j \neq i}}^{n} \left(\frac{\lambda_j}{\lambda_j - \lambda_i}\right)^{\mu_j+1} (-1)^{\mu_j+1} \sum_{\omega=0}^{\infty} \binom{\omega + \mu_j + 1}{\mu_j + 1} \left(\frac{m\Delta_i}{\lambda_j - \lambda_i}\right)^{\omega} \tag{5.36}$$

5.5.2 Reduction from a limit of two variables to a limit of a single-one.

Considering (5.29) and (5.36), the original limit has been reduced to a single-one variable limit:

$$\sum_{i=1}^{n}\sum_{m=0}^{\mu_i}\lim_{\Delta_i\to 0}\prod_{\substack{j=1\\ j\neq i}}^{n}\left(\frac{\lambda_j}{\lambda_j-\lambda_i}\right)^{\mu_j+1}(-1)^{\mu_j+1}\sum_{\omega=0}^{\infty}\binom{\omega+\mu_j+1}{\mu_j+1}\left(\frac{m\Delta_i}{\lambda_j-\lambda_i}\right)^{\omega}$$
$$\times\frac{\lambda_i^{\mu_i}e^{-\lambda_i}}{\mu_i!}\left(\lim_{\Delta_i\to 0}\prod_{\substack{k=1\\ k\neq m}}^{\mu_i}\frac{1}{\Delta_i}\right)\lim_{\Delta_i\to 0}(-1)^{m-1}\binom{\mu_i}{m}\sum_{z=0}^{\infty}\frac{(-m\Delta_i t)^z}{z!} \tag{5.37}$$

The product of the inverse of increments can be written as:

$$\prod_{\substack{k=1\\ k\neq m}}^{\mu_i}\frac{1}{\Delta_i}=\Delta_i^{1-\mu_i} \tag{5.38}$$

Using this last expression and reordering the terms inside the products it follows that:

$$\prod_{\substack{r=1\\ r\neq i}}^{n}\lambda_j^{\mu_j+1}\sum_{i=1}^{n}\frac{\lambda_i^{\mu_i}e^{-\lambda_i}}{\mu_i!}\sum_{m=0}^{\mu_j}(-1)^{m-1}\binom{\mu_i}{m}$$
$$\times\lim_{\Delta_i\to 0}\Delta_i^{1-\mu_i}\sum_{z=0}^{\infty}\frac{(-m\Delta_i t)^z}{z!}\left[\prod_{\substack{j=1\\ j\neq i}}^{n}\left(\frac{1}{\lambda_j-\lambda_i}\right)^{\mu_j+1}\sum_{\omega=0}^{\infty}\binom{\omega+\mu_j+1}{\mu_j+1}\left(\frac{m\Delta_i}{\lambda_j-\lambda_i}\right)^{\omega}(-1)^{\mu_j+1}\right] \tag{5.39}$$

5.5.3 Analysis of the power series and the final solution.

The formal procedure to show that the limit in (5.39) exists depends on the analysis of the product of series inside the brackets. It is possible to carried out such step using the generalized Cauchy product (Apostol, 1997):

$$\sum_{k_1=0}^{\infty}\sum_{k_2=0}^{k_1}\cdots\sum_{k_n=0}^{k_{n-1}}a_{1,k_n}\,a_{2,k_{n-1}-k_n}\cdots a_{n,k_1-k_2}x^{k}=\prod_{j=1}^{n}\left(\sum_{k_j=0}^{\infty}a_{j,k_j}x^{k_j}\right) \tag{5.40}$$

Strictly speaking, Cetnar did not show this in an explicit way, but he describes the main steps that are included. Firstly, he considers that when Δ_i approaching zero only the terms of the sum products that contain $\Delta_i^{\mu_i-1}$ will not vanish, and the terms with smaller order will cancel out.

The author of the present thesis is working in a formal demonstration of this step, which involves a combinatorial analysis, and it represents a topic for a future research.

Considering the above facts, the final expression for the Bateman solution can be written as:

$$X_n(t) = \frac{X_1(0)}{\lambda_n}\sum_{i=1}^{n}\lambda_i\,\alpha_i e^{-\lambda_i t}\sum_{m=0}^{\mu_i}\frac{\lambda_i t}{m!}\Omega_{i,\mu_i-m} \tag{5.41}$$

$$\text{where}\qquad \mu_i = m_i - 1,\ \ \alpha_i = \prod_{\substack{j=1\\ j\neq i}}^{d}\left(\frac{\lambda_j}{\lambda_j-\lambda_i}\right)^{m_j}$$

And:

$$\Omega_{i,\mu_i-m} = \sum_{h_1=0}^{j}\sum_{h_2=0}^{j}\cdots\sum_{h_{i-1}=0}^{j}\sum_{h_{i+1}}^{j}\cdots\sum_{h_d}^{j}\prod_{\substack{k=1\\ k\neq i}}^{n}\binom{h_k+\mu_k}{\mu_k}\left(\frac{\lambda_i}{\lambda_i-\lambda_k}\right)\delta\left(j,\sum_{\substack{l=1\\ l\neq i}}^{d}h_l\right) \tag{5.42}$$

Equation (5.41) will be called as the Cetnar's general solution of the Bateman equation.

5.6 Main aspects of the Dreher Solution.

The procedure followed by Dreher consists of the same basic idea followed by Centar: taking a limit. Nevertheless, Dreher uses a formal definition of the derivative to compute the limit. This procedure is very elemental, and it is related to another formulation of the Bateman's solution. In order to show the main idea behind Dreher's work, a solution where there are two repeated elements will be used. First, it is necessary to use a different formulation of the Bateman Solution given by:

$$X_n(t) = \frac{X_1(0)}{\lambda_n} \sum_{i=1}^{n-1} \lambda_i \frac{e^{-\lambda_i t} - e^{-\lambda_n t}}{\lambda_n - \lambda_i} \prod_{j=1, j\neq i}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_i} \tag{5.43}$$

It is possible to show that (5.43) is equivalent to (1.24). In Appendix A, a formal proof is provided using mathematical induction. In the last equation, the number of "addends" has been reduced from n to $n-1$, but at the same time two operation subtractions have been introduced. Considering that $\lambda_k = \lambda_m + \Delta$, for $k \neq m$, it is possible to separate the summation in (5.43) as follows:

$$\begin{gathered} \sum_{\substack{i=1 \\ i\neq k \\ i\neq m}}^{n-1} \lambda_i \frac{e^{-\lambda_i t} - e^{-\lambda_n t}}{\lambda_n - \lambda_i} \prod_{j=1, j\neq i}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_i} + \lambda_k \frac{e^{-\lambda_k t} - e^{-\lambda_n t}}{\lambda_n - \lambda_k} \prod_{j=1, j\neq k}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_k} \\ + \lambda_m \frac{e^{-\lambda_m t} - e^{-\lambda_n t}}{\lambda_n - \lambda_m} \prod_{j=1, j\neq m}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_m} \end{gathered} \tag{5.44}$$

Due to this representation, increment Δ will not appear in the first term. replace $\lambda_k = \lambda_m + \Delta$ in the last equation:

$$(\lambda_m + \Delta) \frac{e^{-(\lambda_m+\Delta)t} - e^{-\lambda_n t}}{\lambda_n - (\lambda_m + \Delta)} \prod_{j=1, j\neq k}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_m - \Delta} + \lambda_m \frac{e^{-\lambda_m t} - e^{-\lambda_n t}}{\lambda_n - \lambda_m} \prod_{j=1, j\neq m}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_m}$$

It is possible to separate the product, in order to have the same set of subscripts in the product notation. Thus, the last equation can be written as:

$$\begin{gathered} (\lambda_m + \Delta) \frac{e^{-(\lambda_m+\Delta)t} - e^{-\lambda_n t}}{\lambda_n - (\lambda_m + \Delta)} \prod_{\substack{j=1, j\neq k \\ j\neq m}}^{n-1} \frac{\lambda_j}{\lambda_j - (\lambda_m + \Delta)} \left(-\frac{\lambda_m}{\Delta}\right) \\ + \lambda_m \frac{e^{-\lambda_m t} - e^{-\lambda_n t}}{\lambda_n - \lambda_m} \prod_{\substack{j=1, j\neq m \\ j\neq k}}^{n-1} \frac{\lambda_j}{\lambda_j - \lambda_m} \frac{\lambda_m + \Delta}{\Delta} \end{gathered} \tag{5.45}$$

Factoring the term $-(\lambda_m + \Delta)\lambda_m/\Delta$, the equation (5.45) is reduced to:

$$-\frac{(\lambda_m+\Delta)\lambda_m}{\Delta}\left[\frac{e^{-(\lambda_m+\Delta)t}-e^{-\lambda_n t}}{\lambda_n-(\lambda_m+\Delta)}\prod_{\substack{j=1,j\neq k\\ j\neq m}}^{n-1}\frac{\lambda_j}{\lambda_j-(\lambda_m+\Delta)}-\frac{e^{-\lambda_m t}-e^{-\lambda_n t}}{\lambda_n-\lambda_m}\prod_{\substack{j=1,j\neq m\\ j\neq k}}^{n-1}\frac{\lambda_j}{\lambda_j-\lambda_m}\right]$$

Introducing the increment, and taking the limit Δ→ 0 we have the following expression:

$$-\lambda_m^2\, lim_{\Delta\to 0}$$

$$\frac{\left[\frac{e^{-(\lambda_m+\Delta)t}-e^{-\lambda_n t}}{\lambda_n-(\lambda_m+\Delta)}\prod_{\substack{j=1,j\neq k\\ j\neq m}}^{n-1}\frac{\lambda_j}{\lambda_j-(\lambda_m+\Delta)}-\frac{e^{-\lambda_m t}-e^{-\lambda_n t}}{\lambda_n-\lambda_m}\prod_{\substack{j=1,j\neq m\\ j\neq k}}^{n-1}\frac{\lambda_j}{\lambda_j-\lambda_m}\right]}{\Delta} \tag{5.46}$$

Defining the following function:

$$f(\lambda_m): R\to R,\quad f=\frac{e^{-\lambda_m t}-e^{-\lambda_n t}}{\lambda_n-\lambda_m}\prod_{\substack{j=1,j\neq m\\ j\neq k}}^{n-1}\frac{\lambda_j}{\lambda_j-\lambda_m} \tag{5.47}$$

Using the last equation, it is possible to write (5.46) as:

$$\lambda_m^2\lim_{\Delta\to 0}\frac{[f(\lambda_m+\Delta)-f(\lambda_m)\,]}{\Delta}$$

Which involves the formal definition of the derivative. In other words, the original limit is equal to:

$$-\lambda_m^2\left(\frac{df}{d\lambda_m}\right) \tag{5.48}$$

As it can be noted from the last equation, essentially the general solution proven by Dreher involves function's derivates with repeated lambdas as independent variables. The general case with m_E different isotopes, denoted by X_m, and where there are g_m repeated elements, the Bateman Solutions developed by Dreher is given by:

$$X_n(t)=X_1(0)\prod_{j=1}^{i-1}\lambda_j\sum_{m=1}^{m_E}\frac{(-1)^{g_m-1}}{(g_m-1)!}\left(\frac{d^{g_m-1}}{d\lambda^{g_m-1}}\left[\frac{e^{-\lambda t}}{\prod^{i}_{\substack{p=1\\ p|\lambda_p\neq\lambda_m}}(\lambda_b-\lambda)}\right]\right) \tag{5.49}$$

It is clear that equation (5.49) is very different from equation (5.41) and, in fact, the first one might seem easier to use than the Cetnar's equation. Nevertheless, both expressions must be equal, since an elemental calculus theorem guarantees that if a limit exists, it must be unique. Strictly, Cetnar and Dreher compute the same limit, and therefore their Bateman general solutions must be equivalent, even when they are expressed in different ways. To prove that equality can be an interesting mathematical contribution, even when it can be a very difficult task. The manual of ORIGEN code (Bell, 1973), that was published in 1973, contains the same idea that Dreher's use in his procedure. In such publication, the case where two isotopes have equal removal constants is considered. Even when the procedure that was used is not explicit mentioned, it is a straightforward task to show that it was obtained using the derivate function.

5.7 A combinatorial analysis related to the general solutions.

Until now, all the described solutions do not have restrictions related either to the position in which the repeated isotopes appear, neither the number of times that they do it. In other words, the general solutions admit any configuration of a linear chain, even the one where all the isotopes are equal. Nevertheless, as it will be described in the present section, some of the possible configurations are physically impossible, at least in nuclear engineering problems, and therefore the total number of possible configurations can be significantly reduced to a lower number. If this last number is small enough, it is possible to build the set of solutions a priori, having them in a library inside an algorithm, and using in an activation or burnup code.

This last methodology can simplify the use of the general solutions in practical problems. For example, for the integral equation it will be not necessary to compare all the effective lambda decays in each step, and therefore the execution time will be reduced. Using a combinatorial analysis and a study of the main reactions that appear in nuclear engineering, it is possible to propose a set of rules that allow simplifying the analysis of linear chains with repeated isotopes.

5.7.1 Representation of the position using boxes.

It is possible to use a scheme of boxes to represent a sequence of isotopes. For example, the equivalent representation for the sequence given in (1.11) is equal to:

$$\boxed{1}_1 \xrightarrow{r_{1,2}} \boxed{2}_2 \xrightarrow{r_{2,3}} \dots \xrightarrow{r_{i-1,i}} \boxed{i}_i \xrightarrow{r_{i,i+1}} \boxed{i+1}_{i+1} \xrightarrow{r_{i+1,i+2}} \dots \xrightarrow{r_{n-1,n}} \boxed{n}_n \tag{5.50}$$

In the last equation, the boxes represent positions. The right sub-index indicates the position of the boxes, whereas the number inside them represents the isotope that is in such a position in the original sequence of isotopes. For example, the numbers 1 and n represent the isotopes that are in the first and the last position in a linear chain. The terms $r_{k,k+1}$ have the same meaning that in (1.11). The notation provided in (5.23) allows writing the configuration or permutations that the isotopes have in a given linear chain. Thus, if all the isotopes in a linear chain are equal, the corresponding notation is given by:

$$\boxed{1}_1 \xrightarrow{r_{1,1}} \boxed{1}_2 \xrightarrow{r_{1,1}} \dots \xrightarrow{r_{1,1}} \boxed{1}_i \xrightarrow{r_{1,1}} \boxed{1}_{i+1} \xrightarrow{r_{1,1}} \dots \xrightarrow{r_{1,1}} \boxed{1}_n \tag{5.51}$$

Also, the notation allows representing in a more general way the different linear chains. For example, for the two following linear chains:

$$\begin{gathered} {}^{235}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{236}\mathrm{U} \xrightarrow{(n,2n)} {}^{235}\mathrm{U} \xrightarrow{(n,2n)} {}^{234}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{235}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{236}\mathrm{U} \xrightarrow{(n,2n)} {}^{235}\mathrm{U} \\ {}^{238}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{239}\mathrm{U} \xrightarrow{(n,2n)} {}^{238}\mathrm{U} \xrightarrow{(n,2n)} {}^{237}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{238}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{239}\mathrm{U} \xrightarrow{(n,2n)} {}^{238}\mathrm{U} \end{gathered} \tag{5.52}$$

can be represented as:

$$\boxed{1}_1 \xrightarrow{r_{1,2}} \boxed{2}_2 \xrightarrow{r_{2,1}} \boxed{1}_3 \xrightarrow{r_{1,3}} \boxed{3}_4 \xrightarrow{r_{3,1}} \boxed{1}_5 \xrightarrow{r_{1,2}} \boxed{2}_6 \xrightarrow{r_{2,1}} \boxed{1}_7 \tag{5.53}$$

That can be simplified as:

$$\boxed{1}\boxed{2}\boxed{1}\boxed{3}\boxed{1}\boxed{2}\boxed{1} \tag{5.54}$$

5.7.2 Rules for decay and transmutation.

An adequate way to represent the reactions between the isotopes ${}^{A}_{Z}X_i$ and ${}^{A+u_a}_{Z+u_z}Y_j$ is given by:

$${}^{A}_{Z}X_i + p_i \xrightarrow{r_{i,j}} {}^{A+u_a}_{Z+u_z}Y_j + p_j \tag{5.55}$$

Where A and Z are the mass and the atomic numbers, p_i and p_j can represent initial particles (for example, for capture reactions) and products (as α and β^+ particles) respectively. The parameters u_a and u_z are numbers who describe the effects of the reactions over A and Z. For example, in the case of α decay, $u_a = -4$ and $u_z = -2$, p_i is not considered and p_j is equal to an α particle. Pusa (Pusa, 2013) reported the main reactions that are considered in burnup problems, which can be represented in terms of the effects that they produced over the numbers A and Z. Such reactions are listed in Table 5.2, as well as the values u_a and u_z. For the case of fission, it has been considered that the values of u_a and u_z is equal to 1000, since once the isotopes undergo fission it is practically impossible that, through a cyclic chain, a given isotope appears for a second time.

In the present analysis only the terms ${}^{A}_{Z}X$ and ${}^{A+u_a}_{Z+u_z}Y_j$ will be considered, being adequate to ignore p_i and p_j. Using the notation given in (5.55) it is possible to describe the cyclic chains using bidimensional vectors. This allows developing rules for decay and transmutation that can help to answer the problem of: given an isotope in a linear chain in the position i, for what of the following positions k it is possible that such isotope can appear?

Essentially, a cyclic chain begins with an isotope that can be denoted by (Z, A). If this isotope undergoes a reaction, then the parameters u_a and u_z are added to the vector:

$$\underbrace{(Z, A)}_{\text{Initial isotope}} + \underbrace{(u_z, u_a)}_{\substack{\text{Effect of the}\\ \text{reaction}}} = \underbrace{(Z + u_z, A + u_a)}_{\text{Final isotope}} \tag{5.56}$$

Since that only an ordered pair (u_z, u_a) has been introduced in (5.56), it is possible to conclude that isotopes (Z, A) and $(Z + u_z, A + u_a)$ are consecutive. In other words, the number of ordered pair that are included in a given reactions allows finding the number of reactions that are considered. With the reactions described in Table 5.2, it is possible to answer if it is possible that two consecutive isotopes can be equal. In other words, the case where an isotope appears in two immediate positions can be analyzed, setting the following vector equation:

$$\underbrace{(Z, A)}_{\text{Initial isotope}} + \underbrace{(u_z, u_a)}_{\substack{\text{Effect of the}\\ \text{reaction}}} = \underbrace{(Z, A)}_{\text{Final isotope}} \tag{5.57}$$

Whose solution is equal to:

$$\underbrace{(u_z, u_a)}_{\substack{\text{Effect of the}\\ \text{reaction}}} = (0,0) \tag{5.58}$$

Table 5. 2. Main reactions present in burnup problems in nuclear engineering.

Reaction	Daughter	u_z	u_a	Reaction	Daughter	u_z	u_a
α	(Z-2, A-4)	-2	-4	(n,γ)	(Z, A+1)	0	1
Proton emission	(Z-1, A-1)	-1	-1	(n,p)	(Z-1, A)	-1	0
Neutron emission	(Z, A-1)	0	-1	(n,d)	(Z-1, A-1)	-1	-1
β^{-1}	(Z+1, A)	1	0	(n,t)	(Z-1, A-1)	-1	-2
β^{+}	(Z-1, A)	-1	0	$(n,\ ^{3}\mathrm{He})$	(Z-2, A-2)	-2	-2
$(n,2n)$	(Z, A-1)	0	-1	(n,γ)	(Z-2, A-3)	-2	-3
$(n,3n)$	(Z, A-2)	0	-2	Fission	(Z/2, A/2)	1000	1000
$(n,4n)$	(Z, A-3)	0	-3				

From Table 5.2 it is possible to note that there is not reaction with value parameters $u_z = 0$ and $u_a = 0$, and therefore it is not possible, with these reactions, the case where an isotope appears two consecutive times. In the same way it is possible to analyze the case where an isotope that appears in the position i, can appear in the position $i + 4$. For this case it is necessary to set the following vector equation:

$$\underbrace{(Z,A)}_{\text{Initial isotope}} + \underbrace{\left(u_z^{r_1},u_a^{r_1}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 1}}} + \underbrace{\left(u_z^{r_2},u_a^{r_2}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 2}}} + \underbrace{\left(u_z^{r_3},u_a^{r_3}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 3}}} + \underbrace{\left(u_z^{r_4},u_a^{r_4}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 4}}} = \underbrace{(Z,A)}_{\text{Final isotope}} \tag{5.59}$$

In the last equation the super-index over the parameters u_z and u_a indicates the reaction that produces the effect. Using the box scheme described in the past section, equation (5.59) can be represented as:

$$\boxed{1}_{i}\boxed{\&}_{i+1}\ \boxed{\&}_{i+2}\boxed{\&}_{i+3}\boxed{1}_{i+4} \tag{5.60}$$

Where the symbol & has been used to represent an arbitrary isotope, different from 1. Equation (5.59) can be written as:

$$\underbrace{\left(u_z^{r_1},u_a^{r_1}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 1}}} + \underbrace{\left(u_z^{r_2},u_a^{r_2}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 2}}} + \underbrace{\left(u_z^{r_3},u_a^{r_3}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 3}}} + \underbrace{\left(u_z^{r_4},u_a^{r_4}\right)}_{\substack{\text{Effect of the}\\ \text{reaction 4}}} = (0,0) \tag{5.61}$$

Which in turn can be expressed as:

$$\sum_{k=1}^{4}\left(u_z^{r_k},u_a^{r_k}\right) = (0,0), \sum_{k=1}^{4} u_z^{r_k} = \sum_{k=1}^{4} u_a^{r_k} = 0 \tag{5.62}$$

The following step will consist in finding which reactions of Table 5.2 fulfill equation (5.61). A first approach to this problem will be building all the possible combinations with 4 elements, from the total of 14 reactions that are listed in Table 5.2 (discarding the fission reaction) and introducing them in equation (5.61). From combinatorics (Grimaldi, 2003) such number of combinations is equal to:

$$\binom{14+4-1}{4-1} = \binom{17}{3} = 680 \tag{5.63}$$

Not all these configurations fulfill equation (5.61). Even when the 680 combinations can be tested using programming, it is also possible to study equation (5.62) using Number Theory, in order to provide more restrictions. In Table 5.2 there are only two reactions whose parameters u_z^r and u_a^r are positive: the (n,γ) and β^-reaction. Such reactions are represented with the ordered pairs (0,1) and (1,0) respectively. The rest of reactions are described by pair of numbers that are equal to or less to zero.

From the above discussion it can be concluded that any cyclic chain will have positive and negative numbers, in order to fulfill that the sum of the effects will be equal to zero. Therefore, it is necessary that a (n,γ) or a β^-reaction will be present in any cyclic chain. Even more, for each negative parameter $u_z^{r_k}$ equal to a, it is necessary to introduce the β^- reaction a-times in order to have a sum equal to zero. A similar reasoning can be applied for (n,γ). Based on this, it is possible to conclude that the α reaction cannot be part of the solution of the equation (5.62).

In fact, according to Table 5.2, if an α reaction is present in a cyclic chain, the minimum vector equation will be:

$$\underbrace{(-4,-2)}_{\alpha\text{ reaction}} + 4\cdot \underbrace{(1,0)}_{\substack{\beta^-\\ \text{reaction}}} + 2\cdot \underbrace{(1,0)}_{\substack{(n,\gamma)\\ \text{reaction}}} = (0,0) \tag{5.64}$$

As it can be noted such equation involves at least six reactions in addition to the α decay, four β^- and two α reactions. Nevertheless, the equation (5.62) only considers four reactions. Therefore, it is possible to conclude that a scheme as the one described in (5.59) cannot contain α reactions. With a similar reasoning the reactions (n,α), $(n,\ {}^3\mathrm{He})$, (n,d), $(n,3n)$ and the proton emission can be discarded. Therefore, the actual number of combinations that must to be analyzed is reduced to:

$$\binom{6+4-1}{4-1} = \binom{9}{3} = 84 \tag{5.65}$$

5.7.3 Rules for the configurations.

A set of rules for the possible configurations was deduced using a similar combinatorial analysis as the one described in the past sections. Additionally, with the library “itertools” of the Python programming language was possible to test all the possible configurations for linear chains. The rules can be summarized as follows:

1) The case where two consecutive isotopes have the same lambda constant is not possible.
2) Most of the cases where singularities appear have their origin in a combination of the (n,γ) and $(n,2n)$ reactions.
3) There are some cyclic chains where the reaction $(n,2n)$ is not included, but where the reaction β^- is present.

Finally, from the study of the ENDF/B-VII Library it is possible to provide a fourth rule:

4) The case where two different isotopes have the same lambda decay is practically impossible.

5.8 Development of a computer algebra algorithm.

5.8.1 An intuitive definition of computer algebra.

It is possible to divide calculations in two types: the ones that are based on numerical methods, and the ones that depends on the manipulation of symbolical formulas. For example, it will be supposed that the solution of the following integral is searched:

$$G(a,b)=\int_a^b x^2 dx \tag{5.66}$$

It is possible to find an approximate value to such integral using the following equation:

$$G(a,b)=\int_a^b x^2 dx \approx \frac{b-a}{n}\left(\frac{a^2+b^2}{2}+\sum_{k=1}^{n-1}\left(a+k\frac{b-a}{n}\right)\right)^2 \tag{5.67}$$

Where n is the number that has been used to divide the interval $[a,b]$, and $k=0,1,\dots,n-1$. This approximation depends on how large is n. On the other hand, another approach to (5.66) will consist in to perform the following algebraic manipulations:

$$G(a,b)=\int_a^b x^2 dx=\left[\frac{x^3}{3}\right]_a^b,\ \text{using}\ \int u^n du=\frac{u^{n+1}}{n+1}+¿c \tag{5.68}$$

There are two main differences between the last methodologies. Firstly, in equation (5.68) it is necessary to know the exact solution of the problem or some methods to determine if such solution exists, as well as carrying out several symbolical manipulations. On the other hand, in equation (5.67) it is not necessary to know what the solution of the problem is, and, instead, it is necessary to perform a greater number of arithmetical operations.

The first type of methodology is called numerical calculus, because it allows approximating with a high degree of precision an operation or a solution. In such calculus it is not necessary to know the exact solution, and the approximation depends on a set of numerical algebraic operations. The second methodology, known as symbolical calculus, requires manipulating symbolical expressions in order to determine if the exact solution exists. In the example given in (5.68), the exact solution was found, but there are cases where the symbolical calculus needs to apply mathematical methods in order to search for such solution.

Since several problems in engineering do not have an exact solution, and the mathematical methods that they require are very complex, the symbolical calculus have a limited range of applications. Instead, the numerical calculus is the predominant methodology to solve problems, and the development of computer science has been guided by the way in which numerical operations can be carried out. Finally, it is worth mentioning that the symbolical calculus requires a different paradigm in terms of software development.

5.8.2 Computer Algebra as an approach to the general Bateman solutions.

It is possible to define the computer algebra or symbolic computation, as the process of automatically transforming mathematical formulas (Damiano and Ghio, 1997). Initially, the development of this computational branch was focus only on algebraic operations, but nowadays this includes every mathematical manipulation technique, including advanced mathematical methods. Strictly speaking, the computer algebra consists of the introduction of a symbolic computation in an algorithm or a software, forming a computer algebraic system, denoted as CAS. Today, it exists several CAS software, being the most popular Mathematica (where Wolfram|Alpha is included) (David, 2010), and Maple (Char et. al., 1983) among others.

Using computer algebra, it is possible to calculate high order derivates, as well as performing multiple integrals in a fast way. Therefore, this computational tool allows including the general Bateman solutions who contains such operations in a burnup or activation code. In other words, computer algebra removes the complexity in the process of building general solutions for the Bateman equation. Nevertheless, instead of using some of these existing CAS to carry out such task, it was considered more useful to develop a reduced CAS who only requires operations related to string variables. Since most of the modern programming languages include strings functions, the developed algorithm can be included easily in a burnup code.

Once the main generals' solutions were studied, it was considered convenient to use the Laplace Inverse Method and the Integral solution, because other solutions involve complex nested sums, as it was showed in the Centar's case.

5.8.3 Algorithm properties and recurrence relationships.

As it was discussed in the last section, equation (1.149) of the method, and equation (1.60) of the integral solution were chosen to develop the computer algebra algorithm. Such solutions are equivalent, but they have different properties in an algorithmic level. The integral approach finds the solution step by step, performing exponential integrals when the effective lambda coefficients are different, and for the case where there are repeated isotopes, such equation requires to find integrals of the type:

$$e^{-\lambda_y^{\text{eff}} t} \int_0^t t_1^n e^{-(\lambda_x^{\text{eff}} - \lambda_y^{\text{eff}}) t_1} dt_1 \tag{5.69}$$

For which the following formula was developed as a part of the present thesis:

$$= -\frac{t^n e^{-\lambda_x^{\text{eff}} t}}{\left(\lambda_x^{\text{eff}} - \lambda_y^{\text{eff}}\right)} + \frac{n!\left(e^{-\lambda_x^{\text{eff}} t} - e^{-\lambda_y^{\text{eff}} t}\right)}{\left(\lambda_x^{\text{eff}} - \lambda_y^{\text{eff}}\right)^{n+1}} + \sum_{k=1}^{n-1}\left[-\frac{n!\, t^{n-k} e^{\lambda_x^{\text{eff}} t}}{(n-k)!\left(\lambda_x^{\text{eff}} - \lambda_y^{\text{eff}}\right)^{k+1}}\right] \tag{5.70}$$

As it can be noted, the complexity of the integration is reduced to a manipulation of algebraic operations with several terms, which is an advantage in computational terms. On the other hand, the main disadvantage of the integral solution, is that it requires to perform a comparison by pars of the effective lambda coefficients, in each step of the integrals.

In addition to the equation (5.70), it is possible to define a set of recurrence formulas that allows reducing the complexity when the integral method is used. Such formulas can be described as:

$$\int_0^t e^{-\left(\lambda_i^{\text{eff}}-\lambda_j^{\text{eff}}\right)t_1} dt_1 = \begin{cases} -\dfrac{1}{\lambda_i^{\text{eff}}-\lambda_j^{\text{eff}}}\left[e^{-\left(\lambda_i^{\text{eff}}-\lambda_j^{\text{eff}}\right)t}-1\right], & \lambda_i^{\text{eff}} \neq \lambda_j^{\text{eff}} \\ t, & \lambda_i^{\text{eff}} = \lambda_j^{\text{eff}} \end{cases} \tag{5.71}$$

The operation described in the last equation can be represented using a binary function given by $\omega(\lambda_i^{\text{eff}}, \lambda_j^{\text{eff}})$. Starting with (5.71), the following common operation has been identified:

$$\begin{aligned} \varphi\left(\lambda_i^{\text{eff}}, \lambda_j^{\text{eff}}\right) &= e^{-\lambda_j^{\text{eff}} t}\omega\left(\lambda_i^{\text{eff}}, \lambda_j^{\text{eff}}\right) \\ &= \begin{cases} -\dfrac{1}{\lambda_i^{\text{eff}}-\lambda_j^{\text{eff}}}\left[e^{-\lambda_i^{\text{eff}} t}-e^{-\lambda_j^{\text{eff}} t}\right], & \lambda_i^{\text{eff}} \neq \lambda_j^{\text{eff}} \\ e^{-\lambda_j^{\text{eff}} t} t, & \lambda_i^{\text{eff}} = \lambda_j^{\text{eff}} \end{cases} \end{aligned} \tag{5.72}$$

Following the procedure of integration, the following expression appears:

$$\begin{aligned} & e^{-\lambda_k^{\text{eff}}} \int_0^t e^{\lambda_k^{\text{eff}} t_1} \varphi\left(\lambda_i^{\text{eff}}, \lambda_k^{\text{eff}}\right) dt_1 \\ &= \begin{cases} -\dfrac{1}{\lambda_i^{\text{eff}}-\lambda_j^{\text{eff}}}\left[\varphi\left(\lambda_i^{\text{eff}}, \lambda_j^{\text{eff}}\right)-\varphi\left(\lambda_j^{\text{eff}}, \lambda_k^{\text{eff}}\right)\right], & \lambda_i^{\text{eff}} \neq \lambda_j^{\text{eff}} \\ e^{-\lambda_k^{\text{eff}} t} \displaystyle\int_0^t e^{-\left(\lambda_j^{\text{eff}}-\lambda_k^{\text{eff}}\right)t_1} t_1 dt_1 & \lambda_i^{\text{eff}} = \lambda_j^{\text{eff}} \end{cases} \end{aligned} \tag{5.73}$$

On the other hand, the Laplace's solution carries out a comparison of the effective lambda constants only one time, because it determines how many isotopes are repeated, and the number of times that they appear. Therefore, it easily computes the coefficients k_i of equation (1.49). Nevertheless, the main difficulty with this solution is the calculation of the high-order derivatives that are involved:

$$A(s)\left(s+\lambda_j^{\text{eff}}\right)^{k_j} = P(s)_j \tag{5.74}$$

Where:

$$P(s)_j = (s+\lambda_1)^{-k_1}(s+\lambda_2)^{-k_2} \dots \left(s+\lambda_{j-1}\right)^{-k_{i-1}}\left(s+\lambda_{j+1}\right)^{-k_{i+1}} \dots (s+\lambda_n)^{-k_n} \tag{5.75}$$

In the present work it was found that such derivatives can be solved using the natural logarithm. If such function is applied in both sides of (5.76):

$$\ln\left(P(s)_j\right) = \sum_{\substack{u=1 \\ u\neq j}}^{n} -k_u \ln\left(s+\lambda_u\right) \tag{5.76}$$

And therefore:

$$\frac{d}{ds}A(s)\left(s+\lambda_j^{\text{eff}}\right)^{k_j} = P'(s)_j = P(s)_j\left[\sum_{\substack{u=1\\u\neq j}}^{n} -\frac{k_u}{s+\lambda_u}\right] \tag{5.77}$$

For the following derivative it follows:

$$P''(s)_j = \left[\sum_{\substack{u=1\\u\neq j}}^{n} -\frac{k_u}{s+\lambda_u}\right]P'(s)_j + P(s)_j\left[\sum_{\substack{u=1\\u\neq j}}^{n} \frac{k_u}{(s+\lambda_u)^2}\right]$$

$$= P(s)_j\left\{\left[\sum_{\substack{u=1\\u\neq j}}^{n} -\frac{k_u}{s+\lambda_u}\right]^2 + \left[\sum_{\substack{u=1\\u\neq j}}^{n} \frac{k_u}{(s+\lambda_u)^2}\right]\right\} \tag{5.78}$$

Defining:

$$S(a,b) = \sum_{\substack{u=1\\u\neq j}}^{n} (-1)^b \frac{a\cdot k_u}{(s+\lambda_u)^b} \tag{5.79}$$

Using the last notation, it is possible to express (5.78) as:

$$= P(s)_j[(S(1,1)^2 + S(1,2)]$$

For the following derivative:

$$P(s)_j''' = P(s)_j[2S(1,1)\cdot S(1,2) + S(2,3)] + [(S(1,1)^2 + S(1,2)]P(s)_j[S(1,1)]$$

$$P(s)_j''' = P(s)_j[S(1,1)^3 + 3S(1,1)\cdot S(1,2) + S(2,3)]$$

Using the notation given in (5.79) it is possible to compute in a very fast recursive way the high order derivates.

5.8.4 Main features of the computer algebra algorithm.

In order to describe the main features of such algorithm, some of the functions related to the integration solution will be described in Table 5.3. As it is possible to note, the functions have a symbolical representation that consist of a capital letter, as well as a parenthesis to close their argument. The letter "l" accompanied with a number will be used to represent the lambda constants. As the Table 5.3 shows, the functions have some properties as commutation or distribution, which allowreducing them.

The algorithm begins solving the following equation:

$$e^{-\lambda_2^{\text{eff}}t}\int_0^t e^{-(\lambda_1^{\text{eff}}-\lambda_2^{\text{eff}})t'}dt' \tag{5.80}$$

Which has the equivalent string expression:

$$M\left(ex(-l2), G\left(ex\big(-d(l2,l1)\big)\right)\right) \tag{5.81}$$

Table 5. 3. Main string functions related to the part of the integral solution.

Function	Symbolical representation	Arguments	Properties
$a-b$	$d(a,b)$	2	$d(a,b)=-d(b,a)$
$a+b$	$S(a,b)$	2	$S(a,b)=S(b,a)$ $S(a,-b)=d(a,b)$
$a\cdot b$	$M(a,b)$	2	$M(a,b)=M(b,a)$ $M(d(a,b),c)=d(M(a,c),M(b,c))$ $M(S(a,b),c)=S(M(a,c),M(b,c)$ $M(a,1)=a$
e^{at}	$ex(a)$	1	$M\big(ex(a),exp(b)\big)=expn(S(a,b))$ $ex\big(d(lx,ly)\big)=1$, si $lx=ly$
t^a	$t(a)$	1	$M\big(t(a),t(b)\big)=t\big(S(a,b)\big)$
$\dfrac{1}{a-b}$	$d_i(a,b)$	2	$d_i(a,b)=-d_i(b,a)$
$\displaystyle\int_0^1 adt$	$G(a)$	1	$G\big(M(a,b)\big)=M\big(a,G(b)\big)$ if a is constant $G\big(S(a,b)\big)=S\big(G(a),G(b)\big)$ $G(d(a,b)=d\big(G(a),G(b)\big)$

Then, it is necessary to identify all the integrals operations that appear in (5.81), which is a straightforward task, because it is only necessary to search for the character "G". Once such identification is carried out, it is necessary to analyze the argument of the integrals and reduce them in order to have a subtraction of the lambda constants inside it. Such task can be carried out using the relationships described in the past section.

For example, for the first stage the integral given in (5.81) has the following possibilities:

$$\begin{cases} a\displaystyle\int_0^t g(t)t & \text{if } f(t)=ag(t) \\ t & \text{if } f(t)=e^{-(\lambda_x^{\text{eff}}-\lambda_y^{\text{eff}})t},\ \text{ and } \lambda_x^{\text{eff}}=\lambda_y^{\text{eff}} \\ -\dfrac{1}{\lambda_x^{\text{eff}}-\lambda_y^{\text{eff}}}\left[e^{-(\lambda_x^{\text{eff}}-\lambda_y^{\text{eff}})t}-1\right] & \text{if } f(t)=e^{-(\lambda_x^{\text{eff}}-\lambda_y^{\text{eff}})t} \text{ and } \lambda_x^{\text{eff}}\neq\lambda_y^{\text{eff}} \end{cases} \tag{5.82}$$

Which have the following symbolical representation:

$$\begin{cases} M(a, G(b)) & \text{for } G\big(M(a,b)\big) \text{ and } a \text{ constant} \\ t(1) & \text{for } G\left(ex\big(-d(lx,ly)\big)\right) \text{ and } lx = ly \\ M\left(-di(lx,ly), d\big(ex\big(-d(lx,ly)\big),1\big)\right) & \text{with } G\left(ex\big(-d(lx,ly)\big)\right) \text{ and } lx \neq ly \end{cases} \tag{5.83}$$

For the last case where $lx \neq ly$ the expression (5.81) becomes:

$$M\left(ex(-l2), M\left(-d_i(l1,l2), d\big(ex\big(-d(l1,l2)\big),1\big)\right)\right) \tag{5.84}$$

The multiplication inside the main product can be expressed as:

$$d\left(M\left(-d_i(l1,l2), ex\big(-d(l1,l2)\big)\right), -d_i(l1,l2)\right) \tag{5.85}$$

Replacing this term in (5.84) it follows that (5.81) is equal to:

$$d\left(M\big(-d_i(l1,l2), ex(-l1)\big), M\big(-d_i(l1,l2), ex(-l2)\big)\right) \tag{5.86}$$

This last expression was found only using the properties and functions given in Table 5.3. If (5.86) is written in a mathematical form, it is equal to:

$$-\frac{e^{-\lambda_1^{\text{eff}} t}}{\lambda_1^{\text{eff}} - \lambda_2^{\text{eff}}} + \frac{e^{-\lambda_2^{\text{eff}} t}}{\lambda_1^{\text{eff}} - \lambda_2^{\text{eff}}} \tag{5.87}$$

Which can be obtained using the standard integration manipulations. This brief example shows how the symbolical algorithm can perform the operation required to find the general solution.

5.8.5 Comparison in computational time.

It is possible to show the utility of the proposed algorithm through a comparison with the use of the Bateman equation for a given cyclic chain. Such comparison could be in terms of the accuracy and in terms of the computational time. Isotalo and Aarnio study the first one, analyzing the results that are obtained when the general solution is applied in comparison with the use of the Bateman equation with modified effective lambda constants (Isotalo and Aarnio, 2011). In the present thesis a comparison of the execution time using the proposed algorithm is showed. For this task the following cyclic chain will be used:

$$^{235}\text{U} \xrightarrow{(n,\gamma)} {}^{236}\text{U} \xrightarrow{(n,2n)} {}^{235}\text{U} \xrightarrow{(n,2n)} {}^{234}\text{U} \xrightarrow{(n,\gamma)} {}^{235}\text{U} \xrightarrow{(n,\gamma)} \ldots \tag{5.88}$$

Typical values for $X_{^{235}\text{U}}(0)$ and the effective lambda coefficients were taken from a thermal burnup problem, but the results are not depending on such values, instead of the time required to build the solution. The analysis was focus on the time that the Bateman equation with modified effective lambda constants were used to obtain a solution, in comparison with the solution obtained with the symbolical algorithm. Such analysis considers as variable the length of the cyclic chain given in (5.88), which repeated the sequence given by the reactions (n,γ), $(n,2n)$, $(n,2n)$, (n,γ), and (n,γ).

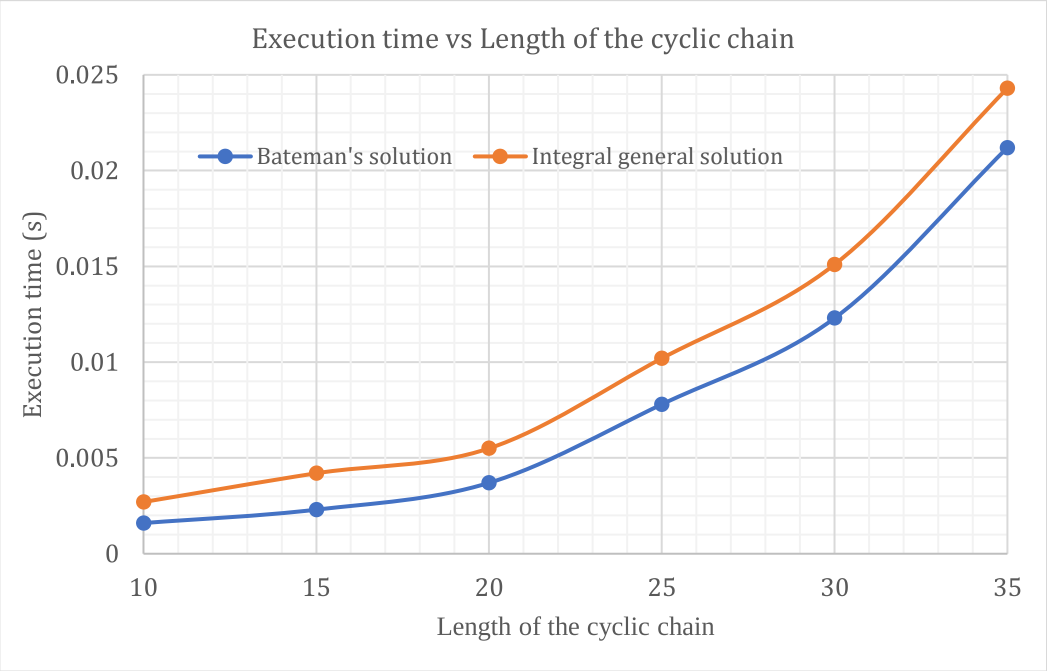

Figure 5. 1. Comparison between the execution time.

The test was run on a 2.6 GHz–3.5 GHz2 Intel i7, 6700 HQ, under a 64-bit Windows, using the Python 3.5 programming language. Figure 5.1 shows the execution time of the methods as a function of the length of the cyclic chain showed in (5.88).

From the Figure 5.1 it is possible to obtain a geometrical mean of the quotient between the execution times. Using as base the time involved when the Bateman equation is used, for the set of values the geometrical mean is approximately 1.4. Clearly, the Bateman solution that uses the modified effective lambda constants is the fastest, because the form of the equation does not change in function of the configuration of the repeated isotopes. Therefore, its execution time is only related to the calculation of the lambda modifications as well as the numerical evaluation. On the other hand, the proposed algorithm requires more time that is related to building the symbolical solution. Nevertheless, the results show that such time is comparable with the numerical evaluation, as well as to the execution of the standard Bateman equation. In other words, these preliminary results show that using general solutions through computer algebra can be suitable for burnup codes. It is clear that a more detailed study is required, where a considerable number of cyclic chains will be included, and where a complete study of the optimization of the algorithm will be carried out, but these topics are part of a future research.

5.9 Theory of increments.

It is possible to represent the Bateman solution in equation (5.3) using the following notation:

$$B(\lambda_1, \lambda_2, \dots, \lambda_n) \qquad (5.89)$$

Where all the lambda constants are different. If the isotope λ_k, $2 \leq k \leq n$, is repeated m times, through the small variations discussed in Section 5.2 it is possible to denote the Bateman equation as:

$$B(\lambda_1, \lambda_2, \dots, \underbrace{\lambda_k, \lambda_k + \Delta_k, \lambda_k + 2\Delta_k, \dots, \lambda_k + (m-1)\Delta_k}_{\text{repeated isotope}}, \dots, \lambda_n)$$

For the case where there are p different isotopes, each of one appear k_p times, it is possible to generalize the last notation in the following way:

$$B\left(\underbrace{\lambda_1, \lambda_1 + \Delta_1, \dots, \lambda_1 + (k_1 - 1)\Delta_1}_{\text{isotope } X_1}, \dots, \underbrace{\lambda_p, \lambda_p + \Delta_p, \dots, \lambda_p + \left(k_p - 1\right)\Delta_p}_{\text{isotope } X_p}\right)$$

Due to the extension of the function's argument, it is possible to reduce the notation as:

$$B(\Delta_1, \Delta_2, \dots, \Delta_p) \tag{5.90}$$

The Cetnar's work that was described in Section 5.5 can be summarized considering that he computed the following limit:

$$\lim_{\Delta_i \to 0, i=1,\dots,p} B(\Delta_1, \Delta_2, \dots, \Delta_p) = B_g\left(\lambda_1, \lambda_2, \dots, \lambda_p\right) \tag{5.91}$$

Where $B_g\left(\lambda_1, \lambda_2, \dots, \lambda_p\right)$ is the general solution, which depends on p arguments. As it will be discussed later, inside equation (5.91) lies a formal procedure to compute the increments in order to have adequate results. As it was discussed in Section 5.2, there is not a formal methodology to propose the modification to the Bateman equation, and therefore it is necessary to develop a theory, which can be called the theory of increments.

5.9.1 Case where only one isotope is repeated k_p times.

Using the symmetry function defined in Section 1.8.3 the Bateman equation can be written as:

$$B(\lambda_1, \lambda_2, \dots, \lambda_n) = X_1(0) \prod_{k=1}^{n-1} \lambda_k \, F(\lambda_1, \lambda_2, \dots, \lambda_n) \tag{5.92}$$

It is important to note that B is not symmetric on the lambda arguments due to the initial product over the index k. The following linear chain can be considered:

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} \dots \xrightarrow{\lambda_{p-1}} X_p \xrightarrow{\lambda_p} X_{p+1} \xrightarrow{\lambda_{p+1}} \dots \xrightarrow{\lambda_{n-1}} X_n \xrightarrow{\lambda_n} X_{n+1} \xrightarrow{\lambda_{n+1}} \dots \xrightarrow{\lambda_{n+k_p-2}} X_{n+k_{p-1}} \tag{5.93}$$

In this structure there will be n different isotopes, and the isotope X_p will be repeated $k_p - 1$ times. In other words, there are k_p isotopes in (5.93) that are equal to X_p. As it will be shown it is not necessary to know the positions where the isotope X_p is repeated. In order to use the Bateman solution, a set of small variation will be introduced. It is possible to carry out this procedure in any order. Then, the Bateman solution for this linear chain is given by:

$$B\left(\lambda_1, \lambda_2, \dots, \lambda_{n+k_p-1}\right) = X_1(0) \prod_{k=1}^{n+k_p-2} \lambda_k \, F\left(\lambda_1, \lambda_2, \dots, \lambda_{n+k_p-1}\right) = PF\left(\lambda_1, \lambda_2, \dots, \lambda_{n+k_p-1}\right)$$

Where F was defined in (1.135) and P is a constant whose value is equal to:

$$P = X_1(0) \prod_{k=1}^{n+k_p-2} \lambda_k$$

Now, due to the symmetry of the function F it is possible to make a new enumeration where the first $n-1$ terms correspond to the different isotopes, and the last k_p terms are the repeated isotopes. Therefore:

$$B\left(\lambda_1, \lambda_2, \ldots, \lambda_{n+k_p-1}\right) = PF\left(\lambda_{\mu_1}, \lambda_{\mu_2}, \ldots, \lambda_{\mu_{n-1}}, \lambda_p, \lambda_{p+1}, \ldots, \lambda_{p+k_p-1}\right) \tag{5.94}$$

The last function F can be expressed as:

$$\sum_{i=1}^{n-1} e^{-\lambda_{\mu_i} t} \prod_{\substack{j=1 \\ j \neq i}}^{n-1} \frac{1}{\lambda_{\mu_j} - \lambda_{\mu_i}} \prod_{\substack{j=p \\ j \neq i}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_{\mu_i}} + \sum_{i=p}^{p+k_p-1} e^{-\lambda_i t} \prod_{\substack{j=1 \\ j \neq i}}^{n-1} \frac{1}{\lambda_{\mu_j} - \lambda_i} \prod_{\substack{j=p \\ j \neq i}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_i} \tag{5.95}$$

It is necessary to make simplifications to the second term of the last equation. One of them is related with the product. Firstly, the constants λ_{p+j} will be rewritten as:

$$\begin{array}{cccc} \lambda_p & \lambda_{p+1} & \ldots & \lambda_{p+k_p-1} \\ \downarrow & \downarrow & \ldots & \downarrow \\ \lambda_p & \lambda_p + \Delta_p & \ldots & \lambda_p + (k_p - 1)\Delta_p \end{array}$$

Using the last relationships, it is possible to deduce an expression for the following product for the index p:

$$\prod_{\substack{j=p \\ j \neq p}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_p} = \frac{1}{\lambda_{p+1} - \lambda_p} \frac{1}{\lambda_{p+2} - \lambda_p} \cdots \frac{1}{\lambda_{p+k_p-1} - \lambda_p}$$

$$= \frac{1}{\Delta_p} \frac{1}{2\Delta_p} \cdots \frac{1}{(k_p - 1)\Delta_p} = \left(\frac{1}{\Delta_p}\right)^{k_p-1} \frac{1}{(k_p - 1)!}$$

In the same way, for the index $p+1$

$$\prod_{\substack{j=p \\ j \neq p+1}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_{p+1}} = \frac{1}{\lambda_p - \lambda_{p+1}} \frac{1}{\lambda_{p+2} - \lambda_{p+1}} \cdots \frac{1}{\lambda_{p+k_p-1} - \lambda_{p+1}}$$

$$= \left(-\frac{1}{\Delta_p}\right)\left(\frac{1}{\Delta_p}\right)\left(\frac{1}{2\Delta_p}\right) \cdots \frac{1}{(k_p - 2)!\Delta_p} = -\left(\frac{1}{\Delta_p}\right)^{k_p-1} \frac{1}{(k_p - 2)!}$$

For the case $i = p + 2$:

$$\prod_{\substack{j=p \\ j \neq p+2}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_{p+2}} = \frac{1}{\lambda_p - \lambda_{p+2}} \frac{1}{\lambda_{p+1} - \lambda_{p+2}} \frac{1}{\lambda_{p+3} - \lambda_{p+2}} \cdots \frac{1}{\lambda_{p+k_p-1} - \lambda_{p+2}}$$

$$= \left(-\frac{1}{2\Delta_p}\right)\left(-\frac{1}{\Delta_p}\right)\left(\frac{1}{\Delta_p}\right)\cdots\left(\frac{1}{(k_{p-3}-3)\Delta_p}\right) = \left(\frac{1}{\Delta_p}\right)^{k_p-1}\frac{1}{2(k_p-3)!}$$

And following the same reasoning, it is possible to infer the case $i = r,\ \ 1 \leq r \leq k_p$:

$$\prod_{\substack{j=p \\ j\neq p+r}}^{p+k_p-1} \frac{1}{\lambda_j - \lambda_{p+2}} = (-1)^r \left(\frac{1}{\Delta_p}\right)^{k_p-1} \frac{1}{r!\,(k_p-r)!} \tag{5.96}$$

Using the last expression, the second term in (5.95) is equal to:

$$\left(\frac{1}{\Delta_p}\right)^{k_p-1} e^{-\lambda_p t} \cdot \sum_{i=p}^{p+k_p-1} e^{-(i-p)\Delta_p t} \prod_{\substack{j=1 \\ j\neq i}}^{n-1} \frac{(-1)^{i-p}}{\lambda_{\mu_j} - (\lambda_p + (i-p)\Delta_p)} \frac{1}{i!\,(k_p+p-1-i)!} \tag{5.97}$$

On the other hand, the first product in the left side of equation (5.95) can be expressed as:

$$\sum_{i=1}^{n-1} e^{-\lambda_{\mu_i} t} \prod_{\substack{j=1 \\ j\neq i}}^{n-1} \frac{1}{\lambda_{\mu_j} - \lambda_{\mu_i}} \prod_{\substack{j=p \\ j\neq i}}^{p+k_p-1} \frac{1}{\lambda_p + (j-p)\Delta_p - \lambda_{\mu_i}} \tag{5.98}$$

Then, the Bateman equation with increments is the sum of equation (5.97) with equation (5.98). Using these last equations, it is possible to obtain a set of inequalities that provides information about the increments Δ.

5.9.2 Formal definition of limit.

In mathematical analysis there is a formal definition of limit, which has the following sentence (Bartle and Sherpert, 2010): Let f a real function defined in the interval (a, b), and a number x_0. It is said that $\lim\limits_{x\to x_0} f(x)$ exists and that it is equal to L, if for all $\epsilon > 0$ there is a $\delta > 0$, such that, $\forall x \in (a, b)$ if:

$$|x - x_0| < \delta, \text{ then } \ |f(x) - L| < \epsilon"$$

It is possible to use this definition to know how to compute the increments in the Bateman equation. First it is necessary to interpret that in terms of the present problem. In first place, if it will be supposed that only one isotope, X_p is repeated k_p times, therefore the limit given in (5.91) can be expressed as:

$$\lim_{\Delta_p \to 0} B(\Delta_p) = B_g(\lambda_1, \lambda_2, \ldots, \lambda_n) \tag{5.99}$$

The left part of (5.99) is equal to the Bateman equation with increments that was developed in (5.21). The right side is the general solution. In this particular case, this is the general solution where the isotope X_p is repeated k_p times. Therefore, from the epsilon-delta definition, we will have the following inequality:

$$\left|B\left(\Delta_p\right) - B_g\left(\lambda_1, \lambda_2, \dots, \lambda_p\right)\right| < \epsilon \tag{5.100}$$

In terms of the present problem, ϵ will be the error between the general solution B_g and the function of increments $B(\Delta_p)$. Then by the definition, if the limit exists, for a given error it will be possible to find a value of δ, such that it is true that:

$$\left|\Delta_p - 0\right| = \left|\Delta_p\right| < \delta \tag{5.101}$$

This value δ allows finding the increments, because it will be an upper bound. Since that several authors showed that the limit exists, and proposed a general equation, it is possible to work with inequalities with the purpose to find an expression for this δ value. In other words, both functions inside (5.100) are known, and the procedure is reduced to solve the inequalities in order to find (5.101). In other words, the procedure to find the value of the increments is reduced to apply the formal definition of limit. A graphical representation of this procedure is shown in Figure 5.2.

5.9.3 Special considerations.

Strictly speaking, the limit described in (5.91) is a multivariable limit, not a single limit as was set in equation (5.100). Nevertheless, the problem is extremely complex if we considered the multivariable case. Therefore, the procedure described in the last section is only valid when only one isotope is repeated, which is a very rare case: in fact, it is more probable that more than one isotope is repeated. Nevertheless, it is possible to suppose that the errors related to the increments are independents. In other words, for the case where more than one isotope is repeated, it will be possible to compute the error due to each increment of the repeated isotope, and finally to compute a total error, which will be defined as:

$$\epsilon_t = \epsilon_{\Delta_1} + \epsilon_{\Delta_2} + \cdots + \epsilon_{\Delta_m} \tag{5.102}$$

Furthermore, in order to deal with inequalities in an easy way, it is necessary to use a general Bateman solution whose form is not complex. In such terms, the Cetnar's solution described in Section 5.5 is discarded because it is expressed in very difficult nested sums, as it was described before. The most appropriate general solution to the present problem is the one obtained by the Inverse Laplace Method, which was described in 1.9.2.

5.9.4 The main inequality.

The author of the present thesis continues working in an adequate way to solve the inequality given in (5.100). It is expected that such solution will be part of a future publication, and therefore in the following lines the main inequality that needs to be solved is described, as well as some of the properties of such problem.

Once the equation (5.95), with the terms given in (5.97) and (5.98), is replaced in (5.100), and the general solution obtained by the Inverse Laplace method is used, the following inequalities need to be solved:

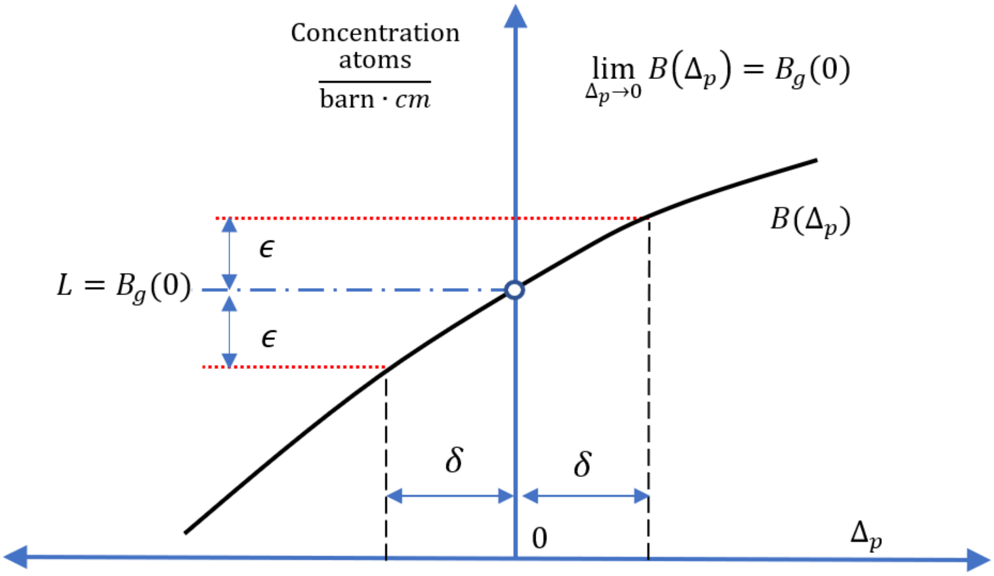

Figure 5. 2. Scheme of the procedure to compute the increment Δ_p using the epsilon-delta definition of limit. In the image, the point in the function $B(\Delta_p)$ does not appear in the vertical axis, because in this point such function is not defined, even when the limit exists.

$$\left| \prod_{\substack{j=p\\ j\neq i}}^{p+k_p-1} \frac{1}{\lambda_p + (j-p)\Delta_p - \lambda_{\mu_i}} - \frac{1}{\left(\lambda_p - \lambda_{u_i}\right)^{k_p}} \right| \tag{5.103}$$

$$\left| \sum_{i=p}^{p+k_p-1} e^{-(i-p)\Delta_p t} \prod_{\substack{j=1\\ j\neq i}}^{n-1} \frac{(-1)^{i-p}}{\lambda_{\mu_j} - \left(\lambda_p + (i-p)\Delta_p\right)} \frac{\left(\frac{1}{\Delta_p}\right)^{k_p-1} e^{-\lambda_p t}}{i!\left(k_p + p - 1 - i\right)!} - \sum_{i=1}^{k_p} c_{p,i} \frac{e^{-\lambda_p t} t^{i-1}}{(i-1)!} \right| \tag{5.104}$$

For the first inequality it is possible to show that:

$$\prod_{\substack{j=p\\ j\neq i}}^{p+k_p-1} \frac{1}{\lambda_p + (j-p)\Delta_p - \lambda_{\mu_i}} = \prod_{\substack{j=p\\ j\neq i}}^{p+k_p-1} \frac{1}{\left(\lambda_p - \lambda_{\mu_i}\right)\left(1 - \frac{(j-p)\Delta_p}{\lambda_p - \lambda_{\mu_i}}\right)}$$

$$= \frac{1}{\left(\lambda_p - \lambda_{u_i}\right)^{k_p}} \prod_{\substack{j=p\\ j\neq i}}^{p+k_p-1} \frac{1}{\left(1 - \frac{(j-p)\Delta_p}{\lambda_p - \lambda_{\mu_i}}\right)}$$

Starting from these equations there have been found several other relationships that can be help us to solve the inequality, and therefore to find the relationship between the error and the increments. The most difficult part is the analysis of the equation (5.104), which strongly depends on the computation of derivatives of the Inverse Laplace method, which was showed in Section 5.8.3. Such topics are part of a future research.

5.10 Conclusions of the Chapter 5.

The present chapter was related to the study of the general solutions of the Bateman equation, and with some important findings that can be deduced from them. Firstly, through a detailed review it was possible to conclude that the general solutions appeared, in a formal way, in the 60's. Since then, other general solutions were obtained, which belong to a wide range of disciplines. Therefore, the solution that was developed by Cetnar in 2006 was not the first one that appeared, as wrongly some authors suggest.

It is possible to divide all the published solutions according to the methodology that was used to find them. Essentially there are four types: the inverse Laplace method, the integral solution, the limit solution and the power series solution. From this study it was possible to conclude that the most straightforward methodology that provides a general solution is the inverse Laplace method. Additionally, it was discussed that some of the solutions can be represented as nested sums, others involve the calculation of high order derivatives, and others require to compute a multiple integration. Due to these elements, it is very complex task to include some of these solution in a burnup code.

Nevertheless, using symbolic calculation and computer algebra, it is possible to develop an algorithm that includes some of the general solutions in a burnup or activation code. After an algorithmic analysis it was concluded that the most adequate solutions that can be solved using these computational tools are the Laplace and integral type. In terms of numerical results, a comparison study was carried out, which demonstrated that the developed algorithm can build the general solution in a time that is approximately equal to the time that is involved when the modified Bateman equation is used.

Since the general solutions were developed without restrictions related to the position of repeated isotopes, it is possible to reduce the number of possible linear chains with repeated isotopes, using a set of rules that were deduced with a combinatorial study. From this combinatorial analysis can be obtained a set of rules that allows building the linear chains that are physically possible in practical problems, and therefore the complexity of the cases is reduced. An important finding was the conclusion that is not possible that in a linear chain two consecutive isotopes can be equal.

In terms of the increments it was possible to propose a methodology to compute such values in a formal way. This methodology is based in the concept of limit, and in the Bateman solution with increments. The main difficulty of this methodology consists of solving inequalities, which is part of a future research. From the above facts it is possible to conclude that the general solutions continue being a topic of interest and where can be obtained important contributions to the Bateman equations.

6. General Conclusions.

The work developed in this thesis addresses several aspects of the Bateman equations, which allow solving them in a more precise and fast way. A set of new algorithms was proposed, which contributes from a programming level until a mathematical modelling a of complex decay and transmutation structures. In the present work the linearization process was improved, developing an alternative algorithm that builds the linear chain in a very fast way. It was possible to conclude from this part, that some of the methodologies that the linear chain uses can be improved using new approaches to the topology of decay and transmutation networks.

Regarding the cyclic chains, the present work contains a detailed study of these structures, proposing a classification to them, and developing a mathematical background to this solution. From this part has been concluded that the study of cyclic chains is very important, because the solution of these structures allows reducing the number of linear chains that need to be solved with the Bateman equation. On the topic of general solutions, the present work shows that a theory of increments can be developed from it. This represents an important contribution to the development of burnup codes, because it can provide a formal way to introduce small modifications in the Bateman equation. Another important conclusion from this topic was the development of a computer algebra algorithm, which allows including general solutions in burnup codes.

In terms of the yield's assignation, it has been concluded that is very important to use only independent yields and using complete and transmutation networks. Additionally, in order to make adequate comparison between two different burnup codes, it is necessary to study the way in which the yields are assigned to a given structure.

The contributions described before will be part of the burnup code Szilard, belonging to the AZTLAN Platform project. From the above discussion it is possible to conclude that all the objectives defined at the beginning of this doctoral research were fulfilled, and that it is possible to find improvements to the linear chain method and to the solution of the Bateman equations. As product of the present doctoral research, the following papers were published in an international journal as well as in international congress.

Journal Papers:

Cruz-López, C. ; François, J., 2018. An alternative algorithm for the linearization process for transmutation and decay networks. Computer Physics Communications. Vol. 231, pp 122-139.

Cruz-López, C.; François, 2019. Two alternative approaches to the solution of cyclic chains in transmutation and decay problem. Submitted to Computer Physics Communications (December, 2019).

Conference Papers:

Cruz-López, C.; François, J., 2018. An alternative algorithm for the linearization process for transmutation and decay networks. The PHYSOR 2018 conference: Reactor Physics Paving The Way Towards More Efficient Systems. Mexico, April 22-26, 2018.

Cruz-López, C.; François, J., 2019. An Algebraic Computation Approach to the General Solutions of the Bateman Equation. Submitted to “The PHYSOR 2020 conference: Transition to a Scalable Nuclear Future” (November, 2019).

Cruz-López, C.; François, J., 2017. Desarrollo de un Algoritmo Computacional para la Linealización de Cadenas de Decaimiento y Transmutación. XXVIII Congreso Anual de la Sociedad Nuclear Mexicana-2017 LAS/ANS-Symposium, New Technologies for a Nuclear Power Expansion Program. Ciudad de México, México, June 2017.

Cruz-López, C.; François, J., 2019. Desarrollo de un Algoritmo de Cálculo Simbólico para la Resolución de las Ecuaciones de Decaimiento y Transmutación de Bateman. XXX Congreso Anual de la Sociedad Nuclear Mexicana-2019. Aplicaciones Pacíficas de la Energía Nuclear en Beneficio de México. Nuevo León, México, August 2019.

Appendix A.

A.1 Solution of the Bateman equation by Mathematical Induction.

The strong mathematical induction will be used to formulate the hypothesis as:

1) (1.24) is valid for $n = 1, 2, 3$
2) It will be supposed that is valid for $n - 1$
3) Finally, it will be proved that (1.24) it is valid for n (using supposition 2)

Starting with the following differential equation:

$$\frac{dX_n}{dt} = \lambda_{n-1} X_{n-1}(t) - \lambda_n X_n \tag{A.1}$$

Using supposition 2) the term $X_{n-1}(t)$ will be replaced in the last equation:

$$\frac{dX_n}{dt} = \lambda_{n-1} \left[\lambda_1 \lambda_2 \ldots \lambda_{n-2} X_1(0) \sum_{i=1}^{n-1} \prod_{\substack{j=1 \\ j \neq i}}^{n-1} \frac{1}{\lambda_j - \lambda_i} e^{-\lambda_i t} \right] - \lambda_n X_n \tag{A.2}$$

Both sides are multiplied by $e^{\lambda_n t}$, and the term $e^{\lambda_n t} \lambda_n X_n$ is added:

$$e^{\lambda_n t} \lambda_n X_n + e^{\lambda_n t} \frac{dX_n}{dt} = e^{\lambda_n t} \lambda_{n-1} \left[\lambda_1 \lambda_2 \ldots \lambda_{n-2} X_1(0) \sum_{i=1}^{n-1} \prod_{\substack{j=1 \\ j \neq i}}^{n-1} \frac{1}{\lambda_j - \lambda_i} e^{-\lambda_i t} \right] \tag{A.3}$$

Rearranging the terms is follows that:

$$e^{\lambda_n t} \lambda_n X_n + e^{\lambda_n t} \frac{dX_n}{dt} = \frac{d}{dt} \left(X_n e^{\lambda_n t} \right) \tag{A.4}$$

Therefore, the resultant expression is:

$$\frac{d}{dt} \left(X_n e^{\lambda_n t} \right) = \lambda_{n-1} \left[\lambda_1 \lambda_2 \ldots \lambda_{n-2} X_1(0) \sum_{i=1}^{n-1} \prod_{\substack{j=1 \\ j \neq i}}^{n-1} \frac{1}{\lambda_j - \lambda_i} e^{-(\lambda_i - \lambda_n) t} \right] \tag{A.5}$$

After the variable t is changed for t_1, it is necessary to perform the following integration:

$$\int_0^t \frac{d}{dt_1}\left(X_n e^{\lambda_n t_1}\right)dt_1 = \int_0^t \lambda_{n-1}\left[\lambda_1\lambda_2 \dots \lambda_{n-2}X_1(0)\sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}e^{-(\lambda_i-\lambda_n)t_1}\right]dt_1 \tag{A.6}$$

Since that $\lambda_i, 1 \leq i \leq n$ is constant with respect to t_1, it is only necessary to analyze the following integrals:

$$\int_0^t \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}e^{-(\lambda_i-\lambda_n)t_1}\,dt_1 = \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\int_0^t e^{-(\lambda_i-\lambda_n)t_1}dt_1 \tag{A.7}$$

$$\sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\int_0^t e^{-(\lambda_i-\lambda_n)t_1}dt_1 = \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\frac{1}{(\lambda_n-\lambda_i)}\left[e^{-(\lambda_i-\lambda_n)t_1}\right]_0^t \tag{A.8}$$

After carrying out the evaluation:

$$= \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\frac{1}{(\lambda_n-\lambda_i)}\left(e^{-(\lambda_i-\lambda_n)t}-1\right) \tag{A.9}$$

The last term is expressed in the following way:

$$= \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\frac{1}{(\lambda_n-\lambda_i)}e^{-(\lambda_i-\lambda_n)t} - \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\frac{1}{(\lambda_n-\lambda_i)} \tag{A.10}$$

If the term $1/(\lambda_n-\lambda_i)$ is included in the product, the index can be extended until n in (A.10):

$$\sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{\lambda_j-\lambda_i}e^{-(\lambda_i-\lambda_n)t} - \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{\lambda_j-\lambda_i} \tag{A.11}$$

Finally, we can apply (1.120) in the second term in (A.11), therefore:

$$-\sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{1}{\lambda_j-\lambda_i} = \prod_{j=1}^{n-1}\frac{1}{\lambda_j-\lambda_n} \tag{A.12}$$

Therefore (A.9) can be written as:

$$\sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\frac{1}{(\lambda_n-\lambda_i)}\left[e^{-(\lambda_i-\lambda_n)t_1}\right]_0^t = \sum_{i=1}^{n-1}\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{e^{-(\lambda_i-\lambda_n)t}}{\lambda_j-\lambda_i} + \prod_{j=1}^{n-1}\frac{1}{\lambda_j-\lambda_n} \tag{A.13}$$

Since the term n it is not considered in $\prod_{j=1}^{n-1}\frac{1}{\lambda_j-\lambda_n}$, it is possible to rewrite this term as:

$$\prod_{j=1}^{n-1}\frac{1}{\lambda_j-\lambda_n}=\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{\lambda_j-\lambda_n}$$

If the term (A.13) is replaced in (A.8), it follows that:

$$\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n-1}\frac{1}{\lambda_j-\lambda_i}\int_0^t e^{-(\lambda_i-\lambda_n)t_1}dt_1=\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{e^{-(\lambda_i-\lambda_n)t}}{\lambda_j-\lambda_i}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{\lambda_j-\lambda_n} \tag{A.14}$$

Then, through (A.6) and (A.7):

$$\int_0^t \frac{d}{dt_1}\left(X_n e^{\lambda_n t_1}\right)dt_1=\lambda_1\lambda_2\ldots\lambda_{n-2}\lambda_{n-1}X_1(0)\left[\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{e^{-(\lambda_i-\lambda_n)t}}{\lambda_j-\lambda_i}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{\lambda_j-\lambda_n}\right] \tag{A.15}$$

The integration of the left side is straightforward, considering that $X_n(t=0)=0$:

$$\int_0^t \frac{d}{dt_1}\left(X_n e^{\lambda_n t_1}\right)dt_1=X_n(t)e^{\lambda_n t} \tag{A.16}$$

Therefore:

$$X_n(t)e^{\lambda_n t}=\lambda_1\lambda_2\ldots\lambda_{n-2}\lambda_{n-1}X_1(0)\left[\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{e^{-(\lambda_i-\lambda_n)t}}{\lambda_j-\lambda_i}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{\lambda_j-\lambda_n}\right] \tag{A.17}$$

Multiplying both sides of (A.17) by $e^{-\lambda_n t}$:

$$X_n(t)=\lambda_1\lambda_2\ldots\lambda_{n-2}\lambda_{n-1}X_1(0)\left[\sum_{i=1}^{n-1}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{e^{-(\lambda_i-\lambda_n)t}}{\lambda_j-\lambda_i}+\prod_{\substack{j=1\\j\neq n}}^{n}\frac{1}{\lambda_j-\lambda_n}\right]e^{-\lambda_n t} \tag{A.18}$$

Finally, (A.18) can be written as:

$$X_n(t)=\lambda_1\lambda_2\ldots\lambda_{n-2}\lambda_{n-1}X_1(0)\left[\sum_{i=1}^{n}\prod_{\substack{j=1\\j\neq i}}^{n}\frac{e^{-\lambda_i t}}{\lambda_j-\lambda_i}\right] \tag{A.19}$$

Comparing (A.19) with (1.24) it is possible to conclude that it was obtained the case $k=n$, and therefore the validity of the equation has been showed. Then, the mathematical induction process is complete.

A.2 Proof of Properties in section 3.3.

Proposition:

“In a linear chain, the dashes number in consecutive elements increases. Therefore, if there are two consecutive elements in V_2^*, with a growing dashes number, then these belong to the same linear chain; even more, they are ordered.”

Importance and proof:

The first property is very important, because it guarantees that vector V_2^* can be divided using the decrease in the dashes number. Then, the vector can be cut based on this, with the guarantee that all the resultant elements belong to a linear chain.

The proof of the first property can be carried out starting from the definition given in (3.10). It will be considered that there are two consecutive elements in V_2^* with an increasing dashes number, which can be denoted by:

$$\underbrace{P0 - \cdots - j}_{k-dashes}, \qquad \underbrace{P0 - \cdots - l}_{k-dashes} - h \tag{A.20}$$

In order to show that these two elements belong to the same linear chain, it is only necessary to demonstrate that:

$$P0 - \cdots - j = P0 - \cdots - l \tag{A.21}$$

Now, under the lexicographical ordering, and using the fact that the elements are consecutive, it follows that:

$$P0 - \cdots - j < \ P0 - \cdots - l - h \tag{A.22}$$

These last elements can be seen as ordered pairs, in the form:

$$P0 - \cdots - j = \ (P0 - \cdots - j, \text{empty space}) \tag{A.23}$$

$$P0 - \cdots - l - h = < (P0 - \cdots - l, -h) \tag{A.24}$$

Since the elements are consecutive and due to the fact that the algorithm produces strings with a continuous dashes number, it follows that $P0 - \cdots - j$ and $P0 - \cdots - l$ have the same number of characters. Therefore, the “empty space” has been added to compare the elements of the ordered pairs, one to one, considering that the empty space is “minor” (under a lexicographical order) that any other character. In other words, adding the empty space does not modify the problem. Using the definition given in (3.10), the inequality in (A.22) and the expressions as ordered pairs given in (A.23) and (A.24), only one of the following sentences is true:

i) $P0 - \cdots - j < P0 - \cdots - l$

ii) $P0 - \cdots - j = P0 - \cdots - l$, empty space $< -h$

For the proof, it is necessary to show that we can discard the first possibility. It will proceed by contradiction. It will be assumed that the first condition is true. Then:

$$P0 - \cdots - j < P0 - \cdots - l \tag{A.25}$$

Now, it is necessary to observe that the string $P0 - \cdots - l$ is the immediate ancestor of $P0 - \cdots - l - h$, then under a lexicographical ordering this element must be to the left of it. In other words:

$$P0 - \cdots - l < P0 - \cdots - l - h \tag{A.26}$$

Nevertheless, because $P0 - \cdots - j$ and $P0 - \cdots - l - h$ are consecutive elements under a lexicographical ordering, it follows that the only possibility for the position of $P0 - \cdots - l$, the immediate ancestor of $P0 - \cdots - l - h$, must be to the left of $P0 - \cdots - j$, i.e.:

$$P0 - \cdots - l < P0 - \cdots - j \tag{A.27}$$

But this is a contradiction the supposition in (A.25). Then, the only possibility is that $P0 - \cdots - j = P0 - \cdots - l$ and *an* empty space $< -h$, therefore it has been proved.

Proposition:

"If k −consecutive elements in V_2^* have the same dashes number from the position j, then it is necessary to duplicate $k - 1$ times the previous "segment" of a linear chain until the position $j - 1$."

Importance and proof:

As mentioned in the Chapter 3, this property allows duplicating chains with several branches. If there are k consecutive elements with the same dashes number, denoted by j, then it is necessary to build the linear chains under the following scheme:

$$\begin{cases} X_0 \to X_1 \to \cdots \to X_{j-1} \to X_1 \\ X_0 \to X_1 \to \cdots \to X_{j-1} \to X_2 \\ \cdots \\ X_0 - X_1 \to \cdots \to X_{j-1} \to X_k \to \end{cases} \tag{A.28}$$

The right arrow in the final chain indicates that this chain can continue, whereas the previous chains do not.

The proof for this property must show two facts:

i) When there are elements in V_2^* with the same dashes number, denoted by j, it is necessary to duplicate the segment of the linear chain, because each of these elements is a branch or ramification.

ii) Only the last element with a repeated dashes number can have more descendants.

For the first fact it will be supposed that there are k consecutive elements in the vector V_2^*, with a dashes number j:

$$P0, \dots, \underbrace{P0 - \cdots - n}_{j-1, dashes}, \underbrace{P0 - \cdots - m - 1}_{j\ dashes}, \underbrace{P0 - \cdots - m - 2}_{j\ dashes}, \dots, \underbrace{P0 - \cdots m - k}_{j\ dashes}, \dots \qquad \text{(A.29)}$$

First, it is necessary to show that $P0 - \cdots - n$ is a common ancestor for all the k items with the same dashes number.

It is a straightforward task, because $P0 - \cdots - n$ and $P0 - \cdots - m - 1$ are consecutive elements, and they have an increasing dashes number. Therefore, by the previous theorem, these elements belong to the same linear chain. Therefore, it follows that:

$$P0 - \cdots - n = \ P0 - \cdots - m \qquad \text{(A.30)}$$

Then this element is a common ancestor (in fact the immediate ancestor) of the k elements. This imply that the k elements are branches in this point, and therefore in order to build the linear chains it is necessary to duplicate the segment:

$$P0, \dots, P0 - \cdots - m$$

Then, the first fact is proved.

For the second fact it will be assumed that there is at least one element after $P0 - \cdots f - u$ with a dashes number given by $j + 1$:

$$P0 - \cdots - f - u$$

Again, by the previous theorem, this element belongs to the same linear chain that $P0 - \cdots - m - k$, and thus it can be written as:

$$P0 - \cdots - k - u \qquad \text{(A.31)}$$

Finally, since the k index that appears before the u, all the others $k - 1$ items with the same dashes number do not belong to the same linear chain. Therefore, only the last with a repeated dashes number can have more descendants, that is that was wanted to be proved.

Proposition:

"If the dashes number decreases between two consecutive elements, j and $j + 1$ in V_2^*, then the element j is the final of the current linear chain, and the $j + 1$ item is where the following linear chain begins."

Importance and proof:

This sentence allows to divide the vector V_2^* based on the decreases in the dashes number. Its proof guarantees that the "segment" cut belongs to distinct linear chains.

The proof is very simple. If it is assumed that under lexicographical ordering, two consecutive elements have a decreases dashes number:

$$\underbrace{P0 - \cdots - i - j}_{k\ dashes},\ \underbrace{P0 - \cdots - l - m}_{n\ dashes},\quad n < k \tag{A.32}$$

Such elements can be written as:

$$P0 - \cdots - i - j = (\underbrace{P0 - \cdots - u}_{n\ dashes}, \underbrace{- \cdots - i - j}_{k-n\ dashes}) \tag{A.33}$$

$$P0 - \cdots - l - m = (\underbrace{P0 - \cdots - l - m}_{n\ dashes}, \underbrace{\qquad\qquad\qquad}_{k-n\ empty\ spaces}) \tag{A.34}$$

According to (3.10), there are only two cases:

i) $P0 - \cdots - u < P0 - \cdots - l - m$

$ii)$ $P0 - \cdots - u = P0 - \cdots - l - m$ and $- \cdots - i - j < \underbrace{\qquad\qquad\qquad}_{k-n\ empty\ spaces}$

The last case is always false, because the empty space is less than any other character. Then the only valid case is $P0 - \cdots - u < P0 - \cdots - l - m$. This implies that:

$$P0 - \cdots - u < P0 - \cdots - u - 1 < P0 - \cdots - u - 1 - 1 < \cdots < P0 - \cdots u - \cdots - i - j$$
$$< P0 - \cdots - l - m$$

All the elements before the string $P0 - \cdots - l - m$ belong to the same linear chain, nevertheless, this final element is not part of the linear chain, because of the first theorem, this would imply that its dashes number is greater than the dashes number of $P0 - \cdots - i - j$. Therefore, this element does not belong to the same linear chain, and the proposition is proved.

A.3 Proof of the Equation (4.130)

It will be proved that:

$$Y_p = X_1(0) b^{\mathrm{eff}}_{j,Y_1} \lambda^{\mathrm{eff}}_j \prod_{h=1}^{p-1} b^{\mathrm{eff}}_{Y_h,Y_{h+1}} \lambda^{\mathrm{eff}}_{Y_h} \prod_{m=1}^{j-1} b^{\mathrm{eff}}_{m,m+1} \lambda^{\mathrm{eff}}_m$$
$$\left(\sum_{i=1}^{n} F\left(\beta_i, \lambda^{\mathrm{eff}}_{Y_1}, \dots, \lambda^{\mathrm{eff}}_{Y_p}\right) \prod_{l=j+1}^{n} \left(\lambda^{\mathrm{eff}}_l - \beta^*_i\right) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{\left(\beta^*_j - \beta^*_i\right)} \right)$$

Proof

It will be used induction over p. Considering that

$$F(a_1, a_2, \dots, a_n) = \sum_{i=1}^{n} e^{-a_i t} \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{(a_j - a_i)}$$

The case $p = 1$ was obtained in (4.129), being equal to

$$Y_1^R = X_1(0) b_{j,Y_1}^{\text{eff}} \lambda_j^{\text{eff}} \prod_{m=1}^{j-1} b_{m,m+1}^{\text{eff}} \lambda_m^{\text{eff}}$$

$$\left(\sum_{i=1}^{n} \frac{e^{-\beta_i^* t} - e^{-\lambda_{Y_1}^{\text{eff}} t}}{\lambda_{Y_1}^{\text{eff}} - \beta_i^*} \prod_{l=n-j+1}^{n} \left(\lambda_l^{\text{eff}} - \beta_i^*\right) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{\left(\beta_j^* - \beta_i^*\right)} \right)$$

It is clear that:

$$\frac{e^{-\beta_i^* t} - e^{-\lambda_{Y_1}^{\text{eff}} t}}{\lambda_{Y_1}^{\text{eff}} - \beta_i^*} = F(\beta_i^*, Y_1)$$

Therefore, case $p = 1$ confirm equation (4.130). The next step is assuming that equation is valid for $p = u$, and finally it is necessary that is valid to $p = u + 1$.Using the forward method, it follows:

$$Y_{u+1} = e^{-\lambda_{Y_{u+1}}^{\text{eff}} t} b_{Y_u,Y_{u+1}}^{\text{eff}} \lambda_{Y_u}^{\text{eff}} \int_0^t Y_u(t') e^{\lambda_{Y_{u+1}}^{\text{eff}} t'} dt'$$

Using the induction assumption:

$$Y_{u+1} = \Lambda e^{-\lambda_{Y_{u+1}}^{\text{eff}} t} \sum_{i=1}^{n} \prod_{l=j+1}^{n} \left(\lambda_l^{\text{eff}} - \beta_i^*\right) \prod_{\substack{j=1 \\ j \neq i}}^{n} \frac{1}{\left(\beta_j^* - \beta_i^*\right)} \int_0^t F\left(\beta_i, \lambda_{Y_1}^{\text{eff}}, \dots, \lambda_{Y_u}^{\text{eff}}\right) e^{\lambda_{Y_{u+1}}^{\text{eff}} t'} dt'$$

With

$$\Lambda = X_1(0) b_{j,Y_1}^{\text{eff}} \lambda_j^{\text{eff}} \prod_{h=1}^{u} b_{Y_h,Y_{h+1}}^{\text{eff}} \lambda_{Y_h}^{\text{eff}} \prod_{m=1}^{j-1} b_{m,m+1}^{\text{eff}} \lambda_m^{\text{eff}}$$

If it is considered that $a_1 = \beta_i^*, a_2 = \lambda_{Y_1}^{\text{eff}}, a_3 = \lambda_{Y_2}^{\text{eff}}, \dots, a_{u+1} = \lambda_{Y_u}^{\text{eff}}, a_{u+2} = \lambda_{Y_{u+1}}^{\text{eff}}$, then:

$$\int_0^t F\left(\beta_i, \lambda_{Y_1}^{\text{eff}}, \dots, \lambda_{Y_u}^{\text{eff}}\right) e^{\lambda_{Y_{k+1}}^{\text{eff}} t'} dt' = \int_0^t F(a_1, a_2, \dots, a_{u+1}) e^{-a_{u+2} t'} dt'$$

$$= \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j \neq i}}^{u+1} \frac{1}{(a_j - a_i)} \int_0^t e^{-(a_i - a_{u+2}) t'} dt' = \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j \neq i}}^{u+1} \frac{1}{(a_j - a_i)} \frac{e^{-(a_i - a_{u+2}) t} - 1}{(a_{u+2} - a_i)}$$

$$= \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j \neq i}}^{u+2} \frac{e^{-(a_i - a_{u+2}) t} - 1}{(a_j - a_i)} = \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j \neq i}}^{u+2} \frac{e^{-(a_i - a_{u+2}) t}}{(a_j - a_i)} - \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j \neq i}}^{u+2} \frac{1}{(a_j - a_i)}$$

Using (1.120) we will have:

$$\int_0^t F\left(\beta_i, \lambda_{Y_1}^{\text{eff}}, \dots, \lambda_{Y_u}^{\text{eff}}\right) e^{\lambda_{Y_{u+1}}^{\text{eff}} t'} dt' = \sum_{i=1}^{u+1} \prod_{\substack{j=1 \\ j\neq i}}^{u+2} \frac{e^{-(a_i - a_{u+2})t}}{(a_j - a_i)} + \prod_{\substack{j=1 \\ j\neq u+2}}^{u+2} \frac{1}{a_j - a_{u+2}}$$

$$= \sum_{i=1}^{u+2} \prod_{\substack{j=1 \\ j\neq i}}^{u+2} \frac{e^{-(a_i - a_{u+2})t}}{(a_j - a_i)}$$

Therefore

$$Y_{u+1} = \Lambda \sum_{i=1}^{n} \prod_{l=k+1}^{n} \left(\lambda_l^{\text{eff}} - \beta_i^*\right) \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\left(\beta_j^* - \beta_i^*\right)} e^{-\lambda_{Y_{u+1}}^{\text{eff}} t} \sum_{i=1}^{u+2} \prod_{\substack{j=1 \\ j\neq i}}^{u+2} \frac{e^{-(a_i - a_{u+2})t}}{(a_j - a_i)}$$

$$= \Lambda \sum_{i=1}^{n} F(a_1, a_2, \dots, a_{k+2}) \prod_{l=j+1}^{n} \left(\lambda_l^{\text{eff}} - \beta_i^*\right) \prod_{\substack{j=1 \\ j\neq i}}^{n} \frac{1}{\left(\beta_j^* - \beta_i^*\right)}$$

Which it was searched to proof.

A.4 Proof of the Equation (5.43)

Equation (1.24) is equivalent to the following expression:

$$N_n(t) = \frac{N_1(0)}{b_n} \sum_{i=1}^{n-1} b_i \frac{e^{-d_i t} - e^{-d_n t}}{d_n - d_i} \prod_{j=1, j\neq i}^{n-1} \frac{b_j}{d_j - d_i} \tag{A.35}$$

The equation (1.24) will be written as follows:

$$\frac{N_1(0)}{b_n} \left[\sum_{i}^{n-1} b_i\, e^{-d_i t} \prod_{j=1, j\neq i}^{n} \frac{b_j}{d_j - d_i} + b_n e^{-d_n t} \prod_{j=1, j\neq n}^{n} \frac{b_j}{d_j - d_n} \right] \tag{A.36}$$

Adding and subtracting the following terms (they had been highlighted in red):

$$\begin{aligned} &= \frac{N_1(0)}{b_n} \sum_{i}^{n-1} b_i\, e^{-d_i t} \prod_{j=1, j\neq i}^{n} \frac{b_j}{d_j - d_i} + \frac{N_1(0)}{b_n} b_n e^{-d_n t} \prod_{j=1, j\neq n}^{n} \frac{b_j}{d_j - d_n} \\ &\quad + \frac{N_1(0)}{b_n} \sum_{i}^{n-1} b_i\, e^{-d_n t} \left(\prod_{j=1, j\neq i}^{n} \frac{b_j}{d_j - d_i} \right) \\ &\quad - \frac{N_1(0)}{b_n} \sum_{i}^{n-1} b_i\, e^{-d_n t} \left(\prod_{j=1, j\neq i}^{n} \frac{b_j}{d_j - d_i} \right) \end{aligned} \tag{A.37}$$

If the first and the fourth term are grouped (they had been highlighted in blue), it follows:

$$= \frac{N_1(0)}{b_n} \sum_{i}^{n-1} b_i \left(e^{-d_i t} - e^{d_n t}\right) \prod_{j=1, j \neq i}^{n} \frac{b_j}{d_j - d_i} + \frac{N_1(0)}{b_n} b_n e^{-d_n t} \prod_{j=1, j \neq n}^{n} \frac{b_j}{d_j - d_n} \tag{A.38}$$

$$+ \frac{N_1(0)}{b_n} \sum_{i}^{n-1} b_i\, e^{-d_n t} \left(\prod_{j=1, j \neq i}^{n} \frac{b_j}{d_j - d_i} \right)$$

The first term is the expression (A.35), then it is only necessary to show that the sum of the second and third term is equal to zero. In other words:

$$= \frac{N_1(0)}{b_n} \left[b_n e^{-d_n t} \prod_{j=1, j \neq n}^{n} \frac{b_j}{d_j - d_n} + \sum_{i}^{n-1} b_i\, e^{-d_n t} \left(\prod_{j=1, j \neq i}^{n} \frac{b_j}{d_j - d_i} \right) \right] = 0$$

It is possible to observe that the following product appears in both terms:

$$b_1 b_2 b_3 \dots b_n$$

Additionally, the exponential is common. Then, the equation is simplified as:

$$= \prod_{j}^{n} b_i\, e^{-d_n t} \left(\prod_{j=1, j \neq n}^{n} \frac{1}{d_j - d_n} + \sum_{i}^{n-1} \prod_{j=1, j \neq i}^{n} \frac{1}{d_j - d_i} \right) = 0 \tag{A.39}$$

The problem finally is reduced to show that:

$$\prod_{j=1, j \neq n}^{n} \frac{1}{d_j - d_n} + \sum_{i}^{n-1} \prod_{j=1, j \neq i}^{n} \frac{1}{d_j - d_i} = 0$$

Which was showed in (1.120). Therefore, the proof is complete.

Appendix B.

Appendix B. 1 Table I. Linear chains for the U-235 case. The “F.P.” is the link for the linear chains of fission products.

0	U-235	U-236	U-237	Np-237	Np-238	Pu-238
1	U-235	U-236	U-237	Np-237	Np-238	Np-239
2	U-235	U-236	U-237	Np-237	Np-238	FP
3	U-235	U-236	U-237	Np-237	Np-236	U-236
4	U-235	U-236	U-237	Np-237	Np-236	Pu-236
5	U-235	U-236	U-237	Np-237	Np-236	Pa-232
6	U-235	U-236	U-237	Np-237	Np-236	Np-237
7	U-235	U-236	U-237	Np-237	FP	
8	U-235	U-236	U-237	U-238	U-239	Np-239
9	U-235	U-236	U-237	U-238	U-239	U-240
10	U-235	U-236	U-237	U-238	U-237	Np-237
11	U-235	U-236	U-237	U-238	U-237	U-238
12	U-235	U-236	U-237	U-238	U-237	FP
13	U-235	U-236	U-237	U-238	FP	
14	U-235	U-236	U-237	FP		
15	U-235	U-236	U-235	U-236	U-237	Np-237
16	U-235	U-236	U-235	U-236	U-237	U-238
17	U-235	U-236	U-235	U-236	U-237	PF
18	U-235	U-236	U-235	U-236	U-235	U-236
19	U-235	U-236	U-235	U-236	U-235	U-234
20	U-235	U-236	U-235	U-236	U-235	FP

Appendix B. 2. Table II. Isotopes and reactions considered for the U-235 and U-238 comparative cases

U-235, α , (n,γ) , $(n,2n)$, f
U-238, α , (n,γ) , $(n,2n)$, f

N	Isotope	Reactions	N	Isotope	Reactions	N	Isotope	Reactions
1	Th-231	β^-	2	Pa-231	α , (n,γ) , f	3	Rn-220	α
4	Ac-227	α, β^-	5	Th-227	α, f	6	Bi-213	α,β^-
7	Pa-232	$CE,\beta^-,(n,\gamma)$	8	Th-232	α ,(n,γ) ,f	9	Po-214	α
10	U-232	α , (n,γ), f	11	Pa-233	β^-, (n,γ)	12	Pu-241	α ,(n,γ) ,$(n,2n)$,f
13	U-236	α ,(n,γ) ,$(n,2n)$, f	14	Th-233	β^-, (n,γ)	15	Tl-210	β^-
16	Th-234	β^-	17	U-237	β^-, (n,γ) , f	18	Bi-211	α,β^-
19	Np-237	α , (n,γ) , $(n,2n)$, f	20	Np-238	β^- ,(n,γ) , f	21	Pb-212	β^-
22	Np-236	EC, β^-, α , (n,γ)	23	U-238	α , (n,γ) , $(n,2n)$, f	24	Pu-242	α ,(n,γ) ,$(n,2n)$,f
25	U-239	β^-, (n,γ)	26	U-234	α , (n,γ) , $(n,2n)$ f	27	Am-240	α,EC
28	Th-230	α , (n,γ)	29	U-233	α , (n,γ) , f	30	Bi-210	α,β^-
31	Th-229	α, f	32	Ra-225	β^-	33	Hg-206	β^-
34	Ra-228	β^-	35	Ac-228	β^-	36	Po-211	α
37	Ra-226	α	38	Rn-222	α	39	Tl-207	β^-
40	Fr-223	$\alpha, \beta^-,$	41	Ra-223	α	42	Bi-209	α
43	At-219	$\alpha, \beta^-,$	44	Th-228	α	45	Pb-207	STABLE
46	Pa-234	β^-,(n,γ)	47	Pu-238	α ,(n,γ) ,$(n,2n)$,f	48	Po-212	α
49	Np-239	β^-,(n,γ)	50	Pu-239	α ,(n,γ) ,$(n,2n)$,f	51	Tl-210	β^-
52	U-240	β^-	53	Pu-236	α ,(n,γ)	54	Cm-243	$\alpha,EC,(n,\gamma)$,f
55	Ac-225	α	56	Po-218	α,β^-	57	Pb-208	STABLE
58	Rn-219	α	59	Bi-215	β^-	60	Cm-244	$\alpha,(n,\gamma), f$
61	Pa-235	β^-, (n,γ)	62	Pu-237	EC, α , (n,γ)	63	Cm-246	α,f
64	Np-240	β^-	65	Ra-226	α	66	Cm-245	$\alpha,(n,\gamma)$,f
67	Pb-214	β^-	68	At-218	α,β^-	69	Pb-208	STABLE
70	Ra-224	α	71	Fr-221	α	72	At-217	α,β^-
73	Rn-219	α	74	Bi-215	β^-	75	Pb-211	β^-
76	Pa-235	β^-, (n,γ)	77	Pu-237	EC, α , (n,γ)	78	Po-216	α
79	Np-240	β^-	80	Ra-226	α	81	Rn-217	α
82	Pb-214	β^-	83	At-218	α ,β^-	84	Am-241	α, (n,γ) ,$(n,2n)$, f
85	Ra-224	α	86	Fr-221	α	87	Pb-210	α,β^-
88	Pa-236	β^-	89	Pu-240	α ,(n,γ) ,$(n,2n)$,f	90	Po-213	α
91	Ra-223	α	92	Bi-214	α,β^-	93	Tl-209	β^-
94	Rn-218	α	95	Po-215	α	96	Am-242	$EC,\beta^-,(n,\gamma)$
97	Bi-212	α,β^-	98	Pb-209	β^-	99	Pu-243	β^-
100	Am-243	$\alpha,(n,2n),f$	101	Cm-242	$\alpha,(n,\gamma)$,f	102	Po-210	α
103	Tl-206	β^-	104	Tl-208	β^-	105	Pb-206	STABLE
106	Tl-205	STABLE	107	Cm-245	$\alpha,(n,\gamma)$,f	108	Pb-208	STABLE
109	Cm-246	α,f						

Appendix B. 3. Table III. List of all the isotopes and their reactions that appear in the network related with Se-87

Se-87, β^-								
N	Isotope	Reactions	N	Isotope	Reactions	N	Isotope	Reactions
1	Br-087	β^-	29	Kr-087	β^-	16	Rb-087	β^-,(n,γ)
3	Sr-087	(n,γ)	31	Sr-088	(n,γ)	18	Sr-089	β^-,(n,γ)
5	Y-089	(n,γ)	33	Y-090	β^-,(n,γ)	20	Zr-090	(n,γ)
7	Zr-091	(n,γ)	35	Zr-092	(n,γ)	22	Zr-093	β^-, (n,γ)
9	Nb-093	(n,γ)	37	Zr-095	β^-, (n,γ)	24	Nb-095	β^-, (n,γ)
11	Mo-095	(n,γ)	39	Mo-096	(n,γ)	26	Mo-097	(n,γ)
13	Mo-098	(n,γ)	41	Mo-099	(n,γ), $(n,2n)$	28	Tc-099	(n,γ), $(n,2n)$
15	Ru-099	STABLE	2	Tc-100	CE,β^-	30	Mo-100	(n,γ)
17	Mo-101	β^-	4	Zr-094	(n,γ)	32	Tc-098	β^-
19	Nb-096	β^-,(n,γ)	6	Nb-097	β^-	34	Zr-096	(n,γ)
21	Zr-097	β^-	8	Y-091	β^-, (n,γ)	36	Y-092	β^-
23	Sr-090	β^-,(n,γ)	10	Sr-091	β^-	38	Rb-088	(n,γ)
25	Ru-100	(n,γ)	12	Ru-101	β^-, (n,γ)	40	Ru-098	STABLE
27	Tc-101	β^-						

Appendix B. 4. Table IV. List of all isotopes and their reaction that appear in the network related with Sb-135

Sb-135, β^-								
N	Isotope	Reactions	N	Isotope	Reactions	N	Isotope	Reactions
1	Te-135	β^-	29	Pr-142	(n,γ)	16	Sm-147	α , (n,γ) , $(n,2n)$
3	I-135	β^-	31	Pr-143	β^-,(n,γ)	18	Sm-148	α , (n,γ)
5	Xe-135	β^-,(n,γ)	33	Nd-143	(n,γ) $(n,2n)$	20	Sm-146	α
7	Cs-135	β^-,(n,γ)	35	Nd-144	α, (n,γ)	22	Pm-148	β^-, (n,γ)
9	Ba-135	(n,γ)	37	Pr-144	β^-	24	Pm-149	β^-, (n,γ)
11	Ba-136	(n,γ)	39	Ce-142	(n,γ)	26	Nd-148	(n,γ)
13	Ba-137	(n,γ)	41	Nd-149	β^-	28	Nd-142	(n,γ)
15	Ba-138	(n,γ)	2	Ce-143	β^-,(n,γ)	30	Pr-145	β^-
17	Ba-139	β^-	4	Ce-144	β^-,(n,γ)	32	Cs-136	β^-, (n,γ)
19	La-139	(n,γ)	6	Ce-145	β^-	34	Cs-137	β^-, (n,γ)
21	La-140	β^-	8	Nd-145	(n,γ),$(n,2n)$	36	Cs-138	β^-
23	Ce-140	(n,γ)	10	Nd-146	(n,γ)	38	Xe-136	(n,γ)
25	Ce-141	β^-,(n,γ)	12	Nd-147	β^-, (n,γ)	40	Xe-137	β^-
27	Pr-141	(n,γ)	14	Pm-147	β^- ,(n,γ)			

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