PROBABILISTIC APPROACH OF SOLVING THE STIFF LINEAR DIFFERENTIAL EQUATIONS IN NUCLEAR TRANSMUTATION ANALYSIS

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by

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TABLE OF CONTENTS

LIST OF TABLES

Page

LIST OF FIGURES

Page

LIST OF SYMBOLS

${}_{0}^{1}n$	Neutron with mass number
α	Alpha particle
β	Beta particle
γ	Gamma particle
λ	Decay constant
b_{ij}	Decay branching ratio from nuclide-j into nuclide-i;
$\mathcal{F}_{ik}$	Production yield from nuclide-k into nuclide-i;
A_{ik}	Production rate of nuclide-i due to the removal of nuclide-k from the system
$\mathbf{A}$	Transmutation matrix
n_{max}	Number of sub-steps
Δt	Sub-steps interval
t_f	Total time step
$\pi_i(j)$	Joint Poisson distribution
γ_i	Normalization constant
δ_{vj}	Kronecker delta
w_i	Weight of nuclide
$\tau_{i\to d}$	Transmutation scale factor

LIST OF ABBREVIATIONS

BDF	Backward Differentiation Formula
CNUCTRAN	C++ Code for Solving Nuclear Transmutations
CRAM	Chebyshev Rational Approximation Method
CRPG	Computational Reactor Physics Group
ENDF	Evaluated Nuclear Data File
IRK	Implicit Runge–Kutta
LIL	list-of-list
LPAM	Laguerre Polynomial Approximation Method
MCNP	Monte Carlo N-particle code
MMPA	Mini–Max Polynomial Approximation Method
ODEs	Ordinary Differential equations
ORNL	Oak Ridge National Laboratory
PL	Poisson Distribution
PPL	Parallel Patterns Library
QRAM	Quadrature-based Rational Approximation Method
SpMM	Sparse Matrix-Matrix Multiplication
TTA	Transmutation Trajectory Analysis

LIST OF APPENDICES

PENDEKATAN KEBARANGKALIAN BAGI MENYELESAIKAN PERSAMAAN PEMBEZAAN LINEAR KAKU DALAM ANALISIS TRANSMUTASI NUKLEAR

ABSTRAK

Transmutasi nuklear ialah satu proses yang menukar satu nuklida kepada yang lain yang menyumbang kepada sisa radioaktif dan aplikasi lain. Baki kepekatan nuklida ini berubah mengikut masa, dan ia merupakan solusi sistem persamaan pembezaan linear yang dikenali sebagai persamaan Bateman. Persamaan ini lebih sesuai dalam menganggar kepekatan akhir nuklida dalam masalah transmutasi yang linear dalam rantai. Bagi bukan linear, persamaan menjadi lebih sukar. Beberapa kaedah penyelesaian Bateman, termasuk Kaedah Pendekatan Rasional Chebyshev (CRAM) dan Analisis Trajektori Transmutasi (TTA), dicadangkan untuk menyelesaikan masalah ini. Satu masalah yang tidak dapat diselesaikan ialah mencari algoritma yang mudah dan boleh dipercayai untuk menyelesaikan persamaan pembezaan kaku yang melibatkan nuklida jangka pendek dan jangka panjang. Penyelidikan ini memperkenalkan pendekatan kebarangkalian untuk menyelesaikan persamaan pembezaan linear kaku dalam teori transmutasi nuklear. Kaedah ini menawarkan alternatif unik kepada pengiraan eksponen matriks tradisional. Ia memberikan kesetiaan pengiraan yang tinggi dan berjaya menangani cabaran untuk menyelesaikan masalah transmutasi kaku tanpa menyelesaikan persamaan Bateman yang berkaitan. Kaedah ini dilaksanakan dalam program C++, untuk Transmutasi Nuklear (CNUCTRAN) dan disahkan terhadap algoritma terkini. Dalam sampel pertama, kaedah probabilistik menunjukkan konsistensi yang tinggi dengan formula Analytical Bateman. Keputusan sampel kedua dan ketiga dibandingkan dengan

CRAM pesanan 48 (CRAM48), menunjukkan perbezaan yang boleh diabaikan dan ketepatan yang luar biasa. Di sini, ralat relatif antara CNUCTRAN dan CRAM kebanyakannya $<10^{-5}$ (0.001%) untuk semua nuklida penting dengan kepekatan pecahan $\geq 10^{-15}$. CRAM berprestasi lebih baik daripada CNUCTRAN dari segi masa pengiraan. Walau bagaimanapun, purata masa CPU CNUCTRAN adalah munasabah praktikal, membenarkan tumpuan pada kesetiaan pengiraan dan bukannya kelajuan. CNUCTRAN telah membuktikan kesediaannya untuk pengiraan terbakar merentas pelbagai analisis transmutasi dimensi. Ia menawarkan struktur input yang mesra pengguna dalam format Extensible Markup Language (XML) dan menyediakan fleksibiliti untuk melaraskan pelbagai parameter simulasi. Selain itu, ia boleh berfungsi sebagai alat rujukan yang boleh dipercayai untuk mengesahkan dan mengesahkan keputusan.

PROBABILISTIC APPROACH OF SOLVING THE STIFF LINEAR DIFFERENTIAL EQUATIONS IN NUCLEAR TRANSMUTATION ANALYSIS

ABSTRACT

Nuclear transmutation is a process that changes one nuclide into another that contributes to radioactive waste and other applications. The remaining concentration of these nuclides evolves with time, as described by a system of linear differential equations known as the Bateman equation. The equation is more applicable in estimating the final concentration of nuclides in transmutation problems that are linear in a chain. For non-linear transmutation problems, the equation becomes more difficult. Several Bateman solvers, including Chebyshev Rational Approximation Method (CRAM) and Transmutation Trajectory Analysis (TTA), are proposed to solve these problems. One unresolved problem is to find a simple, reliable algorithm for solving stiff differential equations involving short-lived and long-lived nuclides. This research introduces a probabilistic approach to solving stiff linear differential equations in nuclear transmutation theory. The method offers a unique alternative to traditional matrix exponential calculations. It provides high computational fidelity and successfully tackles the challenges of solving stiff transmutation problems without solving the associated Bateman equations. The method is implemented in a C++ for Nuclear Transmutation (CNUCTRAN) and verified against Analytical Bateman Formula and CRAM. In the first sample, the probabilistic method showed high consistency with the Analytical Bateman formula. The second and third sample results were compared with CRAM of order 48 (CRAM48), showing negligible differences and remarkable accuracy. Here, the relative errors between CNUCTRAN and CRAM

are mostly < 10^{-5} (0.001%) for all important nuclides with a fractional concentration of ≥ 10^{-15} . CRAM performs better than CNUCTRAN in terms of computation time. However, the average CPU time of CNUCTRAN is reasonably practical, allowing the focus on computational fidelity rather than speed. CNUCTRAN has proven its readiness for burnup calculations across various dimensional transmutation analyses. It offers a user-friendly input structure in Extensible Markup Language (XML) format and provides the flexibility to adjust various simulation parameters. Additionally, it can serve as a reliable reference tool for validating and confirming results.

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CHAPTER 1

INTRODUCTION

1.1 Background of the Research

Nuclear transmutation is a process that involves a change in one nuclide into another over a time step. It can occur due to various transmutation events, such as nuclear decay, fission, fusion, neutron capture, etc. These events can only happen independently at a particular time, meaning that the occurrence of one event has nothing to do with the occurrences of other events. Nuclear transmutation is very significant in many aspects, such as nuclear physics, nuclear engineering, and other disciplines of study (Uzhhorod National University, 2024). It enables researchers to examine the behaviours of nuclear nuclei, offering insights essential for advancing our understanding of fundamental nuclear processes. Scientists can produce novel nuclides with significant applications in industry and medicine by studying these processes (Z. Talip, 2020). For instance, certain transmuted nuclides are used in medical imaging and cancer treatments, providing critical tools for healthcare professionals. In the industrial sector, transmutation can lead to new materials with unique properties that enhance various technologies.

Additionally, nuclear transmutation is pivotal in exploring the feasibility of nuclear energy generation. By transmuting certain nuclides, it is possible to create more efficient fuel cycles, potentially leading to cleaner and more sustainable energy production. This is especially relevant in reducing greenhouse gas emissions and addressing global energy demands. Moreover, transmutation offers promising solutions for nuclear waste disposal. By converting long-lived radioactive nuclides into stable nuclides. This mitigates the environmental impact and enhances the safety

and sustainability of nuclear energy. Therefore, the research and development of methods for effective nuclear transmutation, such as those tackled using the point mode or transport mode, are essential for these advancements, as they provide robust framework for solving nuclear transmutation problems. These codes aim in the practical applications of theoretical concepts, ultimately contributing to progress across various fields.

The nuclear transmutation process can manifest either naturally or artificially. In natural or spontaneous transmutation, nuclides predominantly transform into stable ones through decay or decay chains. A notable example is the transmutation of U-235, illustrated in Fig. 1.1 Conversely, artificial transmutations occur when a nuclide is bombarded by smaller particles in devices such as linear accelerators, cyclotrons, or synchrotrons. This process produces daughter nuclides known as spalls. These newly generated nuclides follow their decay chain, undergoing natural transmutation after spallation. For example, when a U-238 nuclide is bombarded with a neutron beam, it splits to generate a U-239 nuclide. The U-239 then decays via beta decay to form Np-239. This sequence of events illustrates how artificial transmutation can create new elements and isotopes through a combination of induced and natural processes.

$$^{238}_{92}U + ^{1}_{0}n \rightarrow ^{239}_{92}U$$

$$^{239}_{92}U \rightarrow ^{239}_{93}Np + ^{0}_{-1}\beta$$

Moreover, transmutation can range from simple to complex, particularly in burnup analysis within nuclear reactors during fission reactions. These processes are often characterized by their inflexibility, necessitating a numerical approach that takes small steps to yield satisfactory results. In light of these challenges, there is a clear need for innovative strategies, leading to various codes and algorithms to address these

complexities. As such, many of these solutions are mathematically difficult due to the complexity of the problems, including nuclides with very small half-lives, which make the differential equations stiff, which will be further explored in Chapter 2 of this research.

Scientists typically estimate the final concentration of nuclides by solving a set of linear differential equations formulated by (Ernest Rutherford, 1904). These equations determine the final nuclide concentrations. Henry Bateman, was the first to tackle this problem, obtaining a solution using the Laplace transform of the equations (BATEMAN, 1910). However, this analytical solution is limited to radioactive decay in linear chains and often fails when dealing with non-linear chains. In transmutation processes, two or more daughter nuclides can be generated, making the problem complex and non-linear.

This study presents an unconventional approach to tackle stiff transmutation problems, involving short and long-lived nuclides. Rather than directly solving the Bateman equations, the proposed method successfully simulates the transformation of nuclides based on probabilistic theory, making it a self-possessed and valuable feature. Additionally, the new approach does not directly intend to challenge the state-of-the-art algorithm's robustness. By deviating from conventional approaches, we hope to shed new light on this complex field and pave the way for novel advancements in nuclear transmutation research.

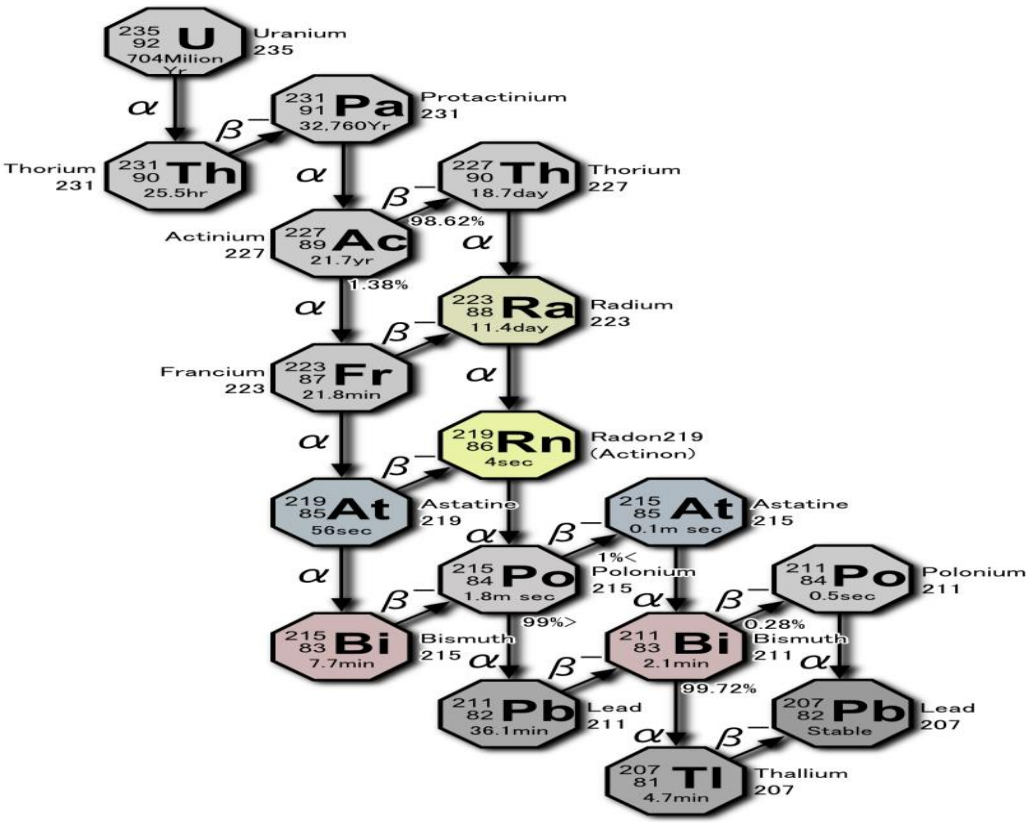

Figure 1.1 An Example of U-235 Transmutation
Source: (Tosaka, 2008)

1.2 Scope and Limitations

This study focuses on developing the probabilistic approach for solving stiff linear differential equations in nuclear transmutation theory. The new algorithm developed simulates the transformation of nuclide concentrations without directly solving the Bateman equation. This method has been implemented in CNUCTRAN, a C++ code for solving nuclear transmutation problems, including sample decay reactions with and without fission from benchmark problems. The nuclide data used in the study are obtained from the Evaluated Nuclear Data File (ENDF) version B-VIII.O (Brown et al., 2018).

Additionally, Python code derived from the OpenMC burnup feature (P. K. Romano et al., 2021) was used compared to some of the methods during the validation. All simulations and calculations in this study were conducted on a Windows Subsystem for HP (2.6 GHz). It is vital to acknowledge that the results of a performance evaluation may differ based on the choices of operating systems, computational hardware, and programming languages utilized throughout the study. However, the key limitations of this study are as follows.

Firstly, the method accurately tracks the transformation of nuclides by considering only decay reactions and fission, which effectively addresses the key processes involved in nuclide transformation. However, it does not account for other reactions, such as fusion, pair production, and electromagnetic interactions. This limitation means that the method may not comprehensively understand the full range of processes occurring in the system being studied. To achieve this limitation, a more complete analysis must incorporate these additional reactions, ensuring that all potential processes are considered. This inclusive approach would enhance understanding of the system's behaviour, leading to more accurate and reliable results.

Secondly, the method assumes constant reaction rates, which limits its applicability to non-linear problems where reaction rates vary with time. In most reactor cores, the thermal power generated is considered constant over a specific time interval. However, solving the equation with variable flux introduces significant complexity. Burnup codes address this by identifying an average flux that effectively represents the entire interval, a process known as flux normalization. This simplification helps manage the complexity but may not accurately capture the dynamic behaviour of systems with time-dependent reaction rates, thus limiting the precision of the analysis in such scenarios.

Lastly, when addressing complex transmutation problems, the method's efficiency is generally practical for achieving its objectives. However, the limitations of current computational technology, such as processing power, memory, and software capabilities, often become significant obstacles to reducing computational time. This implies that while the method is theoretically effective, the available computational resources usually hinder its practical implementation. Consequently, delays or inefficiencies in the computational process can occur, impacting the method's overall effectiveness in real-world applications. These technological constraints highlight the need for continued advancements in computational technology to fully realize the method's potential and improve its practical applicability in handling complex transmutation problems more efficiently.

1.3 Problem Statement

The change in nuclide concentration over time during transmutation is described by a set of first-order linear differential equations known as the Bateman equations. One major issue in solving the Bateman equation is finding a straightforward, trustworthy approach to solving transmutation problem containing over 1500 short- and long-lived nuclides, a problem that remains unsolved (Cai et al., 2019; Li & Cai, 2018). The existing methods for solving these equations are complicated, computationally demanding, and require rational approximation functions (Cai et al., 2019; Li & Cai, 2018). Therefore, an effective strategy for handling these equations is needed. The involvement of short-lived and long-lived nuclides makes the Bateman equations stiff. Interestingly, the concentration of short-lived nuclides varies rapidly with time, whereas long-lived nuclides exhibit more modest variations in concentration over time. The numerical approach of solving stiff

Bateman equations requires a reduced temporal sub-step size for accurate results. However, the variation of long-lived nuclides within the small sub-step is very subtle, leading to a loss of accuracy. The Bateman equation also gained its notoriety due to its stiffness, where achieving high-accuracy nuclide concentrations remains a challenge today. Recent works have proposed numerous complex algorithms that can only solve stiff Bateman equations with concentration errors of >0.001% for most nuclide species (Bakin et al., 2018; Cai et al., 2019; Josey et al., 2017; Lahaye et al., 2017; Li & Cai, 2018; Tavakkoli et al., 2021; Xia et al., 2021).

Another challenge in solving the Bateman equation is reducing CPU run time for transmutation problems. Previous research has demonstrated the efficiency and speed of various methods, such as the Chebyshev Rational Approximation Method (CRAM) and Transmutation Trajectory Analysis (TTA), in handling complex transmutation problems. However, TTA is too slow for routine calculations, with impractical computational times even when run on different CPUs (A. E. Isotalo & Aarnio, 2011). Despite these limitations, techniques derived from mathematical approaches to solving the Bateman equations offer potential solutions for improving efficiency. One promising direction is the development of new algorithms and computational techniques that can handle the stiffness of the Bateman equation more effectively, thereby reducing run times. For instance, adaptive step-size methods and parallel computing approaches could significantly enhance computational speed and accuracy.

Hence, a novel method is needed to archieve the limitations of conventional approaches by eliminating the reliance on rational approximation functions. The new method will employ advanced mathematical techniques, such as Sparse Matrix-Matrix Multiplication (SpMM) and exponentiation by squaring, to speed the computation

process and accurately estimate nuclide concentrations. This proposed method must undergo validation against CRAM48 solver and Analytical Bateman formula to ensure reliability and effectiveness. Doing so promises significant advantages in implementation, computational efficiency, and adaptability to stiff problems, addressing the current challenges in solving the Bateman equation.

1.4 Objectives

This research proposes a probabilistic method for solving stiff linear differential equations in nuclear transmutation analysis. Two objectives that contribute to the achievement of the main objectives are listed as follows:

1. Determine if the probabilistic approach achieves sufficient accuracy to ensure that all nuclide concentration errors for stiff Bateman equations remain below the target threshold of 0.001%.
2. Using the rapid sparse matrix multiplication (SpMM) technique, the proposed method reduces computing time while maintaining accuracy with a relative error consistently below 1%, as verified against standard benchmarks

1.5 Thesis Outline

This thesis comprises five chapters: introduction, theory and literature review, methodology, results and discussions, and conclusion. **Chapter 1** provides the background of the research, the problem statement, the research objectives, and the thesis outline. **Chapter 2** discusses the theory relevant to this research: the Bateman equation and the probability distribution. This chapter also summarizes the previous study that addressed the same problem: proposing a new method to solve the stiff linear

differential equation occurring in nuclear transmutation. **Chapter 3** presents the probabilistic approach based on probability theory. The probabilistic method is improved by incorporating sparse matrix multiplication (SpMM). **Chapter 4** highlights sample problem results using the proposed method and validates them using a benchmark problem to establish their reliability and efficiency in determining the final nuclide concentrations. Finally, **chapter 5** summarizes the findings and suggests a perspective to advance the research field.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter illustrates the theory behind this research, transitioning from the foundational principles to pioneering concepts. The Bateman equation is initially derived based on linear decay reaction, followed by its non-linear transformation, and further developed into nuclear transmutation analysis. The nuclear transmutation calculation can be addressed using linear decay chain, matrix exponential method and Ordinary Differential equations (ODEs), which are explained in detail from its fundamental concepts to its application in nuclear transmutation. Lastly, a review of the statistical theory of nuclear transmutation proposed by a recent study is outlined in the last section of this chapter.

2.2 Bateman Equation

A set of ordinary differential equations governs the remaining concentration of nuclides upon transmutation and is called the Bateman equation. These nuclides vary in terms of their chemical and physical properties. Predicting the time evaluation of nuclide concentrations is vital in nuclear applications. However, the solution of the Bateman equation is quite complicated, especially for solving complex nuclear transmutation problems, such as the burnup process in nuclear reactor analysis. The burnup process is coupled together with both neutronics and thermal-hydraulic techniques. The burnup process in nuclear reactor analysis refers to the gradual depletion of nuclear fuel and the buildup of fission products and transmutation elements within a nuclear reactor. Burnup is an important factor in understanding how

fuel is utilized during reactor operation and the evolution of the reactor core over time. It is a measure of how much energy is extracted from the nuclear fuel as a function of time, typically expressed in gigawatt-days per metric ton of uranium (GWd/MTU) or as a percentage of the initial fuel's fissionable material that has undergone fission.The neutron transport problem is often handled by the Monte Carlo N-particle code (MCNP) or MCNPX and its implementation is restricted to calculations involving neutrons, photon, and electrons (Pelowitz et al., 2008). The Oak Ridge National Laboratory (ORNL) established the code ORIGEN (Croff, 1980) which solves the Bateman equation by updating the nuclides inventory by power series approximation and secular equilibrium approximation. The key to the burnup code is to solve the Bateman equations. Currently, there are three primary techniques for resolving Burnup equations: Linear chain, matrix, and Runge-kutta or linear multi-step methods.

The linear chain method (BATEMAN, 1910) offers a direct approach to evaluating final nuclide concentrations in decay chains. With minimal approximations, this technique explicitly measures the chain and evaluates all significant nuclides within it. In this context, a linear chain refers to a simple decay process where each nuclide has only one predecessor and one successor. These linear chains are governed by the Bateman equations, which are analytically solvable. The overall solution to the burnup equations is derived by superimposing these individual analytical solutions. Transmutation problems are categorized into decay problems and burnup problems (Isotalo, 2013.) based on the presence or absence of neutron flux. In a pure decay system, the linearization of the directed graph representing the burnup matrix is exact. This is because a finite number of linear chains of finite length corresponds precisely to the original burnup equations, adhering to the superposition principle and the absence of closed transformation cycles. This is ensured because a nuclide cannot

undergo a series of energy-emitting decay processes and return to its original state with the same mass. However, closed transformation cycles may exist in a burnup system where neutron-induced nuclear reactions occur. Consequently, an infinite or very large number of linear chains with infinite lengths is required to maintain equivalence between the original burnup equations and the linear chains. Practically, not all these linear chains are significant enough to be considered, as the number of nuclides transferred through the chain decreases rapidly along its length. Therefore, a cutoff check determines where each linear chain effectively ends. Determining linear chain solutions constitutes the bulk of the computational cost. (Huang et al., 2016) Efficiency can be improved by reducing the number and length of calculated linear chains or by developing more efficient analytical formulas to determine nuclide number densities for these chains. Such efficiency improvements would significantly reduce computational effort in applications requiring numerous burnup calculations. Recently, Cetnar introduced a general analytical solution called Transmutation Trajectory Analysis (TTA) (Cetnar, 2006). This method successfully addresses key issues in solving burnup problems using analytical Bateman equations. The TTA method provides explicit analytical expressions for each nuclide in a decay chain. However, when cycles are present in the burnup chain, the TTA method must break down a cycle into a longer chain, adversely impacting its efficiency and accuracy. This is because burnup equations fall within the category of ordinary differential equations.

Another technique is the matrix exponential approach, where the answer to the matrix exponent is crucial. Numerous techniques have been developed and proposed based on various numerical approximations of matrix exponent. Taylor's method with scaling and squaring was implemented in the ORIGEN code (Cai et al., 2019). However, some special care should be taken for the short-lived nuclides in this method.

Other methods include the Chebyshev Rational Approximation Method (CRAM) (Pusa, 2011), the Quadrature-based Rational Approximation Method (QRAM) (She, Wang, et al., 2013), the Laguerre Polynomial Approximation Method (LPAM) (She, Zhu, et al., 2013), the Mini–Max Polynomial Approximation Method (MMPA) (Kawamoto et al., 2015a), the Krylov Subspace Method (Li & Cai, 2018), and Magnus expansion techniques for time-ordered exponentials (S. Blanes a, 2009). The CRAM method has the advantages of fast and excellent solution accuracy, making it appropriate for resolving complex nuclide burnup systems (A. E. Isotalo & Aarnio, 2011). Codes like Serpent (Leppänen, 2011), GNIAC (Ma & Huang, 2018a), and and OpenMC (Leppänen, 2015; P. K., & F. B. Romano, 2023).

The Burnup equations may be solved using Ordinary Differential Equation techniques (ODEs), such as the Backward Differentiation Formula (BDF) approach and the Runge-Kutta method. The ALEPH2 (Stankovskiy & Van Den Eynde, 2012) Monte Carlo burnup algorithm utilizes the fifth-order Implicit Runge–Kutta (IRK) approach in the burnup solver RADAU5. Step size must be changed, and iteration convergence must be accelerated via the IRK approach. Furthermore, the IRK technique is restricted in its use due to its substantial dependence on the size of the burnup matrix in terms of solution speed. Regarding the BDF approach, the LSODE package, whose kernel is the backward differentiation technique, is used by the FISPACT-II code to solve first-order ordinary differential equations (Sublet et al., 2017a). The speed and stability of the numerical solutions are further issues with this approach. The IRK methods offer significant advantages over BDF methods. Being single-step, IRK methods avoid issues with system discontinuities and changes in order or time step. Additionally, they are self-starting and require only the initial conditions as starting values. However, after time discretization, IRK methods result

in larger and more complex sets of linear equations. Numerous tests have demonstrated that the Radau IIA IRK method (with 3 stages and accuracy order 5), as implemented in the RADAU5 solver, is more stable and faster than the BDF method used in the DLSODA solver. These solvers are especially beneficial for time-dependent reactivity or external source factors, as examined in reactor kinetics applications (Fridman, 2019). Semi-analytical techniques, like the Adomian Decomposition Method (ADM) and the Homotopy Perturbation Method (HPM), have been employed to approximate stiff systems using symbolic-numeric formulations (Wazwaz, 2000). Recently, Neural-ODE frameworks have surfaced as AI-driven alternatives for dynamic reactor simulations, presenting possibilities for real-time predictive modelling (Ghosh, 2021).

Another technique for solving the burnup equations is the Monte Carlo method. This method evaluates the number of nuclei in the system and probabilistically decides their creation or removal based on interaction probabilities and nuclear cross-sections. However, applying the Monte Carlo method to full-scale assembly or core calculations remains challenging due to its significant computational demands and inherent stochastic fluctuations, which introduce substantial errors. Consequently, the Monte Carlo method provides limited practical advantage for efficiently resolving large-scale burnup calculations compared to deterministic methods. Additionally, Table 2.1 provides the summaies of existing methods with limitations.

Table 2.1 Comprehensive Summaries of Burnup Solver

Method	**Type**	**Best for**	**Limitations**
Bateman Equations	Analytical	Simple decay chains	Only for simple, linear chains; no feedback or branching
TTA	Numerical	Trajectory-based decay networks	Inaccurate for complex branching; approximation only

Table 2.1 (Continued)

Method	Type	Best for	Limitations
CRAM	Numerical	Matrix exponential in stiff systems	Requires accurate rational coefficients; dense matrix ops
Padé Approximation	Numerical	Matrix exponential in stiff systems	Scaling and squaring needed; less efficient for large sparse matrices
Krylov Subspace Methods	Numerical	Large sparse systems	Memory-intensive; complex implementation
Magnus Expansion	Semi-analytical	Time-ordered matrix exponentials	Slow convergence for stiff systems; time-ordered integration needed
Runge-Kutta (Implicit)	Numerical	General stiff ODE solvers	Requires small step size for stiff systems; can be slow
BDF / CVODE	Numerical	Adaptive stiff system integration	Sensitive to time step and error control settings
Adomian Decomposition	Semi-analytical	Nonlinear series solution	Slow convergence; complex term computation
Homotopy Perturbation	Analytical	Approximate stiff ODEs	Primarily AI research; lacks industry adoption
Neural-ODEs	Machine Learning	AI-assisted time evolution	Requires large training data; interpretability concerns
Monte Carlo + Depletion	Hybrid	Coupled transport-burnup codes	Computationally expensive; stochastic noise

2.3 Equations governing nuclides transmutations

In 1904, Ernest Rutherford demonstrated that the quantity of the initial nuclide and other nuclides generated within the decay chain, comprising n-nuclides, varies according to a sequence of time-dependent differential equations.

$$\frac{dN_1(t)}{dt} = -\lambda_n N_1(t), \tag{2.2}$$

$$\frac{d\,N_i(t)}{dt} = \lambda_{i-1} N_{i-1}(t) - \lambda_i N_i. \tag{2.3}$$

$$\frac{d\,N_n(t)}{dt} = -\lambda N_n(t), \tag{2.4}$$

Where N_i, is the amount of initial nuclide and other nuclides at a given time t, and λ_i, is the decay constant for each of the respective nuclides. Though this sequence of equations is accessible and more applicable to simple linear chains problems, the equations differ from linear chains for complex radioactive decay or transmutation in particle flux, and the solution appears to be more complicated.

However, (BATEMAN, 1910) proposed a series of differential equations using Laplace transformation, which can be extremely useful for complex radioactive decay chains by breaking them down into linear chains. Bateman equation is given as:

$$N_i(t) = c_1 e^{-\lambda_1 t} + \ c_2 e^{-\lambda_2 t} + \cdots + c_n e,^{-\lambda_n t}, \tag{2.5}$$

Where c_i, is the coefficient of the equation and is given as,

$$c_i = \frac{\lambda_1 \lambda_2 \dots \lambda_{n-1} N_1(0)}{\prod_{j=1, i \neq j}^{n} (\lambda_j - \lambda_i)}, \tag{2.6}$$

In general form,

$$N_i(t) = N_1(0) \bullet \prod_{i=1}^{n-1} \lambda_i \bullet \sum_{i=1}^{n} \frac{e^{-\lambda_i t}}{\prod_{j=1, i \neq j}^{n} (\lambda_j - \lambda_i)}. \tag{2.7}$$

Nevertheless, Eq. (6) has its limitations. When two or more decay constants have values close to each other $\lambda_j \approx \lambda_i$, it experiences catastrophic cancellation (Cetnar, 2006). The denominator of the Bateman equation involves subtracting decay constants. Insufficient precision in the computational solution of the equation leads to a substantial loss of precision digits in the difference between λ_j and λ_i. As a result, the computed results deviate significantly from the actual result. Therefore, methods discussed in the literature review section are introduced to generate accurate computational results based on the Bateman equation.

Suppose a system with I number of nuclides undergoing transmutations, changing into one another per a particular transmutation chain. The Bateman equations for a system with i as the index of nuclides of interest and I as the index of other nuclides exclusive of i are as follows:

$$\frac{dy_i}{dt} = \sum_{j=1}^{I} b_{ij} \lambda_{ij} y_j + \sum_{k=1}^{I} \mathcal{F}_{ik} A_{ik} y_k - (\lambda_i + A_i) y_i, \tag{2.8}$$

Where y_i is the atom density of nuclide-i;

λ_j is the radioactive decay constant of nuclide-i causing its removal from the system (the decay constant);

λ_{ij} is the decay constant of nuclide-j causing the production of nuclide-i in the system;

b_{ij} is the decay branching ratio from nuclide-j into nuclide-i;

$\mathcal{F}_{ik}$ is the production yield from nuclide-k into nuclide-i; and,

A_{ik} is the production rate of nuclide-i due to the removal of nuclide-k from the system.

Eq. (2.8) is a system of I linear differential equations dependent on one another. The solution of Eq. (2.8) is the concentration of all I nuclides. The term on the left-hand side of the equation is known as the rate of change of the nuclide-I concentration. The left-hand side of the equation is balanced with the terms on the right-hand side of the equation. Here, each term describes the various transmutations of nuclides that lead to the production and removal of nuclide-I within a system.

In particular, the first term on the right-hand side of the equation includes the natural decay and various reactions that transform nuclide-I into other nuclides, causing its removal from the system. Likewise, the second term of the right-hand side describes the formation of nuclide-I via different transmutation reactions of all nuclides, including their natural decay reaction. Alternatively, if we let $y = (y_1\ y_2\ \cdots\ y_I)^{\mathrm{T}}$, Eq. (2.8) can be expressed in its compact matrix form given by,

$$\frac{d\mathbf{y}}{dt} = \mathbf{A}\mathbf{y}, \tag{2.9}$$

Where **A** is the transmutation matrix such that its elements correspond to all coefficients defined in Eq. (2.8). In general, **A** is a sparse $I \times I$ square matrix such that its elements are zeros. On the other hand, **y** is a column vector corresponding to the concentration of all I nuclides defined in the calculation. Technically, the norm of **A** is in order of ~$\mathbf{10^{21}}$ It contains both off-diagonal elements and diagonal elements. This large magnitude of **A** implies significant numerical difficulties, as stiff system exhibits

varying timescales that make them particularly sensitive to numerical instability caused by short-lived nuclides.

2.4 Stiff linear differential equation in nuclear decay

According to Moler (2018.), an ordinary differential equation problem is considered stiff if it involves short-lived and long-lived nuclides. In such cases, the concentration of long-lived nuclides changes slowly over time, while the concentration of short-lived nuclides changes rapidly. Accordingly, the numerical method must take small steps to obtain efficient and accurate results. In particular, the system of equations (2.9) is identified as a stiff problem due to the significant variance between matrix eigenvalues resulting from the large differences in the removal rates of short-lived and long-lived nuclides. Therefore, solving Eq. (2.9) requires specialized numerical methods capable of handling stiffness to ensure accuracy and reliability. For Example, when evaluating the final concentration of nuclides in transmutation problems from $t = 0\ s$ to $t = t_{f,}$ If one wishes to evaluate the final concentration of nuclides in transmutation problems from $t = 0\ s$ to $t = t_{f,}$ The numerical calculation must carefully select step size. The number of steps, n_{max}, is given by $n_{max} = {}^{t_f}/_{\Delta t}$, where Δt is the step size. Notably, the smaller the value Δt, the larger the number of step n_{max}, thereby enhancing accuracy.

A similar procedure applies to a simple nuclear decay problem. Generally, a faithful simulation of the nuclear decay phenomenon requires iterating over all nuclei at each step n, where $n \in \{0,1,2,\ldots,n_{max}\}$. For instance, suppose a system consists of N_i Nuclei of nuclide-i. To determine the final concentration, we let $n_i^{(n)}$, be the nuclei count of nuclide-i after n steps, then we create and initialize $n_i^{(0)} = N_i$, nuclei of

nuclide-i in the system. However, this process is continued until $n = n_{max}$ step for e.g from $n = 0$ to $n = 1$, the nuclei count of nuclide-i, $n_i^{(1)}$, is obtained by iterating over $n_i^{(0)}$ nuclei of nuclide-i. For each nucleus, we choose an event-j, from a list of possible outcomes the nucleus might experience. This list includes no-reaction, decay, neutron absorption, fission, and fusion. Events that result in the nucleus disappearing from the system are termed removal events.

We index these events using integers j within the range $\{0,1,2,3,..J_i\}$, where $j = 0$, represents a no-removal event, and J_i, denotes the total number of possible removal events for nuclide-i. For Example, $j = 1$ corresponds to radioactive decay, $j = 2$ to neutron absorption, etc. The list must include all probable reactions defined for nuclide-i.

To select an event for nuclide-i, we sample from a joint Poisson distribution, $\pi_i(j; \Lambda_1, .. \Lambda_{j_i}) \equiv \pi_i(j)$, which we will derive in section 2.2. Here, $\pi_i(j)$, represents the discrete probability of each event occurring for the nuclei of nuclide-i during each sub-step. If the selected event is a removal event, the nucleus is removed from the system, and its daughter nuclide-k is created in the simulation. This process continues until no nuclei of nuclide-i remain in the current step. Although the method is quite complicated and computationally expensive, it generates results with random noise, which is undesirable. Nevertheless, understanding this basic algorithm is beneficial for grasping the basics of a faithful simulation of the phenomenon. A more sophisticated simulation closely following the physical process will always yield a reliable solution

2.5 Conventional Methods for Solving Stiff Linear Differential Equations

Several methods have already been proposed to solve Eq. (2.9), three of which are the major ones discussed in the previous literature. Firstly, The Markov chain approach involves solving the burnup system by linearizing depletion chains and applying the Bateman solution (BATEMAN, 1910; Cetnar, 2006; Halász & Szieberth, 2018; Ma & Huang, 2018b). This method, also known as Transmutation Trajectory Analysis (TTA), effectively mitigates the stiffness of equations. However, it requires extensive time to solve problems involving numerous nuclides subjected to intense irradiation or lengthy time steps due to the necessity to track longer burnup chains. The CINDER code (CINDER 1962) employs this approach for solving burnup equations. Secondly, a numerical method for the comprehensive system of ordinary differential equations (ODEs), such as Euler and Runge-Kutta methods (Sublet et al., 2017b), is often unsuitable for resolving complex burnup problems characterized by stiffness. This approach may result in divergence. The third method employs numerical techniques to estimate the exponential of the transition matrix. The solution to Eq. (2.1) can be represented as follows:

$$\mathbf{y} = \exp(\mathbf{A}t)\mathbf{y}_0, \tag{2.10}$$

Where $\mathbf{y}_0$ is a list of initial concentrations of nuclides considered in the calculation. Finally, evaluating the matrix exponential in Eq. (3.1) is difficult. Naively, one may approximate the matrix exponential by using a Taylor series,

$$\exp(\mathbf{A}t) \approx \sum_{k=0}^{\infty} \frac{1}{k!} \mathbf{A}^k t,^k \tag{2.11}$$

However, burnup problems are stiff, such that matrix **A** consists of elements with minimal and significant numeric values. Thus, evaluating Eq. (2.11) for a transmutation matrix is impractical and does not always yield an accurate and reliable result. Several other methods include the Padé approximation of the matrix exponential (Higham, 2009), mini-max polynomial approximation (Kawamoto et al., 2015b), Krylov subspace (Yamamoto et al., 2007), and so on., which unfortunately suffers the identical drawback as the Taylor series approximation.

Pusa (2011) discovered that the eigenvalues of the matrix **A** lie near the negative real axis. This approach enables the approximation of the matrix exponential by employing Chebyshev approximants. Here, the technique is known as the Chebyshev Rational Approximation Method (CRAM), where the matrix exponential approximation is given by,

$$\exp(\mathbf{A}t) = a_o + 2\mathrm{Re}\sum_{i=1}^{k/2} a_i(\mathbf{A}t - \theta_i\mathbf{I})^{-1} \quad , \tag{2.12}$$

Where a_i and θ_i Are the complex numbers published by Pusa (2016) for the approximation order $K \in \{16, 32, 48\}$. Here, the computation of Eq. (2.12) requires solving. $k/2$ linear systems. Due to the sparsity pattern of the burnup matrix, the linear systems can be solved efficiently using sparse Gaussian elimination (Pusa, 2016). To date, CRAM is the most efficient and can handle significant burnup problems. Researchers have verified CRAM by comparing it with different burnup solvers like the mathematically precise Transmutation Trajectory Analysis (TTA) (A. E. Isotalo & Aarnio, 2011; Pusa, 2016) conducted TTA calculations with 250-bit precision. The duration for a single step varied significantly, ranging from one second to ten days, depending on the specific test case; step length demonstrates that TTA is

computationally demanding compared to the CRAM method. However, TTA computational speed is inadequate for routine calculations (A. E. Isotalo & Aarnio, 2011) and remains impractical across different CPUs. It is essential to highlight that these methods originated from theoretical mathematical approaches designed to solve burnup equations.

2.6 Statistical Theory of Nuclear Transmutations

In general, a single nucleus undergoes various transmutation events that can transform it into daughter nuclei, with only one event being eventually selected to occur at a particular time; this implies that the occurrence of one event does not influence the occurrence of others. According to Garcia-Torano et al. (2019), nuclear transmutation events occur randomly. However, a general statistical model that estimates the probabilities of these events from the perspective of a single nucleus has never been discussed. Recently, (Omar & Karim, 2022) derived a general probability distribution that accurately predict the probability of transmutation event. This distribution has successfully aided in predicting which of the listed transmutation events is more likely to occur.

2.7 Poisson Distribution; Probability Distribution

In probability theory and statistics, the Poisson distribution is a discrete probability distribution used to predict the number of events occurring a certain number of times (k) within a given interval. Generally, there are two conditions for the Poisson distribution (P-L): firstly, individual events must happen randomly and independently, meaning that the occurrence of one event has nothing to do with other events, and secondly, the mean λ of these events must be known. In the context of

nuclear transmutation, events occur independently of each other and at a fixed mean rate λ. Therefore, the probabilities of these events can be effectively described using the Poisson distribution by dividing the total time into interval of time Δt. This approach allows for accurate modeling of the occurrence of nuclear transmutation events over time.

$$\Pr(X = l) = \frac{\lambda^l e^{-\lambda}}{l!}, \tag{2.13}$$

Where l is the number of times an event occurs.

$\Pr(X = l)$ is the probability that an event will occur l times,

λ is the average number of times an event occurs.

2.8 Probability of Transmutation Events

Suppose we select a nucleus at a particular time, t. The time taken t is divided into L steps where $L \in \{1, 2, 3, \dots, \infty\}$ of time interval $(t, t + \Delta t)$ and the nucleus of nuclide-k is selected during step l. This nucleus can transform into a daughter nucleus due to the option of transmutation events. These occurrences can be conveniently indexed as $j \in \{1, 2, 3, \dots, J_i\}$. It is convenient to designate neutron absorption for event-1, fission for event-2, radioactive decay for event-3, etc. Here, J_i, Represents the total number of events defined for nuclide-k. To simplify, we designate $j = 0$ to indicate no transmutation occurring within the interval $(t, t + \Delta t)$. In this study $\pi_i(j)$, is the probability of nuclide-i undergoing reaction-j.

For a system of a single nucleus of nuclide-i is given by Eq. (2.13),