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RECYCLING ACTINIDES AND DEPLETED URANIUM AS AN ALTERNATIVE TO ENVIRONMENTAL CHALLENGES AND PUBLIC ACCEPTANCE IN THE NUCLEAR SECTOR

Belo Horizonte
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Recycling Actinides and Depleted Uranium as an Alternative to Environmental Challenges and Public Acceptance in the Nuclear Sector

GUILHERME VALDIR MARCHIORI DA SILVA

Dissertação submetida à Banca Examinadora designada pelo Colegiado do Programa de Pós-Graduação em CIÊNCIAS E TÉCNICAS NUCLEARES, como requisito para obtenção do grau de Mestre em CIÊNCIAS E TÉCNICAS NUCLEARES, área de concentração ENGENHARIA NUCLEAR E DA ENERGIA.

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I dedicate this work to the first person who believed in my master's degree, my dear grandmother Maria Trindade Marchiori.

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"*Progress is the realisation of Utopias.*"
(Oscar Wilde)

RESUMO

A emergência climática global destacou a necessidade de reduzir a dependência de combustíveis fósseis e de transitar para fontes de energia mais limpas. A energia nuclear surge como uma alternativa devido às suas baixas emissões de carbono, embora ainda esteja cercada de preocupações relacionadas à segurança, ao gerenciamento de resíduos e à proliferação nuclear. Esta pesquisa investiga o reprocessamento de combustível nuclear como uma estratégia para aumentar a sustentabilidade e a segurança do ciclo do combustível nuclear. O estudo avalia diferentes métodos de reprocessamento (PUREX, GANEX e UREX+) combinados com materiais de mistura, como urânio empobrecido e tório natural. Os resultados mostram que a quantidade de urânio empobrecido e material reprocessado necessária para fabricar o combustível de cada método está disponível após um ciclo do reator. Esse uso pode eliminar 100% da necessidade de mineração de urânio e proporcionar um destino para parte do combustível gasto. Em contraste, enquanto o tório reduz a demanda por mineração (cerca de 83%), ainda é necessária a sua extração. Além disso, os métodos GANEX e UREX+ apresentam vantagem sobre o PUREX ao evitar a separação de plutônio puro, mitigando, assim, os riscos de proliferação. As análises de radiotoxicidade e decaimento mostram que os combustíveis reprocessados começam com níveis mais altos devido aos actinídeos menores e ao plutônio, mas ao longo de 1000 anos seus valores se aproximam do combustível UOX. Essa estabilidade a longo prazo é relevante para o gerenciamento de resíduos, pois reduz a complexidade e os custos de descarte. Os cálculos das quantidades de materiais de mistura e reprocessados foram realizados por meio de equações específicas. As simulações foram realizadas considerando a inserção do combustível reprocessado de Angra 1 no reator Angra 2, utilizando o código SERPENT 2.2.1. O decaimento e a radiotoxicidade foram analisados após um queima de 33 GWd/t.

Palavras-chave: reprocessamento; urânio empobrecido; actinídeos menores; redução de mineração; não proliferação.

ABSTRACT

The global climate emergency has highlighted the need to reduce dependence on fossil fuels and transition to cleaner energy sources. Nuclear energy is an alternative due to its low carbon emissions, yet it remains surrounded by safety, waste management, and nuclear proliferation concerns. This research investigates nuclear fuel reprocessing as a strategy to enhance the sustainability and security of the nuclear fuel cycle. The study evaluates different reprocessing methods (PUREX, GANEX, and UREX+) combined with spiking materials such as depleted uranium and natural thorium. The results show that the amount of depleted uranium and reprocessed material required to fabricate the fuel for each method is available after a reactor cycle. This use can save 100% the need for uranium mining and give a destination to part of the spent fuel. In contrast, while thorium reduces mining demand (around 83%), it still requires extraction. Additionally, GANEX and UREX+ methods present an advantage over PUREX by avoiding the separation of pure plutonium, thereby mitigating proliferation risks. Radiotoxicity and decay analyses show that reprocessed fuels start with higher levels due to minor actinides and plutonium, but over 1000 years, their values approach those of standard fuels. This long-term stability is relevant for waste management, as it reduces complexity and disposal costs. The calculations of the spiking and reprocessed material quantities used specific equations. The simulations were performed considering the insertion of the reprocessed fuel from Angra 1 into the Angra 2 reactor using the SERPENT 2.2.1 code. Decay and radiotoxicity were analyzed after a burnup of 33 GWd/t.

Keywords: reprocessing; depleted uranium; minor actinides; reducing mining; non-prolifearation.

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LIST OF ABBREVIATIONS AND ACRONYMS

COP	Conference of the Parties
FSAR	Final Safety Analysis Report
GANEX	Group Actinide Extraction
GHG	Greenhouse gases
GWd/MTHM	Gigawatt-days per Metric Ton of Heavy Metal
IAEA	International Atomic Energy Agency
MA	Minor Actinides
MOX	Mixed Oxide Fuel
MTHM	Metric Tons of Heavy Metal
MTU	Metric Tons of Uranium
NPT	Non-Proliferation Treaty
PUREX	Plutonium Uranium Recovery by Extraction
STD	Standard
TBP	Tri-n-butyl phosphate
TRU	Transuranic
TRU(Th)	Transuranic Actinides with Thorium
TRU(DU)	Transuranic Actinides with Depleted Uranium
TRU(U)	Transuranic Actinides with Natural Uranium
TTC	Twice-Through Cycle
UREX+	Uranium Extraction Plus

CONTENTS

1 INTRODUCTION

Since the Industrial Revolution, fossil fuels have played an important role in global economic and social development. Since then, the growing energy demand has increased the use of coal, oil, and natural gas, which, despite enabling technological advances and improving the quality of life, has led to an increase in the emission of greenhouse gases (GHG), especially CO_2 (carbon dioxide), which consequently increases the temperature of the planet due to the greenhouse effect. As a result, the adverse effects have culminated in the current climate crisis, which could worsen if the release of GHG continues to be intense (NASH; NILSSON, 2015).

Some efforts have been made through global agreements over the past decades. The Paris Agreement, for example, was adopted during the 21st Conference of the Parties (COP21) of the United Nations Framework Convention on Climate Change (UNFCCC) in 2015. Although other COP conferences have occurred over the last ten years, it remains the most important global climate treaty, setting targets to limit global warming and promoting coordinated actions among signatory countries. The agreement's main goal is to keep the increase in global average temperature well below 2°C above pre-industrial levels and to pursue efforts to limit it to 1.5°C. The Paris Agreement also acknowledges that developed countries bear greater responsibility for greenhouse gas emissions and should lead mitigation efforts and support developing countries (United Nations Framework Convention on Climate Change (UNFCCC), 2015).

The Paris Agreement recognizes the importance of decarbonizing the global energy system. Although it does not specify which technologies should be invested in or avoided, it indirectly encourages the energy transition in developed and developing countries (United Nations Framework Convention on Climate Change (UNFCCC), 2015). Nuclear energy emerges as a viable alternative in this context because it can generate electricity with low carbon emissions and operate stably and continuously. However, despite its strong performance as an energy source, nuclear energy still faces environmental, political, and social barriers (NASH; NILSSON, 2015).

Due to the atomic bombs at the end of the Second World War and the accidents of Chernobyl and Fukushima, nuclear energy is sometimes remembered as an unsafe energy resource. Also, using nuclear material, waste management, and mining to constitute its fuel hinders public acceptance due to environmental concerns.

To achieve the sustainability goals and be efficient, an energy source must preserve the natural resources and not harm life in all forms. It must also be economical and have good public acceptance. Progress in the use of nuclear energy must seek to minimize GHG emissions, preserve natural resources, and ensure social acceptance through informed consensus, maintaining stable production costs as much as possible (POINSSOT; BOULLIS; BOURG, 2015).

Nuclear energy can have its fuel cycle in two ways. In the once-through cycle, used fuel

is discarded after a single use, which simplifies management but requires isolation for many years due to the presence of plutonium and other actinides. After removal from the reactor, nuclear fuel remains extremely radioactive and emits high amounts of heat, which requires it to be stored in active cooling systems for years. Alternatively, the closed nuclear fuel cycle involves recycling uranium and plutonium from reprocessed spent fuel. It permits using another cycle, providing a way to dispose of spent fuel (NASH; NILSSON, 2015).

Some methods are applied to reprocess spent fuel in the closed nuclear fuel cycle. Due to the amount of fissile material after applying a reprocessing method, a spiking step is necessary. In the context of nuclear fuel cycle, spiking refers to the intentional addition of natural uranium, depleted uranium, or thorium to reprocessed nuclear fuel. This practice adjusts the fissile content of the reprocessed fuel, improves handling characteristics, enhances proliferation resistance, and optimizes the isotopic composition for reactor performance. The spiking with depleted uranium can decrease the need for mining because the tailings in the first cycle are still available and stored, offering a more sustainable future for the nuclear sector. Studies have demonstrated that reprocessed fuels spiked with depleted uranium provide environmental benefits, such as reducing greenhouse gas emissions and minimizing the demand for natural uranium extraction. Moreover, the efficiency of advanced proliferation-resistant recuperation methods can assist in nuclear disarmament policies, and this, along with less radioactive waste, can improve public acceptance of nuclear energy (POINSSOT; BOULLIS; BOURG, 2015). These findings underscore the importance of further exploring reprocessing methods to mitigate the environmental effects of nuclear activities, reinforcing the argument that a closed nuclear fuel cycle is key to enhancing sustainability in the energy industry (ESTANISLAU *et al.*, 2023).

By closing the fuel cycle, nuclear power becomes more efficient and aligned with sustainability and societal goals. These technological advances may help reshape public discourse, improve transparency, and increase public acceptance in the safety and viability of nuclear energy as a part of the future energy transition scenario. The MOX (Mixed Oxide Fuel) cycle allows the partial reuse of plutonium but generates a high amount of high-level radioactive waste (ESTANISLAU *et al.*, 2021). The TRU(DU) (Transuranic Actinides with Depleted Uranium) cycle, which reprocesses actinides together with depleted uranium, eliminates the need for natural uranium and enrichment, consumes part of the depleted uranium inventory, and reduces waste generation, resulting in greater fuel cycle sustainability (ESTANISLAU *et al.*, 2021). Other fuel alternatives that do not separate actinides include TRU(U) (Transuranic Actinides with Natural Uranium) and TRU(Th) (Transuranic Actinides with Thorium); however, they still require the mining of natural uranium or thorium, although to a lesser extent than in the open fuel cycle.

Previous studies have evaluated the neutronic parameters of models using reprocessed fuels, demonstrating the feasibility of transuranic fuels (TRUs) usage. Neutronic evaluations of coprocessing fuels have shown that proliferation-resistant reprocessing methods produce fuels exhibiting intermediate behavior between standard UO_2 and MOX (Mixed Oxide Fuel)

fuels, with acceptable reactivity and safety characteristics (COTA; PEREIRA, 1997). Analysis including thorium in the reprocessing confirmed that TRU(Th) (Transuranic Actinides with Thorium) fuels from alternative reprocessing cycles can maintain good neutronic and environmental performance, reducing plutonium and actinides formation (PEREIRA; LEITE, 1998). Reprocessed fuels containing transuranics exhibited lower total activity and radiotoxicity than conventional MOX cycles, particularly when thorium was included. The presence of thorium reduced the long-lived actinides, such as plutonium, americium, and curium, thereby decreasing the long-term storage of spent fuel (PEREIRA; LEITE; FARIA, 2000). The inclusion of thorium in particular has attracted attention because of its favorable neutronic properties and ability to reduce the decay activity.

1.1 Goals

This work aims to assess how reprocessing nuclear fuel can environmentally and politically impact the nuclear fuel cycle, contributing to a more sustainable energy source and the public acceptance of nuclear energy. To achieve this aim, the specific objectives include:

- Evaluate the impact of fuel reprocessing on uranium and thorium mining savings through the use of depleted uranium;
- Examine how the use of proliferation-resistant methods, that recycle actinides, and the use of depleted uranium contribute to the international agenda, particularly in light of the objectives of the Nuclear Non-Proliferation Treaty;
- Assess the composition, specific activity, decay heat, and radiotoxicity of spent recycled fuel in a closed fuel cycle using depleted uranium, when processed by PUREX or proliferation-resistant reprocessing methods.

To achieve the goals of this work, an analysis of the entire reprocessed fuel cycle, from fuel fabrication to final disposal, was carried out. First, the reprocessed fuel's composition was determined using the SERPENT Monte Carlo code. The goal was to identify a composition of fissile material, spiked with thorium or depleted uranium, that reproduces the k value of Angra 2's fresh fuel under similar conditions (standard value). Simulations were performed iteratively until a composition capable of achieving the k standard value was found. Once the appropriate isotopic composition was obtained, analytical equations were applied to calculate the quantities of fissile material, thorium, and depleted uranium needed to fabricate reprocessed fuel assemblies. These steps were performed for three different reprocessing methods: PUREX, GANEX, and UREX+, directly comparing a conventional method (PUREX) and two proliferation-resistant alternatives (GANEX and UREX+). The fuel considered for reprocessing was assumed to have the same composition as the burned fuel from Angra 1. Decay and

radiotoxicity analyses were conducted using SERPENT for the three reprocessing schemes. The study covered key parameters such as the multiplication factor at the beginning of life (BOL) and end of life (EOL), and isotopic inventories over a 1000-year cooling period after a 33GWd/MTHM burnup. The specific activity, decay heat, and ingestion and inhalation radiotoxicity were assessed to provide a view of the waste management implications of using reprocessed fuel.

1.2 Contents

The remainder of this work is organized as follows:

- **Chapter 2** provides a theoretical analysis of the sustainability and economics of the nuclear fuel cycle. It conceptualizes the reprocessing, presents the methods PUREX, GANEX, and UREEX+, and discusses the impact of reprocessing on mining, non-proliferation, waste management, and the economy;
- **Chapter 3** is the methodology employed in this work. It presents how the impacts will be measured using a reprocessed fuel based on the spent fuel of Angra 1 inserted in the Angra 2 model reactor;
- **Chapter 4** introduces the results and interprets how they respond to the goals of this work;
- **Chapter 5** concludes the dissertation.

At the end, the references used to sustain this work have also been added and the papers published as part of this research have been attached: a presentation at the 7th SENCIR and the paper in the journal Progress in Nuclear Energy (PNE).

2 REPROCESSING OF THE NUCLEAR SPENT FUEL

The closed nuclear fuel cycle involves recovering and reusing materials present in spent fuel through reprocessing. Unlike the open cycle (or once-through cycle), in which the spent fuel is discarded after a single use, the closed cycle allows these materials to be reused in the form of new fuels, such as MOX produced by the PUREX (Plutonium Uranium Recovery by Extraction) method. The most widespread form of this cycle is the so-called TTC (Twice-Through Cycle), in which the irradiated fuel is reprocessed only once, and the resulting material is used in thermal reactors. Although the new fuels are not reprocessed again in the TTC, they can be stored for future use in more advanced reactors, such as fast neutron reactors (FNRs). Within the closed cycle, advanced processes can aid in the recycling of minor actinides (americium, curium, and neptunium) as the GANEX (Group Actinide Extraction) and UREX+ (Uranium Extraction Plus) methods, offering a safer alternative for the nuclear sector compared to single recycling for MOX production. Methods like GANEX and UREX+ do not separate plutonium, ensuring that the spent fuel cannot be used for military purposes and can also help reduce the volume of radioactive waste (POINSSOT; BOULLIS; BOURG, 2015).

Comparisons between reprocessed fuel as MOX, TRU(U), TRU(DU), and TRU(Th) cycles show that it is possible to optimize the use of natural resources and reduce waste volumes. Recent studies indicate that closed cycles, in addition to reducing the demand for natural uranium, also minimize emissions associated with the nuclear fuel cycle. Compared to the traditional cycle, these alternatives prove to be promising in terms of sustainability (ESTANISLAU *et al.*, 2023). The flowchart in Figure 2.1 can represent the desired reprocessing strategy using depleted uranium as spike material.

Military purposes drove the development of the PUREX reprocessing technology, providing a strategy to separate plutonium, which is then used to fabricate new fuel (POINSSOT; BOULLIS; BOURG, 2015). Recent developments have expanded reprocessing strategies, including processes like GANEX and UREX+. GANEX, for example, addresses the coextraction of actinides, allowing for efficient recycling of plutonium and minor actinides while reducing proliferation risks (POINSSOT; BOULLIS; BOURG, 2015). On the other hand, UREX+ employs multiple extraction steps to achieve selective separation of uranium and technetium, followed by plutonium and minor actinides, ultimately producing separated materials suitable for reuse in different reactor types (ARUQUIPA *et al.*, 2017).

The following subsections of this chapter will present the PUREX and the proliferation resistant methods (GANEX and UREX+). It will show the options for the thorium in the nuclear fuel cycle, and it will be followed by an analysis of how reprocessing can impact mining, nuclear non-proliferation, and waste management. Finally, a discussion on the economics of reprocessing will be presented.

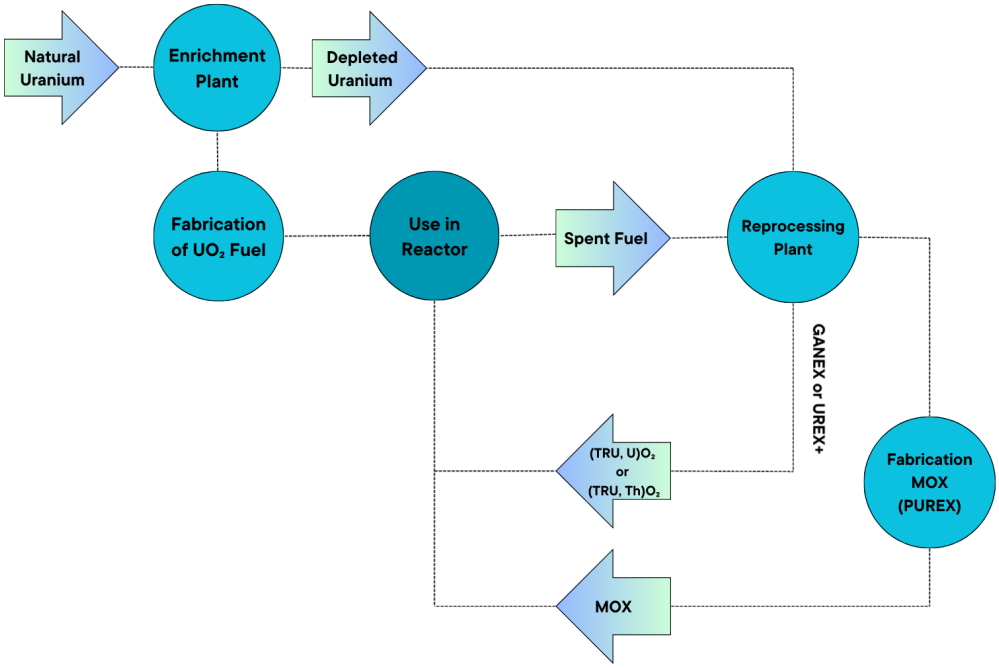

Figure 2.1: Alternatives for reprocessing (From Author, 2025).

2.1 PUREX

The PUREX (Plutonium Uranium Recovery by Extraction) process is a reprocessing technique designed to recover valuable fissile materials from spent nuclear fuel. Initially, it was developed for military purposes due to the separation of plutonium; nowadays, it supports the sustainability of the nuclear fuel cycle. This method employs Tri-n-butyl phosphate (TBP) as a selective solvent to separate uranium and plutonium from fission products and minor actinides (POINSSOT; BOULLIS; BOURG, 2015).

In the initial phase, the spent fuel is chopped and dissolved in concentrated nitric acid, resulting in a solution where uranium and plutonium become chemically available for extraction. During the liquid-liquid extraction stage, TBP preferentially binds to uranium and plutonium ions, transferring them into an organic phase and effectively isolating them from other radioactive byproducts (SHARRAD; WHITTAKER, 2015).

A subsequent separation step is then carried out to isolate plutonium from uranium, using additional solvents and chemical adjustments to achieve high purity levels. Finally, in the stripping stage, the extracted materials undergo purification to remove residual radioactive contaminants (SHARRAD; WHITTAKER, 2015).

The PUREX process enhances the recycling potential of spent nuclear fuel. It also helps reduce the environmental and safety concerns associated with long-term nuclear waste storage, making it a key technology for sustainable nuclear energy management. On the other hand, the plutonium separation process can influence the diversion of material for weapons construction,

increasing stigma with nuclear energy.

2.2 Proliferation-resistant recuperation methods

Some alternatives to advanced reprocessing methods have been studied and designed as proliferation-resistant recuperation methods. Their goal is to recycle uranium, plutonium, and the minor actinides without separation, which does not allow the materials to be used for the fabrication of nuclear weapons. In this work, the processes GANEX and UREX+ will be considered as these methods.

GANEX (Group Actinide Extraction) process is an advanced nuclear fuel reprocessing method designed to enhance the separation and recycling of actinides and other valuable materials from spent nuclear fuel. It is a more sustainable option as a reprocessing method than PUREX. Generally, it involves a first solvent extraction cycle, which removes the amount of uranium, and the second cycle recovers the transuranic elements (CARROTT *et al.*, 2014). The first step aims to partition actinides as a group, minimizing long-lived radioactive waste (TAYLOR *et al.*, 2016). The first solvent extraction stage involves removing uranium using N-dialkylamide extractants. DEHiBA (N, N-di-(ethyl-2-hexyl)isobutyramide) is usually selected as this extractant due to the extraction of uranium without the addition of any reducing or complexing agents for plutonium (MODOLO; GEIST; MIGUIRDITCHIAN, 2015). The second stage consists of separating TRU (Neptunium, Plutonium, Americium, and Curium) through an organic solvent containing a combination of complexing ligands (TAYLOR *et al.*, 2016). For example, a mixture of a malonamide (DMDOHEMA) and an alkylphosphoric acid (HDEHP) can extract almost all actinides from concentrated nitric acid solution (MODOLO; GEIST; MIGUIRDITCHIAN, 2015). The solvents used in GANEX are designed for selectivity and efficiency. They often include complexing agents that bind specifically to actinides, distinguishing them from other elements (TAYLOR *et al.*, 2016).

The UREX+ (Uranium Extraction Plus) process involves modular solvent extraction and separation steps designed to recover valuable materials and reduce waste from spent nuclear fuel. It was designed to separate the components of spent nuclear fuel without isolating plutonium in its pure form, thereby increasing proliferation resistance and enabling the recycling of actinides to reduce long-lived radioactive waste (REGALBUTO, 2011).

According to Regalbuto (2011), the process involves some solvent extraction steps after the dissolution of spent nuclear fuel. The UREX+3a is used as an example, considering the following steps:

- UREX module: Uranium (U) and Technetium (Tc) are extracted using tributyl phosphate (TBP) dissolved in an organic solvent like n-dodecane;
- CCD-PEG or FPEX module: Cesium (Cs) and Strontium (Sr) are separated using special-

ized solvents. CCD-PEG uses chlorinated cobalt dicarbollide and polyethylene glycol, while FPEX uses calixarene and crown ethers. FPEX is more usual in UREX+3a;

- NPEX module: Neptunium (Np) and Plutonium (Pu) are co-extracted with some remaining Uranium. The co-extracted product is stripped to yield a mixed uranium-plutonium-neptunium product;
- TRUEX module: Transuranic elements (americium, curium) and lanthanides are extracted using a mixture of CMPO (octyl(phenyl)-N, N-diisobutylcarbamoylmethyl-phosphine oxide) and TBP in n-dodecane. Fission products, such as zirconium and noble metals, are left in the raffinate;
- TALSPEAK module: Americium and curium (actinides) are separated from lanthanides using HDEHP (bis(2-ethylhexyl)phosphoric acid) and a buffered aqueous phase containing DTPA (diethylenetriaminepentaacetic acid).

After these steps, all recovered products are processed into appropriate forms for reuse or disposal. Uranium and actinides can be recycled into new nuclear fuel, while separated waste is conditioned to meet the specifications for long-term storage (REGALBUTO, 2011).

Both GANEX and UREX+ give options for a sustainable nuclear fuel cycle. The resistance to proliferation advantage can also reinforce the commitment to society to be safe and peaceful. These arguments can improve the societal acceptance of nuclear energy.

However, the co-extraction processes of plutonium can generate products such as Pu+Np, which are attractive for military purposes. Furthermore, as done in GANEX, the lanthanide mixture only makes plutonium separation more difficult but not impossible (BATHKE *et al.*, 2012). Therefore, it is important to highlight that advanced methods only provide an additional barrier to prevent nuclear products from being diverted for weapons proliferation, given that PUREX separates plutonium and was initially designed for military purposes. Adopting proliferation-resistant technologies does not completely prevent proliferation, so implementing advanced methods must be accompanied by reprocessing safeguards and agreements that oversee and regulate their use.

2.3 The options for the thorium in the nuclear fuel cycle

Thorium (Th-232) is a fertile material that, after neutron capture, transforms into U-233, a fissile isotope capable of sustaining chain reactions in thermal reactors. Thorium is about three to four times more abundant in the Earth's crust than uranium, is distributed in minerals such as monazite and titanite, and exhibits favorable chemical and thermodynamic properties (EKBERG, 2015). The thorium fuel cycle has been studied since the early decades of nuclear energy. Still, it has gained renewed interest in recent years due to its potential to

reduce plutonium production and long-lived waste, in addition to supposedly offering greater resistance to nuclear proliferation.

Thorium fuel can be used in thermal and fast reactors when spiked with enriched uranium. In thermal systems, thorium is combined with U-235 to initiate the reaction, and the subsequent conversions of Th-232 into U-233 sustain fission. Reprocessing the spent fuel would also be possible through processes such as THOREX, analogous to the PUREX method used for uranium. However, the possible reprocessing of thorium presents technological challenges due to the low solubility of ThO_2 and the presence of U-232, a strong gamma emitter that requires shielding and automation during handling (EKBERG, 2015).

Beyond using thorium as fresh fuel, combining thorium with reprocessed uranium fuels is possible, forming the TRU–Th cycle. Incorporating thorium into matrices containing recycled actinides reduces plutonium and improves long-term behavior (PEREIRA; LEITE, 1998; PEREIRA; LEITE; FARIA, 2000). The presence of thorium in reprocessing cycles decreases radiotoxicity and the inventory of minor actinides, contributing to a smaller environmental and radiological impact of the final waste. Therefore, the TRU-Th cycle represents a hybrid alternative between the closed uranium cycle and the pure thorium cycle, combining thorium sustainability with the recycling of transuranic materials from conventional reactors. In comparison, the reprocessing cycle that uses depleted uranium (TRU–DU) eliminates the need for natural uranium and enrichment, thus reducing mining demands and consuming part of the existing depleted uranium stored. This cycle generates a smaller volume of high-level waste and exhibits proliferation resistance compared to MOX fuel, making it a sustainable transitional option toward closed fuel cycles. While TRU–DU represents an efficient solution for recycling and waste reduction in the short and medium term, TRU–Th stands out for its potential to reduce plutonium and actinides in the waste within a fully closed nuclear fuel cycle (ESTANISLAU *et al.*, 2021). Furthermore, the thorium cycle is more proliferation-resistant because it produces less plutonium and fewer minor actinides. Still, the U-233 present in the cycle is a material that can be used in nuclear weapons.

On the other hand, technological and economic limitations are obstacles to using thorium. Nevertheless, advances in concepts such as Molten Salt Reactors (MSR) and policies for nuclear waste reutilization have increased interest in the thorium cycle, both as a standalone system and combined with reprocessed fuels (EKBERG, 2015).

2.4 Reprocessing as alternative for mining in the nuclear sector

Mining is of great economic importance to Brazil because it generates employment and income. However, mineral extraction has caused problems such as soil and water contamination by heavy metals, ecosystem degradation, deforestation, and risks to the safety of nearby populations. Initially, mining activities began without environmental concerns, but today, more

legislation is in place to mitigate the adverse effects of this practice (BARRETO, 2001).

Mining has negative impacts on the environment and human communities. It affects the soil due to deforestation, removal of fertile soil, and destruction of vital ecological functions, such as microclimate regulation, in addition to the irreversible changes to landscapes. Besides the high water consumption, mining operations can contaminate the water with heavy metals, devastate aquatic ecosystems, disrupt water supplies, and harm the air. It can also harm communities in protected lands, and the large amount of waste in dams can cause accidents (MILANEZ, 2017).

Uranium mining can affect both the environment and human health. In some cases, intensive extraction and inadequate waste management have contaminated surrounding mining areas, with groundwater and soil showing uranium concentrations above the World Health Organization's 30 µg/L guideline. People living close to these sites may have higher levels of uranium in their bodies and may face increased health risks (MA *et al.*, 2020). Within the nuclear fuel cycle, uranium mining is estimated to be responsible for about 25% of the GHG emissions in the French nuclear fuel cycle, as shown in Figure 2.2, a pattern that may also be observed in the fuel cycles of other regions (POINSSOT; BOULLIS; BOURG, 2015). Although nuclear energy harms the environment far less than fossil fuels, as shown in Figure 2.3, reducing these impacts is important to avoid the climate crisis.

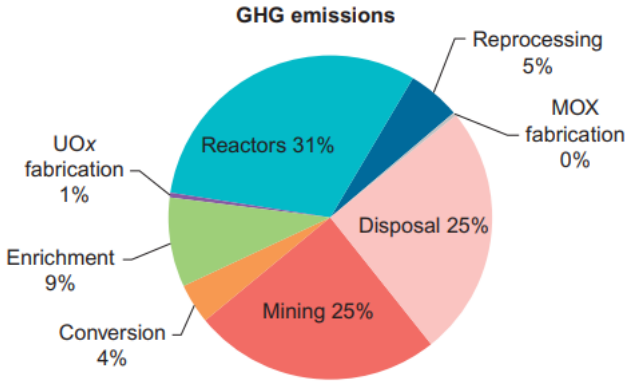

Figure 2.2: Estimates of the GHG emissions distribution over the different steps of the French nuclear fuel cycle (POINSSOT; BOULLIS; BOURG, 2015).

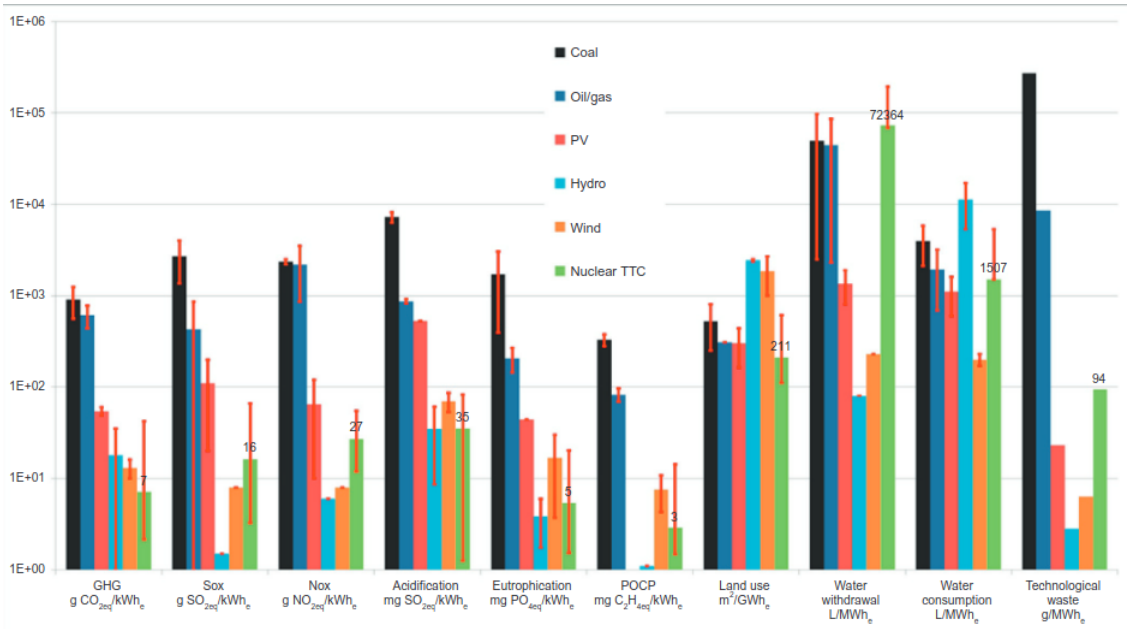

Figure 2.3: Comparison of the environmental footprint indicators for the different forms of energy (POINSSOT; BOULLIS; BOURG, 2015).

According to the graph in Figure 2.4, mining in the nuclear fuel cycle can emit carbon dioxide, sulfur oxides (SOX), and nitrogen oxides (NOX), contributing to greenhouse gases, acid rain formation, worsening respiratory diseases, etc. Furthermore, mining is the main contributor to water pollution during the nuclear fuel cycle and to land use (POINSSOT; BOULLIS; BOURG, 2015). Nuclear energy is seen as a clean energy source during operation, but mining still causes negative impacts on the environment.

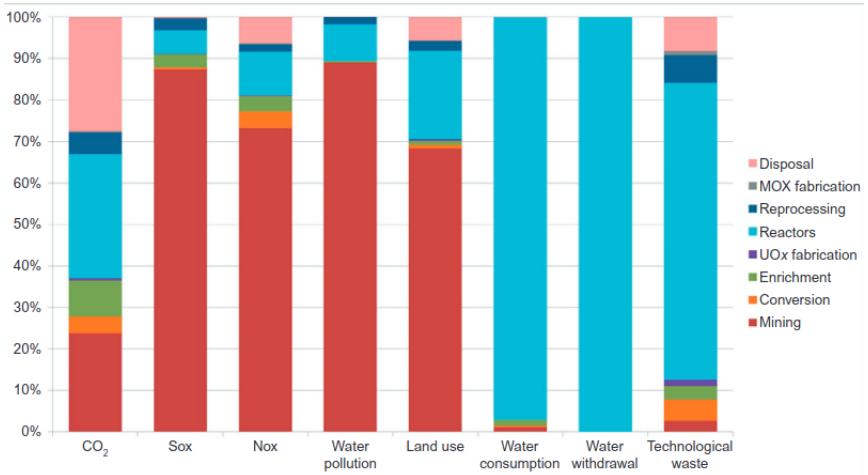

Figure 2.4: Contribution of the different fuel cycle steps on the main environmental footprint indicators (POINSSOT; BOULLIS; BOURG, 2015).

Reprocessing spent nuclear fuel offers a solution by reducing the demand for mining. During the burnup of a cycle in a nuclear reactor, not all the fissile material is burned. Then

it is possible to utilize the amount of fissile material and plutonium produced in a new cycle if the correct reprocessing operation is applied. After applying a reprocessing method such as PUREX, GANEX, UREX+, or others, diluting this solution with less fissile material is necessary. This spiking step can be made with natural uranium, natural thorium (Th-232), or depleted uranium. Even if less than in the first cycle, using natural uranium or thorium would require an additional mining step. However, spiking the depleted uranium (called tailings, which were unused at the beginning of the cycle) with the reprocessed material to fabricate a recycled fuel is an effective approach that can reduce the need for mining.

This approach can conserve resources and avoid the problems related to mining. By decreasing the need for mining, almost 25% of the GHG emissions in the nuclear fuel cycle can be avoided. This decrease in emissions can contribute to the sustainable goals of nuclear energy, making it a good alternative for the energy transition. Using this reprocessed material, it is also possible to reduce the need for enrichment and conversion, which are other steps that harm the environmental footprint.

Mining impacts the environment, and it is an important step in the nuclear fuel cycle due to the need for uranium to fabricate the fuel. It is necessary to think about new strategies to minimize this necessity and make nuclear energy more compliant with the Paris Agreement, achieving a decrease in GHG emissions. Therefore, reprocessing spent fuel, combined with the proper dilution in materials such as depleted uranium, represents an efficient solution to conserve natural resources and reduce the need for mining. This practice makes better use of the residual fissile material. Also, it contributes to reducing greenhouse gas emissions, minimizing the environmental impact of the nuclear fuel cycle, and promoting a more sustainable energy transition.

2.5 Reprocessing as alternative for nuclear weapons proliferation

In 1945, the United States of America attacked Japan with two nuclear bombs. The first one, in Hiroshima, was called “Little Boy”, a bomb with highly enriched uranium. The second, used in Nagasaki, was called “Fat Man”, a plutonium implosion-type weapon. The world witnessed the horror of nuclear weapons and the danger. Nuclear attacks can cause long-term health challenges, including increased rates of cancer and other illnesses such as heart disease and cataracts. Mental health can also be prejudiced, because witnessing an atomic bomb explosion can cause depression and a diminished sense of life expectancy. Economic disparities also emerged, prejudicing the local trade and politics. These impacts extended to future generations, as it happened in Hiroshima and Nagasaki (EMICO, 2015).

Since the attack, the world has worried about the proliferation of nuclear weapons, considering the connection between nuclear energy and the popular vision. Thus, the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) was drafted to prevent the spread of nuclear

weapons. Signed in 1968, it has three main objectives: non-proliferation, disarmament, and the peaceful use of nuclear energy. Its signatories are divided into nuclear-weapon states (China, the United States, France, the United Kingdom, and Russia) and non-nuclear-weapon states. The first group commits to reducing its arsenals and not transferring nuclear weapons to other countries. In contrast, the second group agrees not to develop such weapons and, in return, receives support for the peaceful use of nuclear energy (United Nations, 1968). The NPT has been criticized for its discriminatory nature since its inception, as it recognized only five states as nuclear powers and the permanent UN Security Council members. Additionally, the provisions related to disarmament were seen as vague and difficult to implement, raising concerns about the treaty's effectiveness (SILVA; CULPI, 2017).

In addition to this and many other treaties, the International Atomic Energy Agency (IAEA) has a set of technical measures to verify whether States comply with their legal commitments not to divert nuclear materials or technologies for military purposes. To this end, the IAEA conducts on-site inspections, monitoring through cameras and seals, nuclear material accounting, verifying information provided by States, and environmental sampling. States accept these measures by signing safeguards agreements with the IAEA, which may be comprehensive or item-specific (IAEA, 2022). With these tools, the IAEA provides credible assurances to the international community regarding the peaceful use of nuclear energy.

In the context of nuclear fuel reprocessing facilities, safeguards are technically more complex due to the large volume of material and the risk of diversion of plutonium and other sensitive elements. To address this, the IAEA adopts the "safeguards by design" approach, which recommends considering safeguard requirements from the early stages of facility design to minimize costs, avoid rework, and efficiently integrate verification systems (IAEA, 2019).

Using reprocessing methods such as GANEX and UREX+, it is possible to avoid plutonium separation and thus ensure that fissionable materials are not used to manufacture weapons. They are better alternatives than PUREX in this situation since PUREX separates plutonium that can be diverted to weapons manufacturing and therefore requires greater inspection by the IAEA.

Considering the objectives of non-proliferation and the peaceful use of nuclear energy of NTP, with reprocessing, especially using GANEX and UREX+, the nuclear states could guarantee the use of plutonium only in the reprocessed fuel, not diverting it to military purposes, and the nuclear-weapon states can reinforce their commitment to NPT of not using nuclear energy to fabricate weapons. Therefore, using these methods instead of PUREX could guarantee more credibility with the IAEA, since the plutonium is not being separated.

Furthermore, the objective of disarmament specified in the treaty can also be achieved using depleted uranium to dilute the warheads already manufactured. It happened during the program "Megatons to Megawatts", which was a cooperative initiative between the United States and Russia that converted Russian nuclear warheads into fuel for civilian nuclear power plants. Over the 20 years of the program, more than 20,000 warheads were dismantled, and

500 tons of highly enriched uranium were diluted and used in U.S. nuclear reactors, generating about 10% of the country's electricity. The program was beneficial, as the U.S. gained access to reliable and low-cost fuel, while Russia received financial resources and reentered the international nuclear market (YULISH, 2002). Thus, such initiatives using the already depleted uranium from the first cycle and the enriched uranium from the warheads could support the disarmament of nuclear states and help ensure fuel supply for power plants while reducing steps such as natural uranium mining, conversion, and enrichment.

Therefore, reprocessing nuclear spent fuel offers a solution to non-proliferation commitments. To reinforce the commitment to using nuclear energy solely for peaceful purposes, advanced methods such as GANEX and UREX+ can be adopted, as they make it impossible to isolate pure plutonium by processing it alongside other radioactive materials. Not separating plutonium from the minor actinides (MA) is a proliferation-resistant strategy as it makes the material significantly more challenging to handle and process for weapon creation. Moreover, diluting the existing warheads in depleted uranium to constitute new fuel for reactors is a good strategy to align with the disarmament objectives.

2.6 Reprocessing as alternative for waste management

Radioactive waste management is an important step in the nuclear fuel cycle. Although the amount of waste generated by nuclear power plants is small compared to other types of energy sources, their high level of radioactivity requires complex and long-term solutions. The waste is classified into three categories: low, intermediate, and high level, according to their degree of radioactivity and decay time. Low-level waste (LLW) accounts for most of the generated volume and includes protective clothing, tools, and lightly contaminated filters. This waste is generally treated and disposed of in surface or near-surface facilities, through compaction, incineration, and packaging in secure containers. Intermediate-level waste (ILW), such as reactor parts or used resins, requires shielding but does not generate significant heat. High-level waste (HLW) mainly results from spent fuel and, although it represents less than 1% of the total volume, it contains about 95% of the radioactivity generated by a plant (WORLD NUCLEAR ASSOCIATION, 2022).

The most commonly adopted options for high-level waste involve temporary storage in cooling pools, followed by transfers to dry casks. However, the safest and most definitive solution is deep geological disposal, which isolates the waste in stable rock formations. Countries such as Finland, Sweden, and France are working to develop the implementation of these repositories, with the Onkalo project in Finland being the most advanced, with operations expected to begin in 2026 (WORLD NUCLEAR ASSOCIATION, 2022).

Reprocessing is key in improving nuclear waste management by reducing the volume of high-level radioactive waste that requires long-term storage. By separating usable materi-

als from spent fuel, reprocessing decreases the amount of waste that must be isolated in deep geological repositories, thereby reducing the physical space and the complexity of storage infrastructure. This, in turn, can help ease public concerns and shift the perception of nuclear energy as a cleaner and more manageable power source. In addition, reprocessing enables the recovery of valuable fissile materials such as uranium and plutonium, which can be recycled into new fuel. It extends the utility of natural uranium resources and reduces the environmental footprint of nuclear energy production (POINSSOT; BOULLIS; BOURG, 2015).

2.7 Economics in reprocessing

The economics of nuclear fuel reprocessing represent a complex balance between technological feasibility, resource optimization, and long-term waste management costs. Reprocessing involves high capital investment, operational expenses, and stringent safety and safeguards requirements. Figure 2.5 shows the cost distribution in each step of the nuclear fuel cycle. Most of the costs (61%) are concentrated in the initial investment in infrastructure and construction of plants. Other significant portions include operations (21%) and the supply of uranium ore (7.2%). Costs related to enrichment (5.22%), fuel fabrication (1.98%), conversion (0.54%), final repository (0.14%), and recycling (2.88%) are relatively small within the total (POINSSOT; BOULLIS; BOURG, 2015).

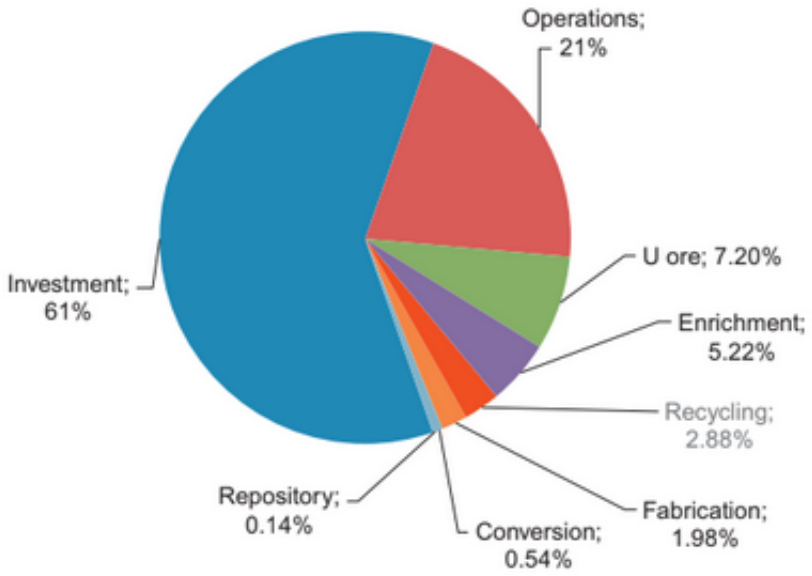

Figure 2.5: Relative share of the various cost components in the final price of electricity in the French system (POINSSOT; BOULLIS; BOURG, 2015).

The reprocessing, although adding an extra cost to the cycle, is low, predictable, and stable in the long term. Costs can also be reduced as applications occur, since new studies may

drive technological innovation in reprocessing by creating more alternative and economically viable methods (POINSSOT; BOULLIS; BOURG, 2015).

An official report commissioned by the French Prime Minister in 2000 (the CDP Report) concluded that the adoption of reprocessing increased the average cost of electricity generation by approximately 5.5%, when compared to a once-through fuel cycle. This increase is primarily driven by the back-end of the fuel cycle, whose total costs rise by up to 85% relative to direct disposal. Despite attempts to economically justify reprocessing, such estimates were criticized for lacking technical and operational realism (SCHNEIDER; MARIGNAC, 2008).

The United States data show that using a closed fuel cycle with MOX increases the total cost by around 4 times. Even with the assumption of free disposal of reprocessed waste, the closed fuel cycle remains economically unfavorable, as the additional reprocessing costs represent only a small fraction of the total cost of nuclear electricity generation, however, under more favorable estimates for reprocessing, which suggest cost parity between open and closed cycles when more optimistic assumptions are adopted, such as high natural uranium prices, lower reprocessing costs, and credits for material reuse. According to Moniz (2003), however, these estimates have significant uncertainty due to the scarcity of reliable public data, the lack of practical implementation of final disposal for reprocessed waste, and the sensitivity of costs to factors such as scale, productivity, and rates of return.

The data for GANEX and UREX+, which are advanced reprocessing methods, are even more uncertain. Both are still in development and demonstration stages; no commercial plants use this fuel. It limits the availability of real data about the costs on a large scale.

Despite this, reprocessing has indirect benefits that justify its adoption from a strategic and long-term perspective. By allowing the reduction of the volume and toxicity of radioactive waste, reprocessing reduces the complexity and cost of definitive geological storage. Furthermore, reducing the need for new mining and enrichment stages contributes to the sector's sustainability. Moreover, adding environmental benefits and using proliferation-resistant reprocessing methods improves societal acceptance, thus overcoming social and political barriers, which is important at this energy transition moment. Therefore, reprocessing should be understood not as an isolated cost but as a central element of a circular nuclear economy, with positive impacts on energy security, waste management, and preserving natural resources.

Although sustainable practices often face resistance due to their higher initial costs, especially in sectors such as energy, they need to be understood as strategic long-term investments. The economic impacts of climate inaction can cost more than mitigating carbon emissions. Furthermore, transitioning to more sustainable practices can create green jobs, reduce public health expenses due to decreased air and water pollution, and lower dependence on finite and imported resources (STERN,).

2.8 Final remarks

This chapter has presented the concept of reprocessing and its importance for a sustainable nuclear fuel cycle. It has presented the method PUREX and the proliferation-resistant methods GANEX and UREX+. It showed the advantages and challenges of the thorium fuel cycle, particularly in reprocessing and fuel fabrication. It discussed how reprocessing can help nuclear energy with sustainability and societal goals, through reducing mining, applying proliferation-resistant methods, and enhancing waste management, provided that proliferation-resistant methods spiked with depleted uranium stored at the beginning of the cycle are employed. Finally, it presented a discussion about the economy in the nuclear sector, bringing some results that were no longer profitable at first but were important in the long term to avoid spending on the consequences of climate change.

The following chapter presents the methodology adopted for the development of this study. As an example, the use of spent fuel from the Angra 1 reactor for the fabrication of reprocessed fuel and its use in the Angra 2 reactor is examined, providing insight into the effects of reprocessing on the nuclear fuel cycle. The results obtained aim to support the assessment of the social and environmental implications of nuclear fuel reprocessing.

3 METHODOLOGY

The nuclear fuel cycle can be conducted using reprocessing to achieve societal and environmental goals. To achieve the objectives, the spent fuel must be reprocessed using a proliferation-resistant recuperation method spiked with the depleted uranium stored in the first mining step, and the composition of the new fuel must sustain the reaction in the reactor, maintaining the k value as the fresh fuel.

Initially, it is necessary to define the composition of the reprocessed fuel according to the selected method (PUREX, GANEX, or UREX) and the spiking material (depleted uranium or natural thorium). To this end, heuristic target values of fissile material were established based on the composition of the spent fuel from Angra 1, the recovery factors of each method, and the fissile fraction of each spiking material. Using the processed composition generated by the Renu software, the resulting fuel was modeled as being loaded into the Angra 2 reactor, replacing the fuel rods with 3.2% enrichment. After a burnup of 33 GWd/t, if the k is consistent with that in the standard cycle of Angra 2, this is the fissile material adopted for the method. The required quantities of spiking material and reprocessed material were then calculated using appropriate equations. Also, the decay and radiotoxicity over a period of 1000 years for each fuel type were analyzed after the same burnup conditions, through simulations with the SERPENT 2.2.1 code, a Monte Carlo-based tool for burnup calculations and reactor physics (LEPPÄNEN *et al.*, 2015). All simulations considered the Angra 2 reactor operating with three typical enrichment levels (1.9%, 2.5%, and 3.2%), and the nuclear data employed were taken from the ENDF/B-VII library (IAEA, 2011).

3.1 Reactor and fuel design basis

To design the reprocessed fuel to be simulated, a reference simulation with the reactor core in its standard configuration is necessary to establish the benchmark k-eff value. This reference case represents the operational baseline against which all subsequent analyses are compared. The methodology for tuning the fissile material percentage for each candidate can be verified in Figure 3.1

The reprocessing technique used a spent fuel matrix from the Angra I PWR reactor, with 3.1% enrichment, burned at 33 GWd/MTHM (Eletrobras Termonuclear S.A., 2013). Considering that this study focuses on the Brazilian nuclear fuel cycle, the reactor's extended operational history can justify the choice of irradiated fuel from Angra 1 as a candidate for potential reprocessing. Angra 1 began operation in 1985, and most of its fuel elements have already been stored for decades. This extended decay period results in lower residual heat and reduced radi-

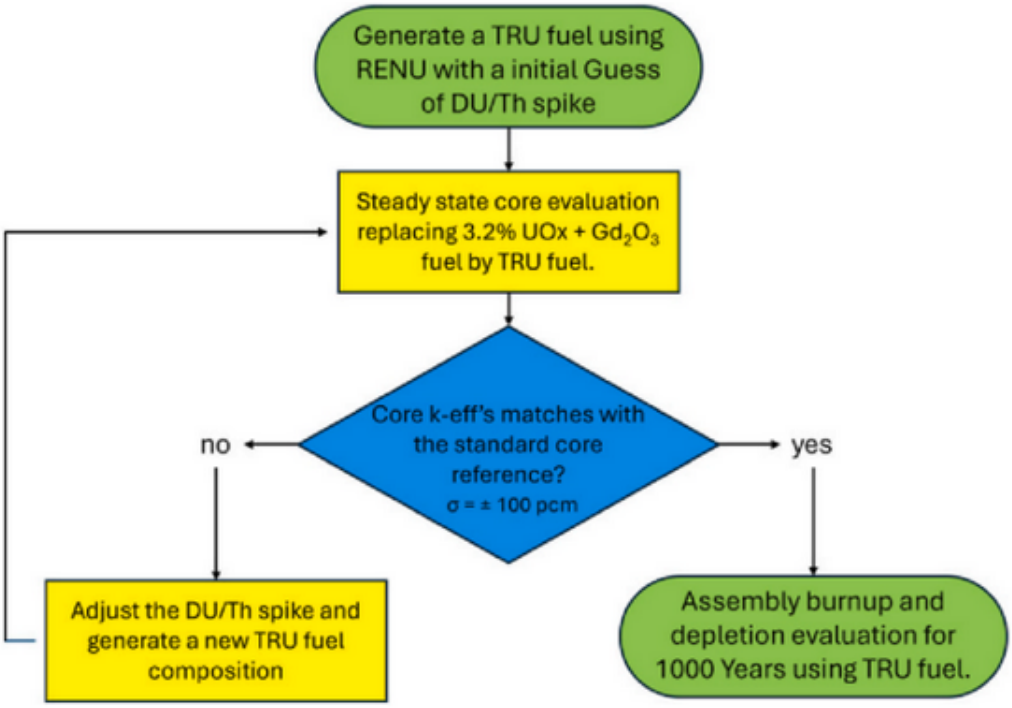

Figure 3.1: Flowchart for methodology of fuel design (From Author, 2025).

ation intensity, conditions that facilitate handling, transport, and eventual chemical processing compared to the more recent fuel from Angra 2, which started operating in 2001. Currently, part of Angra 1's irradiated fuel is also stored at the Complementary Dry Storage Unit (UAS), where it is kept in stainless-steel canisters placed inside concrete-and-steel modules, ensuring shielding and enabling the continued operation of the plants until a final repository is built. Using older stored fuel also represents the management of a long-term environmental liability, so its reprocessing would enable the recovery of nuclear materials with energetic value and contribute to responsible nuclear waste management practices. Table 3.1 shows the matrix of the Angra 1's spent fuel.

Table 3.1: Angra 1 spent fuel isotopic weight fractions.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
H-1	3.27875E-08	Li-6	1.15993E-10	Be-9	9.42560E-11
C-12	1.65701E-11	Co-59	5.74480E-19	Ni-58	2.05669E-16
Cu-63	1.81890E-15	Zn-64	2.54990E-11	Ga-69	1.41750E-11
Ge-74	4.14735E-07	As-75	1.26786E-07	Se-80	3.51525E-05
Br-79	1.35321E-05	Kr-84	2.27470E-04	Rb-85	2.13581E-04
Sr-88	5.40080E-04	Y-89	2.87885E-04	Zr-90	2.21665E-03
Nb-93	1.32849E-05	Mo-92	2.06314E-03	Tc-99	4.86330E-04
Ru-96	1.47125E-03	Rh-103	2.74125E-04	Pd-102	8.39790E-04
Ag-107	5.12990E-05	Cd-106	7.02405E-05	In-113	1.49963E-06
Sn-112	5.77490E-05	Sb-121	1.81202E-05	Te-120	3.03795E-04
I-127	1.51296E-04	Xe-123	3.28305E-03	Cs-133	1.75290E-03
Ba-130	9.23210E-04	La-138	7.66690E-04	Ce-136	1.62239E-03
Pr-141	6.90365E-04	Nd-143	2.40800E-03	Pm-147	7.18530E-05
Sm-150	4.82890E-04	Eu-153	1.02684E-04	Gd-152	5.55345E-05
Tb-159	1.75139E-06	Dy-156	8.68385E-07	Ho-165	9.39120E-08
Er-162	3.70660E-08	Tm-168	3.89580E-11	Yb-170	7.90340E-12
Am-241	1.46903E-06	Am-242	2.70349E-09	Am-243	1.10179E-04
Cm-242	4.57432E-07	Cm-244	5.24399E-07	Cm-245	1.82477E-08
He-4	2.12949E-08	Np-237	4.65668E-04	Np-238	1.38133E-08
Np-239	8.72168E-07	Pu-238	1.81120E-04	Pu-239	4.74279E-03
Pu-240	1.62137E-03	Pu-241	1.52450E-03	Pu-242	5.76141E-04
Th-230	6.03591E-11	U-233	3.66548E-11	U-234	1.51276E-04
U-235	7.87301E-03	U-236	4.02457E-03	U-237	1.03817E-07
U-238	9.57071E-01				

Source: Cota e Pereira (1997).

For the reference simulation, the reactor used is Angra 2. Angra 2 is a commercial nuclear reactor in Angra dos Reis, RJ. It is a Pressurized Water Reactor (PWR), has been in service since 2001, and has a nominal electrical power of about 1,350 MW. According to the FSAR (Eletrobras Termonuclear S.A., 2013), in the first cycle, three typical enrichment levels are employed (1.9%, 2.5%, and 3.2%) in Angra 2 to optimize fuel burnup and power distribution. The reactor has been extensively studied at DEN UFMG (Reis *et al.* (2015), Chaves *et al.* (2022), Silva *et al.* (2018)), including studies involving the application of reprocessed fuel, as reported by Monteiro *et al.* (2015), and Castro (2020).

The core parameters of Angra 2 can be seen in Table 3.2. Angra 2 uses uranium dioxide (UO_2) fuel pellets in cylindrical rods, grouped into rectangular fuel assemblies. The arrangement follows a 16x16 configuration, where each assembly contains 236 fuel rods, guide tubes, and positions for burnable poison rods.

Figure 3.2 presents the XY cross-section of a model representing the standard (STD) core with its different fuel elements. The fuel enriched to 1.9% is shown in red, the fuel enriched to 2.5% in yellow, and the fuel enriched to 3.2% in blue. The small zoomed-in view represents

the rods in the inner part of the assembly with 3.2% fuel enriched, where the light-blue points indicate the guide tubes and the green points represent the burnable poison rods.

Table 3.2: Angra 2 active part model according to FSAR.

Parameter	**Value**
Assembly Matrix	16x16
Number of fuel pins	236
Fuel pin pitch	1.43 cm
Cladding material	Zircaloy-4
Cladding temperature	618 K
Cladding outer radius	0.5385 cm
Fuel gap material	He
Fuel gap outer radius	0.4659 cm
Fuel pellet	UOX+Gd/TRU(DU) or TRU(Th)
Fuel enrichment (UOx)	1.9%, 2.5%, 3.2%
Fuel temperature	873 K
Fuel pellet outer radius	0.4583 cm
Number of tube guides	20
Assembly height	391.6 cm
Assembly pitch	22.88 cm
Assembly types	6
Number of Assemblies	193
Number of UOX+Gd pins per assembly	0/8/12
Gadolinium concentration	7%*
Top/bottom reflector material	Water
Top/bottom reflectors height	30 cm
Coolant/moderator material	Water
Coolant temperature	583 K
Coolant pressure	15.8 MPa

* Gadolinium concentration before pin homogenization.

Source: Eletrobras Termonuclear (Eletrobras Termonuclear S.A., 2013).

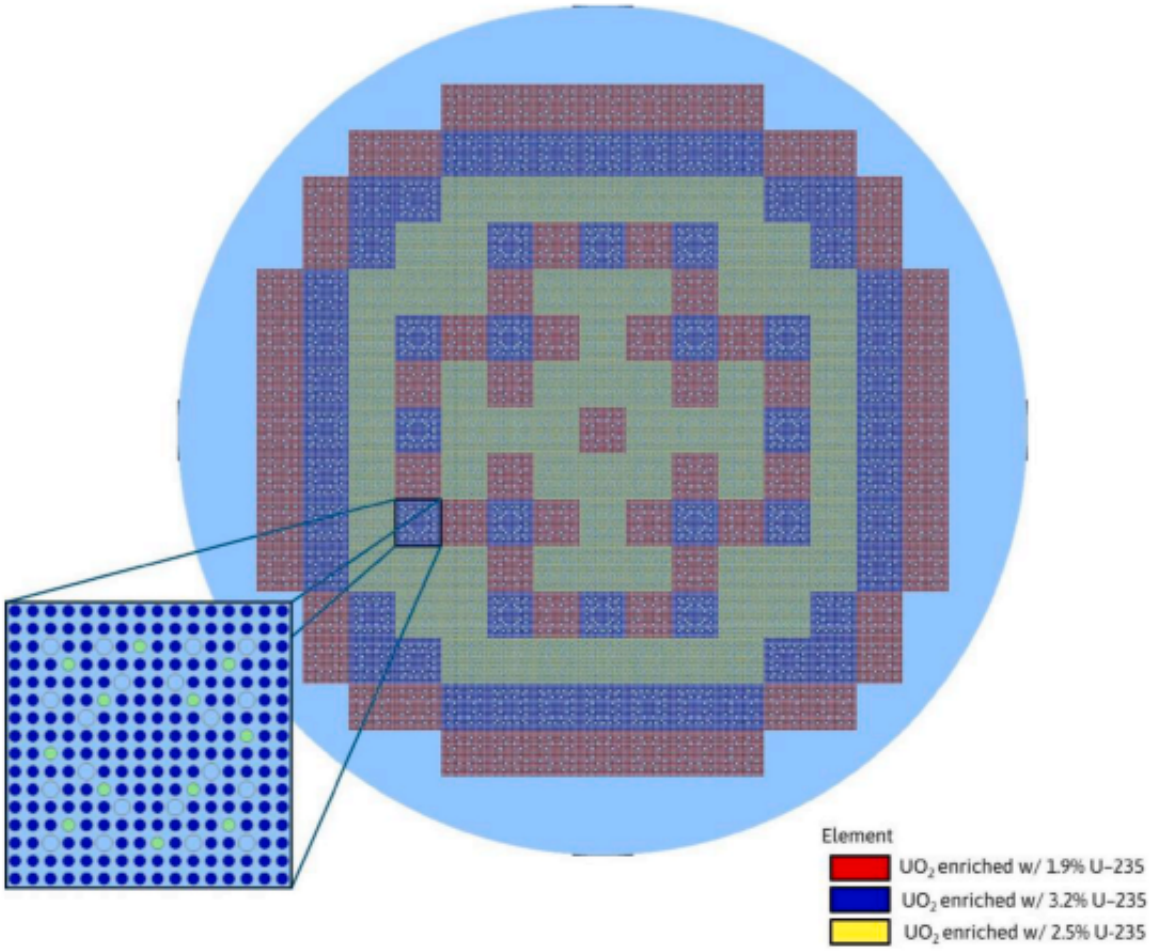

Figure 3.2: XY cross section view from Angra 2 like core (From Author, 2025).

As stated in the FSAR by Eletrobras Termonuclear Eletrobras Termonuclear S.A. (2013), the Angra 2 core consists of six different fuel assembly types, each characterized by specific enrichment levels and varying numbers of burnable poison rods (gadolinium). The simulation was conducted only for the active core region, assuming fully withdrawn control rods. The model was developed using the SERPENT code under working temperature (WT) conditions.

Some aspects of the core design were modified from the FSAR specifications for this analysis, including top and bottom moderator reflectors. The configuration of the gadolinium rods was also modified. Initially, the active region of each gadolinium rod was segmented into three parts: the top and bottom sections contained UO_2 with the same enrichment as the surrounding fuel assemblies. In contrast, the central section consisted of a mixture of natural UO_2 and Gd_2O_3. For the model, all gadolinium rods were homogenized, as illustrated in Figure 3.3, while preserving the original enrichment levels of their respective assemblies.

In the next step, a heuristic approach was adopted in the MATLAB code from Aruquipa *et al.* (2017), Renu, where different quantities of fissile material were introduced into the model until the resulting k-eff matched, as closely as possible, the benchmark value obtained from the standard configuration. The Renu code provides the fuel composition after the user specifies

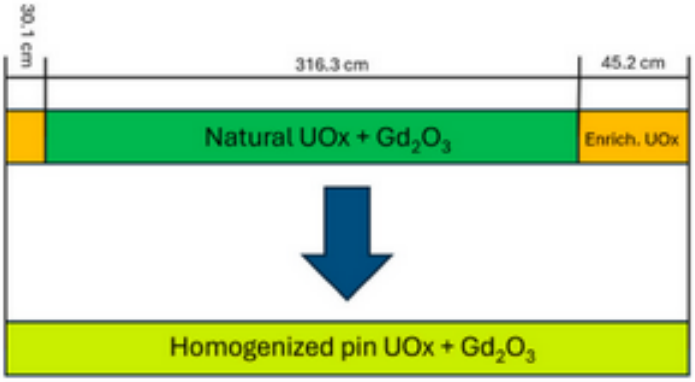

Figure 3.3: Gadolinium rods homogenization (From Author, 2025).

the desired amount of fissile material, the recovery factor of uranium and transuranics, and the fissile content of the material to be used in the spiking. This iterative procedure was applied systematically to the three reprocessing strategies considered (PUREX, GANEX and UREX+). In addition, for each reprocessing route, the simulations were repeated using the two alternative spiking materials under study, natural thorium and depleted uranium.

The recovery factors for the three methods (GANEX, PUREX and UREX+) are listed in Table 3.3.

Table 3.3: Recovery factors for different reprocessing methods.

Element	**PUREX**	**GANEX**	**UREX+**
U	0.9990	0.9350	0.9995
Np	0.9500	0.9980	0.7100
Pu	0.9980	0.9960	0.9950
Am	-	0.9980	0.9800
Cm	-	0.9980	0.7900

Source: Aruquipa *et al.* (2017).

The Renu code calculates the composition of reprocessed fuel based on the burned fuel's composition (available in Table 3.1), the recovery factor of elements for each reprocessing method (available in Table 3.3), and the percentage of fissile material in depleted uranium (0.2%) or thorium, considering the Th-232 (0.0%), depending on the required spike. The interface of the Renu software is shown in Figure 3.4.

Using the exemplified reactor and the compositions given by the Renu code, the adjustment process was repeated iteratively. As presented in Figure 3.1, the fissile material fraction at each step until a satisfactory agreement with the reference k-eff was achieved. For better convergence, the steady-state simulation on SERPENT was carried out using 50000 neutrons during 1000 cycles, skipping the first 400. The boundary conditions are set as "black", neutrons can escape the model.

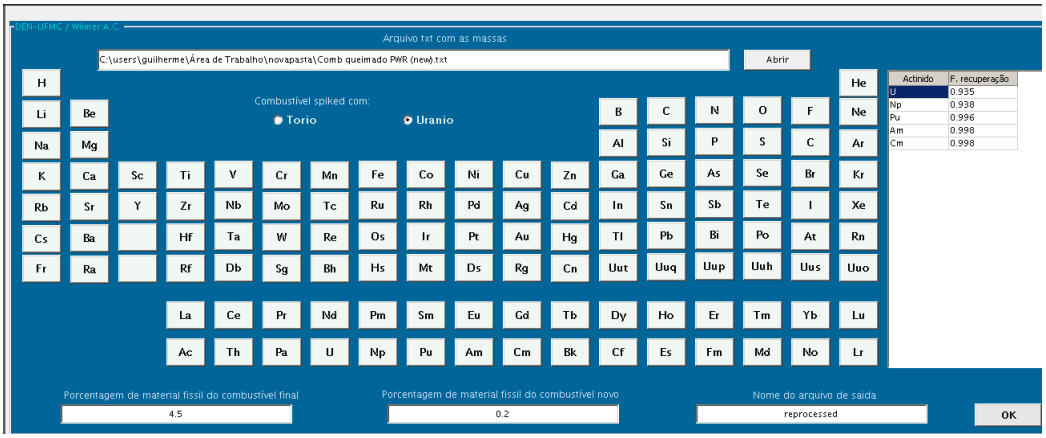

Figure 3.4: Interface of the Software Renu developed by Aruquipa *et al.* (2017).

The reactor's core is refueled every 12 months, replacing approximately one-third of the assemblies. The refueling scheme using fresh enriched uranium fuel follows a logic in which new assemblies (with higher U-235 content) are placed in regions of higher power demand. In contrast, the more burned assemblies are moved to peripheral areas, ensuring a uniform power distribution and extending the cycle. Reprocessed fuel may also be inserted into fresh assemblies, provided that its fissile content maintains the same criticality standard of the reactor as in a cycle with fresh fuel.

Due to the enrichment and number of rods, the 3.2% U-235 enrichment assemblies with 12 Gd rods were chosen for fuel replacement analysis. This assembly has the highest enrichment and number of Gd rods permitted on FSAR. This choice is justified because reprocessed fuel tends to exhibit an intrinsic absorption effect. In this context, inserting reprocessed fuel in positions usually reserved for fresh elements better compensates for this absorption effect, helping to maintain the reactor's reactivity pattern and the uniformity of power distribution. In addition, the fuel assemblies fabricated with 3.2% enrichment incorporate gadolinium (Gd_2O_3) rods. Gadolinium is a burnable poison used to control the initial excess reactivity. Thus, inserting 3.2% assemblies accompanied by Gd rods helps decrease the reactivity peak at the beginning of the cycle, reduces the need for soluble absorbers, and extends core efficiency throughout operation. Consequently, a larger number of gadolinium rods leads to increased neutron absorption. Reprocessed fuels also exhibit high neutron absorption due to the presence of specific isotopes, such as americium (STACEY, 2018).

Therefore, reprocessed fuel assemblies can be introduced into the core in a way that maintains similar fission power and absorption rates compared to fresh UO_2 fuel with gadolinium. The simulation did not include refueling, which is considered an optimization approach. The burnup lasted 868 days without fuel relocation or performing replacements to account for the worst-case scenario. Based on this approach, six reprocessed fuel designs were developed in this study, incorporating thorium (Th) and depleted uranium (DU) as spike materials. The aim was to replace UO_2 assemblies enriched to 3.2% U-235 and containing 12 gadolinium rods.

The methodology used to adjust the fissile material content for each reprocessing strategy was based on achieving the same neutron multiplication factor (k).

3.2 Amount of reprocessed and spike material

According to Aruquipa *et al.* (2017), the percentage of fissile isotopes in GANEX, UREX+, and PUREX is 64.132%, 65.188%, and 65.800%, respectively. These values are higher than those to be inserted in a reactor. The simulations described in the last section will provide the necessary fissile material for each method. So, to achieve them, a spiking step is required. According to Table 3.3, reprocessing techniques present different recovery rates. The PUREX process has recovery rates of 99.9% for uranium, 99.8% for plutonium, and 95% for neptunium (BAETSLÉ; RAEDT, 1997). The GANEX process has recovery rates of 93.5% for uranium, 99.8% for neptunium, 99.6% for plutonium, 99.8% for americium, and 99.8% for curium (ANEHEIM, 2012). The UREX+ process has recovery rates of 99.95% for uranium, 71% for neptunium, 99.5% for plutonium, 98% for americium, and 79% for curium (BARROS *et al.*, 2016). For example, Table 3.4 shows the composition of a reprocessed fuel from Angra 1 before the spiking steps.

Table 3.4: Percentage composition of the processed material.

Isotope	**PUREX**	**GANEX**	**UREX+**
Np-237	5.328	5.467	3.957
Pu-238	2.643	2.576	2.618
Pu-239	52.064	50.744	51.580
Pu-240	20.767	20.240	20.573
Pu-241	13.736	13.388	13.608
Pu-242	5.462	5.323	5.411
Am-241	-	0.624	0.623
Am-242	-	0.015	0.015
Am-243	-	1.581	1.579
Cm-245	-	0.040	0.032
Cm-246	-	0.004	0.003

Source: From Author (2025).

Reprocessing needs a spiking step with natural uranium, depleted uranium, or natural thorium. Although the need for mining decreases when natural uranium or thorium is used in spiking with reprocessed fuel, when it spikes with depleted uranium, it is unnecessary to take another mining step, given that the tailings from the first cycle are used.

These reprocessing approaches reduce the demand for uranium mining, mitigating environmental impacts such as habitat destruction, water contamination, and greenhouse gas emissions. They also provide essential information on nuclear energy's social and safety dimensions,

contributing to its greater societal acceptance. Moreover, when proliferation-resistant recuperation methods, such as GANEX and UREX+, are used, they can contribute to non-proliferation commitments, guaranteeing a more peaceful nuclear fuel cycle and increasing the widespread acceptance of nuclear energy.

The Renu code developed by Aruquipa *et al.* (2017) will calculate the fuel composition, achieving the fissile material necessary for the three methods (GANEX, UREX+, and PUREX). Some equations will be solved and compared to demonstrate the required amount of reprocessed material, depleted uranium, and natural thorium to constitute a new fuel.

To demonstrate that the amount of depleted uranium can be used for reprocessing, the Angra 2 reactor described in the last section is considered. The initial enrichment of the fuel in the reactor is 3.2%, and it has a burnup of 33 GWd/MTHM. The fuel loading is 103 MTU (ELETRONUCLEAR, 2025b). Still, considering that one-third of the reactor is reloaded every 12 months, the amount of heavy metal in the new fuel to be inserted is approximately 34.33 MTU. Using the following Equation 3.1 from Cochran e Tsoulfanidis (1993):

$$\frac{F}{P} = \frac{x_p - x_w}{x_f - x_w} \tag{3.1}$$

Where F is the quantity in kilograms of feed material (per unit time), and P is the quantity in kilograms of enriched material (per unit time). X_f, x_p, and x_w are the weight fractions of U-235 in feed material, product enriched, and tails assay, respectively.

For saving mining, a spiking step with depleted uranium is a better option, since this was already mined at the beginning of the cycle (tailings). With thorium or natural uranium, one more mining step would be necessary.

According to Aruquipa *et al.* (2017), with the following Equations 3.2 and 3.3, it is possible to normalize the values of the new mixture to a percentage of fissile material:

$$NF_R = \frac{f - RAFM}{f - f_R} \tag{3.2}$$

$$NF = \frac{RAFM - f_R}{f - f_R} \tag{3.3}$$

Equation 3.2 is the normalization factor for the reprocessed material (NF_R), where f is the percentage of fissile material present in depleted uranium (0.2%) or thorium Th-232 (0.0%), f_R is the percentage of fissile material in the reprocessed material, and RAFM is the required amount of fissile material. Values must be given in percentage.

Equation 3.3 is the normalization factor (NF) for the depleted uranium or natural thorium and follows the same values of f, f_R, and RAFM. Values must be given in percentage. With these factors, it is possible to analyze the amount of each material in the final reprocessed fuel.

3.3 Decay and radiotoxicity

The last analysis focused on comparing the isotopic composition and radiotoxicity of recycled nuclear fuel spiked with depleted uranium and natural thorium. The composition was tracked for 1,000 years after a complete burnup cycle for each fuel to evaluate its long-term radiological behavior. The assessment considered specific activity, decay heat, and radiotoxicity by inhalation and ingestion.

Assembly burnup calculations were performed with SERPENT 2.2, using the same core model parameters in Table 3.2. The burnup was simulated up to 33 GWd/MTHM, corresponding to an operational period of approximately 868 days (2.37 years) at a specific power of 38 MW/MTHM, consistent with data from the FSAR of Eletrobras Termonuclear Eletrobras Termonuclear S.A. (2013). Each calculation employed 100,000 neutron histories over 1,500 cycles, with the first 400 cycles discarded to ensure statistical convergence. Reflective boundary conditions were applied to the assembly during the burnup phase.

The composition of the fuels used will be calculated as presented in the last sections. The analysis will evaluate the standard fuel and the fuels obtained by PUREX, GANEX, and UREX+ methods. Each reprocessing technique product will be spiked with depleted uranium and thorium to get the new fuel.

Following irradiation, the spent fuel assemblies were subjected to a depletion calculation over 1,000 years to capture the evolution of isotopic inventories. Decay heat, specific activity, and radiotoxicity (for actinides and fission products) were quantified and compared with those of standard UO_2 assemblies. This approach enables a systematic evaluation of the impact of depleted uranium and thorium spiking on the radiological characteristics of reprocessed fuel throughout the post-irradiation period.

This analysis is a strategy to understand the long-term implications of reprocessing on waste management and nuclear fuel cycle sustainability. By quantifying radiotoxicity, decay heat, and activity over 1,000 years, it becomes possible to compare the performance of depleted uranium and thorium spiking in terms of reducing the radiological hazard of spent fuel. These results provide input for optimizing fuel cycle design, improving repository safety assessments, and supporting decisions on adopting proliferation-resistant reprocessing technologies.

3.4 Final remarks

This chapter has presented the methodology of this work. It can be summarized in the following steps:

- Design the fuel and the reactor: the base of the reprocessed fuel is the burned Angra 1's fuel, which will be inserted in the Angra 2 reactor. So, it is necessary to find the keff

in Angra 2 standard operation and find with the Renu code which composition reaches close to the same k for each method and spike material. The burnup simulations use the software SERPENT 2.2.1;

- Amount of fissile material and spiking material: with the percentage of fissile material necessary for each method, it is possible, with equations, to calculate the amount of fissile material and spiking material required to fabricate the new fuel. With this, it is possible to analyze whether the entire amount is available;
- Decay and radiotoxicity analyses: the fuels of each method will be simulated in SERPENT 2.2.1, considering the Angra 2 reactor. After burning in a cycle of 868 days in 33 GWd/t, the decay and radiotoxicity will be simulated through 1000 years.

The next chapter will show the results of this methodology. The composition of each fuel, the amount of material, and the analyses to discuss how reprocessing can be an alternative to the challenges in the nuclear sector.

4 RESULTS

This chapter presents the main results obtained. First, the compositions of the fuels are detailed, followed by the required amounts of fissile and spiking materials. Finally, the burnup, decay, and radiotoxicity results are discussed, allowing for an evaluation of the potential of reprocessing as an alternative for the nuclear sector.

4.1 Simulation of the reactor and fuel

Considering the methodology in section 3.1, the first step is to achieve k-eff values closer to the standard in Angra 2 using a reprocessed fuel from Angra 1 (COTA; PEREIRA, 1997). The final results of these simulations using SERPENT 2.2.1, including the corresponding k-eff values obtained for the reprocessed-fuel cores, are summarized and compared with the reference case in Table 4.1. The validation of the neutron multiplication factor was performed through comparison with reference studies previously developed at the Nuclear Engineering Department (DEN - UFMG), which adopted similar core configurations, fuel enrichment, burnup levels, and modeling assumptions (Cota e Pereira (1997), Monteiro *et al.* (2015), Castro (2020)). The agreement between the reference k-eff values reported in the literature and those obtained in this work confirms the consistency and adequacy of the adopted modeling approach.

Table 4.1: Steady state core evaluation using the designed TRU fuels replacing the 3.2% enrichment. UOx with 12 Gd rods.

Recovery Method	**Model Type**	**k-eff+σ (pcm)**
	Standard	1.25457 ± 14
PUREX	UOx+TRU(DU)	1.25495 ± 14
	UOx+TRU(Th)	1.25371 ± 14
GANEX	UOx+TRU(DU)	1.25548 ± 14
	UOx+TRU(Th)	1.25416 ± 14
UREX+	UOx+TRU(DU)	1.25476 ± 15
	UOx+TRU(Th)	1.25356 ± 15

Source: From Author (2025).

The heuristic method found these values using the composition given by the Renu code. It calculated the compositions of the reprocessed fuel based on the burned fuel's composition of Angra 1 (available in Table 3.1), the recovery factor of elements for each reprocessing method (available in Table 3.3), and the percentage of fissile material in depleted uranium (0.2%) or thorium, considering the Th-232 (0.0%), depending on the required spike. By considering

these inputs and the user-defined target for fissile material (according to Table 4.2), the software determines the final composition of the reprocessed fuel. The composition of each method is shown in Tables 4.5, 4.6, 4.3, 4.4, 4.7, and 4.8.

Table 4.2: Percentage of fissile material in the reprocessed fuel for insertion in the reactor.

Spiked with	**PUREX**	**GANEX**	**UREX+**
Depleted uranium	1.40	1.50	1.40
Natural thorium	2.30	2.60	2.30

Source: From Author (2025).

Table 4.3: TRU(DU) fuel for the PUREX method, Isotopic weight fractions with 1.40% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	0.000000E+00	U-234	0.000000E+00	U-235	1.732317E-03
U-236	0.000000E+00	U-237	0.000000E+00	U-238	8.644264E-01
Pu-238	3.066016E-04	Pu-239	8.028622E-03	Pu-240	2.744672E-03
Pu-241	2.580688E-03	Pu-242	9.752946E-04	O-16	1.184536E-01
Np-237	7.503723E-04	Np-238	2.225862E-08	Np-239	1.405402E-06

Source: From Author (2025).

Table 4.4: TRU(Th) fuel for the PUREX method, Isotopic weight fractions with 2.30% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	0.000000E+00	U-234	0.000000E+00	U-235	0.000000E+00
U-236	0.000000E+00	U-237	0.000000E+00	U-238	0.000000E+00
Th-232	8.496309E-01	Pu-238	5.842238E-04	Pu-239	1.529840E-02
Pu-240	5.229924E-03	Pu-241	4.917454E-03	Pu-242	1.858406E-03
O-16	1.210482E-01	Np-237	1.429821E-03	Np-238	4.241340E-08
Np-239	2.677968E-06				

Source: From Author (2025).

Table 4.5: TRU(DU) fuel for the GANEX method, Isotopic weight fractions with 1.50% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	6.74976E-14	U-234	2.78566E-07	U-235	1.73970E-03
U-236	7.41101E-06	U-237	1.91173E-10	U-238	8.62640E-01
Sm-150	4.44607E-05	Pu-239	8.68990E-03	Pu-242	1.05562E-03
Pu-241	2.79325E-03	Pu-240	2.97074E-03	Pu-238	3.31855E-04
O-16	1.18473E-01	Np-239	1.52574E-06	Np-238	2.41646E-08
Np-237	8.14625E-04	Nd-143	2.21709E-04	Eu-153	9.45432E-06
Cm-242	8.38122E-07	Cm-244	9.60822E-07	Cm-245	3.34340E-08
Am-241	2.69160E-06	Am-242	4.95343E-09	Am-243	2.01874E-04

Source: From Author (2025).

Table 4.6: TRU(Th) fuel for the GANEX method, Isotopic weight fractions with 2.60% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	1.34156E-13	U-234	5.53668E-07	U-235	2.88151E-05
U-236	1.47299E-05	U-237	3.79968E-10	U-238	3.50286E-03
Th-232	8.41326E-01	Sm-150	8.83684E-05	Pu-238	6.59583E-04
Pu-239	1.72717E-02	Pu-242	2.09812E-03	Pu-241	5.55176E-03
Pu-240	5.90453E-03	O-16	1.21061E-01	Np-239	3.03251E-06
Np-238	4.80287E-08	Np-237	1.61912E-03	Nd-143	4.40662E-04
Eu-153	1.87911E-05	Cm-242	1.66582E-06	Cm-244	1.90970E-06
Cm-245	6.64523E-08	Am-241	5.34973E-06	Am-242	9.84526E-09
Am-243	4.01237E-04				

Source: From Author (2025).

Table 4.7: TRU(DU) fuel for the UREX+ method, Isotopic weight fractions with 1.40% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	0.000000E+00	U-234	0.000000E+00	U-235	1.732320E-03
U-236	0.000000E+00	U-237	0.000000E+00	U-238	8.644277E-01
Pu-242	9.752933E-04	Pu-241	2.580684E-03	Pu-240	2.744669E-03
Pu-239	8.028612E-03	Pu-238	3.066012E-04	O-16	1.184531E-01
Np-239	1.053518E-06	Np-238	1.668552E-08	Np-237	5.624947E-04
Cm-242	6.148049E-07	Cm-244	7.048112E-07	Cm-245	2.452552E-08
Am-241	2.449285E-06	Am-242	4.507488E-09	Am-243	1.836999E-04

Source: From Author (2025).

Table 4.8: TRU(Th) fuel for the UREX+ method, Isotopic weight fractions with 2.30% of fissile material.

Isotope	**%WF**	**Isotope**	**%WF**	**Isotope**	**%WF**
U-233	0.000000E+00	U-234	0.000000E+00	U-235	0.000000E+00
U-236	0.000000E+00	U-237	0.000000E+00	U-238	0.000000E+00
Th-232	8.496333E-01	Pu-242	1.858405E-03	Pu-241	4.917451E-03
Pu-240	5.229921E-03	Pu-239	1.529839E-02	Pu-238	5.842235E-04
O-16	1.210471E-01	Np-239	2.007462E-06	Np-238	3.179399E-08
Np-237	1.071824E-03	Cm-242	1.171500E-06	Cm-244	1.343006E-06
Cm-245	4.673298E-08	Am-241	4.667072E-06	Am-242	8.588944E-09
Am-243	3.500370E-04				

Source: From Author (2025).

With these compositions, it is possible to constitute the next steps of this research:

- The next section uses the value of Table 4.2 to calculate the amount of fissile and spiking material necessary to make the fuel;
- Then, the decay and radiotoxicity analyses can be conducted considering the burn of the fuels shown in Tables 4.5 to 4.8.

4.2 Analyzing how reprocessing can contribute to saving mining and proliferation resistance

For the reprocessed fuel to be inserted into the reactor, it must be spiked with depleted uranium, natural uranium, or thorium. This work will analyze the amount of reprocessed material, depleted uranium, and natural thorium necessary to make a reprocessed fuel with each reprocessing method (PUREX, GANEX, UREX+), complementing the earlier studies by highlighting the feasibility associated with the environmental impact. Previous studies have evaluated the neutronic parameters of inserting reprocessed fuels, demonstrating the possibility of their inclusion in the core (COSTA *et al.*, 2009).

Table 3.4 shows that after reprocessing by any method, the fissile material content is high, making its direct insertion into the reactor unfeasible. To maintain a stable reaction, it was shown in the previous section that the amount of fissile material in the fuel to be inserted into the Angra 2 reactor must be in accordance with the values of Table 4.2.

Initially, it is necessary to calculate the amount of material involved in fabricating Angra 2 standard fuel. The reactor fuel has an initial enrichment of 3.2% and reaches a burnup of 33 GWd/MTHM. The total fuel loading is 103 MTU (ELETRONUCLEAR, 2025b). Since one-third of the core is replaced every 12 months, the heavy metal in the fresh fuel loaded is about 34.333 MTU. To apply Equation 3.1, the enriched product mass P was set to 34.333 MTU,

with a fissile content of 3.2%. The fissile content of the feed material was assumed to be 0.711%, while the tailings assay was fixed at 0.2%. Solving the equation indicates that, for each annual reload of 34.333 MTU, approximately 201.563 MTU of natural uranium must be mined. This value is adopted in the subsequent calculations to verify whether the amount of depleted uranium available at the beginning of the cycle is sufficient to fabricate a new reprocessed fuel and to estimate the associated mining savings.

With the values in Table 4.2 for depleted uranium with 0.2% of fissile material, and knowing that the percentage of fissile isotopes in GANEX, UREX+, and PUREX is 64.132%, 65.188%, and 65.800%, respectively, Equations 3.2 and 3.3 were applied to obtain the values for each reprocessing method spiked with depleted uranium. Similarly, using the values in Table 4.2 for natural thorium (Th-232) with 0.0% of fissile material and the same fissile isotope fractions, the corresponding values for each reprocessing method spiked with natural thorium were obtained.

The resume of the calculated percentages is shown in Table 4.9 for spiking with depleted uranium and Table 4.10 for spiking with thorium. With the values found, it is possible to calculate the amount of reprocessed material and material for spiking (depleted uranium or natural thorium), because the values give the percentage of the amount fuel should be reprocessed material (NFR) and material for spiking (NF) to constitute the new fuel to reach the necessary fissile material in each method with each spiking material.

Table 4.9: The percentage of reprocessed material and depleted uranium to constitute the new fuel.

Method	**Fissile material**	**Reprocessed material**	**Depleted Uranium**
PUREX	1.40%	1.85%	98.15%
GANEX	1.50%	2.03%	97.97%
UREX+	1.40%	1.83%	98.17%

Source: From Author (2025).

Table 4.10: The percentage of reprocessed material and thorium to constitute the new fuel.

Method	**Fissile material**	**Reprocessed material**	**Thorium**
PUREX	2.30%	3.53%	96.47%
GANEX	2.60%	4.05%	95.95%
UREX+	2.30%	3.50%	96.50%

Source: From Author (2025).

Considering the values for a burnup of 33 GWd/MTHM, the following values are given for the amount of feed material, fresh fuel, and spent fuel:

- Feed material (natural uranium): 201.563 MTU/year;

- Fresh fuel: 34.333 MTHM/year;
- Spent fuel: 34.333 MTHM/year.

The spent fuel is 34.333 MTHM/year because it is the amount of fuel that entered the reactor and will approximately be the amount that will exit.

Considering the heavy metal in fresh fuel is only uranium, the tailings (depleted uranium) can be regarded as approximately 167.230 MTHM/year, according the solution of the Equation 4.1, where F_M is feed material, F_F is fresh fuel, and W is tailings:

$$F_M - F_F = W \tag{4.1}$$

If the reactor exemplified needs 34,333 MTHM/year of fresh fuel for a cycle burnup of 33 GWd/MTHM, the reprocessed fuel needs the exact amount and should be constituted by the percentages of reprocessed material and depleted uranium for each method, as shown in Tables 4.9 and 4.10.

Considering Equations 4.2 and 4.3 to calculate the amount of each material to constitute the new reprocessed fuel. R_M is the reprocessed material and S_M is spike material:

$$R_M = F_F * NF_R \tag{4.2}$$

$$S_M = F_F * NF \tag{4.3}$$

Considering the values in Tables 4.9 and 4.10, the fuel with a quantity of 34.333 MTHM/year, and the Equations 4.2 and 4.3, the following Tables 4.11 and 4.12 contain the amount of each material in each method:

Table 4.11: Amount of reprocessed material and depleted uranium for the constitution of the new fuel.

Material	**PUREX**	**GANEX**	**UREX+**
Depleted uranium (MTU/year)	33.699	33.635	33.704
Reprocessed material (MTHM/year)	0.634	0.698	0.628

Source: From Author (2025).

Considering the amount of uranium mined in the first cycle (201.563 MTU/year) and the need for mining to make a reprocessed fuel, it is possible to see in Table 4.13 the total saved for mining, because if a new fuel were produced, the feed material would be the same as in the first

Table 4.12: Amount of reprocessed material and thorium for the constitution of the new fuel.

Material	PUREX	GANEX	UREX+
Thorium (MTHM/year)	33.121	32.941	33.133
Reprocessed material (MTHM/year)	1.211	1.392	1.200

Source: From Author (2025).

cycle. The values result from the subtraction between the total mined in the first cycle (201.563 MTU/year) and the material for spiking shown in Tables 4.11 and 4.12.

Table 4.13: Total saved for mining in each method.

Material		PUREX	GANEX	UREX+
DU (MTU/year)	Need for mining	0*	0*	0*
	Total saved	201.563	201.563	201.563
	Total saved (%)	100%	100%	100%
Th (MTHM/year)	Need for mining	33.122	32.941	33.133
	Total saved	168.442	168.622	168.431
	Total saved (%)	83.57%	83.66%	83.56%

*The need for mining with depleted uranium is 0, because the tailings mined in the first cycle would be used.
Source: From Author (2025)

Since the reprocessed fuel to be inserted was the Angra 1 spent fuel, it is possible to analyze if the amount of spent fuel is sufficient to constitute the reprocessed fuel. The Angra 1 has a fuel loading of 49.5 MTU/year (ELETRONUCLEAR, 2025a), and considering the third part, the result of spent fuel is 16.5 MTHM/year. As shown in Tables 4.11 and 4.12, the amount of reprocessed material requested to fabricate a new reprocessed fuel is much less than the spent fuel of Angra 1 (16.5 MTHM/year) if it were used for reprocessing. Table 4.14 shows the amount of spent fuel from Angra 1 that would be used for reprocessing for each method and the percentage this amount represents of the total 16.5 MTHM/year burned. It can be observed that, in both cases, only a portion is utilized, thus ensuring the feasibility of reprocessing. These results also indicate that this reuse contributes to reducing the volume of nuclear waste generated, which would impact the 25% reduction in GHG emissions related to disposal, as shown in Figure 2.2.

Table 4.14: Amount of spent fuel allocated for reprocessing.

Material	Mining	PUREX	GANEX	UREX+
DU(MTU/year)	Spent fuel required	0.634	0.698	0.628
	Percentage of spent fuel used	3.84%	4.23%	3.80%
Th (MTHM/year)	Spent fuel required	1.211	1.392	1.200
	Percentage of spent fuel used	7.34%	8.43%	7.27%

Source: From Author (2025).

The value of depleted uranium is lower than that of tailings in the first cycle. So, using the reprocessing methods, the need for mining decreases. As shown in Tables 4.11 and 4.12, when thorium is used as a material for spiking, the need for mining decreases compared to a step with natural uranium to fabricate a new fuel. But, the absence of the need for mining when using depleted uranium makes it a more sustainable option for the nuclear fuel cycle, saving 201.563 MTU/year compared to the values around 168 MTU/year when spiked with thorium, since the amount of depleted uranium is available. Additionally, prioritizing the use of depleted uranium in the spiking process can support proliferation-resistant strategies, since the use of natural thorium (Th-232) still leads, through its decay chain, to the production of U-233, which, despite being considered safer due to the presence of U-232, remains attractive for nuclear weapons (BATHKE *et al.*, 2010).

The data from Table 4.11 show that the GANEX and UREX+ methods require amounts of depleted uranium and reprocessed material similar to those of the PUREX process for the constitution of new fuel, with only minor differences. GANEX, for example, requires 33.635 MTU/year of depleted uranium and 0.698 MTHM/year of reprocessed material, values that, although slightly higher, are justified by its commitment to non-proliferation. The UREX+ method presents figures even closer to PUREX, with a slight reduction in the amount of reprocessed material required. These minimal differences in input consumption indicate that, from a technical and material efficiency standpoint, GANEX and UREX+ are viable alternatives to PUREX.

The most relevant aspect lies in the fact that both GANEX and UREX+ are considered proliferation-resistant processes, unlike PUREX, which separates pure plutonium and increases the risk of nuclear proliferation. Therefore, the fact that these methods can produce new fuel with material performance practically equivalent to that of PUREX supports their adoption in nuclear reprocessing policies aimed at global security. The combination of technical viability and reduced proliferation risk makes GANEX and UREX+ options more aligned with the goals of non-proliferation and sustainability in the nuclear fuel cycle.

The calculations confirm that only a portion of spent fuel and depleted uranium must be reprocessed each year, allowing a single initial fresh fuel cycle to support multiple reactors or reactor lifetimes. This also reinforces the use of depleted uranium in the fuel spike compared to thorium and natural uranium. Regardless of the material used in the spiking, the amount of fis-

sile material would be reached, but only with the use of depleted uranium that has already been mined and not used, is it possible to achieve the objective of reducing mining and thus having a more sustainable cycle. Additionally, the use of proliferation-resistant recuperation methods such as GANEX and UREX+, when combined with depleted uranium, contributes to minimizing the risks of nuclear proliferation. The results show the possibility of using reprocessing technologies to improve fuel sustainability when used with depleted uranium. Moreover, the amount of depleted uranium in the first step of the cycle is stored. As shown, for one year, the high amount can increase the possibility of fabricating reprocessed fuel, and there is still left. So, to a disarmament commitment, as it was "Megatons to Megawatts" (YULISH, 2002), there is an amount of depleted uranium that could be used to dilute the existing warheads in the countries that still have weapons.

4.3 Analyzing the decay and radiotoxicity

For each reprocessing method, the radiotoxicity of the fuel was evaluated after a single burnup cycle in a core modeled on Angra 2. Based on the decay data, decay heat, specific activity, the inhalation and ingestion radiotoxicity were analyzed for all reprocessed assemblies. These results were compared with those of reference UOx assemblies over 1000 years of decay, following a burnup of 33 GWd/MTHM. This evaluation aims to compare the compositional characteristics of fuels spiked with depleted uranium against those spiked with natural thorium. In Table 4.15, the k-eff values at the beginning of life (BOL) and end of life (EOL) for the STANDARD (STD) and TRU assemblies, during the burnup phase, can be seen.

Table 4.15: Effective multiplication factor at BOL/EOL and the mean value of computational error σ during the burnup phase.

Recovery Method	**Assembly Fuel**	**BOL**	**EOL**	**σ (pcm)**
	STD	1.22914	0.91944	9
PUREX	TRU(DU)	1.23771	0.74397	11
	TRU(Th)	1.22732	0.79861	11
GANEX	TRU(DU)	1.24173	0.74700	11
	TRU(Th)	1.23148	0.80159	11
UREX+	TRU(DU)	1.23735	0.74379	11
	TRU(Th)	1.22606	0.79942	11

Source: From Author (2025).

The reprocessed fuels with the lowest k-eff at the end of life (EOL) are the TRU(DU) fuels. These are also the fuel types with the lowest percentage of fissile materials, as shown in Table 4.2. No reprocessed fuels reach the multiplication factor of the standard fuel at EOL, which

is expected, given the absorbers. All fuel types become subcritical at EOL (33 GWd/MTHM).

During the burnup phase, composition tracking was performed for the actinides of interest across all models. Only actinides were monitored in this evaluation. The analysis did not reveal significant differences in the behavior of fission products, in terms of activity, between the different models and fuel types. Over long-term decay, fission products exhibit a marked decrease in activity after about 100 years, which aligns with expectations. For this reason, the focus here is placed solely on the behavior of actinides.

For a more detailed assessment, a set of nuclides of particular relevance was selected for the evaluation of composition evolution. According to the Spent Nuclear Fuel Assay Data for Isotopic Validation (OECD Nuclear Energy Agency (NEA), 2011), the isotopes Pu-238, Pu-239, Pu-240, Np-137, Am-241, Cm-242, and Cm-244 are the principal contributors to radiological safety concerns in dry spent fuel storage facilities over time frames up to 1000 years, for both UOx and MOX fuels. These nuclides dominate the long-term radiation source terms, especially neutron and gamma emissions, with Pu-238, Pu-239, and Am-241 being the most significant contributors. In addition, given the use of thorium and depleted uranium as spiking materials, it is also relevant to monitor the evolution of U-238 and Th-232. Monitoring the U-233, a fissile isotope produced through neutron absorption in Th-232, is also important.

The following tables present the absolute mass at each burnup phase and the mass difference between the irradiated fuel and the fuel after 1000 years of decay, while the figures illustrate the evolution of uranium, thorium, and actinide compositions. Table 4.16, Table 4.17, and Figure 4.1 refer to the standard UOx fuel, Table 4.18, Table 4.19, and Figure 4.2 to the fuel reprocessed by PUREX, Table 4.20, Table 4.21, and Figure 4.3 to the fuel reprocessed by GANEX, and finally, Table 4.22, Table 4.23, and Figure 4.4 to the fuel reprocessed by UREX+. ΔM is the mass variation.

Table 4.16: Fuel Evolution during burnup and decay for the standard UOx fuel.

Isotope	BOL (kg)	EOL (kg)	1000 years (kg)
U-238	5.3963E+02	5.2900E+02	5.2900E+02
Th-232	0.0000E+00	9.0490E-08	6.6710E-05
U-233	0.0000E+00	2.1558E-07	1.1711E-04
Pu-238	0.0000E+00	3.9468E-02	1.7349E-05
Pu-239	0.0000E+00	1.7324E+00	1.7359E+00
Pu-240	0.0000E+00	1.1835E+00	1.0713E+00
Np-237	0.0000E+00	1.2841E-03	6.5481E-03
Am-241	0.0000E+00	9.6306E-03	9.2120E-02
Cm-242	0.0000E+00	4.7466E-03	1.4960E-09
Cm-244	0.0000E+00	7.2201E-03	1.7324E-19

Source: From Author (2025).

Table 4.17: Mass variation of main isotopes during irradiation and 1000-year decay for the standard UOx fuel.

Isotope	ΔM **burnup (kg)**	ΔM **1000 years decay (kg)**
U-238	-1.0628E+01	3.1008E-05
Th-232	9.0490E-08	6.6619E-05
U-233	2.1558E-07	1.1690E-04
Pu-238	3.9468E-02	-3.9450E-02
Pu-239	1.7324E+00	3.4142E-03
Pu-240	1.1835E+00	-1.1215E-01
Np-237	1.2842E-03	5.2639E-03
Am-241	9.6306E-03	8.2490E-02
Cm-242	4.7466E-03	-4.7466E-03
Cm-244	7.2201E-03	-7.2201E-03

Source: From Author (2025).

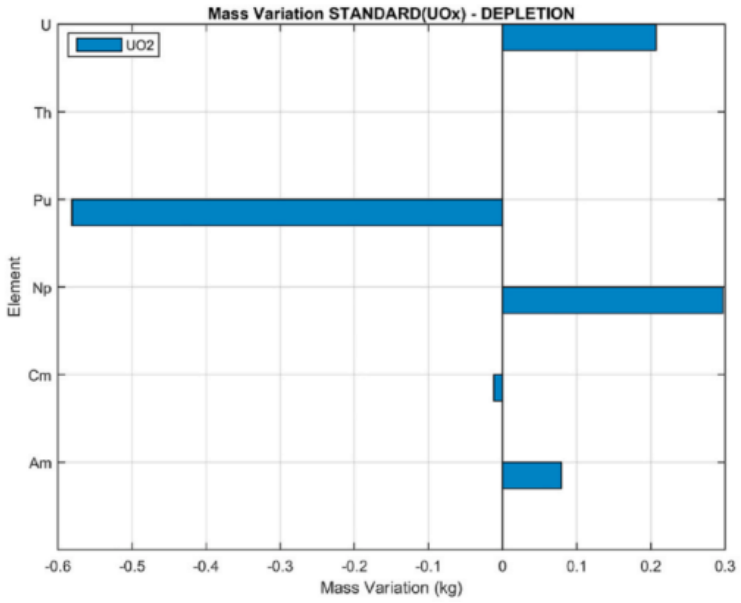

Figure 4.1: Mass variation for the actinides during 1000 years of decay - STD UOX (From Author, 2025).

Table 4.18: Fuel Evolution during burnup and decay for the reprocessed fuels using the PUREX method.

Isotope	REP-fuel	BOL (kg)	EOL (kg)	1000 years (kg)
U-238	TRU(DU)	5.4823E+02	5.3288E+02	5.3289E+02
	TRU(Th)	0.0000E+00	1.4868E-05	4.2927E-03
Th-232	TRU(DU)	0.0000E+00	6.6860E-09	7.5290E-06
	TRU(Th)	5.3885E+02	5.2399E+02	5.2399E+02
U-233	TRU(DU)	0.0000E+00	8.2181E-08	1.7063E-04
	TRU(Th)	0.0000E+00	6.3121E+00	7.1021E+00
Pu-238	TRU(DU)	1.9445E-01	1.7186E-01	7.2449E-05
	TRU(Th)	3.7052E-01	4.2095E-01	1.7428E-04
Pu-239	TRU(DU)	5.0919E+00	1.7065E+00	1.7622E+00
	TRU(Th)	9.7025E+00	1.7221E-01	2.1502E-01
Pu-240	TRU(DU)	1.7407E+00	1.9427E+00	1.9540E+00
	TRU(Th)	3.3169E+00	1.4012E+00	1.4762E+00
Np-237	TRU(DU)	7.8039E-03	3.4477E-03	1.2199E-02
	TRU(Th)	1.4870E-02	6.6953E-03	1.7257E-02
Am-241	TRU(DU)	0.0000E+00	1.9093E-02	2.4716E-09
	TRU(Th)	0.0000E+00	4.0781E-02	1.7260E-01
Cm-242	TRU(DU)	0.0000E+00	1.9093E-02	2.4716E-09
	TRU(Th)	0.0000E+00	3.7389E-02	7.4539E-09
Cm-244	TRU(DU)	0.0000E+00	2.3207E-01	5.5668E-18
	TRU(Th)	0.0000E+00	2.4271E-01	5.8219E-18

Source: From Author (2025).

Table 4.19: Mass variation of main isotopes during irradiation and 1000-year decay for the reprocessed fuels using the PUREX method.

Isotope	REP-fuel	ΔM burnup (kg)	ΔM 1000 years decay (kg)
U-238	TRU(DU)	-1.5350E+01	3.0491E-03
	TRU(Th)	1.4868E-05	4.2779E-03
Th-232	TRU(DU)	6.6860E-09	7.5223E-06
	TRU(Th)	-1.4859E+01	0.0000E+00
U-233	TRU(DU)	8.2181E-08	1.7054E-04
	TRU(Th)	6.3121E+00	7.9003E-01
Pu-238	TRU(DU)	-2.2592E-02	-1.7179E-01
	TRU(Th)	5.0428E-02	-4.2078E-01
Pu-239	TRU(DU)	-3.3854E+00	5.5762E-02
	TRU(Th)	-9.5303E+00	4.2815E-02
Pu-240	TRU(DU)	2.0194E-01	1.1361E-02
	TRU(Th)	-1.9157E+00	7.5021E-02
Np-237	TRU(DU)	-4.3562E-03	8.7511E-03
	TRU(Th)	-8.1748E-03	1.0562E-02
Am-241	TRU(DU)	1.5777E-02	1.2700E-01
	TRU(Th)	4.0781E-02	1.3182E-01
Cm-242	TRU(DU)	1.9093E-02	-1.9093E-02
	TRU(Th)	3.7389E-02	-3.7389E-02
Cm-244	TRU(DU)	2.3207E-01	-2.3207E-01
	TRU(Th)	2.4271E-01	-2.4271E-01

Source: From Author (2025).

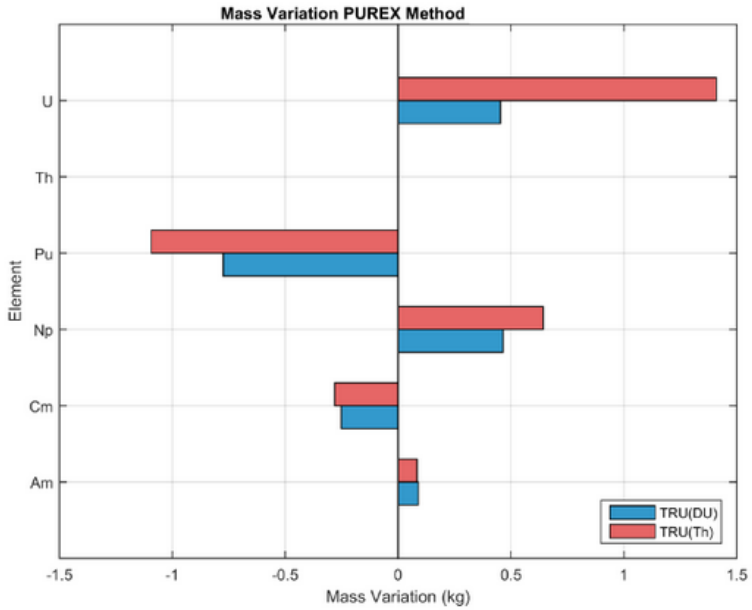

Figure 4.2: Mass variation for the actinides during 1000 years of decay - PUREX (From Author, 2025).

Table 4.20: Fuel Evolution during burnup and decay for the reprocessed fuels using the GANEX method.

Isotope	REP-fuel	BOL (kg)	EOL (kg)	1000 years (kg)
U-238	TRU(DU)	5.4710E+02	5.3228E+02	5.3229E+02
	TRU(Th)	2.2216E+00	1.9550E+00	1.9596E+00
Th-232	TRU(DU)	0.0000E+00	6.8245E-09	7.8767E-06
	TRU(Th)	5.3358E+02	5.2034E+02	5.2034E+02
U-233	TRU(DU)	4.2808E-11	9.6115E-08	1.8380E-04
	TRU(Th)	8.5084E-11	6.0717E+00	6.8000E+00
Pu-238	TRU(DU)	2.1047E-01	1.9443E-01	8.1812E-05
	TRU(Th)	4.1832E-01	5.1197E-01	2.1334E-04
Pu-239	TRU(DU)	5.5113E+00	1.7403E+00	1.7981E+00
	TRU(Th)	1.0954E+01	3.3910E-01	3.8879E-01
Pu-240	TRU(DU)	1.8841E+00	2.0337E+00	2.0946E+00
	TRU(Th)	3.7448E+00	2.0772E+00	2.1813E+00
Np-237	TRU(DU)	8.4721E-03	3.8198E-03	1.3074E-02
	TRU(Th)	1.6839E-02	8.2110E-03	2.2671E-02
Am-241	TRU(DU)	1.7071E-03	1.7865E-02	1.5107E-01
	TRU(Th)	3.3929E-03	6.0538E-02	2.3636E-01
Cm-242	TRU(DU)	5.3155E-04	2.1210E-02	2.8224E-09
	TRU(Th)	1.0565E-03	4.6372E-02	1.1255E-08
Cm-244	TRU(DU)	6.0937E-04	2.9812E-01	7.1509E-18
	TRU(Th)	1.2112E-03	3.5180E-01	8.4381E-18

Source: From Author (2025).

Table 4.21: Mass variation of main isotopes during irradiation and 1000-year decay for the reprocessed fuels using the GANEX method.

Isotope	REP-fuel	ΔM burnup (kg)	ΔM 1000 years decay (kg)
U-238	TRU(DU)	-1.482E+01	3.049E-03
	TRU(Th)	-2.666E-01	4.690E-03
Th-232	TRU(DU)	6.824E-09	7.870E-06
	TRU(Th)	-1.324E+01	-6.098E-04
U-233	TRU(DU)	9.607E-08	1.837E-04
	TRU(Th)	6.072E+00	7.283E-01
Pu-238	TRU(DU)	-1.604E-02	-1.943E-01
	TRU(Th)	9.365E-02	-5.118E-01
Pu-239	TRU(DU)	-3.771E+00	5.775E-02
	TRU(Th)	-1.061E+01	4.968E-02
Pu-240	TRU(DU)	1.496E-01	6.085E-02
	TRU(Th)	-1.668E+00	1.041E-01
Np-237	TRU(DU)	-4.6523E-03	9.2545E-03
	TRU(Th)	-8.6278E-03	1.4460E-02
Am-241	TRU(DU)	1.616E-02	1.332E-01
	TRU(Th)	5.715E-02	1.758E-01
Cm-242	TRU(DU)	2.068E-02	-2.121E-02
	TRU(Th)	4.532E-02	-4.637E-02
Cm-244	TRU(DU)	2.975E-01	-2.981E-01
	TRU(Th)	3.506E-01	-3.518E-01

Source: From Author (2025).

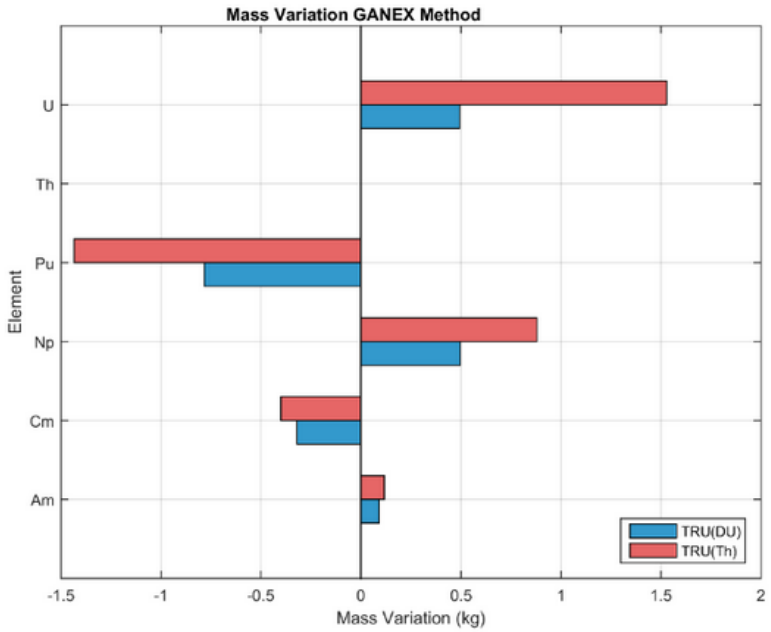

Figure 4.3: Mass variation for the actinides during 1000 years of decay - GANEX (From Author, 2025).

Table 4.22: Fuel Evolution during burnup and decay for the reprocessed fuels using the UREX+ method.

Isotope	REP-fuel	BOL (kg)	EOL (kg)	1000 years (kg)
U-238	TRU(DU)	5.4710E+02	5.3228E+02	5.3229E+02
	TRU(Th)	2.2216E+00	1.9550E+00	1.9596E+00
Th-232	TRU(DU)	0.0000E+00	6.8245E-09	7.8767E-06
	TRU(Th)	5.3358E+02	5.2034E+02	5.2034E+02
U-233	TRU(DU)	4.2808E-11	9.6115E-08	1.8380E-04
	TRU(Th)	8.5084E-11	6.0717E+00	6.8000E+00
Pu-238	TRU(DU)	2.1047E-01	1.9443E-01	8.1812E-05
	TRU(Th)	4.1832E-01	5.1197E-01	2.1334E-04
Pu-239	TRU(DU)	5.5113E+00	1.7403E+00	1.7981E+00
	TRU(Th)	1.0954E+01	3.3910E-01	3.8879E-01
Pu-240	TRU(DU)	1.8841E+00	2.0337E+00	2.0946E+00
	TRU(Th)	3.7448E+00	2.0772E+00	2.1813E+00
Np-237	TRU(DU)	5.8499E-03	2.8112E-03	1.1530E-02
	TRU(Th)	1.1147E-02	5.0173E-03	1.5543E-02
Am-241	TRU(DU)	1.7071E-03	1.7865E-02	1.5107E-01
	TRU(Th)	3.3929E-03	6.0538E-02	2.3636E-01
Cm-242	TRU(DU)	5.3155E-04	2.1210E-02	2.8224E-09
	TRU(Th)	1.0565E-03	4.6372E-02	1.1255E-08
Cm-244	TRU(DU)	6.0937E-04	2.9812E-01	7.1509E-18
	TRU(Th)	1.2112E-03	3.5180E-01	8.4381E-18

Source: From Author (2025).

Table 4.23: Mass variation of main isotopes during irradiation and 1000-year decay for the reprocessed fuels using the UREX+ method.

Isotope	REP-fuel	ΔM burnup (kg)	ΔM 1000 years decay (kg)
U-238	TRU(DU)	-1.5368E+01	2.4393E-03
	TRU(Th)	1.4877E-05	4.2787E-03
Th-232	TRU(DU)	6.6881E-09	7.5990E-06
	TRU(Th)	-1.4876E+01	-6.0982E-04
U-233	TRU(DU)	6.7925E-08	1.5786E-04
	TRU(Th)	6.3141E+00	7.9155E-01
Pu-238	TRU(DU)	-4.9053E-02	-1.4534E-01
	TRU(Th)	-1.2296E-02	-3.5808E-01
Pu-239	TRU(DU)	-3.3956E+00	5.9183E-02
	TRU(Th)	-9.5444E+00	5.0501E-02
Pu-240	TRU(DU)	1.9664E-01	6.6410E-02
	TRU(Th)	-1.9194E+00	1.6972E-01
Np-237	TRU(DU)	-3.0387E-03	8.7192E-03
	TRU(Th)	-6.1297E-03	1.0526E-02
Am-241	TRU(DU)	1.4145E-02	1.2662E-01
	TRU(Th)	3.7839E-02	1.3137E-01
Cm-242	TRU(DU)	1.8798E-02	-1.9188E-02
	TRU(Th)	3.6953E-02	-3.7696E-02
Cm-244	TRU(DU)	2.9306E-01	-2.9350E-01
	TRU(Th)	3.4814E-01	-3.4899E-01

Source: From Author (2025).

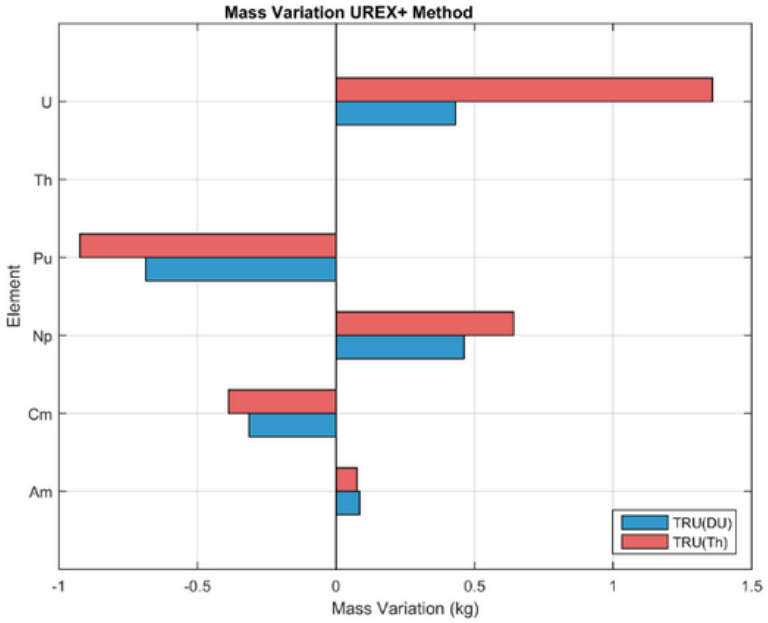

Figure 4.4: Mass variation for the actinides during 1000 years of decay - UREX+ (From Author, 2025).

The differences between the long-term spent fuel analysis methods became evident in the composition evaluation. For comparison, the evolution of UOx fuel was also assessed. As expected from the plutonium decay series, uranium formation was observed, which is consistent with plutonium's negative mass balance. From a radiological safety perspective, the presence of uranium is not a significant concern, apart from waste management requirements. Thorium, however, exhibited a small mass variation. In the TRU(Th) model, a more pronounced formation of curium and neptunium was identified, raising relevant radiological safety concerns. By contrast, americium showed only minor variation between the different reprocessing methods.

Analyzing the results for the sensitive nuclide group previously mentioned, Cm-244 is almost zero, while Cm-242 appears only in trace amounts after 1000 years of decay in all reprocessed fuels. Among the plutonium isotopes, similar behavior is observed across all reprocessed fuels in this evaluation. Pu-239, although present at higher concentrations in thorium spiked fuels, undergoes more significant depletion, resulting in final values an order of magnitude lower than those spiked with depleted uranium fuels. Pu-240 shows similar behavior across the different fuel types, whereas Pu-238 exhibits slightly higher values in depleted uranium spiked fuels after 1000 years. Am-241 is more prominently formed in thorium-spiked fuels.

When summing the masses of nuclides relevant to radiological safety, fuels spiked with depleted uranium retain higher total masses after 1000 years of decay. For example, the GANEX method, due to its simplified separation process, shows a difference between TRU(DU) and TRU(Th) fuels for these nuclides: 4.043 kg for TRU(DU) against the 2.806 kg for TRU(Th). However, as noted in previous analyses, thorium-spiked fuels exhibit higher activity, decay heat, and radiotoxicity values. The formation of U-233 explains this behavior.

In the evaluation, U-233 is not a radiological safety concern but remains a waste management issue. Th-232, a fertile material, is converted into fissile U-233 through neutron capture (primarily thermal), forming Th-233, followed by two beta decays via Pa-233, then producing U-233, which has a half-life of 159,200 years. As shown in the Tables, a significant amount of U-233 is produced over 1000 years of decay (6.8 kg in GANEX TRU(Th) compared to 0.184 g in TRU(DU)). The presence of high-activity, neutron-emitting nuclides facilitates this U-233 production.

This additional fissile material has an impact on the evaluation. Th-232 is already recognized for its potential in U-233 breeding (BALDOVA; FRIDMAN, 2016), and managing the quantities of U-233 generated in Th-spiked REP spent fuels must be carefully considered.

The behavior of the fission products is shown in the graph in Figure 4.5. They are similar in the evaluation using the SERPENT code. That means the choice of reprocessed fuels, spiked with depleted uranium or thorium, does not affect fission product decay activity. Other parameters associated with the activity, including the radiotoxicity and decay heat, tend to follow the same behavior.

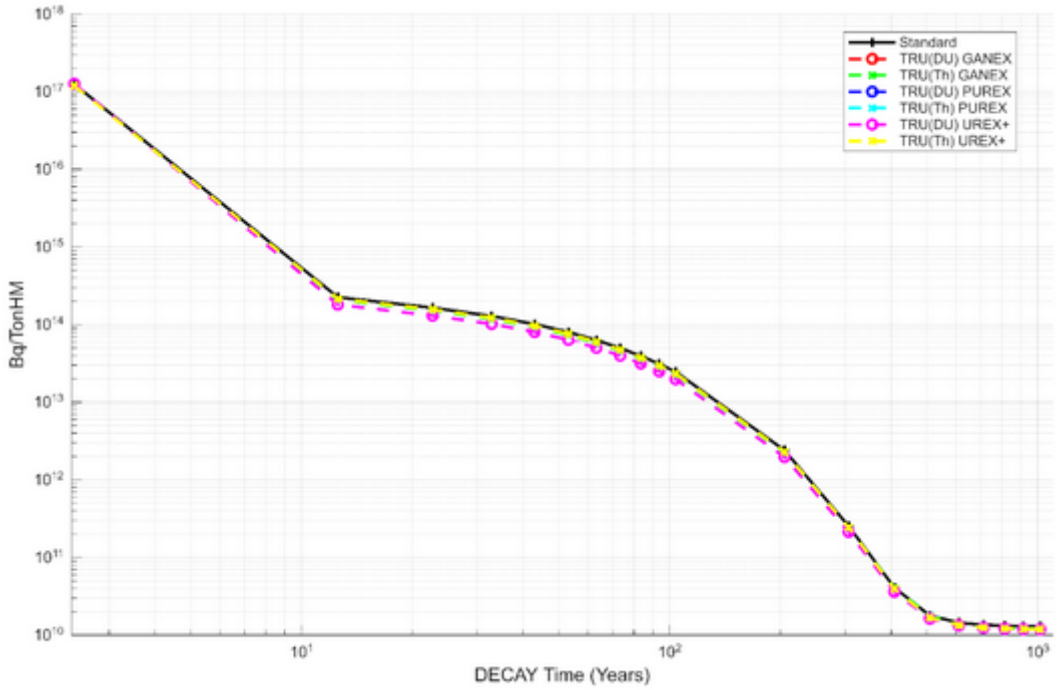

Figure 4.5: Specific activity for the fission products (From Author, 2025).

The specific activity, the decay heat, the radiotoxicity at ingestion and inhalation of the actinides were analyzed during 1000 years. As shown in Figure 4.6, the specific activity of the reprocessed fuels has the same tendency curve as the standard fuel. However, all the reprocessed fuels have a higher value than the standard. The TRU fuels spiked with natural thorium (TRU(Th)) had high activity during the decay time. Among the TRU(Th) fuels, the GANEX method has the highest specific activity, because the GANEX method has a more diverse isotope matrix and/or higher recovery factors compared to the other methods, hence, fewer isotopes are separated. These additional actinides, combined with thorium, have a negative impact on the decay fuel specific activity.

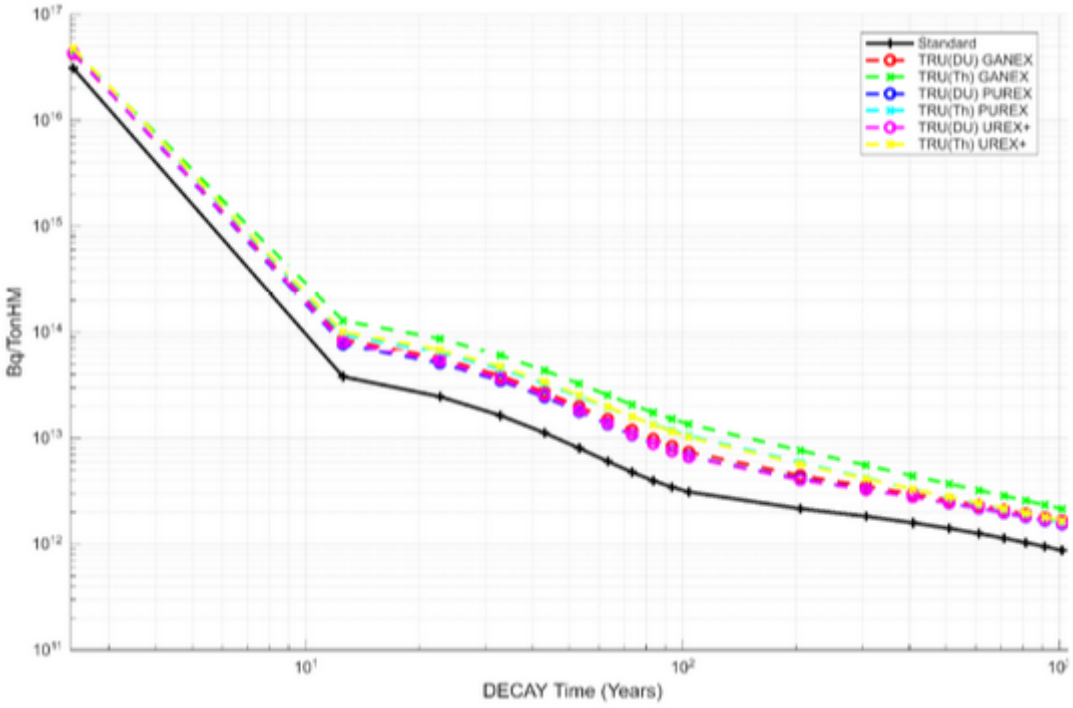

Figure 4.6: Specific activity for the actinides during 1000 years of decay (From Author, 2025).

As shown in Figure 4.7, reprocessed fuels have a more effect on decay heat. During the first 10 years, the decay heat values for standard and reprocessed fuels differ by approximately an order of magnitude. Beyond this period, the values gradually converge. Although this difference is less significant compared to other evaluated parameters, it is notable that the TRU(Th) reprocessed fuels stand out. In particular, the TRU(Th) fuel processed using the GANEX method exhibits the highest decay heat.

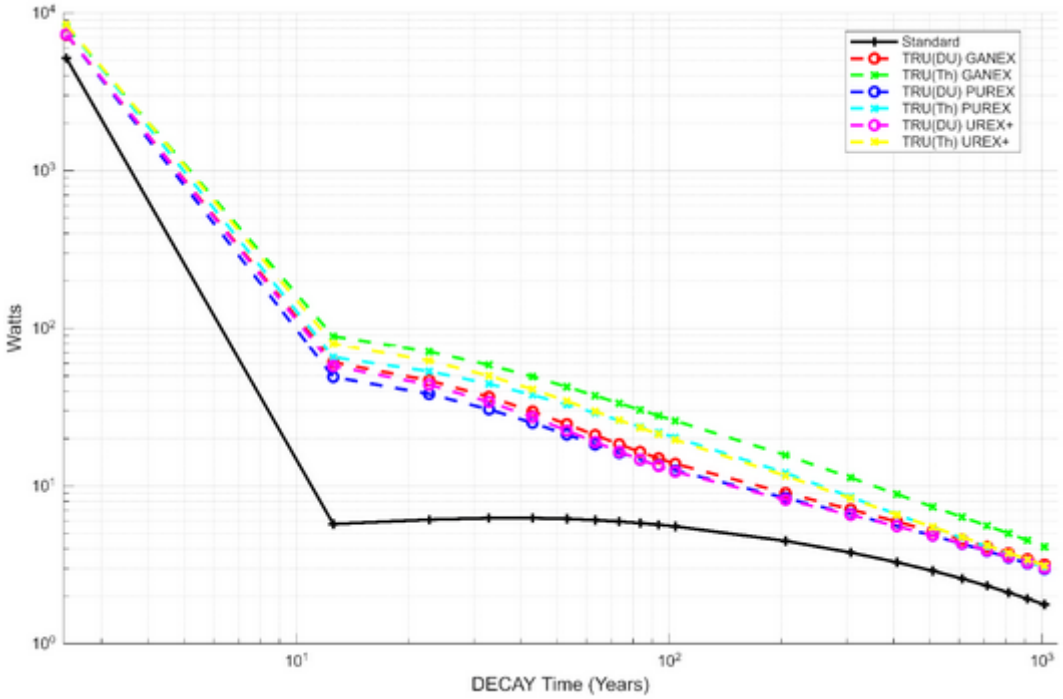

Figure 4.7: Decay heat for the actinides during 1000 years of decay (From Author, 2025).

In Figure 4.8, it is possible to observe 3 different tendencies when analyzing radiotoxicity by ingestion. The standard fuel exhibits a more gradual decline within the first 10 years, followed by a gradual, attenuated decrease over the remaining 990 years. The TRU(DU) fuel shows a reduction during the initial decade, transitioning into an almost linear decline throughout the rest of the period. The TRU(Th) fuel shows an even slower reduction in the first 10 years, forming a more curved profile around the 100-year mark, followed by a decrease in the final 900 years. Among them, the TRU(Th) fuel processed via the GANEX method presents the highest ingestion radiotoxicity.

According to Figure 4.9, the radiotoxicity by inhalation in the reprocessed fuels has more pronounced values compared to standard fuel. Once again, TRU(Th) fuel shows the highest values. The elevated toxicity during the first 100 years for reprocessed fuels is attributed to the greater presence of fertile materials, which increase the potential for harmful deposition in the lungs. The thorium spiking intensifies this effect.

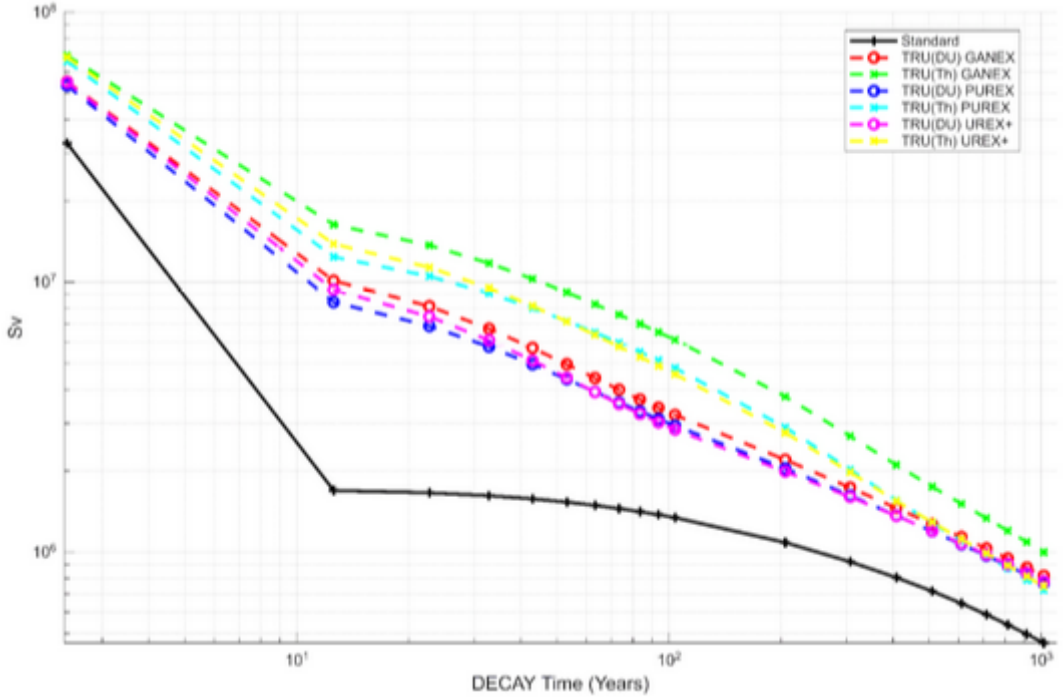

Figure 4.8: Ingestion Radiotoxicity for the actinides during 1000 years of decay (From Author, 2025).

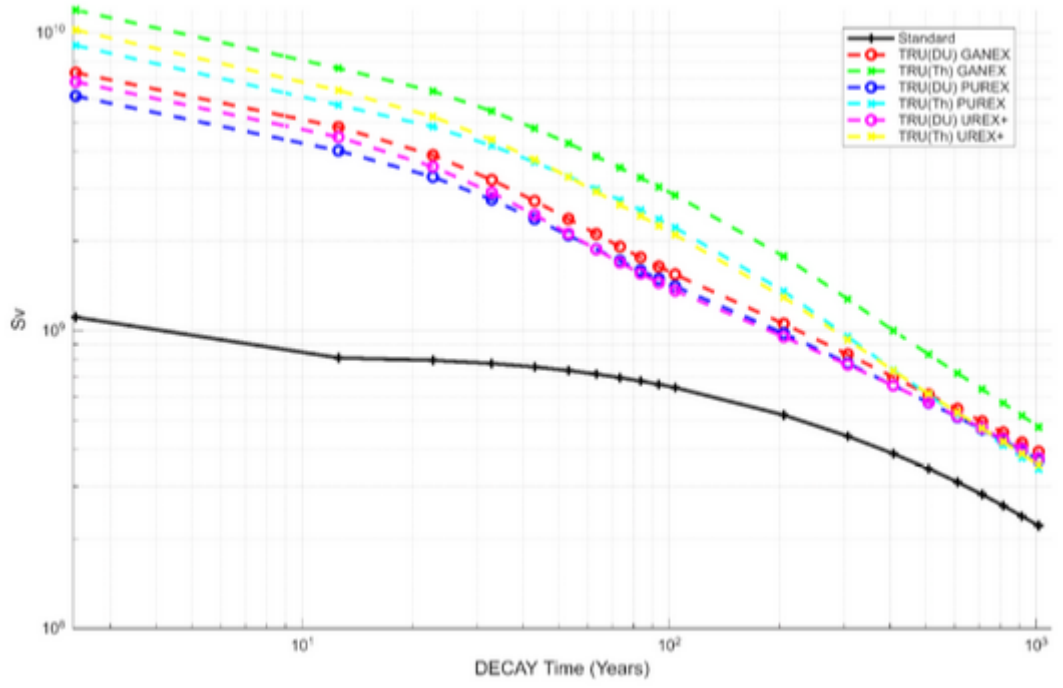

Figure 4.9: Inhalation Radiotoxicity for the actinides during 1000 years of decay (From Author, 2025).

Based on the results obtained, it is possible to observe that, regarding radiotoxicity (ingestion and inhalation), TRU fuels, especially those spiked with natural thorium, exhibit higher

values than the standard fuel in the short term. This behavior is associated with the high presence of minor actinides and plutonium isotopes, which contribute to the radiotoxicity of the irradiated material. However, the analysis over 1000 years shows that reprocessed fuels tend to present a decrease in radiotoxicity, which may represent an advantage for long-term waste management.

The specific activity of the reprocessed fuels was also higher than that of the standard fuel, with the highest values observed in the TRU(Th) fuels, particularly those processed using the GANEX method. This increase is related to the isotopic diversity and the lower efficiency in separating actinides, resulting in a greater inventory of emitting radionuclides. Nevertheless, the decay follows a similar curve among the different fuels, indicating that reprocessed fuels behave predictably over the long term.

Despite the lower amounts of plutonium and minor actinides, the analyses showed that thorium spiked reprocessed fuels exhibit higher decay heat and radiotoxicity curves. This occurs due to the production of U-233 and its decay products, which maintain elevated radiotoxicity and decay heat over long periods. Consequently, TRU–Th cycles are radiologically more complex, presenting higher activity levels and requiring advanced technological solutions for reprocessing and storage. In contrast, TRU–DU cycles, which use depleted uranium, demonstrate lower radiotoxicity and decay values, representing a more viable transitional alternative from both environmental and operational standpoints.

Regarding decay heat, reprocessed fuels show initial values up to one order of magnitude higher than those of the standard fuel during the first 10 years. After this period, the values tend to converge, reducing the thermal impact of temporary and final storage.

The TRU fuels have higher values in all parameters, which is expected due to the actinide's behavior. The plutonium isotopes have about twice the integral absorption cross-section value compared to uranium isotopes on the absorption resonances in the epithermal (0.3 to 1.5 eV) range (STACEY, 2018). Therefore, a higher gamma/beta emission from these isotopes is expected due to transmutation, increasing the decay heat and composition activity. The americium isotopes are excellent absorbers as well. The TRUs spiked with natural thorium have the highest values for actinides, and the TRUs spiked with depleted uranium behave more similarly to UOx fuel from the actinide's point of view.

Despite the higher values in the early years, the results indicate that reprocessed fuels exhibit a favorable or comparable behavior to standard fuel over centuries. This characteristic reinforces the potential of reprocessing as a tool for reducing long-term radiotoxicity, optimizing waste management, and possibly improving public perception of nuclear energy.

4.4 Final remarks

The results show that the use of depleted uranium as a spiking material in reprocessing based fuel fabrication improves the sustainability of the nuclear fuel cycle by reducing the need for uranium mining. While thorium spiking reduces mining requirements compared to natural uranium, only depleted uranium completely avoids additional mining, yielding annual savings of about 201.563 MTU. The material requirements of GANEX and UREX+ are comparable to those of PUREX. However, unlike PUREX, both GANEX and UREX+ offer enhanced proliferation resistance by avoiding the separation of pure plutonium. When combined with depleted uranium, these processes simultaneously support sustainability and non-proliferation goals, demonstrating the potential of reprocessing technologies to improve the long-term efficiency and security of the nuclear fuel cycle.

The long-term spent fuel behavior analysis highlights distinctions between standard UOx fuel and reprocessed TRU fuels. While reprocessed fuels exhibit higher specific activity, decay heat, and radiotoxicity in the short term (driven by minor actinides and plutonium isotopes), these differences decrease over time. After 1000 years of decay, the long-term trends of radiotoxicity, decay heat, and actinide composition indicate that reprocessed fuels follow predictable and manageable behavior patterns.

The formation of U-233 in thorium spiked fuels influences activity and radiotoxicity, emphasizing the importance of waste management considerations for these materials. Over centuries, the overall decrease in radiotoxicity suggests reprocessing, particularly with depleted uranium spiking, may provide advantages for long-term spent fuel management.

The next chapter for the conclusion presents a summarized overview of the research objectives and how the methodology supports the results, addressing them.

5 CONCLUSION

Nuclear fuel reprocessing emerges as an alternative to promote a closed nuclear fuel cycle, thereby enhancing the sustainability and peaceful use of nuclear resources. By enabling the recovery of fissile materials such as plutonium and uranium still present in discharged fuel, reprocessing contributes to greater fuel cycle efficiency, reducing the need for natural uranium mining and enrichment. With the use of methods such as GANEX and UREX+, it is also possible to mitigate the risk of nuclear weapons proliferation, since these methods do not separate plutonium from the actinides, unlike the PUREX method, which was initially developed for military applications.

To address the issue of mining, it is possible to use reprocessed fuel through one of the available methods (PUREX, GANEX, or UREX+), combined with a spiking step using depleted uranium or natural thorium. Although thorium requires less mining, it still must be extracted. In contrast, depleted uranium is stored from the beginning of the nuclear cycle, making it a more efficient option for reducing mining demand. The results show that using depleted uranium as a spiking material is sustainably advantageous, as it eliminates the need for new mining and uses an abundantly available resource. The results also show that the use of depleted uranium can reduce mining by 100% in the refueling of a cycle, while in the reprocessing cycle with thorium it reduces mining by approximately 83% due to the need to extract natural thorium. The spent fuel required is also smaller than the amount of burned fuel, and its use decreases the amount of waste (around 4% for depleted uranium and 8% for thorium). The data also indicate that the amount of depleted uranium required by the GANEX and UREX+ methods is comparable to that of PUREX, with the additional benefit of not separating pure plutonium, thus reducing the risks of nuclear proliferation. Furthermore, the use of available depleted uranium can assist nuclear powers in disarming by diluting weapons-grade materials, in line with the disarmament goals of the NPT.

The analysis of radiotoxicity and activity of reprocessed fuels shows that, although they present higher initial values than standard fuel (especially those enriched with natural thorium), their long-term behavior becomes more stable. The high inventory of minor actinides and plutonium isotopes is responsible for this elevated initial toxicity and specific activity. However, after the first few centuries, a more pronounced decay trend is observed in these fuels, with values approaching those of conventional fuel in terms of reduced toxicity and decay heat. This characteristic is relevant for long-term waste management, as it can reduce the complexity and costs of final disposal.

In conclusion, this research demonstrates that fuel reprocessing, particularly when combined with depleted uranium spiking and proliferation-resistant methods such as GANEX and UREX+, can enhance the sustainability and safety of the nuclear fuel cycle. These findings

contribute to the technical optimization of waste management strategies and the broader global effort of positioning nuclear energy as a viable and responsible solution to the climate crisis.

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ATTACHMENT A – Paper presented at the 7th SENCIR

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RECYCLING ACTINIDES AND DEPLETED URANIUM AS ALTERNATIVE FOR MINING SAVINGS, NON-PROLIFERATION AND WASTE MANAGEMENT

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Palavras-Chave: Nonproliferation; Nuclear fuel reprocessing; Uranium mining savings; GANEX; UREX+.

ABSTRACT

With the beginning of the nuclear industry, studies and debates have also been initiated regarding the possibility of a closed fuel cycle. The reprocessing of spent fuel by PUREX and the recycling of plutonium are already being implemented by France in a process referred to as a twice-through cycle. The next step involves recycling plutonium and minor actinides. With this goal, using GANEX and UREX+, in addition to PUREX, this research aims to demonstrate that reprocessing methods can significantly reduce the need for uranium mining, enhance commitments to non-proliferation, and effectively manage nuclear waste. The role of recycling in reducing waste cannot be overstated, and this research has shown that the reprocessing of spent fuels can play a significant part in this effort. Equations were solved to demonstrate the savings in uranium mining when reprocessed elements from spent fuels are spiked with depleted uranium. As a result, the depleted uranium generated in the first cycle may be sufficient to be blended with the reprocessed material, thereby diminishing the need of mining and reducing greenhouse gas (GHG) emissions. Furthermore, the reprocessing and recycling of spent fuels can significantly contribute to the decrease in the final nuclear waste disposal. The GANEX and UREX+ methods have shown that they contain a comparable amount of fissile material to PUREX. Still, they have advantages as being non-proliferating methods and reducing the toxicity of waste.

1. INTRODUCTION

With the advent of the nuclear industry, discussions and research on the potential of a closed fuel cycle began, focusing on recycling uranium and plutonium recovered from reprocessed spent fuel. This approach not only aims to maximize the use of nuclear fuel and reduce radioactive waste but also holds the potential to significantly decrease the need for uranium mining, offering a more sustainable future for resource management in the nuclear sector. Studies have demonstrated that closed fuel cycles provide environmental benefits, such as reducing greenhouse gas (GHG) emissions and minimizing the demand for natural uranium extraction. Additionally, compared to the open cycle, the closed cycle generates smaller volumes of waste, reduces land use impacts, and simplifies the handling of long-lived radioactive materials. These findings underscore the importance of further exploring reprocessing methods to mitigate the environmental effects of nuclear activities, reinforcing the argument that nuclear fuel recycling is key to enhancing sustainability in the energy industry [1, 2].

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Actinides and depleted uranium as an alternative for sustainable nuclear energy in thermal reactors

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Social acceptance of nuclear energy

ABSTRACT

Spent fuel reprocessing can be an alternative to address the environmental and social challenges associated with nuclear energy. This study investigates the use of depleted uranium, stored at the beginning of the cycle, in the spiking with the reprocessed fuel, eliminating the need for mining new fuel or thorium for spiking. Additionally, the radiotoxicity of burned recycled nuclear fuels is analyzed, and their composition is compared with that of thorium and depleted uranium, demonstrating that the use of both results in similar radiotoxicity characteristics. The work also reinforces the use of methods GANEX and UREX + due to the efficiency of proliferation risks compared to PUREX. The results show that the amount of spent fuel in a cycle and the depleted uranium stored in the beginning of the cycle are sufficient to constitute a reprocessed fuel to be inserted in the reactor. There is no big difference between the material required for the proliferative resistance methods and PUREX. The DU-spiked reprocessed fuels behave more similarly to standard UOx regarding radiotoxicity and decay heat, whereas Th-spiked fuels exhibit higher long-term radiological impact due to U-233 production. Depleted uranium in reprocessing offers environmental and waste management advantages, while proliferation resistant methods such as GANEX and UREX + enhance safety and societal acceptance.

1. Introduction

An energy source must be efficient and economical, preserve the environment, and have good societal acceptance to achieve sustainability goals (Poinssot et al., 2015). Therefore, for nuclear energy to meet these requirements, some achievable points include.

- Maintaining stable production costs to ensure economic efficiency;
- Minimizing greenhouse gas (GHG) emissions, environmental impacts, and preserving natural resources;
- Ensuring social acceptance through informed consensus, while minimizing health and stability risks.

A closed nuclear fuel cycle began focusing on recycling uranium and plutonium recovered from reprocessed spent fuel. In a PWR burnup at 47.5GWd/tHM, the spent fuel matrix has approximately 0.76 % of Pu (239 + 241) and 0.41 % of Pu(236 + 240+242), and 0.75 % of U-235, showing that spent fuel still contains a considerable amount of fissile material for energy output (Poinssot et al., 2015). Due to the amount of fissile material after applying a reprocessing method, a spiking step is necessary. In the context of nuclear fuel cycle, spiking refers to the intentional addition of natural uranium, depleted uranium, or thorium to a reprocessed nuclear fuel. This practice adjusts the fissile content of the reprocessed fuel, improves handling characteristics, enhances proliferation resistance, and optimizes the isotopic composition for reactor performance.

Spiking with depleted uranium not only aims to maximize the use of nuclear fuel and reduce radioactive waste but also holds the potential to significantly decrease the need for uranium mining, offering a more sustainable future for resource management in the nuclear sector. Reprocessed fuels spiked with depleted uranium can provide environmental benefits, such as reducing mining GHG emissions and minimizing the demand for natural uranium extraction. Moreover, the efficiency of advanced proliferation resistant recuperation methods can assist in nuclear disarmament policies, and this, along with less radioactive waste, can improve public acceptance of nuclear energy. These findings underscore the importance of further exploring reprocessing methods to mitigate the environmental effects of nuclear activities,