Id.: EN-41

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

Guilherme V. M. da Silva, Claubia Pereira

Universidade Federal de Minas Gerais – Departamento de Engenharia Nuclear
Av. Antônio Carlos, 6627 Campus UFMG Pampulha
31270-901, Belo Horizonte, MG, Brasil
guivmarch90@gmail.com, claubia@nuclear.ufmg.br

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].

1.1 Savings in Uranium Mining

Despite efforts to manage mining sustainably to balance economic benefits with environmental protection and social well-being, it is important to seek ways to minimize the need for this activity.

Mining can impact water, soil, and air depending on the components and methods employed. In the case of uranium, radiation exposure can harm the local community and the environment if not handled correctly [3].
In the nuclear fuel cycle, mining accounts for 25% of greenhouse gas (GHG) emissions [4]. With reprocessing, the need for mining diminishes as the spent fuel from reactors can still contain fissile material that, once reprocessed, is reintroduced into the reactor. For MOX (Mixed Oxide) fuel, separated plutonium can be spiked with depleted uranium obtained during the first cycle [5]. The importance of using depleted uranium during reprocessing lies in the high availability of this resource due to previous mining activities, which can be used by other generations. This reduces the need for new mining operations, reducing greenhouse gas emissions.

1.2 Non-proliferation

Since 1970, the Treaty on the Non-Proliferation of Nuclear Weapons has been in force, which establishes that countries that did not test nuclear weapons until 1967 cannot build or obtain nuclear weapons, and the five countries that had already tested (China, the United States, France, the United Kingdom, and Russia) must reduce their arsenal [6]. Brazil signed the agreement in 1998, but the 1988 constitution guaranteed that the country would use nuclear science only for peaceful purposes [7].

With reprocessing, it is possible to reinforce this commitment to the Treaty to ensure that weapons of mass destruction are not proliferated. For example, methods such as GANEX and UREX+ do not extract pure plutonium but with minor actinides, making it impossible for this material to be used for military purposes [5].

1.3 Waste Management

Waste from nuclear reactors is usually inserted into pools to cool during decay and provide radiation protection. They are then stored in dry storage systems that use sealed and shielded containers, where they remain until a final long-term warehouse is created. The issue of waste faces criticism from public opinion and must be done correctly to prevent accidents.

However, with reprocessing, especially in a cycle that involves the recycling of actinides (plutonium, neptunium, americium, and curium), it is possible to reduce the volume of waste and also its radiotoxicity, ensuring more safety. Some reprocessing methods reduce the amount of long-lived fission products in the final waste, decreasing radioactivity and toxicity [5].

Public acceptance could also increase, as it would improve safety, limit the risks of nuclear proliferation, and reduce the half-life of radioactive waste [4].

1.4 Reprocessing as an alternative for the spent fuel

Comparisons between nuclear fuel cycles reveal that fuel reprocessing, such as in the MOX, TRU-U, and TRU-Th cycles, can 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. When compared to the traditional cycle, these alternatives prove to be promising in terms of sustainability and economics [1, 2].

At the beginning of the cycle, the uranium mined results in depleted uranium (tailings) and enriched products. Spiking this depleted uranium with the reprocessed material to fabricate MOX is an effective approach that can reduce the need for mining. Depleted uranium is only stored; with this way of reprocessing, it can have a more sustainable cycle without the need to increase costs.

Not separating plutonium from the minor actinides (MA) is a non-proliferation strategy as it makes the material significantly more challenging to handle and process for weapon creation and significantly reduces the long-term radiotoxicity of nuclear waste.

Considering the points discussed in the previous paragraphs, the flowchart in Figure 1 can represent the desired reprocessing strategy.

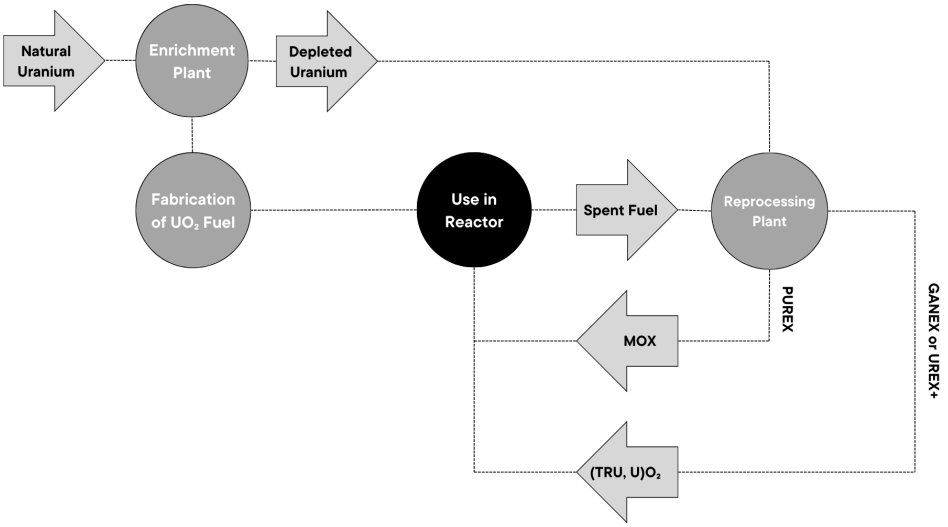

Fig. 1. Alternatives for reprocessing.

1.5 PUREX

The PUREX (Plutonium Uranium Recovery by Extraction) process is a reprocessing method that uses Tri-n-butyl Phosphate (TBP) solvent, liquid-liquid extraction, and oxidation-reduction reactions. The fuel is chopped and then dissolved in nitric acid. Plutonium and uranium are separated from the fission products and minor actinides with solvent extraction [5].

1.6 GANEX

GANEX (Grouped Actinide EXtraction) begins with dissolved spent nuclear fuel in nitric acid. The solution is submitted to a solvent extraction system to remove the uranium. Then, a mixture of complexing agents and extractants co-extract plutonium and the minor actinides from the remaining fission products [8].

1.7 UREX+

UREX+ (Uranium Extraction Plus) is an advanced nuclear reprocessing method that starts with dissolved spent nuclear fuel in nitric acid. It involves five steps. The first step uses a solvent extraction system to extract uranium and technetium from the solution selectively. Subsequent steps involve using specialized chemical agents and extractants to achieve the selective separation of plutonium and minor actinides (americium, curium and neptunium) from fission products that can be used to produce the new fuel [8].

2. METHODOLOGY

To ensure that reprocessing methods contribute to reducing mining, preventing weapons proliferation, and managing waste, it is essential that reprocessed fuel is spiked with depleted uranium, contains an amount of fissile material close to that of fresh fuel and that the minor actinides are not separated. These requirements are necessary because using the depleted uranium stored is not necessarily a new step of mining; an equal amount of fissile material would maintain the reactor's efficiency, and plutonium, together with minor actinides, prevent their use in weapons production.

To demonstrate the amount of fissile material in reprocessed fuel, spent fuel from a reactor with a burnup of 50,000 MWd/t and an initial enrichment of 4.5% is considered.

The recovery factors for the three methods analyzed are listed in Table 1. This factor will be used in the software developed by Aruquipa et al. (2017) to give the quantity of fissile material after the reprocessing and spiking [8].

Tab. 1. Recovery factors for the GANEX, UREX+ and PUREX reprocessing methods.

Element	**GANEX**	**UREX+**	**PUREX**
U	0.935	0.995	0.999
Np	0.938	0.710	0.950
Pu	0.996	0.995	0.998
Am	0.998	0.980	-
Cm	0.998	0.790	-

The composition of the material processed for the different recovery processes is listed in Table 2.

Tab. 2. Percentage composition of the material processed.

Isotope	**GANEX**	**UREX+**	**PUREX**
Np-237	5.467	3.957	5.328
Pu-238	2.576	2.618	2.643
Pu-239	50.744	51.580	52.064
Pu-240	20.240	20.573	20.767
Pu-241	13.388	13.608	13.736
Pu-242	5.323	5.411	5.462
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	-

This composition is just a way to analyze the quantity of fissile material after reprocessing and it can justify the need of spiking. According to Aruquipa et al. (2017), the percentage of fissile isotopes in GANEX, UREX+ and PUREX are 64.132%, 65.188%, and 65.800%, respectively [8]. These values are higher than the 4.5% necessary for the fuel, so spiked with thorium or depleted uranium is necessary. For saving mining, a spike with depleted uranium is a better option once this was already mined at the beginning of the cycle. With thorium, one more mining step would be necessary.

With the following equations is possible normalize the values of the new mixture to a percentage of fissile material [8]:

$$NF_R = \frac{f - 4.5\%}{f - f_R} \qquad (1)$$

$$NF = \frac{4.5\% - f_R}{f - f_R} \qquad (2)$$

Equation 1 is the normalization factor for the reprocessed material, where f is the percentage of fissile material present in depleted uranium (0.2%) and f_R is the percentage of fissile material present in the reprocessed material.

Equation 2 is the normalization factor for the depleted uranium and follows the same values of f and f_R.

With these factors, it is possible to analyze the quantity of each material in the final reprocessed fuel and, compared to fresh fuel, verify if the amount of depleted uranium left at the beginning of the cycle is sufficient.

Using a MATLAB code from Aruquipa et al. (2017), the percentage of fissile material after spiking is given, which makes it possible to compare the methods.

3. RESULTS

3.1 Comparison of the methods in terms of fissile material

The values of fissile material, plutonium, and minor actinides after spike with depleted uranium are shown in Table 3. U-233 is fissile but presents a low percentage. Other isotopes are present in the mixture, but only these are listed:

Tab. 3. Percentage of composition of fissile material, plutonium, and minor actinides for the reprocessed fuels spiked with depleted uranium.

Isotope	**GANEX**	**UREX+**	**PUREX**
U-235	2.097	2.164	2.206
Np-237	0.870	0.138	0.187
Pu-238	0.093	0.090	0.092
Pu-239	1.830	1.777	1.813
Pu-240	0.729	0.707	0.722
Pu-241	0.480	0.466	0.475
Pu-242	0.190	0.184	0.188
Am-241	0.022	0.021	-

Am-243	0.056	0.053	-

Some isotopes of curium and americium are also present. However, they were excluded from the table due to their low percentage in the composition (<0.0001%).
Considering the fissile isotopes U-235, Pu-239, and Pu-241, the spiking resulted in values close to 4.5%. Thus, using the UREX+ and GANEX methods is advantageous because they prevent nuclear weapons proliferation [9] and assist in waste management by processing minor actinides [10]. Moreover, these reprocessing methods do not significantly differ from PUREX's final fissile material values, excluding these minor actinides. Previous studies have evaluated the neutronic parameters of inserting reprocessed fuels, demonstrating the possibility of their inclusion in the core [9-11]. This work complements the earlier studies by highlighting the feasibility associated with the environmental impact, as reprocessing would reduce the two most environmentally critical stages of the nuclear fuel cycle: mining and final waste disposal.

3.2 Calculation of reprocessed material and depleted uranium

Using equations 1 and 2 and the sum of fissile material in Table 2, how to normalize the quantity of reprocessed material and depleted uranium is given as following:

$$NF_R(GANEX) = \frac{0.2-4.5}{0.2-64.132} = 0.067 \qquad (3)$$

$$NF(GANEX) = \frac{4.5-64.132}{0.2-64.132} = 0.934 \qquad (4)$$

$$NF_R(UREX\ +) = \frac{0.2-4.5}{0.2-65.188} = 0.066 \qquad (5)$$

$$NF(UREX\ +) = \frac{4.5-65.188}{0.2-65.188} = 0.934 \qquad (6)$$

$$NF_R(PUREX) = \frac{0.2-4.5}{0.2-65.800} = 0.066 \qquad (7)$$

$$NF(PUREX) = \frac{4.5-65.800}{0.2-65.800} = 0.934 \qquad (8)$$

The values found in equations 3, 5, and 7 and those found in equations 4, 6, and 8 are close. It happens because the percentage of fissile material was close. Therefore, the values 0.066 and 0.934 can be used to calculate the quantity of reprocessed material and depleted uranium for the three methods.

The fuel must have 4.5% enrichment. Considering the values for a burnup of 50,000 MWd/t, the following values are given for the quantities in the cycle [14]:

- Feed material (natural uranium): 306,191 MTU/year
- Fresh fuel: 29,864 MTIHM/year
- Spent fuel: 29,864 MTIHM/year

Considering the heavy metal in fresh fuel is only uranium, the tailings (depleted uranium) can be considered to be approximately 276,327, because:

$$Feed\ material\ -\ Fresh\ fuel = Tailings = 276,327 \qquad (9)$$

If the reactor needs 29,864 MTIHM/year of fresh fuel for burnup of 50,000 MWd/t, the reprocessed fuel needs the same quantity and should be constituted by 6.6% (based on the solutions of equation 1) of reprocessed material and 93.4% (based in the solutions of equation 2) of spiked depleted uranium. Consequently, a 4.5% enriched fuel derived from reprocessing using the GANEX, UREX+, or PUREX methods requires approximately 1,971.024 MTIHM/year of reprocessed material and 27,892.976 MTU/year of depleted uranium. It is showing as following:

$$Reprocessed\ material = Fresh\ fuel * NF_R = 29,864 * 0.066 = 1,971.024 \quad (10)$$

$$Depleted\ uranium = Fresh\ fuel * NF = 29,864 * 0.934 = 27,892.976 \quad (11)$$

The value of depleted uranium is lower than tailings in the first cycle. So, using the reprocessing methods, the need of mining decreases. Only a part of spent fuel and depleted uranium need to be used in reprocessing for one year. So, with one first cycle with fresh fuel, it is possible to create more than one reprocessed fuel for other life cycles of the reactor.

4. CONCLUSION

The reprocessing methods have demonstrated their significant potential in addressing key challenges in the nuclear fuel cycle. Reducing the need for uranium mining and enhancing waste management can solve the most problems with environmental impact in the nuclear fuel cycle.

The methods GANEX and UREX+ showed approximately the same value of fissile material after depleted uranium spinking as PUREX (4.5%). Because GANEX and UREX+ spike together the minor actinides, these methods are more advantageous because they are non-proliferative, different from PUREX, which reprocessed only uranium and plutonium. This characteristic offers an additional advantage, which enhances waste management strategies, reducing the volume and radiotoxicity of nuclear waste

Establishing the proportions of reprocessed material and depleted uranium required, it is possible to observe that the uranium stored at the beginning of the cycle can be used, by the way, more than one time. Then, it reduces the demand for fresh uranium mining and utilizing existing nuclear material; these methods contribute to a more sustainable nuclear energy process.

In conclusion, adopting UREX+ and GANEX in reprocessing spent nuclear fuel presents a viable pathway toward a more secure, sustainable, and responsible nuclear fuel cycle. Their ability to effectively recycle fissile material contributes to the mining reduction, non-proliferation commitment, and better waste management.

ACKNOWLEDGMENTS

The authors are grateful to the agencies Comissão Brasileira de Energia Nuclear (CNEN), Conselho Brasileiro de Desenvolvimento Científico e Tecnológico (CNPq), Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) and Fundação de Apoio à Pesquisa do Estado de Minas Gerais (FAPEMIG) for the financial support. We are also grateful to Wilmer Aruquipa for providing the software and to colleagues Karytha Corrêa and Natália Oliveira for their time answering questions and helping with the software. Grant Project Numbers:

420852/2023-9, 443214/2023-9, 317537/2021-0, 402610/2022-9, 406822/2022-0 – CNPq; APQ-02410-21 - FAPEMIG

BIBLIOGRAPHY

[1] F. B. G. L. Estanislau; C. E. Velasquez; A. L. Costa; C. Pereira. Assessment of alternative nuclear fuel cycles for the Brazilian nuclear energy system. Nuclear Engineering and Design, v. 415, p. 112692, 2023. https://doi.org/10.1016/j.nucengdes.2023.112692.

[2] K. Dungan; R. W. H. Gregg; K. Morris; F. R. Livens; G. Butler. Assessment of the disposability of radioactive waste inventories for a range of nuclear fuel cycles: Inventory and evolution over time. Energy, v. 221, p. 119826, 2021. https://doi.org/10.1016/j.nucengdes.2024.113259.

[3] O. Agboola; D. E. Babatunde; O. S. I. Fayomi; E. R. Sadiku; P. Popoola; L. Moropeng; A. Yahaya; O. A. Mamudu. A review on the impact of mining operation: Monitoring, assessment and management. Results in Engineering, v. 8, p. 100181, 2020. https://doi.org/10.1016/j.rineng.2020.100181.

[4] C. Poinssot; B. Bouullis; S. Bourg. Role of recycling in advanced nuclear fuel cycles. In: Reprocessing and recycling of spent nuclear fuel. Woodhead Publishing, 2015. p. 27-48. https://doi.org/10.1016/B978-1-78242-212-9.00002-2.

[5] R. G. Cochran; N. Tsoulfanidis. The nuclear fuel cycle: analysis and management. (No Title), 1999.

[6] UNITED NATIONS. Treaty on the Non-Proliferation of Nuclear Weapons (NPT). Disponível em: https://treaties.unoda.org/t/npt acessado em 28/05/2024.

[7] BRASIL. Ministério das Relações Exteriores. Desarmamento e Não-Proliferação. Disponível em: https://www.gov.br/mre/pt-br/delbrasonu/paz-e-seguranca-internacional/desarmamento-e-nao-proliferacao acessado em 28/05/2024.

[8] W. Aruquipa; C. E. Velasquez; G. P. Barros; C. Pereira; M. A. F. Veloso; A. L. Costa. Reprocessing techniques of LWR spent fuel for reutilization in hybrid systems and IV generation reactors. 2017.

[9] C. Pereira; E. M. Leite. Non-Proliferating Reprocessed Nuclear Fuels In Pressurized Water Reactors: Fuel Cycle Options. Annals of Nuclear Energy, Grã-Bretanha, v. 25, n.12, p. 937-962, 1998. https://doi.org/10.1016/S0306-4549(98)00008-5.

[10] C. Pereira; E. M. Leite; E. F. Faria. Waste analysis generated by alternative reprocessing fuels from pressurized water reactions. Annals of Nuclear Energy, v. 27, p. 449-464, 2000. https://doi.org/10.1016/S0306-4549(99)00102-4.

[11] A. L. Costa; C. Pereira. A Neutronic Evaluation of the Americium and Neptunium Co-insertion in UO2 Fuel and PUREX Reprocessed Fuel. Annals of Nuclear Energy, Grã-Bretanha, v. 30, n.7, p. 775-783, 2003. https://doi.org/10.1016/S0306-4549(02)00129-9.

[12] A. L. Costa; C. Pereira. An Evaluation of the Am Insertion in the UO2 Fuel. Annals of Nuclear Energy, Gra-Bretanha, v. 29, n.6, p. 767-775, 2002. https://doi.org/10.1016/S0306-4549(01)00072-X.

[13] A. L. Costa; C. Pereira; M. A. F. Veloso; C. A. M. Silva. A neutronic evaluation of the (Pu-U) and (Am-Pu-U) insertion in a typical fuel of Angra-I. Annals of Nuclear Energy, v. 36, p. 1-6, 2009. https://doi.org/10.1016/j.anucene.2008.11.006.

[14] E. Moniz, J. Deutch; S. Ansolabehere; M. Driscoll; H. P. Gray; N. E. Todreas. The future of nuclear power: an interdisciplinary MIT study. Boston, MA: MIT Press, 2003. ISBN 0-615-12420-8.