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PARTITIONING AND TRANSMUTATION OF USED NUCLEAR FUEL IN SUPPORT OF GEOLOGICAL WASTE DISPOSAL

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Idaho National Laboratory

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PARTITIONING AND TRANSMUTATION OF USED NUCLEAR FUEL IN SUPPORT OF GEOLOGICAL WASTE DISPOSAL

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Abstract

Nuclear energy is a low-carbon technology that generates bulk baseload electricity and supports long-lasting eco-friendly sustainability goals. However, it still faces challenges, such as the securing and managing of long-lived nuclear actinides and fission products—most notably the used fuel, which is considered high-level waste (HLW). Direct geological waste disposal would be the most economical option for HLW but requires highly qualified sites whose geophysical behavior will remain stable for 10,000 years. Partitioning and transmutation (P&T) of used fuel reduces radioactivity and decay heat generation, enabling efficient geological waste disposal. P&T research and development, along with the prospects of P&T techniques such as wet/dry separation and accelerator-driven systems, were reviewed to identify critical needs that must be met to foster their successful implementation.

1. Introduction

Global warming and climate change have compelled the international community to focus on reducing or even eliminating carbon emissions from power generation technologies. Ever since 1954, nuclear power reactors have played an instrumental role in providing reliable, environmentally sustainable, and economically viable energy. The nuclear power industry, despite affording one of the lowest-carbon options for bulk baseload electricity generation and only minimally contributing to global warming, faces challenges in regard to safe, economical management of spent nuclear fuel (SNF). SNF management and treatment involves processes to safely store, transport, reprocess, and ultimately dispose of nuclear materials. Initially, SNF management entails storing the SNF in either wet (cooling pools) or dry (air-cooled containers) facilities. Once sufficient radioactive decay is achieved, the SNF can be transported to reprocessing or disposal facilities. Reprocessing separates valuable fissile materials (i.e., U and Pu) and produces long-lived high-level waste (HLW).

Direct geological disposal of HLW is considered one of the most economical alternatives but requires highly qualified sites whose geological behavior will remain stable for over 10,000 years. This long-term radiological burden must be overcome via a feasible solution. The International Atomic Energy Agency (IAEA) estimates that, since the advent of civil nuclear power production, around 370,000 metric tons of heavy metal (MTHM) of spent fuel has been produced, of which 120,000 MTHM has been reprocessed [1]. No country in the world has yet secured a solution for HLW. The nuclear waste inventory, as estimated by IAEA in 2022, reveals that approximately

29,000 cubic meters (m^3) of HLW is currently being stored, with none having ever been disposed of to date [1].

By the end of 2019, around 86,000 MTHM of SNF were being stored onsite at 75 operating or decommissioned commercial nuclear power plants (NPPs) across 33 states. Figure 1 provides a visual representation of this distribution. The majority of the cumulative SNF, totaling more than 17,500 MTHM of uranium mass, is stored in two states: Illinois and Pennsylvania [2]. Close behind, New York, North Carolina, and South Carolina have substantial volumes of stored SNF, each exceeding 4,000 MTHM of uranium. Projections indicate that approximately 2,000 MTHM of commercial SNF will accumulate annually over the next 10 years. Beyond that period, the rate of SNF accumulation is expected to decrease gradually as more reactors begin shutting down in the 2030s. As a result, it is estimated that a total of 140 MTHM of SNF will accumulate over the remaining lifespan of the existing NPPs.

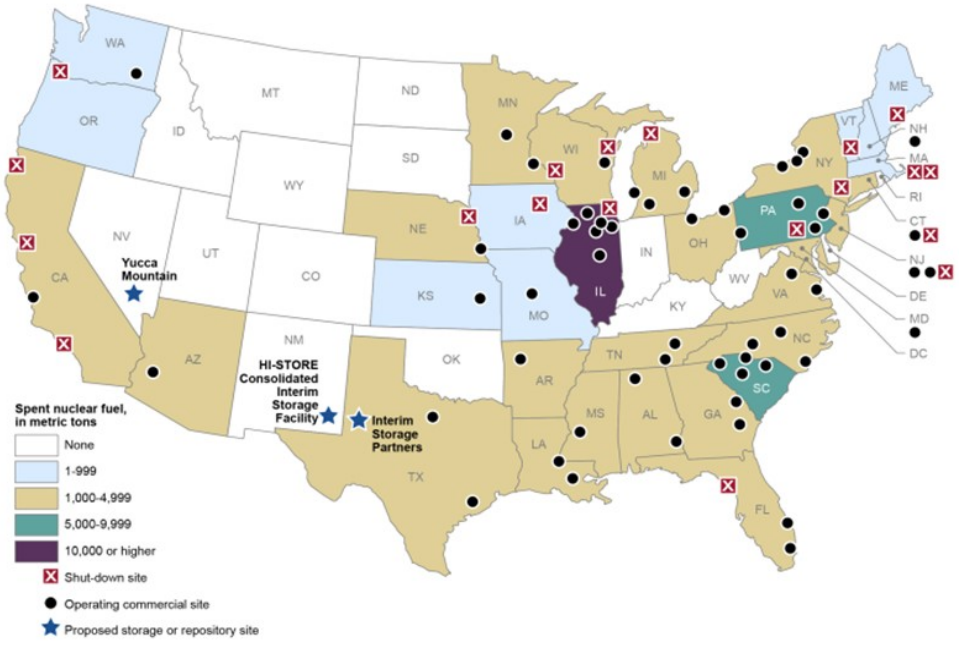

Figure 1 Stored commercial SNF amounts (through 2019) and locations (as of June 2021) [2].

Partitioning and transmutation (P&T) is a promising strategy for treating SNF HLW so as to decrease its waste volume and toxicity. It involves separating the radioactive elements in spent fuel, including fission products and actinides, via chemical procedures or sophisticated techniques such as pyroprocessing or electrorefining. Transmutation alters the attributes of the waste by turning long-lived radioactive elements into shorter-lived or non-radioactive elements via nuclear reactions in fast neutron reactors or accelerator-driven subsystems (ADS). Fast reactors (FRs) and ADS could be used to transmute HLW materials, primarily actinides and fission products.

This study focuses on SNF management—particularly in regard to P&T, even exploring its utilization in diverse reactor systems and its prospects for use in R&D. We discuss global SNF reprocessing trends, decision making for SNF management, and the influence of material composition on P&T, FRs, and ADS for SNF/HLW transmutation. The paper concludes with recommendations for optimal aggregate selection.

2. SNF Basics

The material composition of SNF after fuel burnup depends on the initial fuel enrichment, fuel and reactor type, and operating conditions. The post-burnup material composition of light water reactor (LWR) and CANDU reactor SNF is shown in Figure 2.

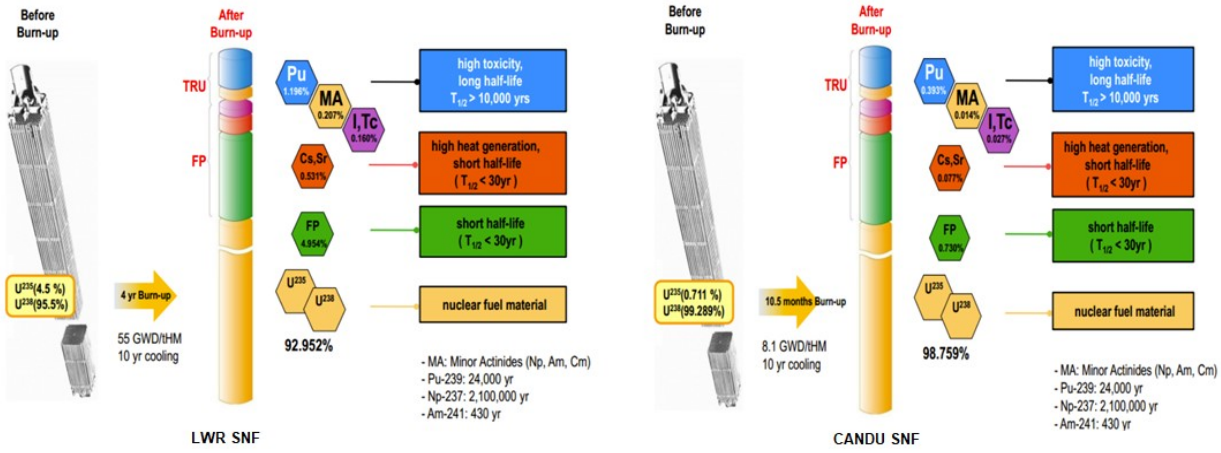

Figure 2 Post-burnup material compositions of SNF.

However, multiple recycling of plutonium and minor actinides can reduce the amount of time necessary to return radioactivity amounts back to their reference levels. Previous studies [3] demonstrated that P&T processes can, with 99% efficiency, recycle plutonium and minor actinides so as to reach the reference levels in approximately 500 years. These results indicate that P&T is useful for reducing the amount of time required for SNF to reach the reference levels, and that it significantly reduces the time needed to decrease it to the radioactivity level of natural uranium ore. Figure 3 compares SNF radiotoxicity levels vs. time when employing either a direct disposal, recycling, or P&T strategy [4].

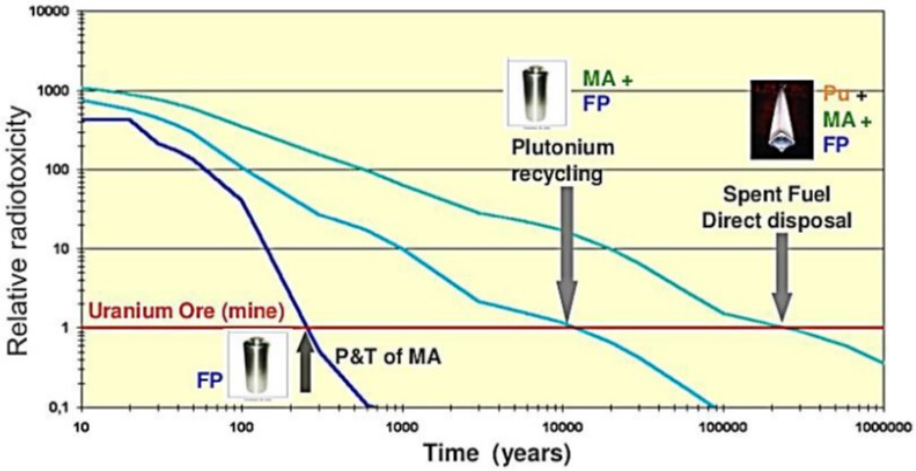

Figure 3 Relative radiotoxicity levels vs. time when SNF is managed using either a direct disposal, recycling, or P&T strategy [4].

For SNF comprised of different materials, Figure 4 shows the variation in radiotoxicity over time. As shown in the figure, the fission products determine the SNF's radiotoxicity during the first 100 years after discharge. Once the plutonium is removed, the minor actinides determine the long-term radiotoxicity. It is important to note that both scales are logarithmic [5].

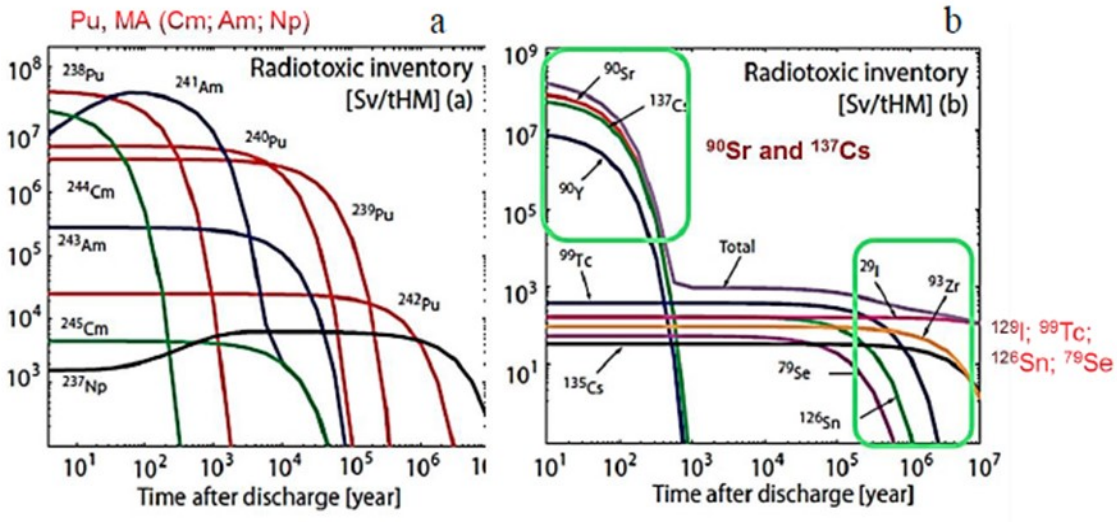

Figure 4 Relative radiotoxicity of different components of SNF, varying over time [5].

The radiological properties of major and minor actinides, as well as their impact on storage facilities and heat generation, further increase the importance of developing an effective treatment system for nuclear waste. Table 1 shows the transmutation needs and isotope separation requirements [5].

Based on Table 1, it is possible to conclude that the isotopes that primarily impact radiotoxicity are ^{79}Sr, ^{135}Cs, and ^{151}Sm; the isotopes with the biggest repository impact are ^{126}Sn, ^{79}Se, ^{135}Cs, 241Am, and ^{237}Np; and the heat generation impact are ^{238}Pu, ^{244}Cm, and ^{241}Am [19].

Table 1: Transmutation needs and isotope separation requirements of transmutable long-lived fission products [5].

Isotope	Transmutation Needs				Isotope Separation Requirement
	Toxicity	Half-life	Repository Impact	Inventory	
^{94}Nb	High	High	Low	Very low	Weak
^{99}Tc	Medium	High	High	High	No
^{107}Pd	Low	High	Low	Medium	Strong
^{129}I	Medium	High	Very high	Medium	Weak
^{135}Cs	Medium	High	Medium	Medium	Strong
^{151}Sm	High	Low	Low	Low	Weak

3. Partitioning of SNF

Partitioning methods aid in managing used nuclear fuel by separating the different constituents for treatment, disposal, and recycling. These methods, which include pyroprocessing, solvent extraction, and acid dissolution, are still in the research phase, due to technical and economic challenges. Pyroprocessing uses high heat and electrochemical processes to separate elements, whereas solvent extraction leverages liquid solvents after the fuel is subjected to acid dissolution. Acid dissolution involves dissolving spent fuel in acid, then separating it chemically.

The separation technique (i.e., partitioning of the radioactive waste) plays a significant role in effective nuclear waste management. The primary goal of partitioning is to separate Pu and minor actinides Np, Am, and Cm from the used nuclear fuel. Dry and wet partitioning are the two most common partitioning methods. Wet partitioning, mainly known as Plutonium Uranium Redox Extraction (PUREX), was established in the early 1940s [4-6]. The PUREX process entails the following steps: fuel dissolution, chemical treatment and separation, off-gas treatment, and waste verification [6]. PUREX is an aqueous chemical method that affords very high separation efficiency (~99.9%) and can be used to process nuclear waste in bulk [6]. The dry partitioning process, primarily known as the pyroprocess, was first developed at Argonne National Laboratory in the 1980s and has only grown in popularity from then until now. The pyroprocess (see Figure 5) entails three steps: oxide reduction of light-water reactor (LWR) fuel and transuranic recovery using electrorefining, actinide removal using an ion exchanger and fuel, and solid waste fabrication.

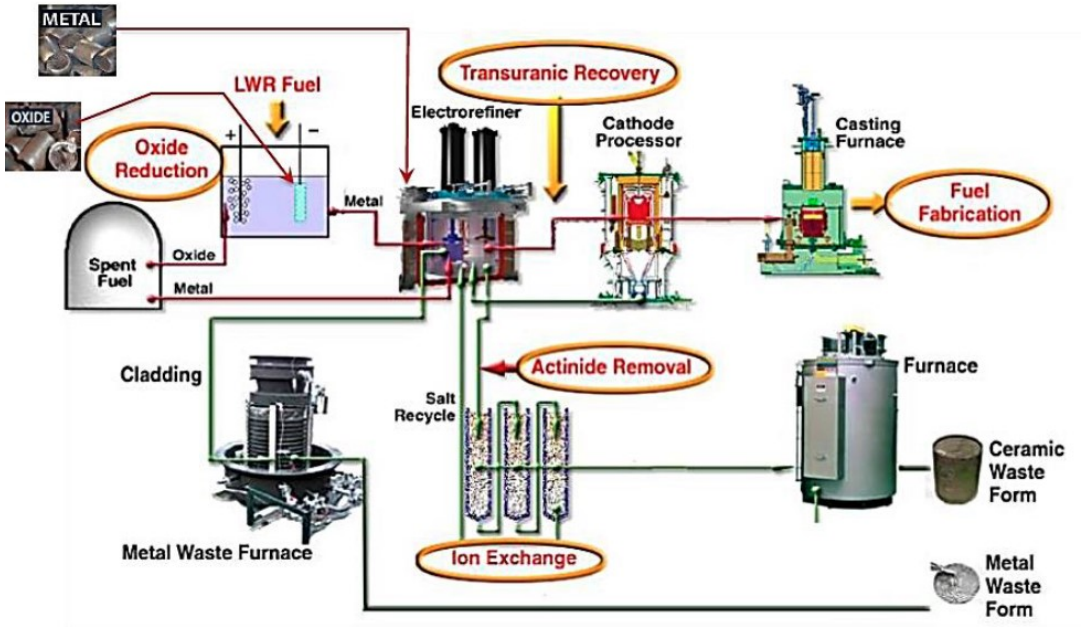

Figure 5 Schematic diagram of the conceptual design for the advanced pyroprocess [6].

This electrorefining process recycles used nuclear fuel [7] and is advantageous in terms of the resulting waste volume, compactness, and proliferation resistance. Wet (or aqueous) processes have several advantages as well, including low operating temperatures, continuous processes, and well-established industrial practices; however, these advantages are outweighed by the fact that the processes produce so much liquid waste. Table 2 compares the pyroprocess to the aqueous process.

Table 2: Aqueous vs. pyrochemical process for waste partitioning [6].

	Aqueous Method	Pyrochemical Method
Solution	Aqueous	Ionic liquid
Operating temperature	Low	High
Process	Continuous	Batch
Product purity	High	Low
Waste type	LLW	HLW
Waste volume	Large	Small
Proliferation resistance	No, but safeguardable	Yes
Compactness	No	Yes
Scale-up	Commercial	Engineering (PRIDE, ANL, INL)

4. Transmutation of SNF

Transmutation is a SNF treatment method that entails converting the fuel's long-lived radioactive elements into shorter-lived or non-radioactive elements by using different FRs and ADS. Transmutation aims to reduce the volume and toxicity of the waste, as well as the length of time that the waste will remain radioactive. Several transmutation methods have been proposed and studied, but most are still at the R&D stage and not yet commercialized. Two potential methods involve FRs and ADS. FRs use fast neutrons to transmute the long-lived radioactive elements in SNF. FRs can burn both actinides and fission products, significantly reducing the volume of the waste. ADS use a proton accelerator to generate high-energy protons so as to transmit the long-lived radioactive elements in SNF. The protons collide with a target, producing neutrons for transmitting the waste.

The level of success of transmutation varies from one reactor system to another. Table 3 compares fractional transmutation performance—in terms of radiotoxicity, decay heat generation, neutron emission, and Np concentration—for the three major FR systems (i.e., molten salt fast reactor [MSR], sodium-cooled fast reactor [SFR], and lead-cooled fast reactor [LFR]). The results prove MSRs to be better than SFRs and LFRs in regard to radiotoxicity, decay heat generation, and neutron emission. Of all the parameters, radiotoxicity and decay heat generation represent the main challenges, as they more greatly impact the environment and repository performance. The radiotoxicity of the treated waste should be as low as reasonably possible. Results show that the PWR waste had the highest radiotoxicity and decay heat generation. The open fuel cycle had higher radiotoxicity over time than the simple closed cycle with Pu recycling. The double strata approach shows lower radiotoxicity compared to the open fuel cycle.

Table 3: Fractional transmutation performance in different FR types [8].

	MSR	SFR	LFR
FT	99.85%	99.78%	99.71%
Radiotoxicity	1	1.19	1.53
Decay heat	1	1.11	1.46
Neutron emission	1	0.043	0.039
Np and precursors	1	1.19	1.5

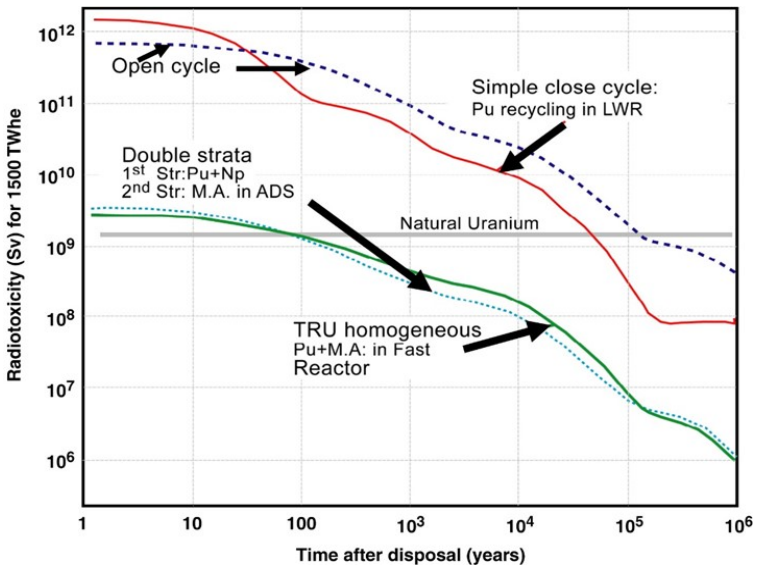

Figure 6 Radiotoxicity reductions achieved via different strategies [3].

Studies show ADS to be a promising option for the transmutation of nuclear waste. ADS is a subcritical reactor system that uses thorium as fuel. A high-energy proton beam from an external proton accelerator makes the reactor core critical, making ADS inherently safe and unique compared to FRs ADS also allow for adjusting the reactivity margin by controlling the energy beam from the external accelerator, ensuring more efficient burning of SNF. Figure 7 compares the subcritical reactor-ADS to a critical reactor.

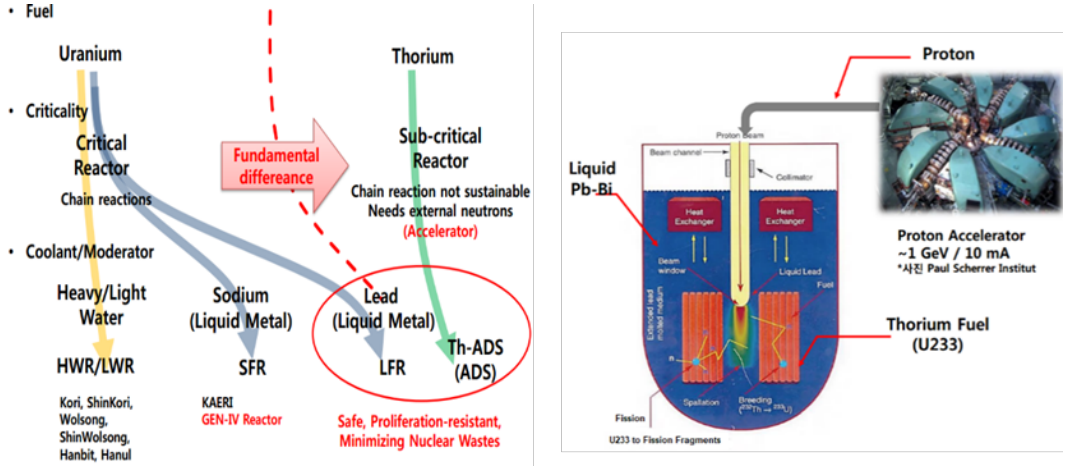

Figure 7 Accelerator driven reactor system [9].

As with other FR systems, ADS burns the minor actinides (MA) and Pu, whereas thermal reactor systems produce them. The Pu and MA generation rate in different reactor systems is presented in Figure 8. Compared to the conventional first reactor system, the burnup of the MA when using ADS is much higher.

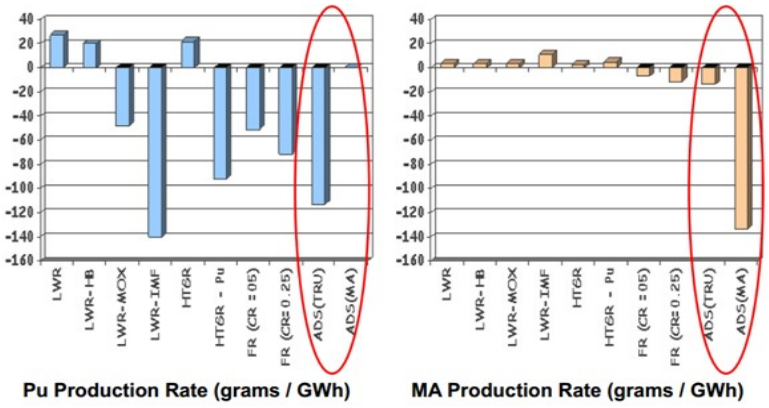

Figure 8 Transmutation performance comparison between fission reactors and ADS [9].

5. Potential for Repository Drift Loading Increase

Separation of Pu, Am, Cs, Sr, and Cm will allow for denser loading of treated waste in the repository, as presented in the Figure 9 below.

- Any system that requires enriched fuel will have a limited uranium utilization of <1% to the fuel burnup limit (~20% for FR fuels)
- Full recycling options can approach full utilization (i.e., >90%), depending on recycle losses
- Radiotoxicity reflects the hazard level of the source materials: TRU dominates after about 100 years; however, fission products dominates in radiotoxicity after 100 years
- Radiotoxicity is not the only parameter in determining how a geologic repository may perform. Engineered and natural barriers isolate waste and/or control radionuclide release
- Cs/Sr (and decay products), Cm, and Pu dominate "early" decay heat
- Am dominates "later" decay heat
- Removal of decay heat producers would enable increased utilization of repository space

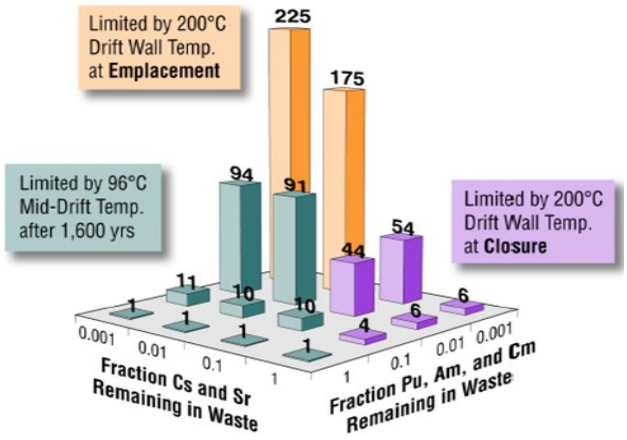

Figure 9 SNF final repository/disposal site performances [10].

- Separation of Pu and Am allows for denser loading of the repository
    - up to a factor of 6 with 99.9% removal
- Subsequent separation of Cs and Sr provides a much greater benefit
    - up to a factor of 50 with 99.9% removal
- Removal of Cm further increases the potential benefit (with Pu and Am)
    - greater than a factor of 100 with 99.9% removal
- Appropriate waste forms are needed to take advantage of this potential.

6. Findings and Discussion

A review of the relevant literature, technical reports, and cutting-edge research was performed to assess state-of-the-art P&T technologies for reducing the effects of HLW prior to geological disposal. It was found that the separation and recycling of Pu are already common practices in the industry. Partitioning of MA has also been demonstrated in the lab and on a pilot scale. Therefore, partitioning Pu and MA is recommended in advanced pyroprocessing so as to reduce the amount of waste produced during wet separation. Partitioned HLW should be transmuted, and partitioning with an ADS system is preferred over partitioning with an FR system. Introducing advanced FRs with in-built transmutation capabilities could reduce HLW inventories.

Furthermore, P&T can afford significant benefits in the management of nuclear waste. It can reduce the long-term radiotoxicity of waste by up to a hundred-fold, making it less hazardous and reducing potential issues such as human intrusion and proliferation interest. Radiotoxicity can reach the level of the original uranium ore in less than 1000 years, as compared to the 100,000 years when P&T is not applied. Waste from P&T processes generates less heat than does typical spent fuel—something that is crucial for geological storage, where thermal limitations determine the disposal density. This reduces the repository's thermal load, potentially delaying the disposal time by 100–200 years. Studies show that employing P&T strategies can significantly reduce the amount of area required for the repository. For example, transmuting MA and partitioning fission products can reduce the repository size by a factor of 4 to 5—even more with extended storage of certain isotopes. P&T lowers the inventory of hazardous radioactive materials, potentially reducing uncertainties about repository safety, especially in scenarios involving disturbed conditions. P&T, as part of future Generation IV reactor operations, can help minimize the amount of produced waste per electricity generated, and lower radiotoxic inventory lifetimes, potentially improving the public perception of nuclear energy. However, implementing P&T would require the operation of reprocessing plants, and represents a significant step forward in nuclear energy technology.

7. Conclusion

P&T potentially mitigates nuclear waste issues by decreasing waste volumes and radioactivity durations and diminishing the environmental impact of nuclear waste by reducing the amount of long-lived radioactive waste needing to be stored. However, P&T is a complicated procedure that demands significant R&D, supportive regulations, and advanced technologies and facilities that are yet to be fully developed or commercialized. Consequently, while P&T is a prospective solution for nuclear waste management, its application is currently limited in practice.

Safety, economy, and environmental justice highly influence the fate of nuclear energy. The waste management issue is vital and challenging, as it is closely related to these three issues—all of which can be addressed via P&T. In this regard, P&T represents a valuable solution for reducing nuclear waste, and would aid in making nuclear power deployment safe, clean, and economically feasible.

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