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

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# Objectives and Motivations

- 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
  - 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,
  - enables efficient geological waste disposal.
- Therefore, research and development (R&D) are needed to foster successful implementation of P&T technologies.

# Objectives and Motivations (cont'd)

- P&T is a promising strategy for treating SNF HLW
  - so as to decrease its waste volume and toxicity.
- Partitioning involves separating the radioactive elements
  - in spent fuel, including fission products and actinides,
  - via chemical procedures or sophisticated techniques
    - e.g., 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), and fast reactors (FRs).

# Objectives and Motivations (cont'd)

- 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].
- The shut-down and operational nuclear power plants (NPPs)
  - at 35 state stored approximately 86,000 metric tons (MT) of commercial spent nuclear fuel (SNF) onsite
  - proposed sites for final disposal or interim storage
  - quantity of SNF is increasing at an annual rate of about 2,000 MT [2]

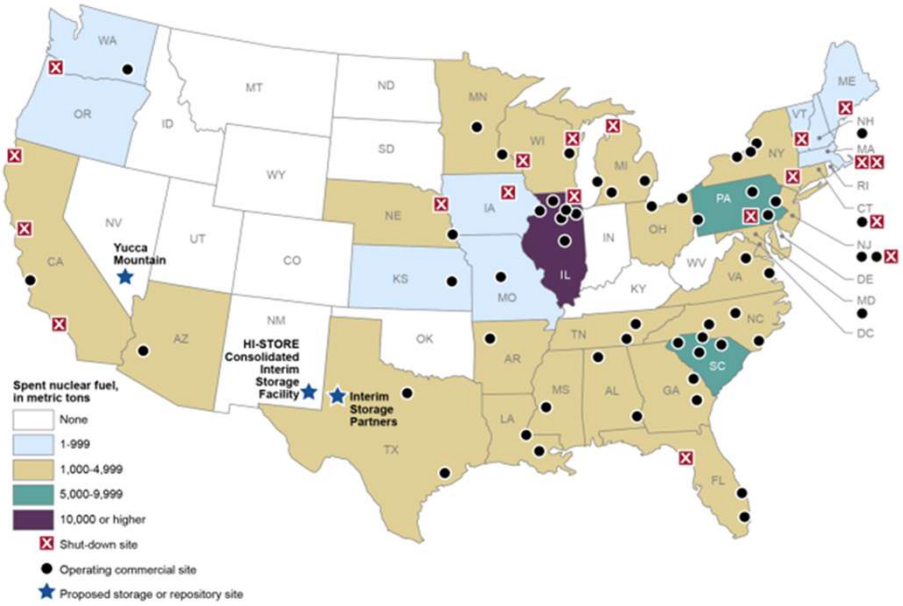


Fig. 1: Stored commercial SNF amounts (through 2019) and locations (as of June 2021) [2]

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# SNF: post-burnup material compositions

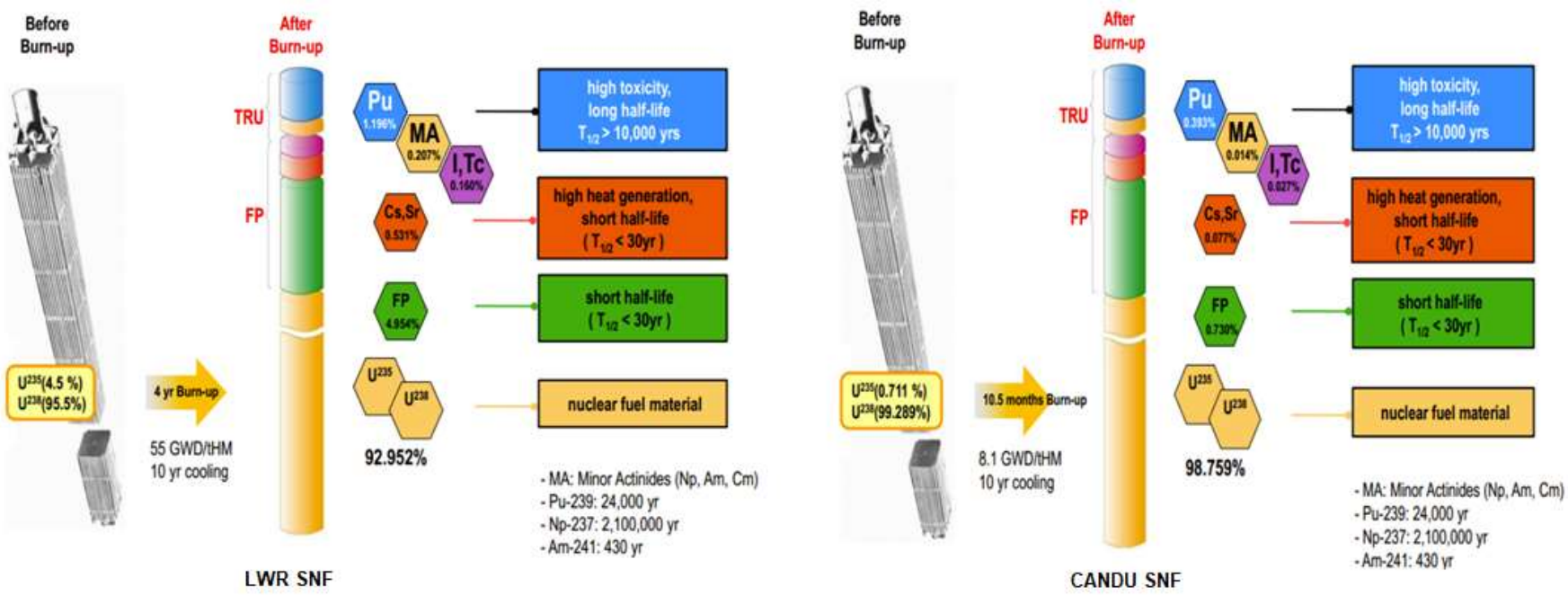


Fig. 2: Post-burnup material compositions of SNF [3]

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# SNF: management challenges

- Multiple recycling of plutonium (Pu) and minor actinides (MA) 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 Pu and MA so as to reach the reference levels in approximately 500 years.

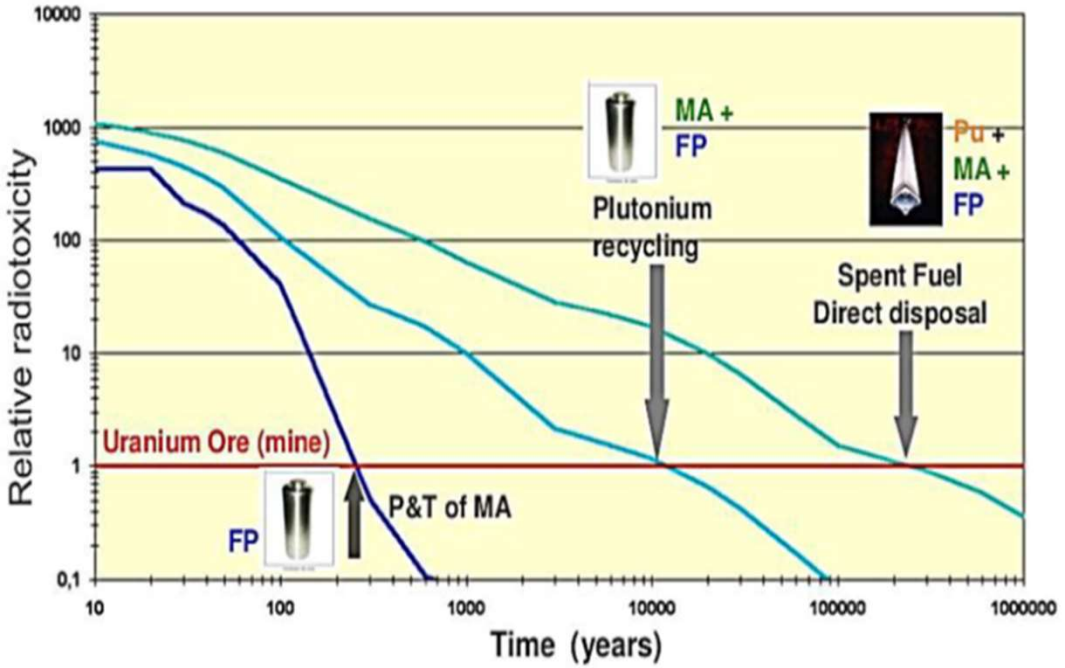


Fig. 3: Relative radiotoxicity levels vs. time when SNF is managed using either a direct disposal, recycling, or P&T strategy [4]

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# SNF: understanding the Needs

- 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].
- Major and minor actinides impacts on
  - storage facilities and heat generation.

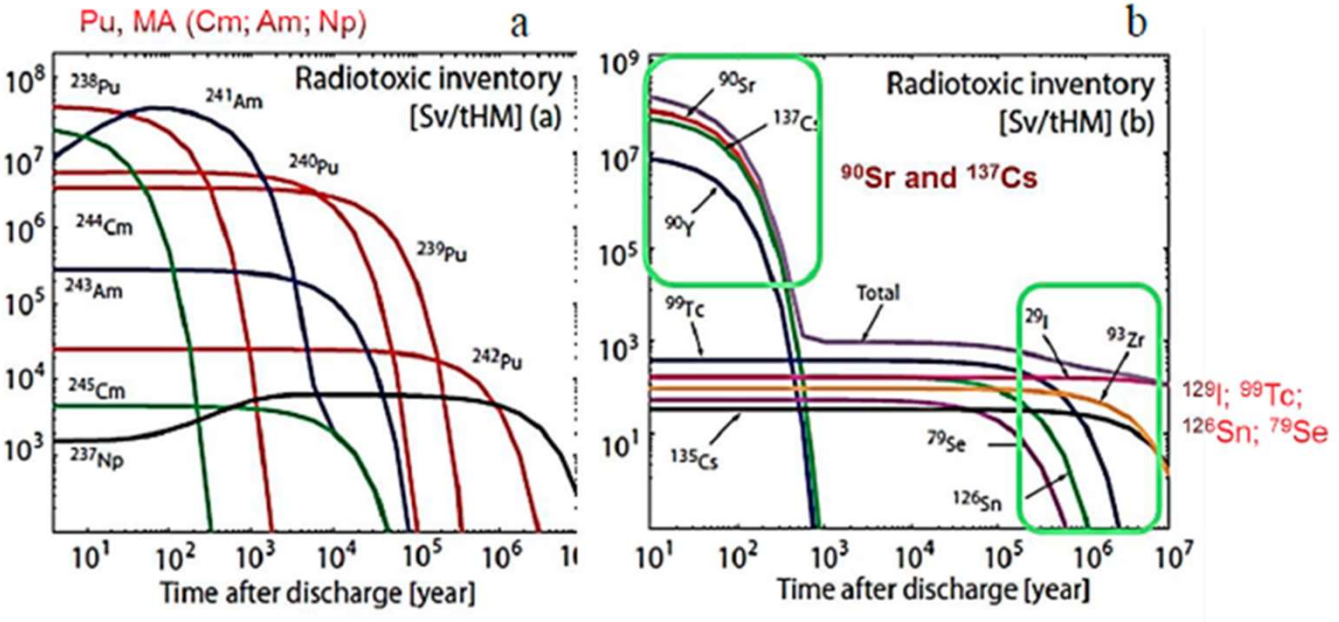


Fig. 4: Relative radiotoxicity of different components of SNF, varying over time [5]

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# SNF: radioisotope impacts on storage

- Radiotoxicity impact
  - $^{79}Sr$, $^{135}Cs$, and $^{151}Sm$;
- Repository impact
  - $^{126}Sn$, $^{79}Se$, $^{135}Cs$, $^{241}Am$, and $^{237}Np$; and
- Heat generation impact
  - $^{238}Pu$, $^{244}Cm$, and $^{241}Am$.

| 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 |

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

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# Partitioning Methods: overview

- Partitioning separates different constituents
  - mainly Pu and MA (Np, Am, and Cm) for treatment, disposal, and recycling.
- Two types:
  - **dry:** Plutonium Uranium Redox Extraction (PUREX), separation efficiency (~99.9%)
    - fuel dissolution, chemical treatment and separation, off-gas treatment, and waste verification [6].
  - **wet:** primarily pyroprocess with three steps [7]
    - 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.

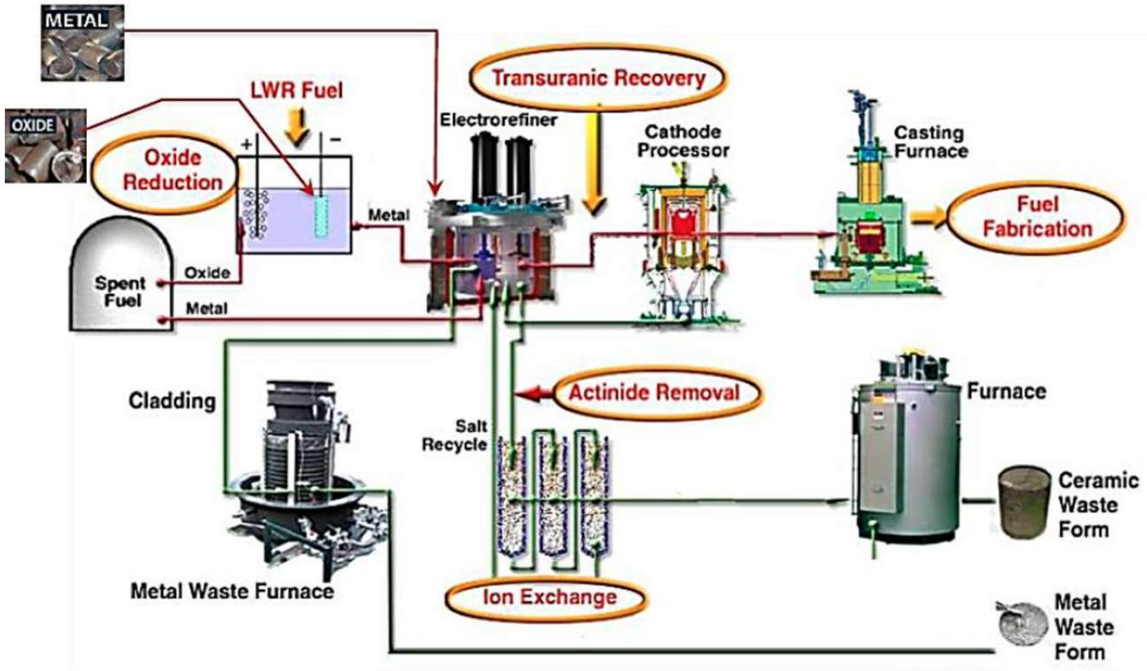


Fig. 5: Schematic diagram of the conceptual design for the advanced pyroprocess [6].

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# Partitioning Methods: comparison

| | 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) |

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

# Transmutation Methods: comparison

- Fast reactor (FR)
  - Molten salt fast reactor (MSR),
  - Sodium-cooled fast reactor (SFR), and
  - Lead-cooled fast reactor (LFR)

| | 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 |

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

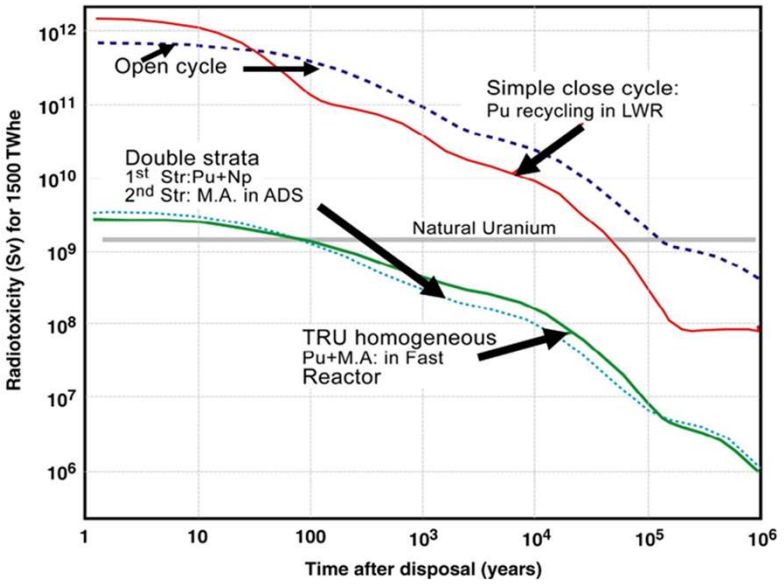


Fig. 6: Radiotoxicity reductions achieved via different strategies [3]

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# Transmutation Methods: comparison (cont'd)

- ADS is a subcritical reactor system that
  - uses thorium as fuel
  - uses high-energy proton beam from an external proton accelerator makes the reactor core critical,
  - ensures more efficient burning of SNF.
- ADS and FR systems
  - burns the minor actinides (MA) and Pu,
    - whereas thermal reactor systems produce them.

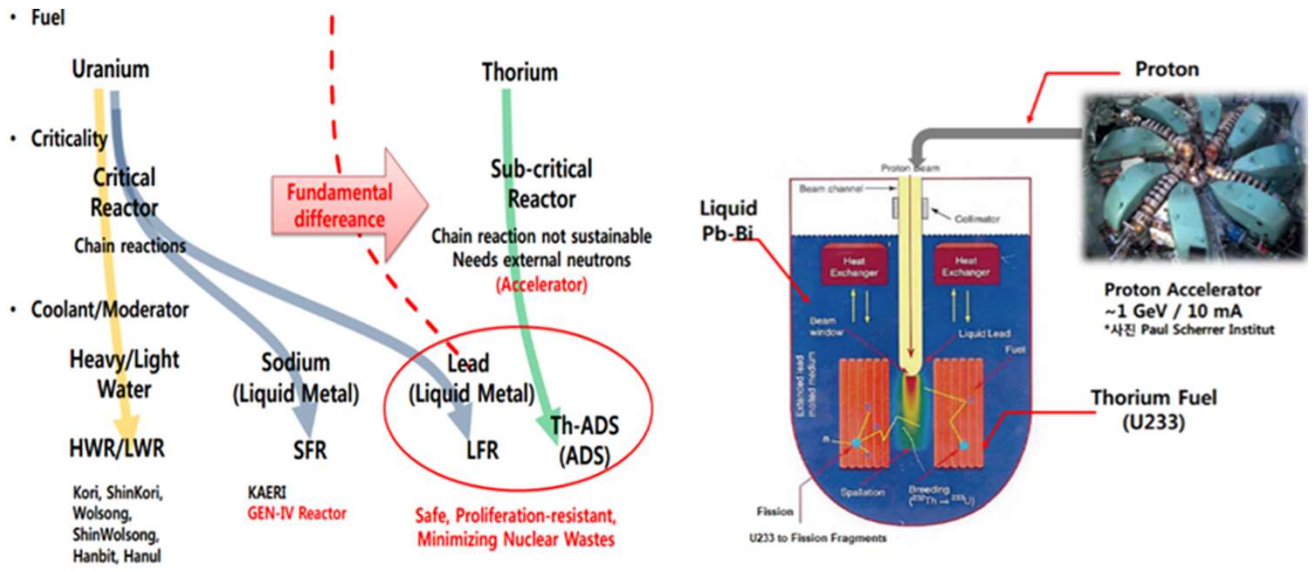


Fig. 7: Accelerator driven reactor system (ADS) [9]

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# Transmutation Methods: comparison (cont'd)

- Compared to the conventional FRs system, the **burnup of the MA when using ADS is much higher.**

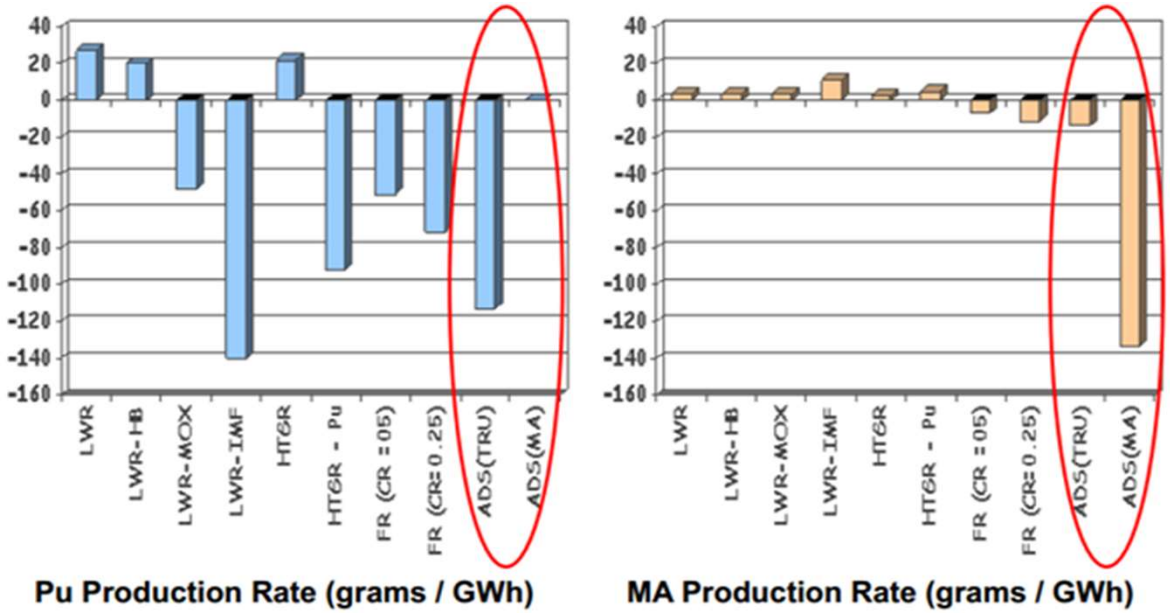


Fig. 8: Transmutation performance comparison between fission reactors and ADS [9].

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# P&T: repository drift loading

- Separation of Pu, Am, Cs, Sr, and Cm will allow for denser loading of treated waste in the repository
  - Radiotoxicity reflects the hazard level of the source materials
    - TRU dominates after about 100 years; however, fission products dominates in radiotoxicity after 100 years.
  - Cs/Sr (and decay products), Cm, and Pu dominate "early" decay heat; whereas Am dominates "later" decay heat
  - 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.

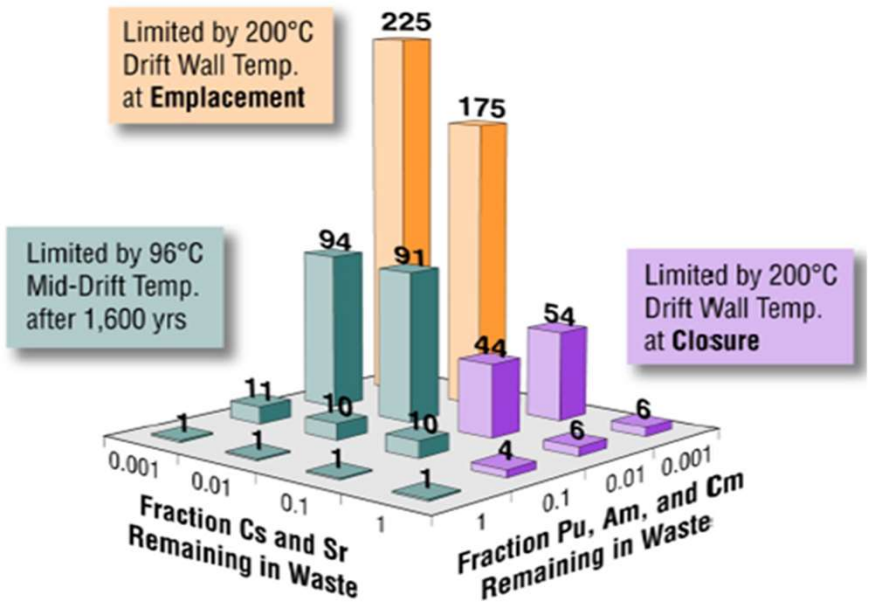


Fig. 9: SNF final repository/disposal site performances [10]

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# Summary: findings and conclusions

- 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.
- ADS system is preferred over an FR system for transmutation
- P&T reduce the
  - long-term radiotoxicity of waste and
    - 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.
  - heat generation—something that is crucial for geological storage
  - amount of area required for the repository
    - e.g., transmuting MA and partitioning fission products can reduce the repository size by a factor of 4 to 5.
- 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.

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## Thank you for your attention!

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