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RECOVERY OF STRATEGIC HIGH-VALUE FISSION PRODUCTS FROM SPENT NUCLEAR FUEL DURING REPROCESSING

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Abstract. Nuclear fission has been identified as a key technology for the Net Zero transition, but high costs and concerns of radioactive wastes hinder wider uptake, and fuel cycle inefficiencies hamper the technology's long-term sustainability. Closing the fuel cycle through spent nuclear fuel (SNF) reprocessing is the primary method of improving sustainability, but high costs and proliferation concerns limit uptake. Current reprocessing technologies recover only the bulk U and Pu in SNF for further energy generation as MOX (mixed oxide) fuel, leaving behind a wide range of untapped, naturally scarce, high-demand, and -value fission product resources. By recovering these resources, such as the platinum group metals (PGMs – Ru, Rh, Pd, Ag), rare earth elements (REEs – Y, La to Dy), and noble gases (He, Kr, Xe), and various other useful isotopes, from SNF during reprocessing operations, the high costs incurred have the potential to be partially or completely offset, in addition to benefits from reducing waste volumes. This paper provides an overview of this concept with exploration of the most promising candidate targets and feeds within reprocessing, and the necessary actions that will be required to see this concept come to fruition in the coming decades.

1. Overview

The Net Zero transition and increasing living standards worldwide are placing exponentially increasing demands on the scarce natural materials (Fig 1) essential for decarbonisation and modern, high-technology applications [1, 2]. As nuclear power has been identified as a key, low-carbon energy source in grid decarbonisation by many nations [3], expanding nuclear capacity will inevitably increase demand for the finite natural uranium and thorium reserves [4]. Combined with the high costs and long build times for reactors and supporting infrastructure, a substantial rethink of the nuclear fuel cycle (NFC) towards a more holistic, and sustainable approach is needed, incorporating comprehensive resource utilisation and advanced waste management [5].

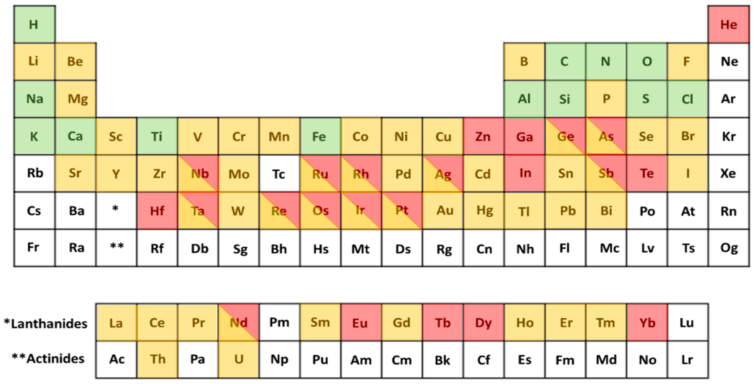


Fig 1: Periodic table of endangered natural resources by element [2].
*Key: **supplies stable**; **supplies at risk**; **supplies endangered**.*

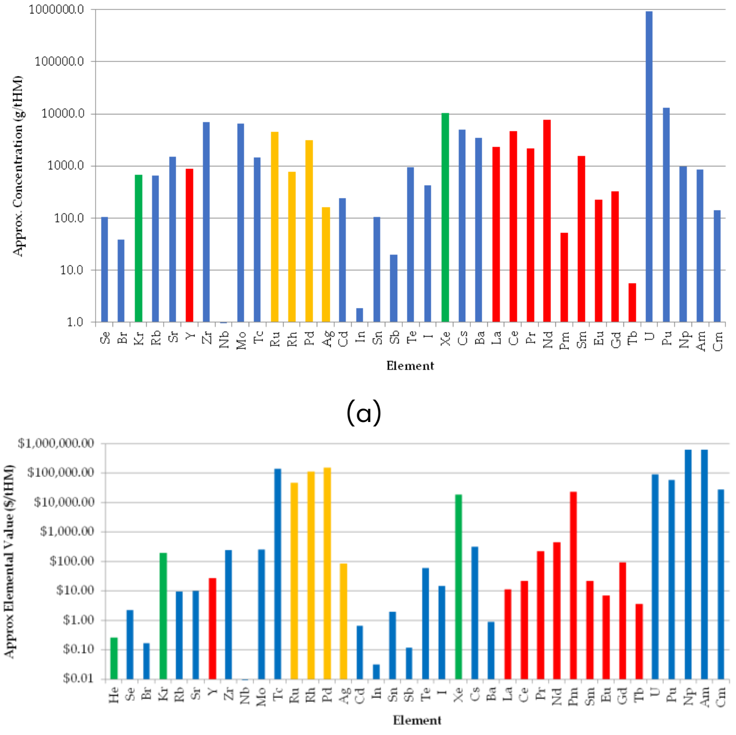
Most of the 7-11 kilotonnes of spent nuclear fuel (SNF) produced annually is not reprocessed, primarily due to limited capacity brought about by high costs, proliferation concerns, and negative image –

most SNF is viewed as a waste rather than a resource [5], leading to the accumulation of more than 280 000 tonnes of SNF around the world [6]. Despite this, reprocessing and responsible management of SNF as a resource will be the key to the NFC's long-term sustainability.

SNF is not just rich in U and Pu for further energy generation, many fission products (FPs) are valuable resources, either as the elements or isotopes [1, 3, 4]. SNF contains resources such as the platinum group metals (PGMs) Ru, Rh, and Pd, and rare earth elements (REEs) Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, and Dy at concentrations (kg/t_{SNF}) far higher than in most natural ores (g/t_{ore}, Fig 2) [7, 8], with approximate values extending from 250 000 USD/t_{SNF} for the PGMs alone [2]. Supplies of many of these elements are limited or nonexistent in the West and becoming increasingly tenuous considering recent geopolitical shifts [3].

SNF thus represents an untapped source of these high-value strategic materials whose recovery would afford a number of primary fuel cycle benefits. These include offseting the high costs of reprocessing, reducing waste volumes, including the demands on and thus the significant costs of deep geological disposal facilities (GDFs), while simultaneously implementing circular economic principles [1, 3, 5], in addition to other benefits[5]. Other potential resources such as the noble gases (He, Kr, and Xe) and a range of isotopes useful for medical (e.g. ^{90}Y, ^{106}Ru), sensing (e.g. ^{85}Kr), power generation (e.g. ^{90}Sr), and irradiation applications (e.g. ^{137}Cs) are also present in significant quantities [5, 7] and represent further targets to be considered for recovery.

Although the concept of recovering FP resources from SNF during reprocessing is not new [1, 7-11], it is under-developed but has received more attention in recent years and is receiving timely attention considering recent geopolitical, technological, and climate shifts [2, 5]. This work highlights the potential FP resources present in SNF, the separation, recovery, and purification routes needed for quantitative recovery and the challenges involved in this [2].



(a)

(b)

Fig 2: Approximate concentrations (a) and values (b) of elements (per t_{HM}) in high-burnup (65 GWd/tHM) pressurised water reactor SNF from an EPR-type reactor or similar (5% ^{235}U initial enrichment, 5 year post-reactor cool) [2]. Key: ***noble gases****;* ***platinum group metals****;* ***rare earths****;* ***remaining fission products and actinides****. NB: y-axes are logarithmic scales. Values are approximated from current market prices, not including those of isotopes [13].*

The probable effects on the NFC should this concept be implemented will be discussed in the context of current and likely future NFC implementations in which advanced hydrometallurgical reprocessing is an essential component [12]. The necessity for decay storage and the end-use applications for these materials are also examined [1, 2, 5].

Focus will be paid to the REEs, targeted to be recovered from SANEX (selective actinide extraction) or GANEX (group actinide extraction) raffinates in advanced reprocessing, and the far more challenging targeted PGM recovery [5], which requires a more comprehensive and complex approach for quantitative recovery at the desired and required purity. The recycling of additional resources to increase fuel cycle sustainability, such as high-purity Zr cladding, is also highlighted [14]. Advanced separations to facilitate segregation of high-heat radionuclides (HHRs – e.g., ^{90}Sr, ^{134}Cs, and ^{137}Cs) for decay storage and advanced waste management, or for value-led recovery is also compatible with this concept [15, 16-18]. Low-level research into this concept is currently ongoing in the UK, USA, France, Russia, Japan, and several other nations.

2. Selective separation of fission products in reprocessing

As the FPs present in SNF are chemically diverse, representing around half of the stable elements on the periodic table [19], achieving targeted, selective separations from the other components of fuel in the high-acidity, high-radiation environment of hydrometallurgical reprocessing is challenging [2, 5, 16-18]. Any new separations targeting the FPs must integrate seamlessly within current (i.e. PUREX) or future (e.g. GANEX) flowsheets as possible, with minimal feed adjustments, acid- and radiation-tolerant processes, and minimised wastes [2, 5, 16-18]. Selective and quantitative FP recovery necessitates the use of techniques established within the nuclear industry such as solvent extraction, gas capture, ion exchange/chromatography, electrochemistry, precipitation and centrifugation, voloxidation, and likely combinations of these [2, 5].

2.1 Selectively separating the REEs in reprocessing

Most proposed SNF reprocessing flowsheets incorporate some form of minor actinide separations (MAs – Np, Am, Cm), such as the GANEX or SANEX processes, the bulk of which also quantitatively co-extract the chemically-similar trivalent (M^{3+}) REEs [12, 20, 21] and thus provide a dedicated feed for targeted recovery [5]. The individual REEs could be separated from these REE-rich feeds using the conventional chromatographic ion exchange resins used for naturally mined materials (Fig 3) [22, 23].

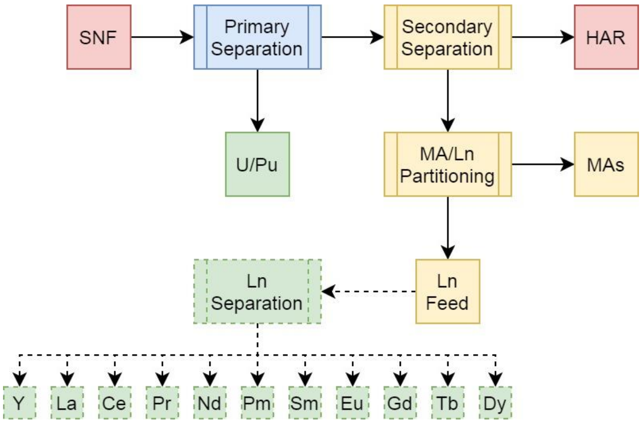


Fig 3: Summary of REE recovery from MA separations in reprocessing. HAR = highly active raffinate.

The necessity to remove all but trace radioactivity for free release of stable or low-activity elements [1] would mean purities several orders of magnitude higher than those accepted for current REE production would be needed [5], unless the acceptable limits for radioactivity could be reevaluated as the current levels are considered to be too limiting by some researchers [24]. Removing the low- or inactive REEs from SNF raffinates in this manner would reduce highly active waste volumes for vitrification by around 25% relative to PUREX with or without MA separations [25]. If separated, Pr, Nd, and Gd, as some of the more valuable and high-demand REEs, could likely be purified to current free-release levels (< 100 Bq/g, above which material is considered very low level waste (VLLW)) [25] with economic viability.

2.2 Selectively separating the PGMs in reprocessing

The PGMs are amongst the most valuable components of SNF after the actinides, making them a prime target for recovery during reprocessing [5, 7, 9-11], however, they are also the most challenging to recover [2]. Due to the variable oxidation states displayed by these elements and the somewhat chaotic partitioning across the solid (undissolved solids), liquid, and gas phases (RuO_4 volatilisation) in reprocessing in addition to undesired co-extraction in primary (U/Pu) and secondary (MA) separations, achieving quantitative recovery will be challenging [2, 26]. As such, a combination of methods will be needed to fully recover the PGMs, but at present, many candidate separative techniques are in a low state of development, work poorly in a highly acidic environment, and/or are poorly selective [2, 5]. The potential locations within an example reprocessing flowsheet that could be targeted for PGM recovery are presented in Fig 4 (adapted from [2]), primarily targeting highly active raffinate (HAR) feeds within reprocessing flowsheets.

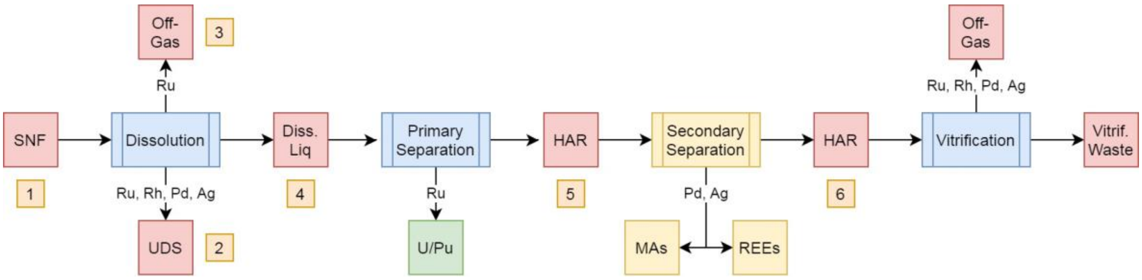


Fig 4: Simplified schematic summarising PGM recovery [2]. 1) Voloxidation (high-temperature (> 500 °C) thermal-oxidative pre-treatment before dissolution) drives off volatile and semi-volatile FPs, including the PGMs; 2) secondary dissolution for undissolved solids (UDS); 3) dissolution off-gas capture of volatile RuO_4*; 4) separation from dissolver liquor prior to primary separations (likely electrochemically); 5) separation on primary HAR before MA/REE separation(s); 6) separation on secondary HAR.*

Rh is the most appealing target PGM as the most valuable element present (price at time of writing is 150 000 USD/kg, having hit 1m USD/kg in 2020-2021), with 0.6-0.7 kg_{Rh}/t_{HM} in high-burnup SNF, with approximately double this in MOX [2]. The presence of trace radionuclides with half-lives of up to 3.3 y means that free release may not be immediately possible, but legacy SNF will have cooled sufficiently to allow this [1]. As with the other PGMs, many separative methods are poorly selective, and as such a combinatorial approach will be needed for quantitative recovery [2].

Ru is of interest for recovery not only due to the element's high value but also because of ^{106}Ru ($t_{0.5} \approx 1$ y), a potentially useful isotope in radiotherapy [26]. This is one of few rationales for the reprocessing of shorter-cooled SNF, as any isotopes with half-lives shorter than a year will have typically decayed to stability after the typical 8-10 y of post-reactor cooling before processing [8]. Oxidation of any Ru present to RuO_4 and volatilisation or extraction into a non-polar solvent system is proposed to recover Ru but this approach requires strongly oxidising conditions and potentially corrosive solvent systems [26-28]. The volumes of PGMs present in SNF have the potential to meet worldwide demand for these elements on their own [1].

2.3 Selectively separating isotopes in reprocessing

As stated above, only isotopes with half-lives of greater than a year can be realistically recovered from SNF during reprocessing, not counting short-lived daughters of longer-lived species, such as ^{90}Y. To recover isotopes within this setting, the chemistry of both parent and daughter elements must be considered. Exhaustive lists of useful isotopes present in SNF have been reviewed elsewhere [5, 7, 29-31] and will not be discussed at any further length here as these are beyond the scope of this work.

The high-heat radionuclides (HHRs – ^{90}Sr, ^{134}Cs, and ^{137}Cs) [15] cause a great many operational challenges in SNF reprocessing [16], which could be mitigated by separating these species early in the process [16-18] and partitioning for either recovery or separate decay storage aside from all remaining wastes [15, 16]. This could potentially allow for reclassification of other waste streams in reprocessing in addition to mitigating many operational challenges of the procedure [25].

2.4 Recovering other resources present in SNF

Materials of value other than the fuel ceramic and its constituents are present in SNF, primarily in the form of high-purity Zr cladding used in most SNF [3, 14]. As Zr has a very low neutron activation potential, even highly irradiated SNF cladding is of low enough activity to be handled in unshielded facilities once any radioactive impurities are separated, allowing for Zr reuse in fresh cladding [14]. This approach could also be applied to similar low-active (i.e., safe to handle without shielding) fission products such as Pd [5].

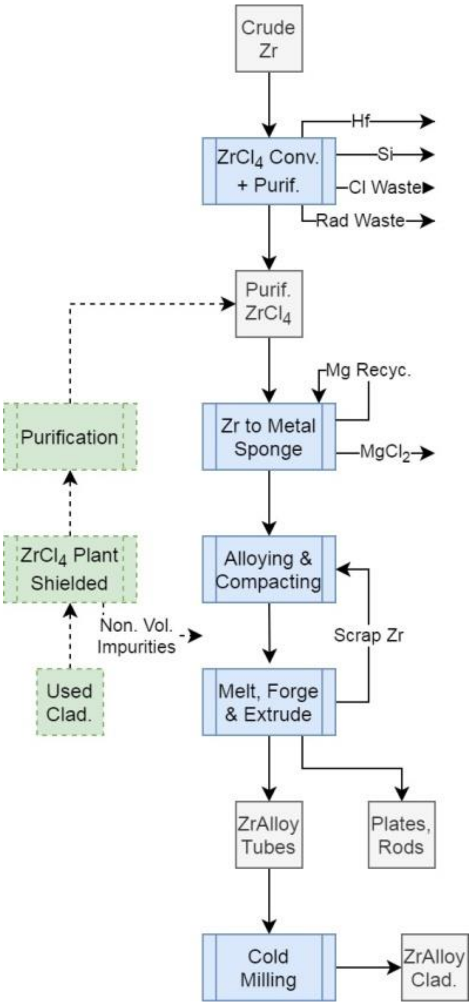


Fig 5: Schematic summary of Zr cladding recycle [14].

3. Challenges and benefits

Despite the promise of unconventional materials recovery from SNF during reprocessing outlined here, several significant challenges remain to be addressed [3, 5]. First and foremost is the development of the necessary selective separations, recovery, and purification methods for operation within a reprocessing environment, the vast majority of which are at very low states of development at present [5]. Second is the necessity for decay storage to allow for shorter-lived isotopes to diminish in concentration sufficient for the free release of recovered materials [1, 2, 5] below the current, somewhat conservative limit of 100 Bq/g; some researchers have argued that this limit could be increased by several orders of magnitude with no ill effects and is a consideration that should be explored further [24]. Thirdly, and likely the largest challenge in the path of this concept's implementation, are the significant regulatory and licensing hurdles that must be overcome to allow for the use of recovered "nuclear" materials in meaningful applications [1, 2, 5]. Finally, due to the complexity of potential recovery operations for these resources, implementation could only realistically be realised in new-build reprocessing facilities given the impossibility of retrofitting to existing plants.

Combined with the limited appetite for closing the fuel cycle at the time of writing, primarily due to low U prices and limited demand [32], implementation soon does not seem likely, despite the well-characterised benefits. In the face of increased nuclear capacity however, which will increase demands on finite natural fissile resources, a closed NFC incorporating advanced reprocessing will likely once again become an attractive proposition over the next several decades (i.e. 2040s-2050s) [12, 33, 45] especially if the benefits of this concept are realised.

These include the partial or perhaps even complete offsetting of the operational costs of reprocessing, the provision of sovereign sources of critical, endangered materials essential to the Net Zero transition, reduced demands on GDF disposal (potentially up to 60% vs. PUREX) [25], and the ability to recover value from large stockpiles of SNF, which are currently stored as wastes. This would allow for the NFC to transition towards a true circular economy if implemented [2, 5].

4. Conclusions and further work

In summary, we have provided an overview of some current thinking and path of travel regarding the recovery of valuable, scarce, and useful FP resources from SNF in a reprocessing environment. This concept holds promise to improve the NFC's long-term sustainability with respect to reduced environmental burdens from mining and waste disposal, lowered costs, and favourable social impacts [3].

For this to be realised, however, several significant steps must be undertaken. Firstly, the necessary separation, recovery, and purification methods for the target FP resources during reprocessing must be developed [2, 5]. In parallel, the regulatory and licensing challenges of this concept must be addressed via close liaison with national and international regulators [2]. Furthermore, the benefits of this concept must be highlighted and disseminated to industries beyond the nuclear field and the wider public, governments must be lobbied to progress towards a closed fuel cycle, and a collaborative, international approach to research in this area be adopted for greatest impact [2].

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