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Article

Spent Nuclear Fuel—Waste or Resource? The Potential of Strategic Materials Recovery during Recycle for Sustainability and Advanced Waste Management

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Abstract: Nuclear fuel is both the densest form of energy in its virgin state and, once used, one of the most hazardous materials known to humankind. Though commonly viewed as a waste—with over 300,000 tons stored worldwide and an additional 7–11,000 tons accumulating annually—spent nuclear fuel (SNF) represents a significant potential source of scarce, valuable strategic materials. Beyond the major (U and Pu) and minor (Np, Am, and Cm) actinides, which can be used to generate further energy, resources including the rare earth elements (Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, and Tb), platinum group metals (Ru, Rh, Pd, and Ag), noble gases (He, Kr, and Xe), and a range of isotopes useful for medical and energy generation purposes are also produced during fission. One reason for the accumulation of so much SNF is the low uptake of SNF recycle (or reprocessing), primarily due to the high capital and operational costs alongside concerns regarding proliferation and wastes generated. This study will highlight the predominantly overlooked potential for the recovery of strategic materials from SNF, which may offset costs and facilitate advanced waste management techniques for minimised waste volumes, thus increasing the sustainability of the nuclear fuel cycle on the path towards Net Zero. Potential challenges in the implementation of this concept will also be identified.

Keywords: nuclear fuel cycle; strategic materials; spent nuclear fuel; recycle; reprocessing; waste management; waste mitigation; resource recovery; circular economy; sustainability



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1. Introduction

Spent nuclear fuel (SNF, also known as used nuclear fuel) is one of the most resource-rich materials known to humans, despite also being one of the most hazardous [1]. Despite well-established (i.e., commercially operating) recycle (the term “recycle” is preferred over the more traditional “reprocessing” as this is envisioned as a holistic approach to UNF management, rather than just the recovery of U and Pu) technology allowing recovery of the major (U and Pu) and substantial progress being made towards delivering minor actinide (MA—Np, Am, and Cm) recovery processes for the generation of further energy and a reduction in long-term radiotoxicity [2], uptake of SNF recycle has been low due to high costs [3], low U prices [4], proliferation concerns [5], and negative public perception of nuclear power generally [6]. This does not include the valuable fission product (FP) resources including the platinum group metals (PGMs—Ru, Rh, Pd, and Ag) [7,8], rare earth elements (REEs—Y and La-Tb) [8], and noble gases (NGs—He, Kr, and Xe) [9] alongside a range of isotopes useful for power and medical applications [10], for which recovery was proposed as far back as the 1960s but has never been implemented [10]. Globally, 7–11,000 tons of SNF are generated annually from ~400 power reactors, and <25% is typically recycled. Currently > 300,000 tons of SNF has accumulated in interim storage

facilities around the world, representing the largest potential stockpiles of some valuable elements, such as Rh [8], and several of those deemed most endangered by limited or unstable supplies (Table 1) [11]. Further depletion of these elements from natural sources is expected to accelerate over the coming decades [12].

Table 1. Periodic table of endangered elements, adapted from [11]. Green indicates plentiful supply; yellow indicates limited availability, future risk to supply; orange indicates rising threat from increased use; red indicates serious threat in the next 100 years; and white indicates synthetic element. Ln * and An * refer to the lanthanide and actinide series presented at the bottom of the table, respectively.

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	Ln *	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	An *	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og
		Ln *	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
		An *	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Not all these SNF reserves may be either suitable or accessible for recycle, however, due to the extent of burnup or the physical state of the fuels themselves or the policies of the nations they are stored within or belong to, amongst other factors. For example, CANDU SNF is produced from natural uranium fissioned to a low burnup resulting in relatively low concentrations of Pu and FPs, which makes this SNF unattractive to recycle under all but the most demanding of circumstances. With this in mind, the stockpile of SNF suitable for recycle is lower than the total amount of accumulated SNF.

Reducing carbon dioxide emissions in order to achieve Net Zero targets will be essential in combating climate change, a transition that will be challenging and intensive in both adapting and replacing energy generation technologies and acquiring material resources that can help deliver this transition [12]. As nuclear power will be an essential technology in reaching Net Zero [13], expanded nuclear power capacity will require a significant paradigm shift in adapting and implementing processes across the entire nuclear fuel cycle (NFC) to address present inefficiencies [14], high costs [15,16], and public, political, and environmental concerns [17,18], especially in light of finite fissile and fertile resources [19]. Although new and likely future reactor designs are more thermally and fuel efficient than most of the established nuclear fleet [20], further NFC improvements will be essential to ensure the necessary supporting infrastructure for a holistic, cradle-to-grave approach to nuclear energy, such as comprehensive SNF recycle [2,17,18] and advanced waste management techniques [21–23] which can meet Net Zero targets.

Recovery of valuable FP resources in addition to the actinides during SNF recycle could address this and contribute to the security of both energy and raw resources critical to modern-day life. Nuclear power can thus potentially contribute in a twofold manner to Net Zero—by providing low-carbon energy and by increasing the availability of other technologies that can utilise resources that are otherwise by-products of fission [12]. The expected concentrations of the key elements in SNF are presented in Figure 1 for Gen III(+) reactor SNF systems (such as the EPR—European Pressurised Water Reactor, Rolls–Royce SMR, Westinghouse AP1000, or Russian VVER-TOI), in which the scarce or valuable classes of materials are highlighted [24].

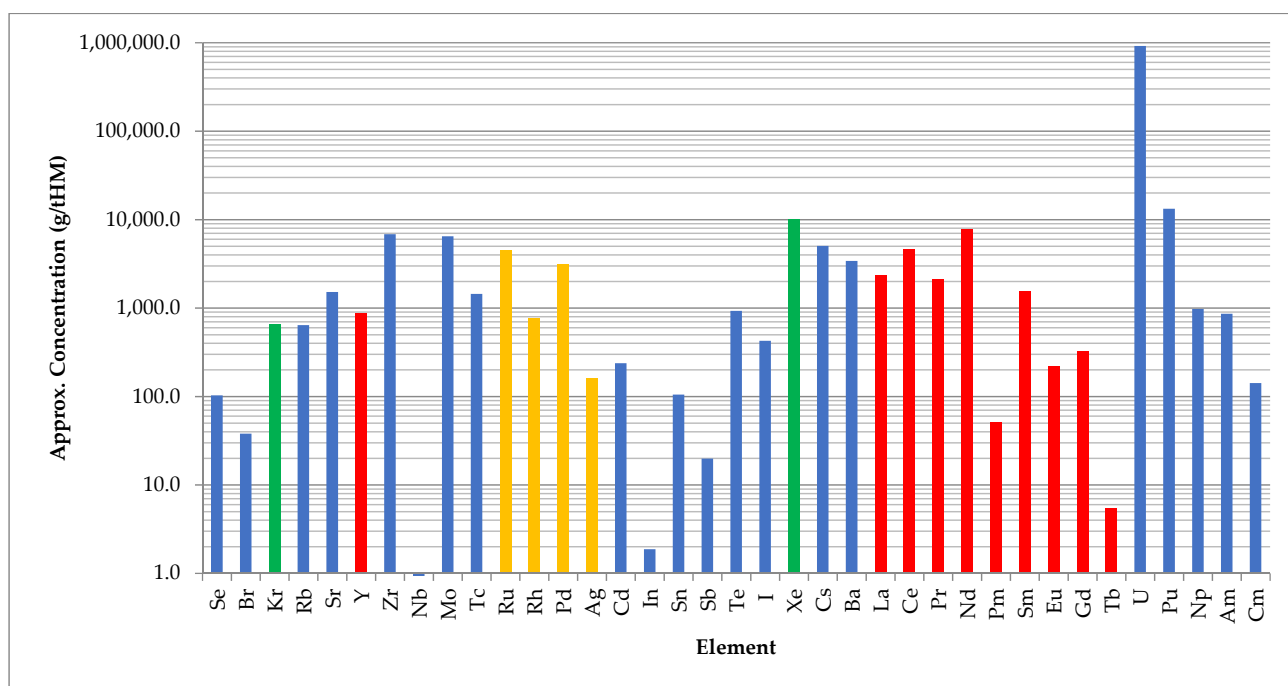


Figure 1. Approximate elemental concentrations in HBU PWR SNF (5% initial ^{235}U , 65 GWd/tHM B/U, 5-year post-reactor cooling). NB: y-axis is logarithmic; He (as ternary fission product) and FP elements produced $\leq 1/\text{g tHM}$ at discharge ($<\text{Se}$ and $>\text{Tb}$) are omitted. PGMs highlighted in orange, NGs in green (not He), and REEs in red. Values calculated using an in-house model, figure amended from Ref [24].

For the purposes of this study, we shall consider high burnup (HBU) pressurised water reactor (PWR) SNF. Legacy SNF from Gen II reactors contains lower concentrations of FPs [25], as they operated at lower burnups (typically 30–45 GWd/tHM; GWd/tHM = gigawatt days per metric ton of heavy metal) [26]. Given the world stockpiles of SNF, the potential untapped resources present could be used to mitigate “peak” element production for those at-risk species identified in Table 1 to provide a secure supply, and offset the high costs of the NFC, particularly recycle [3]. This could also go some way towards addressing demands for these elements from emerging technologies, which will only increase over time [12,14]. Although legacy SNF which has been stored for decades may be less attractive than shorter-cooled, HBU SNF from more modern reactors, the benefits of recovering potential resources from these materials should not be understated as, despite the lower concentrations of Pu and FPs, the recycle of these materials would nonetheless be beneficial in terms of NFC efficiency and environmental impacts.

Recovery of FP resources during SNF recycle represents the most facile and logical point within the fuel cycle to effect separation, purification, and conversion to a useful end-use form, given that SNF is chemically processed into a more readily separable form for actinide recovery [27]. Although the techniques for the recovery of U and Pu and, to a lesser extent, the MAs are well developed [2], those for the recovery of most potential FP resources are in their infancy, in addition to several other technical, regulatory, and societal challenges that must be overcome. In this publication, we shall present an overview of the potential non-actinide resources which could be recovered from SNF during recycle, how these could be recovered, and the challenges that must be addressed before such a concept can be implemented. Where appropriate, other potential, related opportunities for efficiency improvements are highlighted.

2. Resources within SNF

2.1. Elements

2.1.1. Actinides

The recovery of the actinides in SNF recycle is well established, primarily to allow for the generation of further energy from U and Pu (typically as MOX—mixed oxide fuel) [2,17,18], where the extended burning of fissile material still in SNF can reduce the long-term radiotoxicity of SNF requiring disposal (Figure 2). The possible partitioning of the MAs has been explored extensively for the purpose of generating further energy in appropriate (fast) reactors and thereby transmuting the MAs to isotopes of lower long-term radiotoxicity (Figure 2) [2,28]. As such, any further discussion regarding actinide partitioning will be included only where pertinent to the separation and recovery of FP resources. The uses of actinide isotopes for power generation [29], medical applications [30,31], and analytical purposes outside of their potential power usage are well studied and beyond the scope of this study. The actinide and FP isotope contributions to decay heat are presented in Figure 3 to better highlight the sources of these by actinide and fission product isotopes.

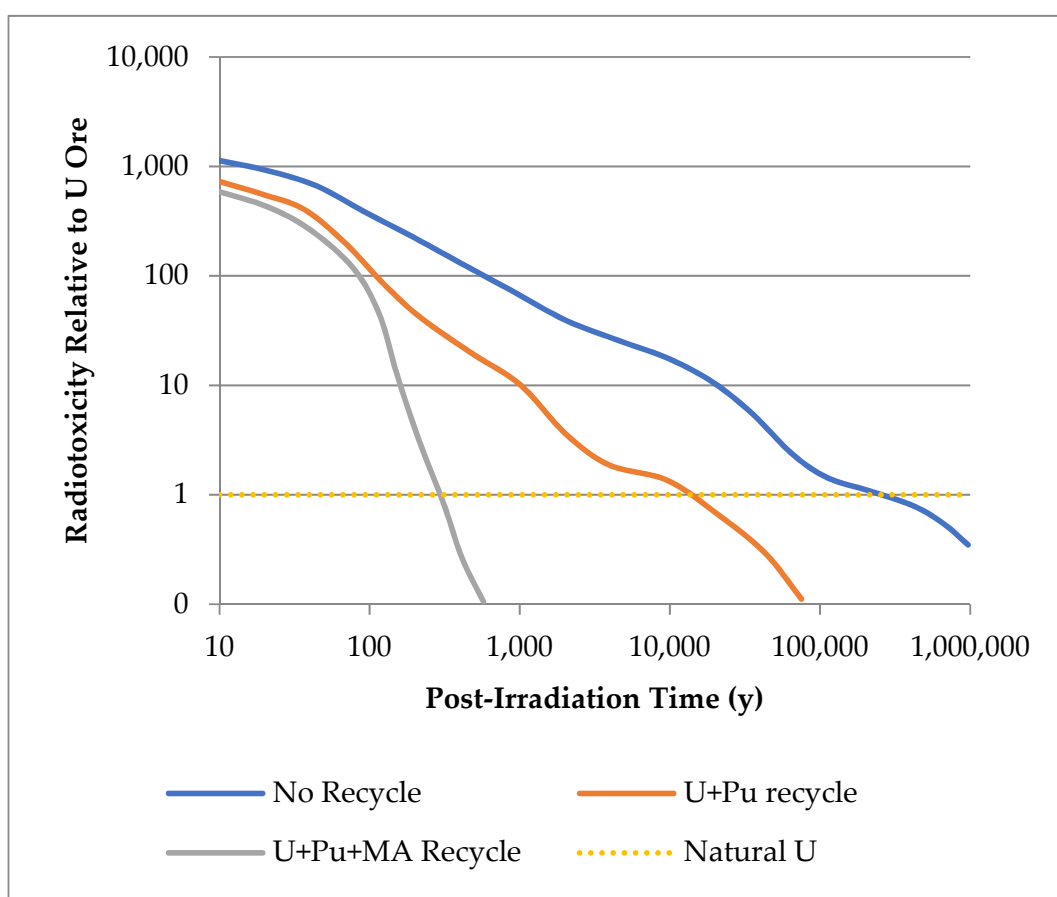


Figure 2. Reduction in length and intensity of radiotoxicity with Pu and U recycle, and additional MA partitioning and transmutation, relative to open fuel cycle (no recycle) and natural uranium [14].

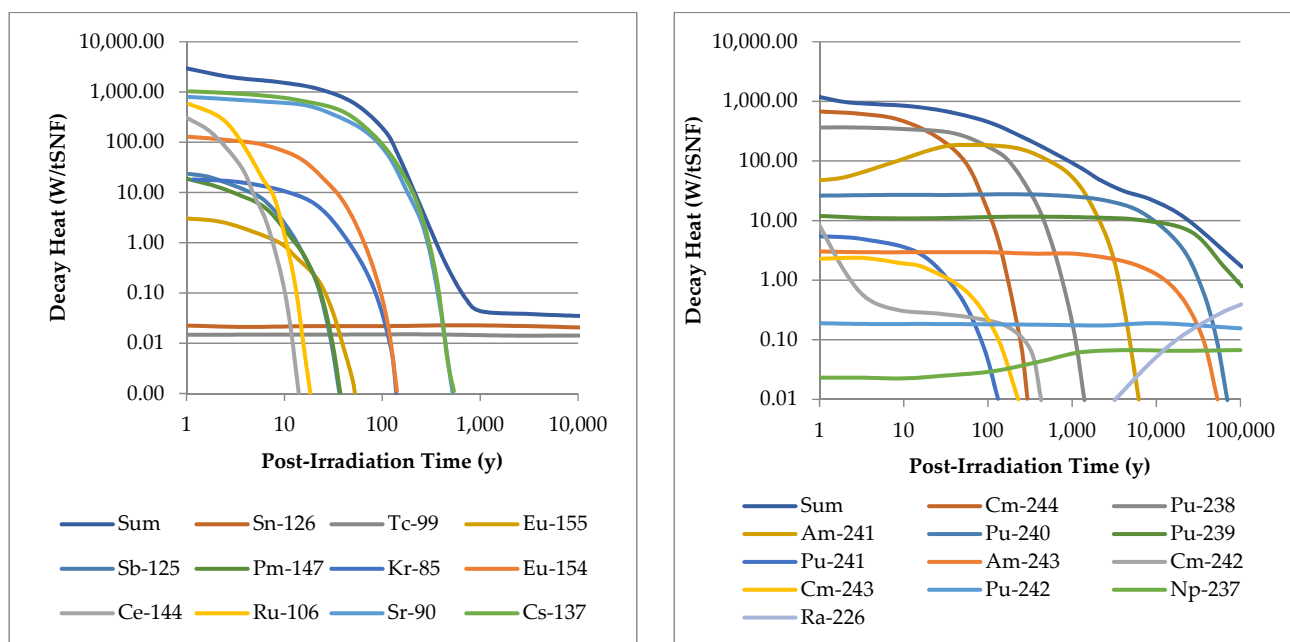


Figure 3. Sources of decay heat for HBU PWR (VVER) fuel with time from primary fission product (left) and actinide (right) isotopes. Graphical data extracted from reference [32]. NB: axes are logarithmic scale.

2.1.2. Fission Products

The highest-value and most naturally scarce FP elements by a significant margin are the PGMs Ru, Rh, and Pd, [8] of which Rh is the most valuable (USD 457/g_{Rh}), followed by Pd (USD 66/g_{Pd}) and Ru (USD 19/g_{Ru}) [33], amounting to some USD 500,000/t_{SNF} for HBU SNF [24]. The yields of PGMs in SNF from MOX fuels are typically higher than those from UO₂ [34]. One source states that if Rh occurred at the same concentrations in natural ores as in SNF, this “would be regarded by miners and economic geologists as nothing less than fantastic” [10]. The PGMs are, therefore, arguably the most attractive FP target for recovery during SNF recycle, though there are several challenges to realising this:

- The PGMs do not completely dissolve in aqueous nitric acid media when SNF is processed in the early stages of SNF recycle, meaning that partitioning and full recovery of these metals using aqueous process technology compatible with current SNF recycle processes is likely to be difficult [27] (Ch. 8). The partitioning of Ru specifically is further complicated by the (partial) formation of volatile RuO₄ [34,35].
- Several radioisotopes of the PGMs are produced during fission, which may limit the end-uses open to PGMs recovered from SNF, or necessitate decay storage [8]:
- Rh is essentially monoisotopic (as Table 1 ¹⁰³Rh), though trace (<0.1 wt% radioactive) ¹⁰¹Rh ($t_{0.5} = 3.3$ y)—which decays by electron capture—and ¹⁰²Rh ($t_{0.5} = 0.57$ y)—a γ emitter—are typically found in SNF [7,8]. These would necessitate decay storage of up to 30 years for the most sensitive applications, such as electronic applications [7,8], but shorter storage times are likely to be needed for the most common use for Rh in catalytic converters in the automotive industry and in other industrial processes.
- Ru contains approx 0.2 wt% ¹⁰⁶Ru (after 5 years of post-reactor cooling, $t_{0.5} = 1.02$ y), which decays to the short-lived but high-decay energy ¹⁰⁶Rh ($t_{0.5} = 30$ s, decay energy = 3.54 MeV). ¹⁰⁶Ru has been proposed for use in brachytherapy [36]. The shorter-lived ¹⁰³Ru ($t_{0.5} = 39.2$ d) will have entirely decayed to stability after the 5 or more years of cooling time needed by HBU SNF before aqueous recycle operations can commence [7].
- Pd contains approx 16 wt% of long-lived ¹⁰⁷Pd ($t_{0.5} = 6.5$ My), a low-energy radioisotope [7,8] which could be considered essentially inactive for most purposes outside

the most sensitive electronic applications [37]. Pd recovered from SNF could be an excellent storage medium for hydrogen [38].

The highest concentration FPs by elemental class are the REEs [24], covering the elements Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, and Tb for most PWR SNF, though for MOX fuel, the yields of the heavier lanthanides beyond Tb are higher [26]. Of these, Pr, Nd, and Gd are the most valuable (~USD 136/kg for Pr_2O_3 and Nd_2O_3 , and USD 69/kg for Gd_2O_3) [39]. These versatile elements find a wide array of uses across modern, advanced technologies including clean energy [40]. Several factors must be taken into consideration for the potential recovery of the REEs during SNF recycle:

- The vast majority of proposed MA separation and recovery processes also partition and separately strip the chemically similar REEs [2,40], thus providing a viable recovery route to these elements. Conventional chromatographic techniques used industrially could then be used to separate the individual REEs from each other [41–43].
- The most valuable REEs outlined above exist only as stable isotopes [8], alongside Y, La, and Tb. Ce, Pm, Sm, and Eu all possess radioisotopes, and thus effective decontamination between elements would be required.

The NGs represent the most inherently separable species from SNF due to their low chemical reactivity and phase differentiation, being completely volatilized during the dissolution step of recycle [27] (Ch. 8). Xe is the highest-yielding FP by element, with >10 kg/tHM being produced in HBU SNF, with lesser amounts (~500 g/tHM) of Kr and further smaller levels of He (~20 g/tHM), the latter of which is produced by ternary fission and α -decay [24]. Only one significant radioisotope is present in SNF NGs— ^{85}Kr ($t_{0.5} = 10.8$ y), which would necessitate ~100 years of decay storage in order to recover and utilise stable Kr from SNF for non-nuclear applications [44], though this isotope does find niche applications in its own right, primarily in materials analysis [45]. It may be possible to separate the lighter, inactive Kr isotopes via gaseous diffusion or centrifugation processes [46,47], though this is an expensive process as per enrichment of U which uses the same techniques. As Xe is worth approximately USD 3000/kg, it would likely be economical to recover this element from SNF during recycle [10]. Extant off-gas scrubbing methods could suffice for this purpose [27] (Ch. 8).

Several other classes of FPs cover useful and valuable materials, such as Zr, Mo, and In, though the recovery of these is likely uneconomical or challenging when compared to more conventional sources. As such, further discussion of these is beyond the scope of this study. Although several other recent studies have provided more comprehensive overviews of this concept in a national context [8], at times, a pessimistic and sometimes overly cautious view of FP utility is taken when compared to the potential values presently not recovered from SNF, particularly with respect to acceptable levels of radioactivity within recovered materials, which could be increased without detrimental harm in many cases [9,37]. A summary of FP concentrations, elemental values, and notes is presented in Table A1.

2.2. Isotopes

For SNF that is sufficiently cooled to allow for aqueous recycle, most of the short-lived isotopes contributing to decay heat have already decayed to stability [24]. Some of these short-lived isotopes are of interest for medical, power generation, or other applications. However, it would likely be unfeasible or impractical to recover any isotope from HBU SNF with a half-life shorter than 1 or 2 years due to the necessary cooling times before aqueous recycle. Longer-lived FPs and their daughters have, however, attracted interest for a number of potential uses perhaps worthy of recovery from SNF during recycle:

- ^{90}Y (the daughter of ^{90}Sr) [48] and ^{106}Rh (the daughter of ^{106}Ru) [49] have been proposed as β - and γ -emitting radiopharmaceuticals, respectively. There is also potential for ^{126}Sb (the daughter of ^{126}Sn) [50] and ^{144}Pr (the daughter of ^{144}Ce) to be used in this role [51]. ^{137}Cs , alongside ^{60}Co , is already used for cancer therapy in developing economies [52].

- ^{90}Sr has been proposed and indeed utilised as a low- γ source for radioisotope thermoelectric generators (RTGs) in place of the actinides ^{238}Pu and ^{241}Am [53]. Other isotopes present in SNF could also be used for heat-generation purposes [8] or beta-voltaics in a similar vein [54].
- ^{137}Cs in particular has found use as a γ irradiation source in place of ^{60}Co for use in sterilisation [55] and analytical processes [56].

Advanced partitioning of SNF beyond those targeting the actinides has the potential to reduce SNF cooling times post-removal from the reactor core, opening the possibility to access the valuable short-lived FPs in SNF. Separation of what are commonly termed high-heat radionuclides (HHRs—usually ^{134}Cs , ^{137}Cs , and ^{90}Sr and occasionally ^{144}Ce and ^{106}Ru) has been proposed by employing radiolytic and thermally resistant partitioning strategies [21,57]. This could lead to the increased availability of short-lived fissile isotopes (particularly ^{241}Pu — $t_{0.5} = 14.1$ y) and possibly a route to some of the shorter-lived FP radionuclides.

Some of our own research has contributed to developing this concept in addition to addressing several of the key operational challenges with present SNF recycle flowsheets [58–61] by removing the HHRs early in the SNF recycle flowsheet to mitigate the majority of downstream radiological issues. This concept was suggested several decades ago for short-cooled fuels [57] but has seen little attention to date [62], despite promising initial results for the Cs separation system. As the dominant source of radioactivity in the remaining raffinate (Figure 2), separation of HHRs could allow for reclassification of SNF recycle raffinate outputs to lower levels or allow for more concentrated disposal of these raffinates due to the lower heat load [58]. Despite the hazards associated with handling HHRs, these isotopes (those of Cs in particular) have been proposed for various applications, including the generation of radiolytic hydrogen [63] and as alternatives to actinide isotopes in RTGs [53], or perhaps even as a source of process heat if sufficiently concentrated [64]. Even if HHRs are considered to be only a waste product, the separation of these isotopes can reduce the high-level waste (HLW) volumes associated with heat emission that need to be disposed in dedicated deep geological disposal facilities [21].

Most elements in SNF comprise several stable isotopes alongside radioactive nuclides. Attempts to separate individual isotopes of the same element, especially those that are not have a readily accessible gaseous form, are deemed unfeasible and uneconomical. Complete recovery of the target element(s) would most likely result in radioisotopes being “diluted” with stable nuclides.

3. Separation, Recovery, and Purification Methods

Established techniques within the nuclear industry present the most likely pathways for the separation of the target resources identified above. Of these, solvent extraction (SX) is the only commercial approach for SNF recycle [2] in the form of the PUREX process and is proposed for use in various advanced SNF-partitioning strategies for U, Pu, and MA recoveries. A simplified schematic overview of current SNF recycle flowsheets is presented in Figure 4. Ion exchange (IX) finds use in uranium milling processes [65,66] and nuclear effluent treatment approaches [67]. Electrochemical methods are used in proposed pyroprocessing of SNF [68,69] for oxidation-state control in nuclear effluents [70] and have been proposed for use in fuel dissolution approaches [71], alongside several conceptual voloxidation (high-temperature oxidation) processes [27] (Ch. 8). Further techniques used in SNF recycle that can be deployed in recovery operations include robust scrubbing for off-gas feeds that would primarily be used to mitigate the release of radioisotopes to the environment [27] (Ch. 8). Innovative uses and combinations of these techniques will likely be required to effect selective separation of the target resources. Here we present an overview of the separations technologies, which will likely be necessary for the high-value targets identified above. Thorough assessments of SNF recycle technologies for actinide recovery have been conducted recently [2], and similar in-depth assessments of these and

other processes for the recovery of other valuable elements/isotopes will be conducted as the development of this concept proceeds.

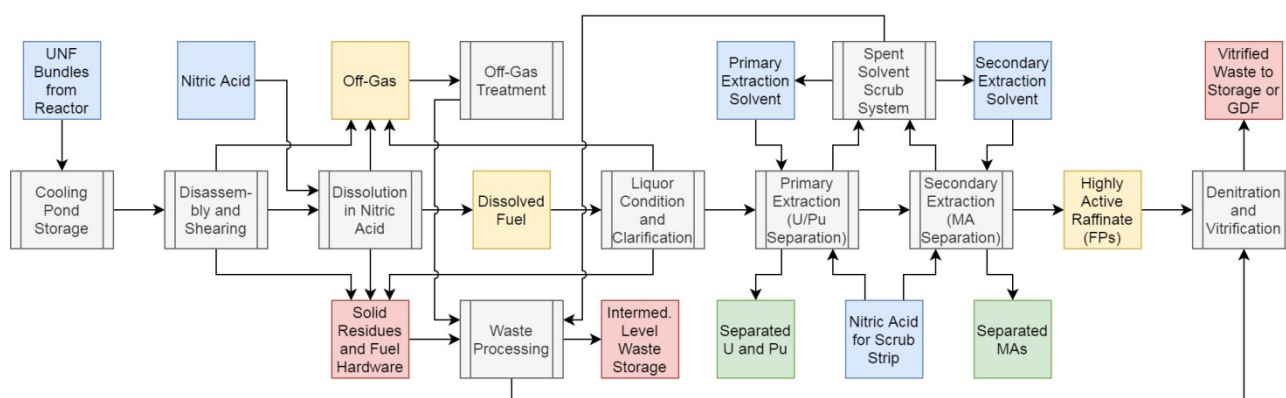


Figure 4. Simplified schematic overview of present concepts for hydrometallurgical SNF recycle. Key: blue—inputs, yellow—intermediates, red—waste outputs, green—targeted outputs, and grey—process boxes. This is based around a PUREX primary separation and SANEX secondary separation, though numerous variations of this flowchart are possible.

3.1. Rare Earth Elements

The easiest of the outlined classes of materials to separate, assuming that the parent partitioning technologies are implemented, are the REEs. A wide range of MA separation processes and flowsheets for implementation during SNF recycle have been explored [2], which also separate the REEs alongside the trivalent actinides, for example GANEX (group actinide extraction) [72], SANEX (selective actinide extraction) [73], TALSPEAK (trivalent actinide lanthanide separation with phosphorus-reagent extraction from aqueous k(c)omplexes) [74], and related processes. Presently, none of these have entered commercial operation, but development is being targeted for deployment at next-generation SNF recycle facilities [2,75] (see Figure 4 for the likely incorporation of these into conceptual flowsheets). As the REEs are separated during most of these processes (not including the one-cycle SANEX flowsheet) [76], separation and recovery of these elements are likely to be the most readily achievable compared to the other groups of elements identified. The remaining challenge is to separate the individual REEs to a high enough purity that any trace radionuclides from other lanthanide elements (given the chemical similarity across the group) are eliminated from final product streams to a satisfactory level [37]. As the lanthanides occur together in nature and are mined as such, the chromatographic (IX-based) separations employed in present industrial purification would likely be the best approach to achieve the required elemental separation [41–43]. Suitable media for storage or output of recovered REEs would likely be as stable metal salts, such as oxides (M_2O_3) or halides (MX_3).

3.2. Platinum Group Metals

As the most valuable of the classes of resources in SNF identified and representing the greatest challenge for selective isolation due to their variable and complex chemistry [27] (Ch. 8), especially in the case of Ru [35], the PGMs warrant the most attention devoted to the development of recovery processes. As stated above, the PGMs only partially dissolve in the head-end of SNF recycling due in part to their chemical inertness [27,34] (Ch. 8). Although approaches such as voloxidation or electrolytically assisted dissolution may increase the amount of the PGMs available for recovery from the aqueous phase for recycle [27] (Ch. 8), a hybrid approach would still likely be required to effect complete recovery of PGMs in SNF. This is likely to involve the separation of PGM residues from the undissolved SNF solids otherwise sent to wastes [7], in addition to solution recovery of Ru, Rh, and Pd [77] and potential gas-phase recovery of Ru [34]. Several SX, IX, and electrolytic approaches

have been tested for the recovery of these PGMs from acidic solutions, such as those in SNF recycle, but all are in their infancy and have not seen significant development beyond small-scale laboratory tests or even concepts [78]. Separation of the PGMs from one another once recovered would require further operations given their relative chemical similarity and inertness [79]. As several of the PGMs (Ru and Pd in particular) are problematic in SNF recycle [78] due to co-partitioning with the actinides, the recovery of PGMs as early as is feasible in the SNF recycle flowsheet would aid in the management of contamination in downstream operations, as has been proposed in the conceptual SREEP flowsheet [70]. The elemental metal is the ideal chemical form of each element of the PGM product stream rather than their compounds, given the inherent low reactivity and stability of these forms, which are best suited for storage and the majority of its uses.

3.3. Noble Gases

Given that the NGs exist as a distinct phase from the majority of other FPs, their separation during SNF recycle would be relatively facile, given the necessity for robust off-gas scrubbing already present in existing processes to prevent the environmental release of volatile radionuclides such as ^{85}Kr , ^{131}I , and ^{129}I , amongst others [27] (Ch. 8). Such sorption and gas separation technologies are well established both in and outside of the nuclear industry, so it would be relatively straightforward to recover Xe outgassed from SNF dissolution. The challenge here arises from the necessity to ensure that produced Xe is decontaminated from Kr sufficiently for all applications, whereas if radioactive ^{85}Kr is required for applications, the Kr could be separately isolated for this reason. The aforementioned gaseous diffusion or centrifugation techniques would allow for fractionation of the NGs although at the expense of significant power and equipment costs [46,47]. Suitable output forms of the NGs would be either as compressed gasses or as the gas sorbed onto a matrix for storage.

3.4. Radioisotopes

When considering the range of potentially useful FP radionuclides in SNF, recovery must factor in not only the radiological aspects of these but also the chemistry of their parent and daughter elements. Our own work investigating separation of Cs isotopes (as HHRs) has utilised an IX approach using Cs-selective AMP-PAN (ammonium phosphomolybdate-polyacrylonitrile composites), demonstrating that selective fission product separations from simulated raffinates are feasible [58–61], even in the presence of dissolver-liquor concentrations of U (300 g/l_U) [80]. For example, several SX approaches for Cs and Sr based on cyclic ethers or more complex extractants have been proposed [81], alongside IX using cyanoferrates [82], polyantimonic acid [83], and zirconium phosphate [84], though many of these perform poorly in the strongly acidic conditions of SNF recycle. Given the diversity of chemistries present across the range of FPs in SNF, a plethora of other approaches has been reported in the literature, many of which would be unsuitable for use in SNF recycle. Consequently, there is substantial scope for novel process development on the recovery of targeted radioisotopes that needs to be compatible with parent SNF recycle strategies.

3.5. Operational Considerations

In contrast to the conventional recovery of minerals in mining, operations in a SNF recycle setting must all be conducted remotely until at least all materials are low enough in radioactivity to be safe for human contact [8,37]. This necessitates comprehensive radiation shielding, remote operations, online monitoring, and thorough accountancy, especially where fissile isotopes must be handled [85]. Decay storage of recovered species would require similar levels of control before releasing materials, though the storage of products in the nuclear industry is well established, and current processes and procedures can be readily adapted to the concept outlined here.

4. Challenges

The scientific record is replete with comprehensive published literature assessing the benefits of actinide recovery, partitioning, and recycle [17,18], but few—if any—have thoroughly explored the environmental and socio-economic implications of recovery of non-actinide resources in SNF recycle [8]. Similarly, a great many promising concepts for the separation of the resources outlined above in both conventional and nuclear settings have been presented, though few of these take a holistic approach beyond the technological and scientific data needed to effect the basic recovery of the target elements. A number of significant challenges and actions must be addressed before resource recovery from SNF during recycle could be implemented:

1. A thorough assessment to identify all elemental and isotopic resources present in SNF, both actinide and FP, and their potential values and acceptable end-use cases accounting for radioactivity while assessing the necessity for decay storage.
2. The technologies necessary to selectively (where possible) separate these resources during SNF recycle using minimally disruptive processes (i.e., with minimal feed adjustments and arising effects on downstream operations).
3. A complete safety case with a technical and economic assessment of these processes and recoveries on SNF recycle operations once technological means for the recovery of economically viable resources are developed.
4. A cradle-to-grave life cycle assessment of these processes on the NFC and effects on supporting and competing industries, including comparisons against the incumbent methods of sourcing the recovered element, i.e., mining operations [12].
5. Compliance with national and international regulatory factors, or development of appropriate frameworks where these are insufficient for emerging technologies.
6. Consideration of emerging concepts in the NFC, including new reactor types (e.g., Gen IV reactors), new SNF types (e.g., advanced technology fuels—ATFs), recycle technologies (e.g., voloxidation), etc.

5. Conclusions

In this publication, we have outlined the potential for the recovery of non-actinide resources from SNF to address, in the first instance, economic factors in the NFC and the increasing national importance of resource security, alongside the well-known benefits to the NFC gained from the recycle of SNF [86]. The most promising of these classes of resources by potential value, concentration in SNF, and viable routes to recovery have been identified, though this overview does not represent an exhaustive list. The possible routes to the recovery of these target elements have been identified, and the challenges that these routes and the (re-)use that these resources may face generally have been highlighted. In summary:

- The PGMs represent the most valuable FP component in SNF (up to USD 500,000 /tHM in HBU SNF) and thus the most appealing to recover. However, given the chemical and radiological complexities involved, these are also likely the most challenging to isolate. Decay storage may be required for certain end-use applications.
- Xe represents a valuable component of SNF (up to USD 30,000/tHM in HBU SNF) which could be readily recovered using current technologies, though decontamination of Kr will be essential to avoid trace radioactivity. He levels present in SNF are likely to be too small to be of consideration for recovery, though this could be reconsidered if global shortages increase in severity and prices increase further.
- The REEs will be isolated if MA partitioning is employed in future SNF recycle, and, as such, are already separated from the bulk raffinate and other FPs. Conventional chromatography can separate these elements, though sufficient decontamination between elements will be required for radiological safety. This would likely require much higher REE prices to be economically viable given the value present in SNF (~USD 1000 s/tHM in HBU SNF), but if shortages increase, this prospect could become more attractive.

- The direct economic values of isotopes are much harder to discern, but increased availability and supply would lower the cost of implementing advanced radioisotope technologies for any species recovered from SNF. The complexities in recovering and handling the range of elements with radioactive isotopes present in SNF represent an additional layer of complexity compared to the simple recovery of the elements.
- Technological, regulatory, environmental, and socioeconomic assessments of all aspects of this concept are required before implementation can be considered.
- The potential to offset the presently high costs of the NFC and reduce current, large waste volumes and thus the potential load on geological repositories by maximising recovery of resources in SNF is one of many drivers behind such research, especially with the necessity to decarbonise power grids as soon as is possible.

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Appendix A

Table A1. Summary of potential FP resources in HBU SNF (65 GWd/tHM PWR fuel, 5% initial ^{235}U , 5-year post-reactor cooling [24], with approximate concentrations, elemental values (not incl. isotope values, g—as gas, ox—as oxide, and m—as metal, correct as of time of writing), key radioisotopes remaining after this cooling period and their half-lives, and notes on uses of these.

	Element	Conc. (g/tHM)	Elem. Value (USD/kg)	Radioisotope(s) ($t_{0.5} - y$)	Notes/Applications of Isotopes
Noble Gases	He	~10	50 (g)	Stable	
	Kr	665	330 (g)	^{85}Kr (10.8)	
	Xe	10,244	3000 (g)	Stable	

Table A1. Cont.

	Element	Conc. (g/tHM)	Elem. Value (USD/kg)	Radioisotope(s) ($t_{0.5} - y$)	Notes/Applications of Isotopes
Rare Earth Elements	Y	874	4 (ox)	Stable	
	La	2332	2 (ox)	Stable	
	Ce	4665	2 (ox)	^{144}Ce (0.78)	^{144}Pr (17.3 m) daughter potential medical isotope
	Pr	2138	136 (ox)	Stable	
	Nd	7792	136 (ox)	Stable	
	Pm	52	-	^{147}Pm (2.62)	Potential betavoltaic source [87]
	Sm	1560	4 (ox)	^{151}Sm (90)	
	Eu	221	29 (ox)	^{154}Eu (8.6), ^{155}Eu (4.8)	
	Gd	326	69 (ox)	Stable	
	Tb	6	2 (ox)	Stable	
Plat. Grp. Metals	Ru	4470	19,000 (m)	^{106}Ru (1.02)	$^{106}\text{Ru}/^{106}\text{Rh}$ (30 s) potential medical isotope
	Rh	767	451,000 (m)	^{101}Rh (3.3), ^{102}Rh (0.56)	
	Pd	3126	61,000 (m)	^{107}Pd (6.5M)	
	Ag	162	1,000 (m)	$^{110\text{m}}\text{Ag}$ (0.68)	
Actinides	U	917,823	-	Various	Fissile/fertile
	Np	974	-	^{237}Np (2.1 M)	Precursor to ^{238}Pu for RTGs [29]
	Pu	13,700	-	Various	MOX fuel
	Am	860	-	^{241}Am	RTG isotope [52], precursor to ^{238}Pu for RTGs (via ^{242}Cm) [29]
	Cm	142	-	Various	Transmutation targets
Useful Isotopes	Cs	5035	-	^{134}Cs (2.1), ^{135}Cs (2.3 M) ^{137}Cs (30.1)	HHR, irradiation
	Sr	1518	-	^{90}Sr (28.9)	HHR, RTG isotope, ^{90}Y (64 h) daughter medical isotope
	Sn	105	-	^{126}Sn (218 k)	^{126}Sb (12.4 d) daughter potential medical isotope

References

- Murray, R. *Understanding Radioactive Waste*; Report PNL-3570; Pacific Northwest Lab.: Richland, WA, USA, 1981. [\[CrossRef\]](#)
- Baron, P.; Cornet, S.M.; Collins, E.D.; DeAngelis, G.; Del Cul, G.; Fedorov, Y.; Glatz, J.P.; Ignatiev, V.; Inoue, T.; Khaperskaya, A.; et al. A review of separation processes proposed for advanced fuel cycles based on technology readiness level assessments. *Prog. Nucl. Energy* **2019**, *117*, 24. [\[CrossRef\]](#)
- Bunn, M.; Holdren, J.P.; Fetter, S.; van der Zwaan, B. The Economics of Reprocessing versus Direct Disposal of Spent Nuclear Fuel. *Nucl. Technol.* **2005**, *150*, 209–230. [\[CrossRef\]](#)
- Kim, S.; Ko, W.; Nam, H.; Kim, C.; Chung, Y.; Bang, S. Statistical model for forecasting uranium prices to estimate the nuclear fuel cycle cost. *Nucl. Eng. Technol.* **2017**, *49*, 1063–1070. [\[CrossRef\]](#)
- Woo, S.M.; Chirayath, S.S.; Fuhrmann, M. Nuclear fuel reprocessing: Can pyro-processing reduce nuclear proliferation risk? *Energy Policy* **2020**, *144*, 111601. [\[CrossRef\]](#)
- Ho, S.S.; Leong, A.D.; Looi, J.; Chen, L.; Pang, N.; Tandoc, E. Science Literacy or Value Predisposition? A Meta-Analysis of Factors Predicting Public Perceptions of Benefits, Risks, and Acceptance of Nuclear Energy. *Environ. Commun.* **2018**, *13*, 457–471. [\[CrossRef\]](#)
- Bush, R.P. Recovery of Platinum Group Metals from High Level Radioactive Waste. *Plat. Group Rev.* **1991**, *35*, 202–208.
- Bourg, S.; Poinssot, C. Could spent nuclear fuel be considered as a non-conventional mine of critical raw materials? *Prog. Nucl. Energy* **2017**, *94*, 222–228. [\[CrossRef\]](#)

9. Koch, H.; Schober, A. On the possibilities of the extraction and application of low-activity fission products from high-activity nuclear fuel wastes. *Isotopenpraxis* **1976**, *12*, 49–56. (In German)
10. Lang, L.W.; Deonigi, D.E.; Rohrmann, C.A. Power Cost Reduction from Isotope Revenues. *Nucl. Appl.* **1967**, *3*, 665–678. [CrossRef]
11. European Chemical Society. Available online: <https://www.euchems.eu/euchems-periodic-table/> (accessed on 20 October 2022).
12. Kouloumpis, V.; Stamford, L.; Azapagic, A. Decarbonising electricity supply: Is climate change mitigation going to be carried out at the expense of other environmental impacts? *Sustain. Prod. Consum.* **2015**, *1*, 1–21. [CrossRef]
13. Bodel, W.; Hesketh, K.; McGlynn, G.; Matthews, J.; Butler, G. Generic Feasibility Assessment: Helping to Choose the Nuclear Piece of the Net Zero Jigsaw. *Energies* **2021**, *14*, 1229. [CrossRef]
14. Poinssot, C.; Rostaing, C.; Greandjean, S.; Boullis, B. Recycling the Actinides, The Cornerstone of Any Sustainable Nuclear Fuel Cycles. *Procedia Chem.* **2012**, *7*, 349–357. [CrossRef]
15. Eccles, H. Nuclear fuel cycle technologies -sustainable in the twenty first century? *Solvent Extr. Ion Exch.* **2000**, *18*, 633–654. [CrossRef]
16. Rodríguez-Penalonga, L.; Soria, B.Y.M. A Review of the Nuclear Fuel Cycle Strategies and the Spent Nuclear Fuel Management Technologies. *Energies* **2017**, *10*, 1235. [CrossRef]
17. Taylor, R.; Bodel, W.; Stamford, L.; Butler, G. A Review of Environmental and Economic Implications of Closing the Nuclear Fuel Cycle—Part One: Wastes and Environmental Impacts. *Energies* **2022**, *15*, 1433. [CrossRef]
18. Taylor, R.; Bodel, W.; Butler, G. A Review of Environmental and Economic Implications of Closing the Nuclear Fuel Cycle—Part Two: Economic Impacts. *Energies* **2022**, *15*, 2472. [CrossRef]
19. Baker, A.; Fells, A.; Carrott, M.J.; Maher, C.J.; Hanson, B.C. Process intensification of element extraction using centrifugal contactors in the nuclear fuel cycle. *Chem. Soc. Rev.* **2022**, *51*, 3964–3999. [CrossRef]
20. Fernández-Arias, P.; Vergara, D.; Orosa, J.A. A Global Review of PWR Nuclear Power Plants. *Appl. Sci.* **2020**, *10*, 4434. [CrossRef]
21. Forsberg, C.W. Rethinking High-Level Waste Disposal: Separate Disposal of High-Heat Radionuclides (^{90}Sr and ^{137}Cs). *Nucl. Technol.* **2000**, *131*, 252–268. [CrossRef]
22. Krall, L.; Macfarlane, A. Burning waste or playing with fire? Waste management considerations for non-traditional reactors. *Bull. At. Sci.* **2018**, *74*, 326–334. [CrossRef]
23. Mallants, D.; Travis, K.; Chapman, N.; Brady, P.V.; Griffiths, H. The State of the Science and Technology in Deep Borehole Disposal of Nuclear Waste. *Energies* **2020**, *13*, 833. [CrossRef]
24. Holdsworth, A.F.; George, K.; Adams, S.J.; Sharrad, C.A. An accessible statistical regression approach for the estimation of spent nuclear fuel compositions and decay heats to support the development of nuclear fuel management strategies. *Prog. Nucl. Energy* **2021**, *141*, 103935. [CrossRef]
25. Francis, M.W.; Weber, C.F.; Pigni, M.T.; Gauld, I.C. *Reactor Fuel Isotopics and Code Validation for Nuclear Applications*; National Technical Information: Springfield, VA, USA, 2015. [CrossRef]
26. Ando, Y.; Takano, H. *Estimation of LWR Spent Fuel Composition, Report: JAERI-Research-99-004*; Japanese Atomic Energy Research Agency: Tokai, Japan, 1999.
27. Nash, K.L.; Lumetta, G.J. (Eds.) *Advanced Separation Techniques for Nuclear Fuel Reprocessing and Radioactive Waste Treatment*; Woodhead: Oxford, UK, 2011. [CrossRef]
28. Salvatores, M.; Palmiotti, G. Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges. *Prog. Part. Nucl. Phys.* **2011**, *66*, 144–166. [CrossRef]
29. Dustin, J.S.; Borrelli, R. Assessment of alternative radionuclides for use in a radioisotope thermoelectric generator. *Nucl. Eng. Des.* **2021**, *385*, 111475. [CrossRef]
30. Vesnovskii, S.P. RFNC-VNIIEF capabilities to production high pure isotopes for scientific and medical applications. *J. Radioanal. Nucl. Chem.* **2003**, *257*, 27–31. [CrossRef]
31. Rirnbauer, E.R.; Fassbender, M.E.; Ferrier, M.G.; John, K.D.; Mastren, T. Actinides in Medicine. In *The Heaviest Metals: Science and Technology of The Actinides and Beyond*; Evans, W.J., Hanusa, T.P., Eds.; Wiley: Oxford, UK, 2007; pp. 445–466.
32. Ternovyykh, M.; Tikhomirov, G.; Saldikov, I.; Gerasimov, A. Decay heat power of spent nuclear fuel of power reactors with high burnup at long-term storage. *EPJ Web Conf.* **2017**, *153*, 7035. [CrossRef]
33. Johnson Matthey. Available online: <https://matthey.com/products-and-markets/pgms-and-circularity/pgm-management> (accessed on 29 October 2022).
34. Swain, P.; Mallika, C.; Srinivasan, R.; Mudali, U.K.; Natarajan, R. Separation and recovery of ruthenium: A review. *J. Radioanal. Nucl. Chem.* **2013**, *298*, 781–796. [CrossRef]
35. Moeyaert, P.; Miguiditchian, M.; Masson, M.; Dinh, B.; Hérès, X.; De Sio, S.; Sorel, C. Experimental and modelling study of ruthenium extraction with tri-*n*-butylphosphate in the purex process. *Chem. Eng. Sci.* **2017**, *158*, 580–586. [CrossRef]
36. Pe’er, J. Ruthenium-106 brachytherapy. In *Current Concepts in Uveal Melanoma*; Jager, M.J., Desjardins, L., Kivela, T., Damato, B.E., Eds.; Karger: Basel, Switzerland, 2012; Volume 49, pp. 27–40. [CrossRef]
37. Allison, W. We Should Stop Running Away from Radiation. *Philos. Technol.* **2011**, *24*, 193–195. [CrossRef]
38. Adams, B.D.; Chen, A. The role of palladium in a hydrogen economy. *Mater. Today* **2011**, *14*, 282–289. [CrossRef]
39. SMM. Available online: <https://www.metal.com/Rare-Earth-Oxides> (accessed on 29 October 2022).
40. Natrajan, L.S.; Langford Paden, M.H. *f-block Elements Recovery*, in *RSC Green Chemistry No. 22—Element Recovery and Sustainability*; Hunt, A.J., Ed.; Royal Society of Chemistry: Cambridge, UK, 2013; Chapter 6.

41. Asadollahzadeh, M.; Torkaman, R.; Torab-Mostaedi, M. Extraction and Separation of Rare Earth Elements by Adsorption Approaches: Current Status and Future Trends. *Sep. Purif. Rev.* **2020**, *50*, 417–444. [\[CrossRef\]](#)
42. Chen, Z.; Li, Z.; Chen, J.; Kallem, P.; Banat, F.; Qiu, H. Recent advances in selective separation technologies of rare earth elements: A review. *J. Environ. Chem. Eng.* **2021**, *10*, 107104. [\[CrossRef\]](#)
43. Opore, E.O.; Struhs, E.; Mirkouei, A. A comparative state-of-technology review and future directions for rare earth element separation. *Renew. Sustain. Energy Rev.* **2021**, *143*, 110917. [\[CrossRef\]](#)
44. Tingey, G.L.; McClanahan, E.D.; Bayne, M.A.; Gray, W.J.; Hinman, C.A. *Krypton-85 Storage in Solid Matrices in Scientific Basis for Nuclear Waste Management*; Northrup, C.J.M., Ed.; Springer: New York, NY, USA, 1980; Chapter 18.
45. Chemistry Learner. Available online: <https://www.chemistrylearner.com/krypton-85.html> (accessed on 29 October 2022).
46. Aisen, E.M.; Borisevich, V.D.; Levin, E.V.; Yumatov, S.V.; Tikhomirov, A.V.; Popov, G.E. Separation of stable isotopes of low, middle, and heavy masses by a gas centrifuge. In Proceedings of the Workshop on Separation Phenomena in Liquids and Gases, Foz do Iguaçu, Brazil, 22–26 September 1996.
47. Marty, B. On the noble gas isotopic fractionation in naturally occurring gases. *Geochem. J.* **1984**, *18*, 157–162. [\[CrossRef\]](#)
48. Williams, S.J.; Rilling, W.S.; White, S.B. Quality of Life and Cost Considerations: Y-90 Radioembolization. *Semin. Interv. Radiol.* **2021**, *38*, 482–487. [\[CrossRef\]](#)
49. Katsaros, N.; Anagnostopoulou, A. Rhodium and its compounds as potential agents in cancer treatment. *Crit. Rev. Oncol.* **2002**, *42*, 297–308. [\[CrossRef\]](#)
50. Robertson, A.K.H.; Kunz, P.; Hoehr, C.; Schaffer, P. Nuclide production cross sections from irradiation of thorium by 438 MeV protons and a comparison to fluka and geant4 simulations. *Phys. Rev. C* **2020**, *102*, 044613. [\[CrossRef\]](#)
51. Amoroso, A.J.; Fallis, I.A.; Pope, S.J. Chelating agents for radiolanthanides: Applications to imaging and therapy. *Coord. Chem. Rev.* **2017**, *340*, 198–219. [\[CrossRef\]](#)
52. Murayama, Y.; Feloa, J.M.; Beach, J.L. HeLa cell tumor response to ⁶⁰Co, Cs-137, Cf-252 radiations and cisplatin chemotherapy in nude mice. *Cancer* **1984**, *52*, 247–252. [\[CrossRef\]](#)
53. Mikhalev, A.V.; Chernov, D.O.; Korobeinikov, V.V. Use of Am-241 in RTGs. *J. Phys. Conf. Ser.* **2020**, *1689*, 012063. [\[CrossRef\]](#)
54. Bykov, A.S.; Malinkovich, M.D.; Kubasov, I.V.; Kislyuk, A.M.; Kiselev, D.A.; Ksenich, S.V.; Zhukov, R.N.; Temirov, A.A.; Chichkov, M.V.; Polisan, A.A.; et al. Application of Radioactive Isotopes for Beta-Voltaic Generators. *Russ. Microelectron.* **2017**, *46*, 527–539. [\[CrossRef\]](#)
55. Boisseau, P. Food Preservation by Irradiation: A Brief Introduction. *Solid State Phenom.* **1992**, *30–31*, 433–450. [\[CrossRef\]](#)
56. Cao, Y.; Zhou, L.; Ren, H.; Zou, H. Determination, Separation and Application of ¹³⁷Cs: A Review. *Int. J. Environ. Res. Pub. Health* **2022**, *19*, 10183. [\[CrossRef\]](#) [\[PubMed\]](#)
57. McKibben, J.M. Chemistry of the Purex Process. *Radiocim. Acta* **1984**, *36*, 3–15. [\[CrossRef\]](#)
58. Bond, G.; Eccles, H.; Kavi, P.C.; Holdsworth, A.F.; Rowbotham, D.; Mao, R. Removal of Cesium from Simulated Spent Fuel Dissolver Liquor. *J. Chromatog. Separ. Tech.* **2019**, *10*, 417.
59. Holdsworth, A.F.; Eccles, H.; Rowbotham, D.; Bond, G.; Kavi, P.C.; Edge, R. The Effect of Gamma Irradiation on the Ion Exchange Properties of Caesium-Selective Ammonium Phosphomolybdate-Polyacrylonitrile (AMP-PAN) Composites under Spent Fuel Recycling Conditions. *Separations* **2019**, *6*, 23. [\[CrossRef\]](#)
60. Holdsworth, A.F.; Eccles, H.; Rowbotham, D.; Brookfield, A.; Collison, D.; Bond, G.; Kavi, P.C.; Edge, R. The Effect of Gamma Irradiation on the Physicochemical Properties of Caesium-Selective Ammonium Phosphomolybdate-Polyacrylonitrile (AMP-PAN) Composites. *Clean Technol.* **2019**, *11*, 20. [\[CrossRef\]](#)
61. Holdsworth, A.F.; Eccles, H.; Bond, G.; Rowbotham, D.; Brookfield, A.; Collison, D.; Kavi, P.C.; Natrajan, L.S.; Spencer, B.; Murny, C.; et al. Heterogeneous Separations of Highly Active Radionuclides for Advanced Decay Heat & Waste Management in Next-Generation Spent Fuel Recycling/Reprocessing. In Proceedings of the 4th Cloud Conference on Nuclear Waste Management and Disposal, Online, UK, 9 July 2020.
62. Venkatesan, K.A.; Sukumaran, V.; Antony, M.P.; Srinivasan, T.G. Studies on the feasibility of using crystalline silicotitanates for the separation of cesium-137 from fast reactor high-level liquid waste. *J. Radioanal. Nucl. Chem.* **2009**, *280*, 129–136. [\[CrossRef\]](#)
63. Bibler, N.E. *Results of Scoping Studies for Determining Radiolytic Hydrogen Production from Moist CST and CST Slurries*; Report WRSC-RP-98-01143; Savannah River Site: Jackson, SC, USA, 1998. [\[CrossRef\]](#)
64. Banks, H.O., Jr. *The Cesium-137 Power Program*; Quarterly Report No. 3, Report TID-17004; Royal Research Corp.: Hayward, CA, USA, 1961. [\[CrossRef\]](#)
65. Amphlett, J.; Ogden, M.; Foster, R.; Syna, N.; Soldenhoff, K.H.; Sharrad, C.A. The effect of contaminants on the application of polyamine functionalised ion exchange resins for uranium extraction from sulfate based mining process waters. *Chem. Eng. J.* **2018**, *354*, 633–640. [\[CrossRef\]](#)
66. Sole, K.C.; Cole, P.M.; Feather, A.M.; Kotze, M.H. Solvent Extraction and Ion Exchange Applications in Africa's Resurging Uranium Industry: A Review. *Solvent Extr. Ion Exch.* **2011**, *29*, 868–899. [\[CrossRef\]](#)
67. Rahman, R.O.A.; Metwally, S.S.; El-Kamash, A.M. Life Cycle of Ion Exchangers in Nuclear Industry: Application and Management of Spent Exchangers. In *Handbook of Ecomater*; Martinez, L., Kharissova, O., Kharisov, B., Eds.; Springer: New York, NY, USA, 2019. [\[CrossRef\]](#)
68. Choi, E.-Y.; Jeong, S.M. Electrochemical processing of spent nuclear fuels: An overview of oxide reduction in pyroprocessing technology. *Prog. Nat. Sci.* **2015**, *25*, 572–582. [\[CrossRef\]](#)

69. Carlson, K.; Gardner, L.; Moon, J.; Riley, B.; Amoroso, J.; Chidambaram, D. Molten salt reactors and electrochemical reprocessing: Synthesis and chemical durability of potential waste forms for metal and salt waste streams. *Int. Mater. Rev.* **2020**, *66*, 339–363. [\[CrossRef\]](#)
70. Yoshida, Z.; Aoyagi, H.; Mutoh, H.; Takeishi, H.; Sasaki, Y.; Uno, S.; Tachikawa, E. Spent fuel reprocessing based on electrochemical extraction process (SREEP). *J. Alloys Compd.* **1994**, *213–214*, 453–455. [\[CrossRef\]](#)
71. Shadrin, A.Y.; Dvoeglazov, K.N.; Mochalov, Y.S.; Vidanov, V.V.; Kashcheev, V.A.; Terentiev, A.G.; Gerasimenko, M.N.; Cheshuyakov, S.A. Hydrometallurgical and combined technologies fast reactor MNUP and MOX UNF reprocessing. *J. Phys. Conf. Ser.* **2020**, *1475*, 012021. [\[CrossRef\]](#)
72. Authen, T.L.; Adnet, J.-M.; Bourg, S.; Carrott, M.; Ekberg, C.; Galán, H.; Geist, A.; Guilbaud, P.; Miguiditchian, M.; Modolo, G.; et al. An overview of solvent extraction processes developed in Europe for advanced nuclear fuel recycling, Part 2—Homogeneous recycling. *Sep. Sci. Technol.* **2021**, *57*, 1724–1744. [\[CrossRef\]](#)
73. Geist, A.; Adnet, J.-M.; Bourg, S.; Ekberg, C.; Galán, H.; Guilbaud, P.; Miguiditchian, M.; Modolo, G.; Rhodes, C.; Taylor, R. An overview of solvent extraction processes developed in Europe for advanced nuclear fuel recycling, part 1—Heterogeneous recycling. *Sep. Sci. Technol.* **2020**, *56*, 1866–1881. [\[CrossRef\]](#)
74. Nash, K.L. The Chemistry of TALSPEAK: A Review of the Science. *Solvent Extr. Ion Exch.* **2014**, *33*, 1–55. [\[CrossRef\]](#)
75. Taylor, R.; Mathers, G. Developments in Advanced Recycle and Sustainability for Future Fuel Cycle Options. In Proceedings of the SNETP Forum, Online, 3 February 2021.
76. Modolo, G.; Wilden, A.; Geist, A.; Magnusson, D.; Malmbeck, R. A review of the demonstration of innovative solvent extraction processes for the recovery of trivalent minor actinides from PUREX raffinate. *Radiochim. Acta* **2012**, *100*, 715–725. [\[CrossRef\]](#)
77. Wu, H.; Kim, S.-Y.; Takahashi, T.; Oosugi, H.; Ito, T.; Kanie, K. Extraction behaviors of platinum group metals in simulated high-level liquid waste by a hydrophobic ionic liquid bearing an amino moiety. *Nucl. Eng. Technol.* **2021**, *53*, 1218–1223. [\[CrossRef\]](#)
78. Malmbeck, R.; Magnusson, D.; Bourg, S.; Carrott, M.; Geist, A.; Heres, X.; Miguiditchian, M.; Modolo, G.; Mullich, U.; Sorel, C.; et al. Homogenous recycling of transuranium elements from irradiated fast reactor fuel by the EURO-GANEX solvent extraction process. *Radiochim. Acta.* **2019**, *107*, 917–929. [\[CrossRef\]](#)
79. Van Loon, J.C. Accurate determination of the noble metals I. Sample decomposition and methods of separation. *TrAC Trends Anal. Chem.* **1984**, *3*, 272–275. [\[CrossRef\]](#)
80. Holdsworth, A.F.; Eccles, H.; Bond, G.; Sharrad, C.A. Selective Separation of Resources in Used Nuclear Fuel (UNF) Recycle for Fuel Cycle Sustainability. In Proceedings of the 51st Journées des Actinides Conference, Santa Margherita Ligure, Italy, 9–14 April 2022.
81. Xu, C.; Wang, J.; Chen, J. Solvent Extraction of Strontium and Cesium: A Review of Recent Progress. *Solvent Extr. Ion Exch.* **2012**, *30*, 623–650. [\[CrossRef\]](#)
82. Vincent, T.; Vincent, C.; Guibal, E. Immobilization of Metal Hexacyanoferrate Ion-Exchangers for the Synthesis of Metal Ion Sorbents—A Mini-Review. *Molecules* **2015**, *20*, 20582–20613. [\[CrossRef\]](#) [\[PubMed\]](#)
83. Baetsle, L.; Huys, D. Structure and ion-exchange characteristics of polyantimonic acid. *J. Inorg. Nucl. Chem.* **1968**, *30*, 639–649. [\[CrossRef\]](#)
84. Nilchi, A.; Maragheh, M.G.; Khanchi, A.; Farajzadeh, M.A.; Aghaei, A.A. Synthesis and ion-exchange properties of crystalline titanium and zirconium phosphates. *J. Radioanal. Nucl. Chem. Artic.* **2004**, *261*, 393–400. [\[CrossRef\]](#)
85. Natarajan, R.; Raj, B. Fast Reactor Fuel Reprocessing Technology in India. *J. Nucl. Sci. Technol.* **2007**, *44*, 393–397. [\[CrossRef\]](#)
86. *Spent Fuel Reprocessing Options*; Report: IAEA-TECDOC-1587; IAEA (International Atomic Energy Agency): Vienna, Austria, 2008.
87. Sachenko, A.; Shkrebtii, A.; Korkishko, R.; Kostilyov, V.; Kulish, M.; Sokolovskyi, I. Efficiency analysis of betavoltaic elements. *Solid-State Electron.* **2015**, *111*, 147–152. [\[CrossRef\]](#)

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