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Hypothesis

Spent Nuclear Fuel–Waste to Resource, Part 2: Radiolytic Hydrogen Production—A Novel Approach to Sustainably Meeting the World's Hydrogen Needs from the Nuclear Fuel Cycle

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Abstract

Nuclear cogeneration is increasingly proposed to support decarbonisation, partly via hydrogen (H_2) production through electrolysis, thermolysis, or a combination of these, but wide-scale adoption is limited by process inefficiencies, high costs, and concerns around nuclear wastes. Recent work has demonstrated that gamma radiation, sourced from ^{137}Cs, splits water into H_2 at high yields with a TiO_2 photocatalyst, extrapolating that the world's spent nuclear fuel (SNF) stockpiles could generate ~60% of world hydrogen demand, worth ~90 billion USD/y. This is achievable using present technology and would valorise SNF, which is commonly viewed as a waste, and decouple H_2 production from reactor operations, but would limit operations to open fuel cycles, replacing the cooling ponds therein. By combining advanced separations in SNF recycle, partitioning the high-heat radionuclides (^{137}Cs, ^{134}Cs, and ^{90}Sr) responsible for most SNF radiation, and forming these into dedicated sources for hydrogen production, a closed fuel cycle option is afforded. This would lower remaining SNF recycle raffinates to intermediate-level waste and free the actinides for further energy generation, turning nuclear wastes from liabilities into resources. In this publication, we discuss the scientific, technological, and regulatory development needed to realise this concept in full. As this H_2 production approach is distinct from others on the "colour spectrum", we propose to call radiolytically-produced H_2 "platinum hydrogen".

Keywords: nuclear waste valorisation; waste-to-resource; nuclear cogeneration; fission product separation; spent fuel recycle; radiolytic hydrogen production; gamma radiolysis; photocatalytic water splitting; low-carbon hydrogen

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1. Introduction and Context

Co-generation or cogeneration (CoGen) is defined as the exploitation of both heat and electrical power from energy-generating sources [1,2], including the recovery of waste heat and combined heat and power (CHP) [3]. In a nuclear context, this means the use of heat produced from reactors for various processes such as saltwater desalination, district and industrial heating, and hydrogen (H_2) production, in addition to conventional electricity generation (Figure 1) [1,2]. For the purposes of this work, we shall designate this concept as reactor co-generation (RCG). The primary goal of RCG is the decarbonisation of carbon-intensive operations currently driven by fossil fuels, by instead employing the vast amounts of heat generated within nuclear reactors [4], in addition to conventional baseload power generation [2]. Secondary goals include load following more transient renewables such as wind and solar [1], and the flexible use of excess electrical or thermal output for other cogeneration applications (including hydrogen production) where renewables are generating at scale [4–8]. Both conventional, large and small modular reactors (SMRs) are noted as being suitable for this concept [9,10], sometimes where these are coupled with high-temperature energy storage for alternative pathways to load following [11]. In concert with nuclear energy, hydrogen is viewed as the optimal energy storage medium as a zero-carbon alternative to fossil fuels [12,13]. It has been forecast that hydrogen and derivatives will make up 12% of worldwide energy consumption by 2050 [14].

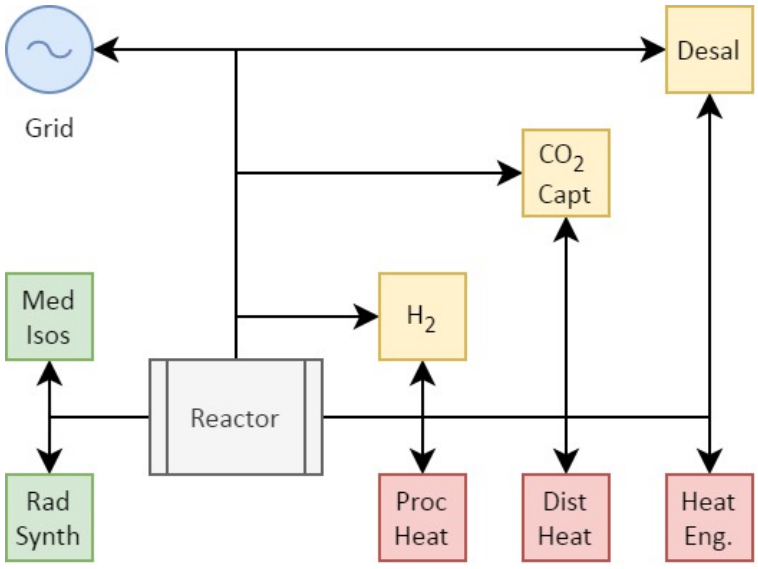


Figure 1. Proposed applications of co-generation from nuclear reactors: radiation used for medical isotopes (Med Isos) or radiation synthesis (Rad Synth), direct heat used to power industrial process heat (Proc Heat), district heating (Dist Heat) or heat engines (Heat Eng., e.g., Sterling engines), and either heat or electrical output used for H_2 production, CO_2 capture (Capt), or saltwater desalination (Desal) in addition to conventional grid outputs. Key: blue = electrical power only; amber = heat or electrical use; red = heat only use; green = radiation use. Adapted from [1].

Despite RCG's potential, several drawbacks have limited uptake beyond small-scale test beds. Firstly, most current reactors are water-cooled types with limited output temperature (≤335 °C), well below the minimum temperatures required to drive many industrial processes [1,3]. Consequently, such high-temperature heat must be provided by resistive heating, with 60–70% losses in converting reactor heat to electricity. Secondly, the advanced, high-temperature reactors capable of driving thermally demanding industrial processes (T > 700 °C [8]) are developmental at the time of writing, with lead times of a decade or more before construction starts, except for a single prototype and twin-reactor first-of-a-kind

station operating/under construction in China [15]. In any case, some of these designs have issues with the long-term sustainability of their fuel cycle, as their spent nuclear fuel (SNF) cannot be readily recycled and lacks turnkey disposal options for open cycles [16]. Regardless, these designs have historically proven troublesome in operation [16,17], with further potential concerns surrounding the development and efficacy of thermochemical cycles for H_2 production in time for high-temperature gas-cooled reactor HTGR deployment [18–21]. This means that even HTGRs will likely rely on electrolysis for hydrogen production, albeit with a significant increase in thermal efficiency over pressurised water reactors (PWRs). Other technologies driven by HTGRs, such as methane pyrolysis (with a carbon by-product) and high-temperature steam electrolysis using solid oxide fuel cells, are under consideration [22]. The round-trip efficiencies of nuclear-electrolysis hydrogen production, factoring in compression/liquefaction for storage, are overall low, around 20–30%.

Realising fuel cycle opportunities will be key to the long-term sustainability of nuclear fission; in this context, the definition of sustainability is the ability to maintain the NFC in the long term (centuries or millennia) [23]. In addition to RCG proposals, increased interest in fission more generally has arisen given urgent imperatives to decarbonise in the face of rapidly accelerating climate change, with ~50 new reactors under construction at the time of writing and many more planned [24]. This will increase demands on finite natural uranium (and thorium) reserves [25] already threatened by recent geopolitical shifts, especially if fuel continues to be utilised only once in the open fuel cycle (Figure 2A) [16]. Closing the fuel cycle by recycling spent nuclear fuel (SNF) to recover the fissile and fertile actinides (U and Pu, Figure 2B, or with the addition of the minor actinides (MAs-Np, Am, Cm), Figure 2C) for further energy generation can greatly increase sustainability [26]. This sustainability improvement is achieved by reducing the environmental demands of mining, as well as by decreasing waste volumes for disposal. Furthermore, careful selection of separation and recycling procedures can reduce radiotoxicity and duration of post-disposal hazards markedly [26,27]. The uptake of SNF recycle is limited, however, due to high costs, proliferation concerns, and historical lack of commercial necessity in the form of low U prices and/or demand for MOX (mixed oxide) fuel [16].

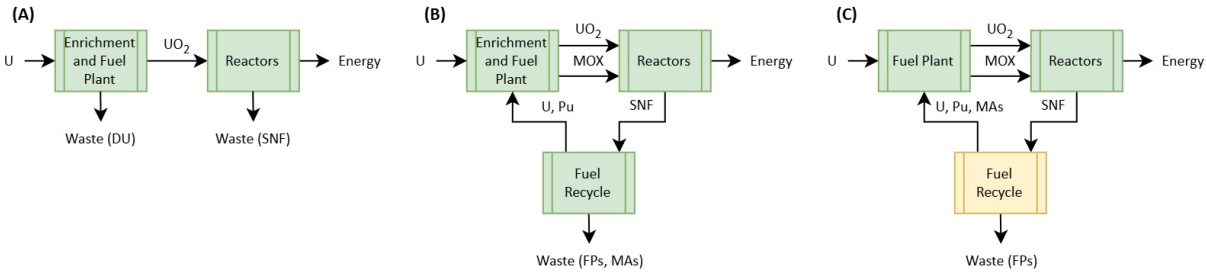


Figure 2. Current and proposed NFC implementations: (**A**) open fuel cycle; (**B**) partially-closed fuel cycle; and (**C**) fully-closed fuel cycle with MA recycle. Key: green = well-developed or operating technology; amber = technology under development. DU = depleted uranium.

More recent proposals for recovery of valuable fission products (FPs) such as the platinum group metals (Ru, Rh, Pd, (and Ag)), rare earth elements (Y, La-Lu), and noble gases (He, Kr, Xe), alongside useful isotopes from SNF during recycle would further increase nuclear fuel cycle (NFC) sustainability. These separations would further reduce waste volumes, whilst offering direct value recovery to offset the high costs of fuel recycle [16,27–34];. These are in addition to further refinements to SNF recycle developed in recent decades [35]. Around 300,000 USD of valuable FPs could be recovered from each ton of SNF, representing an untapped stream of valuable, naturally-scarce resources [27,28,36].

Separation of the high-heat radionuclides (HHRs-^{137}Cs, ^{134}Cs, and ^{90}Sr/^{90}Y as high-yielding fission products [37]) has been proposed several times in the literature [38,39]. This includes in service of advanced waste management [40–42], and more recently in some of the authors' research for additional mitigation of operation challenges in SNF recycle [37,43–50]. This could reduce all other outputs of recycle operations from high-level to intermediate-level wastes [27] at the expense of producing a "screaming hot" Cs-Sr wasteform. This waste stream would then either be stored pending decay or disposed of in a dedicated repository. If these materials could be utilised, however, their disposal would become a secondary consideration, and the radiotoxicity of remaining NFC outputs would reduce to a fraction of present-day values. The use of neutron activation products (e.g., ^{60}Co) could also be considered for waste valorisation; producing these intentionally in a reactor would otherwise negatively affect the neutron economy of the fuel cycle and should ideally be avoided [32].

2. Putting "Waste" Radiation to Use

Although the generation of hydrogen by radiolysis of water has long been recognised as a problematic side-effect of radioactive materials [4,12,51–53], research has proposed harnessing this for energy generation since the 1970s [12,13,51,54–60], a concept we shall henceforth call radiolytic hydrogen production (RHP). The most interesting and likely practical of these concepts proposes using individual SNF assemblies stored in "silos" filled with water containing a TiO_2 photocatalyst to increase the efficiency of this process (Figure 3).

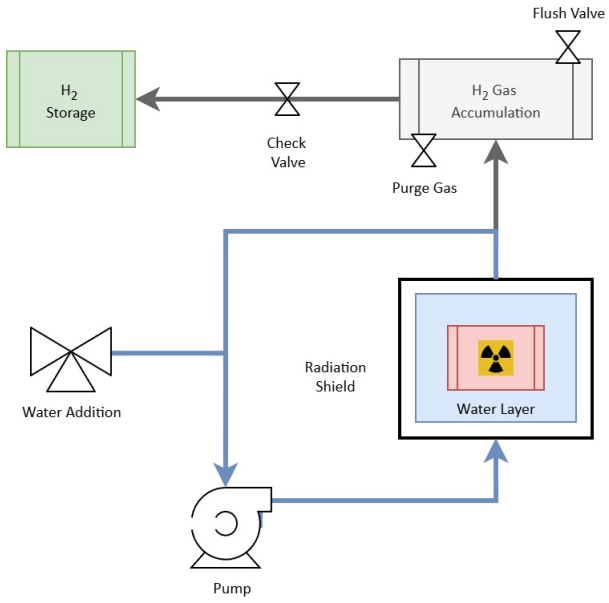


Figure 3. Overview schematic of Vandenborre's proposed hydrogen production using SNF bundles. Adapted from [56].

This would function by harnessing the intense gamma emissions arising from this material [56], most of which are produced by Cs isotopes, and are reported to function by harnessing photoelectric and Compton effects [55–57]. Vandenborre et al. calculated that the current worldwide stockpile of SNF (390,000 t as of 2017) could produce at least 42.9 MtH_2/y (millions of tons of H_2 per year; 60% of world demand) via this approach, or 110 tH_2/tSNF.y (tons of hydrogen per ton of spent fuel per year) in what they reported to be a technologically simple system. This is based on the extrapolation from their reported experimental H_2 production arising from dose rates of 0.42 kGy/h delivered by a ^{137}Cs

source, to a predicted 28 kGy/h from SNF [56]. This remains to be validated independently and has not been verified against a first-principles energy balance, both factors which are planned for future works. We assume that these values are based on the bulk of the world SNF stockpile being both longer-cooled and relatively low burnup; SNF from modern, Gen III(+) reactors will be both higher burnup and shorter cooled, and thus potentially much more effective for H_2 production. We use the conservative values presented for discussions here until this can be more accurately modelled. The radiolysis of water itself is a complex process, producing a range of reactive species in addition to H_2 and H_2O_2, the minutiae of which, including studies with other photocatalysts, have been reported extensively elsewhere and are beyond the scope of this work [12,13,55–60].

In fuel cycle terms, we view this approach as a replacement for traditional SNF cooling ponds. Hydrogen produced in this manner would be essentially passive and "free" as natural radiolysis is exploited to harness energy that is otherwise wasted as heat. In contrast, to produce the same volume of H_2 from current light water reactors, driving electrolysis would require around 164 large current-generation fission plants such as EPRs (European Pressurised water Reactors) devoting their entire electrical output to this operation. This assumes an electrical output of 1650 MWe and a conversion efficiency of 70%, taking 55 kWh to produce 1 kg of H_2, and represents around half of the installed fission-generating capacity at the time of writing.

The H_2 produced from SNF in this manner could be used to generate zero-carbon energy (electricity generation or heating) or for other industrial uses. However, this approach would essentially "lock out" the actinides present in such SNF from fuel cycle use and thus be less sustainable than a closed fuel cycle [26], although this would be beneficial from criticality and security standpoints if individual SNF bundles are used in isolated silos. This is dependent on the availability of fissile and fertile materials from existing stockpiles and the primary economic drivers to reuse U and Pu as MOX, and/or the MAs for waste management purposes. Preliminary results indicate that single assemblies of even fresh fuel in such a silo (representing a severe maloperation) would be sufficiently subcritical ($k_{eff} = 0.832$) to be satisfactorily safe; this can be reduced further by the addition of neutron absorbers to the system (See Appendix A). Other applications for radiation, such as the conversion of bio-feedstocks [61], polymer grafting and cross-linking [62], medical device sterilisation [63], and water purification [64], could also be harnessed.

While it has been demonstrated that gamma radiation, produced by ^{137}Cs, ^{134}Cs, and the like, can effectively produce H_2 from water at significantly enhanced yields (relative to pure water alone) in the presence of a photocatalyst such as TiO_2, the same effects have yet to be verified for beta radiation, although the effects should be similar to gamma [65,66]. The most prominent source of this in SNF is ^{90}Sr, which could further increase the theoretical H_2 yields if effective with the same photocatalyst as demonstrated for gamma radiation. As TiO_2 likely enhances H_2 production yields in water by greater conversion of gamma rays into energetic electrons than the photoelectric and Compton effects, this would seem likely [67]. These, amongst many other optimisations of the system, remain to be explored, as was noted by the authors of that study [56] and other previous proposals [58]. As alpha emitters deposit the majority of their energy within a short range of the source, these are not considered for hydrogen production here. Long-term studies will also be essential to validate catalyst stability under intense radiation fields, cladding corrosion/erosion, water chemistry, decay heat removal, shielding, and many other safety considerations before RHP can be implemented at scale.

3. Recovering and Using the HHRs for H_2 Production

A combination of our own previous work in HHR separations and the H_2 production proposed by Vandenborre et al. could provide a revolutionary approach to the NFC with numerous, unrealised benefits. In contrast to the open fuel cycle (Figure 4A), closed fuel cycle (Figure 4B), and Vandenborre's proposal (Figure 4C), we would combine the closed fuel cycle with two stages of hydrogen production (Figure 4D). Firstly, SNF would be cooled on site at the reactor until it can be safely shipped to a cooling pond. At this time, we would instead transfer individual SNF bundles to individual silos, using their intense radiation fields to produce large quantities of H_2.

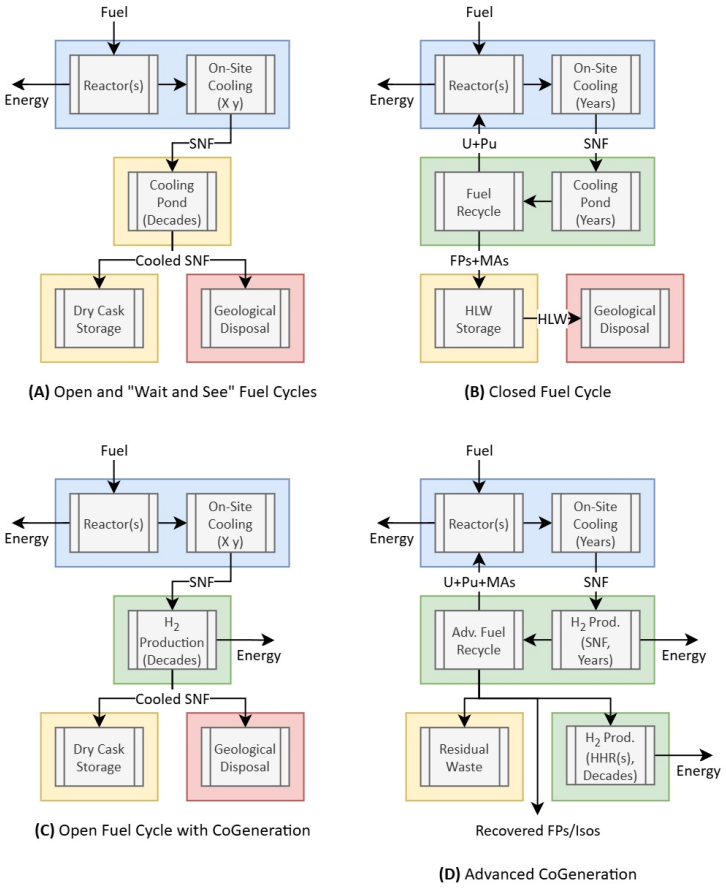


Figure 4. Simplified schematics depicting material flows and energy outputs in open and "wait and see" NFCs (**A**), closed fuel cycles (**B**), open and "wait and see" fuel cycles with RHP (**C**), and finally closed fuel cycles with RHP (**D**).

Once the SNF bundles have cooled sufficiently to permit it (typically 5–10 years depending upon burnup), they would be recycled. This would enable the recovery of the U, Pu, and MAs for further energy generation, ideally alongside valuable FPs, the HHRs, and other useful isotopes. The HHRs would then be processed and concentrated into suitable sources for long-term H_2 production and eventual disposal, returned to the silos or similar apparatus, and retained in-place until radiolysis is no longer effective. The HHRs could be further recycled or "refreshed" midlife to increase activity.

The rationale behind this approach is that most of the radiolysis will arise from only a few kg of Cs and perhaps Sr present within each tonne of SNF [40–42]. By separating these sources from SNF during recycle operations, the actinides and other valuable FP materials are also released for further energy generation and use, respectively. The remaining HHRs would then be converted from a hazardous waste into potentially one

of the most valuable components of SNF and no longer require separate disposal. As demonstrated by our previous work, separating the HHRs from SNF during recycle results in all remaining raffinates being reduced to intermediate-level wastes rather than high-level [27]; this introduces new options in waste disposal and reduces, if not entirely eliminates, the necessity for a geological disposal facility. As demonstrated in our previous work [27], removing Cs and Sr reduces HLW decay heat by 75–80% for 5–10-year-cooled high-burnup PWR SNF, while mass and waste volumes are only modestly reduced. As geological repository design is largely heat-limited, this supports the potential for denser disposal routes.

An intermediate approach, whereby U and the HHRs (and possibly valuable FPs) are recovered from SNF in a pseudo-recycle process to reduce waste volume, could provide a proliferation-resistant fuel cycle approach for nations where such prohibitions exist. It should also be noted that separations for Cs, Sr, and other HHRs would not be isotopically selective, and would also co-extract stable and long-lived species (e.g., ^{133}Cs and ^{135}Cs) alongside the desired radioactive species. The flexibility of this approach affords options for both open and closed fuel cycles, where national regulations prohibit SNF recycle in the latter case. Under the greatest levels of prohibition, H_2 production would continue using SNF bundles until no longer viable, at which point these could be disposed of geologically. Such HHR recovery is also only applicable to new-build SNF recycle facilities, and such a lead time of decades is likely the primary practical barrier, not accounting for economic or regulatory ones, for the full implementation of the concept presented here.

4. Market Potential and Environmental Implications

The market potential for RHP is significant, based on initially available data. From the conservative values utilised by Vandenborre et al. [56] and the potential system optimisations that could be realised, each tonne of SNF, or the HHRs present therein, would produce 110 tH_2 annually, decreasing slightly each year due to radioactive decay. This corresponds to an annual return of 220,000 USD per t_{SNF} at a base price (for H_2 produced by fossil fuels, gas price valuation after the 2008 crisis recovery) of 2 USD/kg of H_2 [68], although prices are often variable and in the EU are often higher than this [19]. For the world SNF stockpile (390 kt), this corresponds to a potential income of 85.8b USD per year, generating 42.9 Mt per year, representing ~60% of world demand in 2024 [56].

While the fuel cycle and other processing costs of RHP are not currently well understood, the potentially low operating costs arising from the ability to generate large volumes of H_2 in a fuel-less, passive system using the Stage 1 approach with SNF bundles make this concept highly desirable compared to incumbent approaches such as methane steam reforming, and may even compete with conventional electrolysis. As additional SNF is produced every year at current rates and with limited recycle capacity, the production potential of RHP increases cumulatively. This continued fuel production has higher burnup than legacy fuels and is thus more radioactive, and therefore potentially capable of greater H_2 production. Stage 2, requiring SNF reprocessing and Cs/Sr separations, carries exponentially higher capital, operational, and regulatory costs, which are not accounted for here, and will be the subject of future studies.

There are several critical use-cases for low-carbon hydrogen: first and foremost, energy storage for later use, either in compressed or liquefied form, and secondly, industrial use for production of important feedstocks, such as ammonia. Direct energy generation could be achieved via fuel cells or combustion. Despite the promise of fuel cell technology, their application to grid-scale energy generation for load following is currently insufficient relative to other technologies due to longer start-up times, challenges in load following, high costs, and the efficiency losses incurred with conversion from DC to AC. In contrast,

combined-cycle gas turbines (CCGTs), which normally run on natural gas, can be readily converted to partially or completely run on hydrogen and directly output 50 or 60 Hz AC at grid voltages with thermal efficiencies over 60% [69].

The wider hydrogen economy faces well-documented challenges. Global H_2 demand exceeded 100 Mt in 2025, arising almost entirely from industry and refining [70]. Low-carbon H_2 production was limited to almost 1 Mt, with growth in this area lagging ambitions: announced expansion of capacity by 2030 has fallen to 27 Mt due to project delays and cancellations, with only 6 Mt likely to be operating by the target date [70]. Commercially, high costs, uncertain demands, unclear regulations, and a lack of infrastructure remain obstacles to wider uptake of H_2 technologies, especially from low-carbon sources [70]. RHP would not remove all of these barriers, but could mitigate several.

In the UK, hydrogen production projects have stalled partly because demand has lagged behind available generation capacity; developers have moved away from speculative capacity towards projects anchored around committed customers [71]. As such, for RHP to be economically viable, it would be sensible to integrate this within a wider energy-generating system, as described below.

The approximate energy contained within a kg of H_2 is 33.3 kWh, which, if continuously turned into electrical energy in a CCGT (at 60% efficiency) at a grid price of 0.12 USD/kWh, would hypothetically afford 264k USD in energy generation per tonne of SNF annually, not accounting for energy consumption in H_2 processing, storage, transfer, overheads, and so on. This would potentially offset the equivalent of 670 tons of CO_2 per year per tonne of SNF from burning natural gas to generate the same volume of energy, assuming CO_2 production of 0.183 $kgCO_2$/kWh of natural gas. One ton of light water reactor fuel is estimated to have a total carbon footprint (including all upstream activities such as mining, enrichment, etc.) of 1.58 tCO_2-equivalent [72]. Therefore, this pathway could effectively offset 424 times the fuel's carbon footprint annually. Individually, these numbers may seem relatively small, but given the potential scalability of this system, a large impact could be reached relatively quickly, especially if integrated alongside other H_2-producing and consuming (i.e., energy or industrial) infrastructure. These values assume continuous operation without significant H_2 storage; use for load following would have a lower capacity factor and higher costs for storage. We assume compressed, rather than liquefied, storage for practicality and energy conservation reasons. This would create an internal demand for H_2 produced by RHP, outputting further, flexible energy from the NFC decoupled from reactor operations.

Moreover, RHP might offer a route to mitigate some of the increasingly clear environmental challenges associated with other hydrogen production routes. Environmental life-cycle assessments of hydrogen production via both electrolysis and steam methane reforming (with and without carbon capture) have suggested that, while hydrogen's carbon footprint is lower than that of fossil fuels such as natural gas, worsening of other environmental impacts is likely: one study found that hydrogen boilers had higher environmental impacts than gas boilers for 18 out of 19 environmental impact categories, with climate change being the exception [73]. Meanwhile, another study analysed scenarios for the production of 500 Mt H_2/y, finding a high likelihood of transgressing four out of nine "safe operating spaces" under the Planetary Boundaries framework, as well as high overall impacts on ecotoxicity and metal depletion [74]. While the life-cycle implications of RHP are currently unknown, its seemingly low material and energy inputs might enable hydrogen production that outcompetes other options on environmental grounds, mitigating the above concerns.

Rock-based routes offer very large theoretical reserves of natural H_2 [75]. Laboratory studies on crushed olivine at 300 °C report rates of up to 182 $bcfH_2/km^3.y$ (bcf = billion cubic feet; 5.15×10^{12} L at STP), while for natural H_2 the total yield modelled for more

realistic conditions and rock species at 200 °C is ~0.9 bcfH_2/km^3.y (2.55×10^{10} L at STP) [76]. RHP, in contrast, could produce 1.232×10^9 lH_2/tSNF.y based on the figures from the work of Vandenborre et al. [56]. These figures are, as with RHP values, idealised ceilings, and their realisation is unproven at the time of writing. H_2 is known to occur at depth; for example, drilling mud from the 12.26 km-deep Kola Superdeep Borehole was reported to be "boiling" with hydrogen [77], but this does not demonstrate a recoverable accumulation, and the commerciality of natural H_2 remains to be established [76]. Recovery from such sources would require H_2 to be generated, migrate, and become trapped. Stimulated production would require water to be injected into and the resultant H_2 recovered from low-permeability rocks at depths of 6–10 km, where conditions for production are optimal, though this is beyond the capabilities of routine drilling; the Kola Borehole stopped when bottom hole temperatures reached in excess of 180 °C and the rock at these depths began to display plasticity [78]. RHP, in contrast, would use existing inventories at known locations, requiring no subterranean exploration, deep drilling, or subsurface infrastructure, while addressing waste challenges simultaneously.

5. Technical and Regulatory Considerations

The necessary development and regulatory challenges for this concept to be implemented in full are manyfold. As noted in their work, Vandenborre et al. state that RHP with SNF bundles could be implemented with little technological development, although further optimisations [56] and validation of performance are required; the second stage we have outlined requires significant further study, advancement, and technological fusion.

Ideally, RHP would be co-sited with SNF recycle operations, augmenting or replacing current cooling ponds as H_2 production from SNF assemblies would essentially fulfil the same role. Once SNF bundles have been sufficiently cooled to allow for recycle (10 years for high-burnup PWR fuel), those operations would be performed and the separated HHRs converted into targets for further RHP. From a security standpoint, this second step would best be performed co-sited with SNF recycle and generation on site. However, given the nuclear industry's extensive expertise in the transport of active sources (especially SNF), it may be possible for RHP to occur on suitably licensed non-nuclear sites, given that such HHR targets would be neither fissile nor fertile. This would mitigate the challenges of compressing and/or liquefying and transporting or piping H_2, but compressed storage for later energy generation or industrial use would be necessary anyway, and could provide the means to achieve localised H_2 production "off-grid" with minimal supporting infrastructure.

A further approach would be integration into clean energy hubs where fission reactors and hydrogen production are co-sited; for example, where excess generation capacity is used to electrolyse water, the facilities for handling H_2 would already be present and could accommodate RHP without duplication of resources, thus lowering costs. This would also negate the need to ship SNF assemblies to a recycle facility prematurely. In this regard, RHP complements, rather than replaces, reactor H_2 production, and could serve to simplify the safety case and economics of such operations.

The separation of Cs in hydrometallurgical SNF recycle has seen significant development by some of the authors, and can be assessed, based on a modified TRL scale [35], to be at TRL 3–4. The Sr separations in this context, and the processing of both Cs and Sr into inert matrices suitable for long-term RHP (and disposal) are all at TRL 1. The various isotopes present in SNF for H_2 production from both beta- and gamma-emitting nuclides are discussed in the Appendix A [65]. The only other isotope in addition to ^{134}Cs, ^{137}Cs, and ^{90}Sr that may appreciably contribute to HHR RHP is ^{154}Eu (see Appendix A), which could be recovered from REE-rich feeds from MA separations [30], if this is economical and practical.

The approximate concentrations of HHRs that could be recovered from a typical SNF recycle facility annually (see Appendix A for further details) are 1750 kg of ^{137}Cs, 9.86 kg of ^{134}Cs, 19.1 kg of ^{154}Eu, and 738 kg of ^{90}Sr. The level of "dilution" to which the HHRs would need to be incorporated in an inert matrix for RHP must also be determined. Industry-standard H_2 purification and handling technologies (TRL 9) are employed worldwide and would not need significant further development for system integration. While it would be desirable to achieve quantitative recovery of the target HHRs, the practicalities of any new separation processes must be balanced with the realities of SNF recycle operations and integrated as seamlessly and with as few process adjustments as possible [28]. Our own initial studies have demonstrated that Cs can be selectively recovered from simulant-dissolved SNF, but further development and refinements of this process are needed to advance this towards deployability.

Vandenborre et al. demonstrated that gamma emissions effectively produce H_2 with a TiO_2 photocatalyst [56], but these results remain to be verified experimentally for beta-emitting isotopes [65], although the yields should be similar [66]. If the radiolytic mechanism by which beta-emitters (^{90}Sr/^{90}Y) produce hydrogen in water is different from gamma, then a different (photo)catalyst may be required, and would logically dictate that Cs and Sr are used separately. Alternatively, ^{90}Sr could (as outlined in the Appendix A) be used as a heat source for low-intensity but high-temperature applications, if not effective in H_2 production, as an isotope with negligible gamma emission (each tonne of high-burnup PWR SNF contains about 700 Wth of ^{90}Sr [79–84]), and as a source of ^{90}Y for medical applications [85](Chakravarty, 2012). A portion of separated ^{137}Cs could similarly be recovered for other industrial processes requiring radiation sources, such as conversion of bio-feedstocks for other co-generation purposes [61] and polymer grafting and cross-linking [62], medical device sterilisation [63], and water purification [64]. Further optimisations in H_2 yields can likely be found in "silo" geometry, photocatalyst loadings and doping, flow rates, water chemistry, and so on. Given the sensitivities around the use of nuclear-derived materials, valuable FPs recovered from SNF, such as Pd and Ag, could be used in remote operations to purify hydrogen produced using this concept [28–30]( ). At all stages, the safety implications of producing hydrogen, as a highly explosive gas, especially in contact with oxygen or air, must be considered; these have been extensively explored in an RCG context elsewhere, but will require specific modifications for the context outlined here in future works [6], especially given the presence of high radiation fields [4,12].

Current regulations in industrial settings require official certification of hydrogen-related technologies, and the financial support from the UK Government (under the Energy Act 2023 law) was restricted to a set of projects on hydrogen transport and storage. A wider approach/consideration for hydrogen production has been taken since 2023, encouraging more technologies to be used, but only if they meet the Low Carbon Hydrogen Standard [86]. A lack of regulations regarding the interface of nuclear (both fission and fusion) and hydrogen is apparent.

Safety considerations around the handling of hydrogen in the presence of radioactive materials must be a foremost concern. Radiolytic production of H_2 occurs naturally in spent fuel pools and within water-cooled reactors and is actively suppressed within existing fuel cycle operations. The explosion risks would be mitigated through proper process design, ensuring that H_2 is never mixed with sufficient oxygen to present a risk, alongside the omission of ignition sources and comprehensive monitoring. This likely means that new regulations will be needed to implement RHP and, as such, early engagement with national and international regulators must be sought to remove roadblocks on the path to implementation.

6. Discussion, Conclusions, and Further Work

Building upon both our own research and that of peers, we have outlined a novel approach to both the NFC and sustainable H_2 production using RHP. This has the potential to drive a substantial paradigm shift in the sustainability of the NFC and decarbonisation of both energy grids and, more generally, industrial hydrogen production. This could facilitate modern, sustainable, fully closed fuel cycles for recovery of valuable resources and, most importantly, utilising materials previously considered as wastes (SNF and, even more importantly, the HHRs) as an incredibly valuable resource.

Our approach would offer further NFC benefits, including mitigating a large proportion of the highly active wastes from SNF recycle, and this minimising or potentially even eliminating the need for costly geological disposal [27], in addition to offsetting a large proportion, if not the entire costs of SNF recycle assuming the full range of valuable FPs are recovered [28–31]. This would hopefully eliminate the barriers to SNF recycle and facilitate a wider closing of the fuel cycle, which will be essential to the long-term sustainability of nuclear fission [16]. The ability to decouple hydrogen production at least partially, if not entirely, from reactor operations could significantly simplify the safety and economic cases of nuclear hydrogen energy storage and production, with the potential for decentralised operations if the security and safety challenges can be addressed.

Significant development is needed to realise our concept in its entirety, despite the enormous potential to the NFC and Net Zero transition more generally. The first stage, where H_2 is produced using SNF assemblies, as proposed by Vandenborre, is achievable using current technology with little further development, albeit with substantial regulatory and licensing barriers to be overcome, and more comprehensive modelling required to better understand the production potential and safety aspects. We invite collaboration to achieve these aims.

The second stage requires comprehensive technological development of HHR separations, recovery, and processing to usable forms, optimisation of hydrogen production with respect to Cs and Sr partitioning (i.e., beta/gamma effectiveness), the potential inclusion of other nuclides, disposal route at the end of life, and so on, though these should all be achievable with present-day technology. A comprehensive life-cycle assessment of this potential approach to hydrogen generation ideally should be undertaken soon, accounting for environmental, socio-economic, and regulatory factors, to inform the following research and best approach to implementation. New regulatory and licensing frameworks may be needed to achieve what has been outlined here alongside a step-change in the attitudes of the nuclear industry, whose conservatism has often held back emerging technologies from having meaningful impact [16]. These will be assessed in future works. We welcome opportunities to collaborate with researchers, industrialists, and regulators from the UK and further afield to realise the development of the RHP concept, especially where experience in and materials relevant to this field lie.

On a final note, as radiolytically-produced H_2 is distinct from the other "colours" of hydrogen (Figure 5)—red, pink, and purple normally being assigned to nuclear-derived hydrogen, from electrolysis, (catalytic) thermolysis, and combinations of these, respectively [20,87]—we propose that this method be designated as **PLATINUM HYDROGEN**. This is to distinguish this method from the other proposed approaches, all of which require some external or natural (underground) chemical and/or energy feedstock in order to function [75,76].

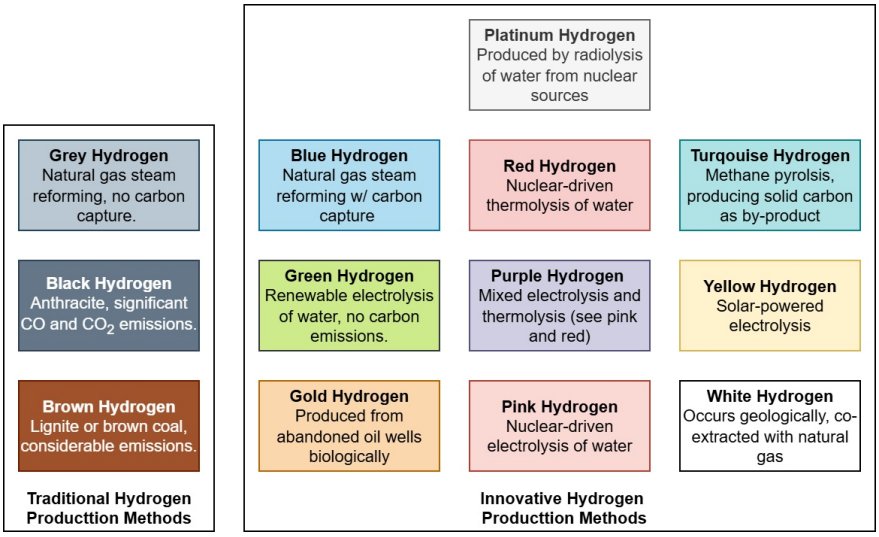


Figure 5. Colours of hydrogen. Adapted from [88].

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Abbreviations

The following abbreviations are used in this manuscript:

CCGT (s)	Combined-Cycle Gas Turbine (s)
CHP	Combined Heat and Power
CoGen	Cogeneration
DU	Depleted Uranium
EPR	European Pressurised water Reactor
FP (s)	Fission Product (s)
FY	Fission Yield
GWd/tHM	Gigawatt-days per Tonne of Heavy Metal
H_2	Hydrogen
H_2O_2	Hydrogen Peroxide
HBU	High-Burnup (fuel)
HHR (s)	High-Heat Radionuclide (s)
HTGR	High-Temperature Gas-Cooled Reactor
IAEA	International Atomic Energy Agency

k_{eff}	Effective Neutron Multiplication Factor
MA (s)	Minor Actinide (s)
MOX	Mixed Oxide (fuel)
NFC	Nuclear Fuel Cycle
OECD	Organisation for Economic Co-operation and Development
PRIS	Power Reactor Information System (IAEA service)
PWR (s)	Pressurised Water Reactor (s)
Q_β	Beta/Gamma Decay Energy per Decay
RCG	Reactor Cogeneration
REE (s)	Rare Earth Element (s)
RHP	Radiolytic Hydrogen Production
SMR (s)	Small Modular Reactor (s)
SNF	Spent Nuclear Fuel
$T_{0.5}$	Half Life
tHM	Tonne (s) of Heavy Metal
TiO_2	Titanium Dioxide
TRL	Technology Readiness Level
UO_x/UO_2	Uranium Oxide (fuel)
USD	United States Dollar

Appendix A

Derivation of K_{eff} for RHP

The reactivity of the proposed storage and hydrogen production system was verified using a SERPENT Monte Carlo model. This model was composed of radially stacked elements of one fuel assembly extending to infinity and was axially infinite. Both of these assumptions are conservative in terms of system reactivity compared to a real, finite, storage silo. No discharge credit was taken for the fuel, which consisted of unirradiated enriched uranium dioxide of 4.75% ^{235}U assay. Furthermore, no credit was taken for the neutron absorption in the titanium dioxide nanoparticles in the water on account of their comparatively low density, despite the comparatively high neutron capture cross section of natural titanium.

The fuel assembly was a 17 × 17 PWR assembly, with integral water filled Zircaloy guide tubes, and was emplaced in a square lattice, surrounded by a 20cm thick blanket of light water (Figure A1), representing a doubling of water thickness than that modelled by Vandenborre for H_2 production. This arrangement ensures that at least 40cm of light water separates adjacent fuel assemblies, not counting for steel or concrete silo barriers or other absorbers. This simultaneously provides ample depth of absorber to interact with the released gamma rays effectively and prevents neutron interactions between assemblies, as this is much greater than the neutron diffusion distance in light water.

For the simple case of a single, fresh fuel assembly in the above configuration, k_{eff} was 0.832, despite the lack of any active neutron absorbers. Although conceptually more assemblies could be arranged in larger lattices (i.e., 2 × 2, 3 × 3, etc.), this would likely require fixed neutron absorbers be placed in the silo, and additionally self shielding of gamma rays in adjacent fuel assemblies would tend to reduce the efficacy of hydrogen generation. That the k_{eff} is substantially less than 1, despite several conservative assumptions, demonstrates that this system should provide the required performance with sufficient criticality safety.

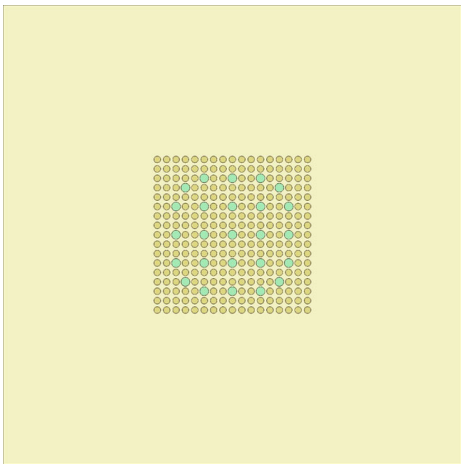


Figure A1. Representation of unit cell of SERPENT model run to calculate k_{eff} of fresh PWR model in storage. Water rods—blue; fuel pins—dark yellow; light water—light yellow.

Isotopes for RHP

We will divide the fission product isotopes for the following discussion into two categories: those that produce significant gamma radiation which are suitable for H_2 production (Table A1/Figure A2), and those primary beta emitters that may more suitable as heat sources (Table A2/Figure A3) if beta emissions are insufficiently effective at splitting water into hydrogen. This distinction arises primarily as beta particles more readily impart their energy and as such cause more localised heating [89] ideally within the matrix containing the isotopes themselves. For the purposes of this work, we consider the recoverable outputs of a 1000 tHM/y SNF recycle facility processing either high burnup UO_2 or MOX. HBU = high burnup 60 GWd/tHM UO_2 LWR UNF with 10 y post-reactor cooling before recycle, or MOX equivalent. Data from Ando and Takano [90] and IAEA isotope browser [91]. Plant size approximation of the former UK THORP UNF recycle plant and UP2 and UP3 operated at La Hague in France.

Table A1. Gamma-emitting isotopes suitable for hydrogen production.

Isotope	$T_{0.5}$ (y)	*FY (%)* [a] ($^{235}U/^{239}Pu$)	*SNF Conc.* [b] (UO_2/MOX)	*Activity* [c] (Bq/g)	*Activity* (Bg/tHM)	*Activity* (% of ^{137}Cs)	Q_b (keV)	*Primary Gamma Emissions* (Energy, keV, Yield, %)
$^{137}Cs/(^{137m}Ba)$	30.08	6.22/6.59	1745/1766	3.2E12	5.6E15/5.7E15	100/100	1175	661.7, 85.1
^{134}Cs	2.07	1.21E-5/6.74E-4 [d]	9.86/8.91	4.7E13	4.6E14/4.2E14	8.2/7.4	2059	604.7, 97.6; 795.9, 65.5; 569.3, 15.4; 802.0, 8.7; 563.2, 8.3, 1365.2, 3.0
$(^{126}Sn)/^{126}Sb$	(2.18E5)/0.03	0.05/0.26	1.33/64.1	4.8E8	6.4E8/3.1E10	0.0/0.0	(378)/3670	(87.5, 36.5); 695.0, 99.6; 666.5, 99.6; 414.7, 83.3; 720.7, 53.8; 697.0, 32.6
$^{106}Ru/^{106}Rh$	1.03/30.1 s	0.41/4.19	0.26/0.43	1.2E14	3.1E13/5.1E13	0.5/0.9	39/3546	511.9, 20.4; 621.9, 9.93
$^{144}Ce/^{144}Pr$	0.78/17.3 min	5.47/5.24	0.058/0.050	1.2E14	6E12/6E12	0.1/0.1	319/2997	(133.5, 11.1); 696.5, 1.3, 2185,7, 0.7, 1489.1, 0.3
^{154}Eu	8.60	1.95E-7/4.90E-5 [e]	19.1/39.5	1.0E13	1.9E14/3.9E14	3.4/6.8	1968	123.1, 40.4; 1274.4, 34.8; 1004.8, 18.0; 873.2, 12.1; 996.3, 10.5; 247.9, 6.89; 591.8, 5.0; 756.8, 4.52
^{155}Eu	4.75	0.09/0.17	3.54/5.35	1.8E13	6.4E13/9.6E13	1.1/1.6	252	86.5, 30.7; 105.3, 21.1; 45.3, 1.3; 60.0, 1.2
^{110m}Ag	0.68	8.15E-9/6.83E-7 [f]	9.02E-5/ 1.92E-4	~1.8E14	1.6E10/3.5E10	0.0/0.0	2891	657.8, 95.6; 884.7, 75.0; 937.4, 35; 1384.3, 25.1; 706.7, 16.7; 1505.0, 13.3;
^{125}Sb	2.76	0.03/0.12	0.96/1.34	3.8E13	3.6E13/5.1E13	0.6/0.9	767	427.9, 29.6; 600.6, 17.7; 656.0, 11.2; 463.4, 10.5; 176.3, 6.8; 606.7, 5.0

$T_{0.5}$ = half life; FY = fission yield; SNF Conc = spent nuclear fuel concentration, g/tHM; Q_b = total decay energy. [a] Cumulative thermal fission yield from ^{235}U and ^{239}Pu. [b] Concentration per tonne of initial heavy metal in 60 GWd/tHM UO_2 and MOX cooled for 10 y post-reactor, 5% ^{235}U initial enrichment for UO_2 and 7.84% initial Pu. Data from [90]. [c] Activity of parent isotope. [d] Shielded by ^{134}Xe. [e] Shielded by ^{154}Sm. [f] Shielded by ^{110}Pd [89].

As shown in Table A1/Figure A1, only ^{137}Cs, ^{134}Cs, and ^{154}Eu generate any significant gamma-emission after 10 y of post-reactor cooling and thus are the most promising targets for separation for RHP use, and rationalises the chemical separations of Cs in reprocessing [37,43,44], separating the ^{137}Cs and ^{134}Cs useful for H_2 production alongside the long-lived ^{135}Cs and stable ^{133}Cs. Further to this, the higher fission and capture yields of ^{154}Eu in MOX SNF mean that the potential gamma energy imparted by this isotope increases from 6.3% of ^{137}Cs to 12.7%. Given most advanced reprocessing flowsheets co-separate the chemically-similar REEs alongside the MAs, partitioning Eu from REE raffinates separate from the other active rare earth isotopes [92] may be appealing for hydrogen production when reprocessing HBU MOX fuels.

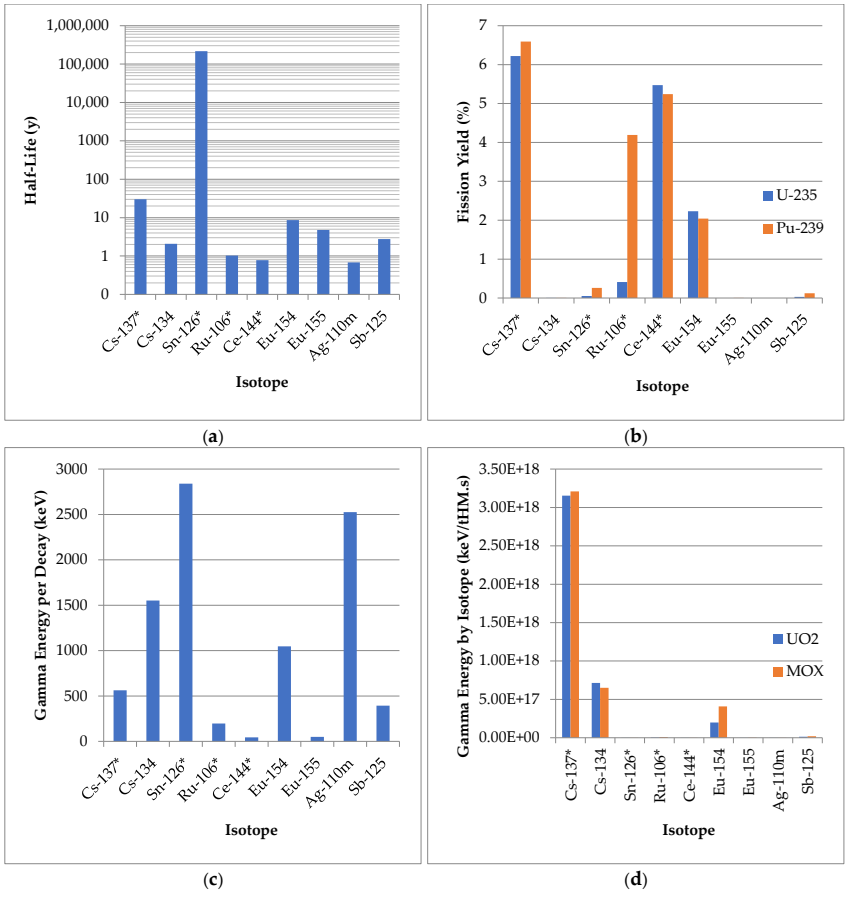


Figure A2. Data for significant gamma-emitting isotopes present in SNF after 10 y of post-reactor cooling, (**a**) half-lives; (**b**) cumulative thermal fission yields from ^{235}U and ^{239}Pu fission; (**c**) the gamma energy released per decay (accounting for branching ratios); and (**d**) the gamma emissions by each isotope per second from both UO_2 and MOX; 1E18 keV = 160.2 J. Data from [90,91]. Isotopes marked with an asterisk (*) have the values of their daughters included with them. Please note this selection is not exhaustive, but represents the primary contributors to gamma emission at time of SNF recycle.

The other isotopes are either too short-lived or too low yielding hydrogen production using them would likely be uneconomical despite, on paper, several being more appealing than ^{137}Cs. Nonetheless, these would generate hydrogen while still contained within SNF in interim storage (typically 10 years) between reactor and recycle, as originally proposed by Vandenborre et al., to maximise the potential yields from this concept. As demonstrated by the data in Figure A2/Table A2, only ^{90}Sr and its ^{90}Y daughter are feasible for heat generation using beta decay. Although ^{151}Sm has a longer half-life, the low decay energy (19.62 keV on average) and lower concentration mean that the energy generated per second

is insignificant in comparison to $^{90}Sr/^{90}Y$, with the shorter half-life and relatively low decay energy of ^{147}Pm countering its viability. The lower fission yield of ^{90}Sr from Pu over ^{235}U, producing just under half the decay heat in MOX over UO_2, mean that recovery of ^{90}Sr from conventional PWR UO_2 would likely be preferable, though this could be offset by higher burnup MOX fuel, especially from fast reactors, which can exceed 100 GWd/tHM. As ^{90}Sr fission yields from ^{233}U are higher still than ^{235}U (6.65% vs. 5.73%), more of this isotope could be recovered from Th-based fuels.

Table A2. Heat-emitting isotopes for heat generation.

Isotope	$T_{0.5}$ (*y*)	*FY* (%) [a] ($^{235}U/^{239}Pu$)	*SNF Conc.* [b] (UO_2/MOX)	*Activity* [c] (Bq/g)	*Activity* (Bg/tHM)	Q_b (keV)	*PrimaryBeta Emissions* (*Energy, keV, Yield, %*)	*Significant Gamma Emission* (*s*) (*Energy, keV, Yield, %*)
$^{90}Sr/^{90}Y$	28.91/64.6 h	5.73/2.01	738/361	5.1E12	3.8E15/1.8E15	526/2776	(195.7, 100); 932.4, 99.9; 184.6, 0.01	2186.2, 1.4E-6
^{147}Pm	2.62	2.23/2.04	16.5/17.4	3.4E13	5.6E14/5.9E14	224	61.8, 99.9; 26.9, 5E-3	121.2, 2.9E-3
^{151}Sm	90	0.42/0.78	17.4/44.4	9.7E11	1.7E13/4.3E13	76.6	19.7, 99.1; 14.0, 0.9	21.5; 0.03

$T_{0.5}$ = half life; FY = fission yield; SNF Conc = spent nuclear fuel concentration, g/tHM; Q_b = total decay energy. [a] Cumulative thermal fission yield from ^{235}U and ^{239}Pu. [b] Concentration per tonne of initial heavy metal in 60 GWd/tHM UO_2 and MOX cooled for 10 y post-reactor, 5% ^{235}U initial enrichment for UO_2 and 7.84% initial Pu. Data from [90]. [c] Activity of parent isotope.

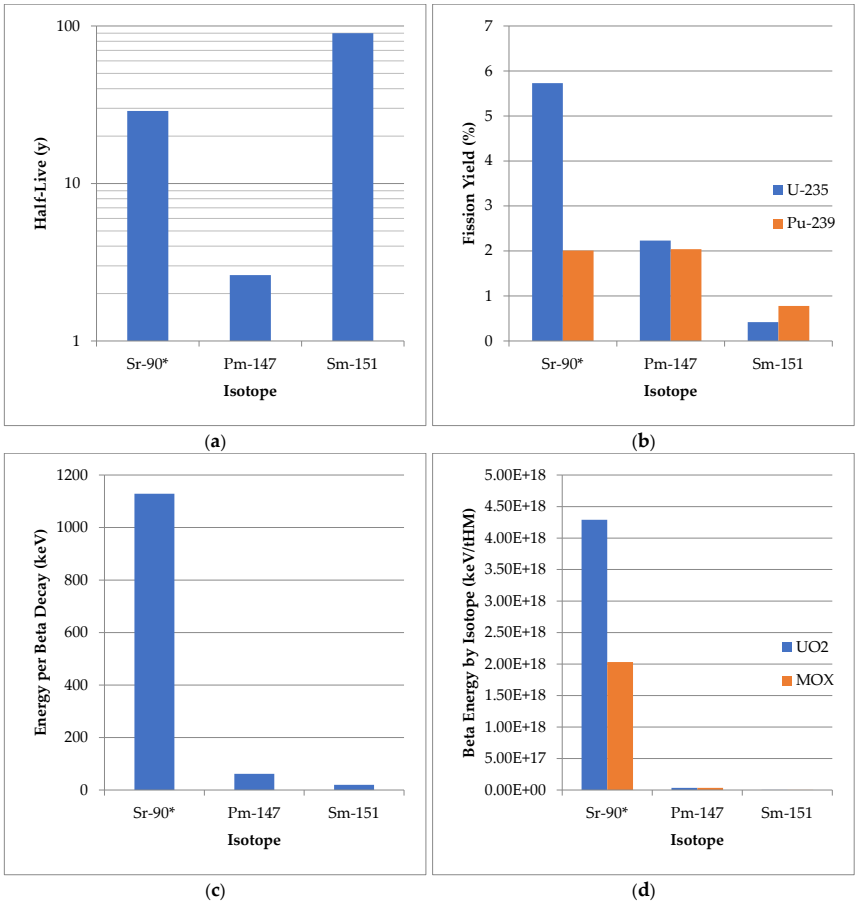


Figure A3. Data for significant gamma-emitting isotopes present in SNF after 10 y of post-reactor cooling, (**a**) half-lives; (**b**) cumulative thermal fission yields from ^{235}U and ^{239}Pu fission; (**c**) the gamma energy released per decay (accounting for branching ratios); and (**d**) the gamma emissions by each isotope per second from both UO_2 and MOX; 1E18 keV = 160.2 J. Data from [90,91]. Isotopes marked with an asterisk (*) have the values of their daughters included with them. Please note this selection is not exhaustive, but represents the primary contributors to gamma emission at time of SNF recycle.

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