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Review

A Review of the Nuclear Fuel Cycle Strategies and the Spent Nuclear Fuel Management Technologies

Laura Rodríguez-Penalonga * and B. Yolanda Moratilla Soria

Cátedra Rafael Mariño de Nuevas Tecnologías Energéticas, Universidad Pontificia Comillas, 28015 Madrid, Spain; ymoratilla@comillas.edu

* Correspondence: lrpenalonga@comillas.edu; Tel.: +34-91-542-2800 (ext. 2481)

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Abstract: Nuclear power has been questioned almost since its beginnings and one of the major issues concerning its social acceptability around the world is nuclear waste management. In recent years, these issues have led to a rise in public opposition in some countries and, thus, nuclear energy has been facing even more challenges. However, continuous efforts in R&D (research and development) are resulting in new spent nuclear fuel (SNF) management technologies that might be the pathway towards helping the environment and the sustainability of nuclear energy. Thus, reprocessing and recycling of SNF could be one of the key points to improve the social acceptability of nuclear energy. Therefore, the purpose of this paper is to review the state of the nuclear waste management technologies, its evolution through time and the future advanced techniques that are currently under research, in order to obtain a global vision of the nuclear fuel cycle strategies available, their advantages and disadvantages, and their expected evolution in the future.

Keywords: nuclear energy; nuclear waste management; reprocessing; recycling

1. Introduction

Nuclear energy is a mature technology that has been developing and improving since its beginnings in the 1940s. However, the fear of nuclear power has always existed and, for the last two decades, there has been a general discussion around the world about the future of nuclear power [1,2]. This discussion has been aggravated since the Fukushima Daiichi accident in 2011 [3,4] and, also, due to the rise of renewable energies. Consequently, several countries have been questioning the continuity of their nuclear power plants (NPPs) on their energy mix.

In this debate, there are arguments both in favour and against nuclear power. On the one hand, some of the arguments in favour are its security of supply, its potential to mitigate climate change [5–7] and the provision of stability to the electricity grid by means of its great inertia, which avoids blackouts and is not replaceable with any type of renewable energies, except solar thermal [8]. On the other hand, the usual arguments against nuclear power are safety concerns, the proliferation risk, its high investment costs, and nuclear waste management.

Therefore, considering these arguments, some countries have finally decided to cease all their NPP activity, either progressively, by not renewing their licenses at the end of their lifetime, or more immediately, such as the case of Germany [9–12]. In contrast, there are other countries which thoroughly support nuclear power, such as China, which is planning to expand its nuclear capacity to 4% of the share (40 GWe) in 2020 and to 20% in 2050 [13,14], and other developing countries which lean on nuclear energy to be able to grow in a more sustainable way. Also, traditional nuclear power supporters, such as France, will continue to produce nuclear energy, but transitioning towards an energy mix model with a higher share of renewable energies [15].

Also, however, regardless of the future of nuclear power, spent nuclear fuel (SNF) management has to be solved around the world in every country that produces or has at some point produced nuclear

energy. Moreover, it is essential to address this issue properly because the periods of time required to manage SNF are extremely long and involve future generations. Hence, the decisions that have to be made today around the world regarding SNF management are of great importance internationally.

In this context, Spain has been facing some challenges. The SNF from Vandellós-I NPP was sent to France for reprocessing and, before 1983, some of the SNF from Santa María de Garoña NPP was sent to reprocess to the UK; the vitrified waste that came from reprocessing all this SNF needed to return to Spain in 2015, but a facility to receive and store this vitrified HLW (high-level waste) is still lacking. Thus, Spain has started to pay a penalty fee and needs to solve this issue before 2020 [16].

However, this problem is not technical, but political. Even though there are technical solutions and a centralized interim storage facility has been presented in the last radioactive waste management general plan [17], its construction has been opposed and postponed for political reasons, because the disagreements between the central and autonomic governments paralysed the project. Furthermore, despite the evidence presented, there are some political parties that have stated that there are no technical solutions for radioactive waste. Additionally, another challenge that Spain has been facing regarding SNF management is the recent legislation surrounding nuclear waste, which has considerably increased its management cost.

Hence, it is very important to know the technologies available for SNF management, their advantages and disadvantages, the technologies that are currently under research and the future R&D tendencies, as well as which alternative suits better the needs for each country and its particular context.

Furthermore, the interactions among legal, economic, and technological issues are essential. Thus, this paper gives an overall view of the technologies available for SNF management, analyses the economic cost that the use of the different technologies could entail, as is key when selecting a strategy for SNF management in each country and, finally, highlights the importance of the legislation and some intangible assets, such as social acceptability of the technology, which can have an influence so important that it may lead to the disappearance of that technology by making it economically unviable.

Therefore, it is essential to know all the technical alternatives available for SNF management and the cost that those technologies entail, while separating political issues from technological needs and while all parties involved cooperate in order to avoid increasing the problems. Thus, this paper seeks not to be only a review about technical and economic aspects, but to also introduce some of the current lines of discussion that are being considered in the political decision-making process in different countries, especially in Spain.

2. Main Strategies for the Nuclear Fuel Cycle

The nuclear fuel cycle is formed by all the steps and processes that nuclear fuel has to be put through before and after its use in an NPP in order to produce electricity. Therefore, two main stages can be distinguished: the front end, which occurs after the arrival of nuclear fuel at the NPP, and; the back end, which takes place once the SNF leaves the reactor.

The exact processes to which nuclear fuel or SNF goes under may vary according to the nuclear fuel cycle strategy that is implemented. Nowadays there are two main nuclear fuel cycle strategies: the once-through cycle (direct disposal or open cycle) and the twice-through cycle (recycling or partially closed cycle). Nonetheless, there aAre currently other strategies under research known as advanced cycles which are not commercially available yet, but which are very promising.

On the one hand, the once-through cycle considers SNF to be high-level waste (HLW) and, consequently, it is directly disposed of in a storage facility without being put through to any chemical processes, where it will be safely stored for millions of years until its radiotoxicity reaches natural uranium levels or another safe reference level.

On the other hand, the twice-through cycle considers SNF to be an energy source due to its composition: approximately 96–97% of its components are recyclable materials, 94–96% of which is uranium (1% approx. of U-235) and 1–1.5% is plutonium [18,19]. Thus, in order to exploit its energy potential, SNF has to be put through a series of chemical processes known as reprocessing. Therefore,

on the twice-through cycle strategy, SNF is reprocessed in order to extract the uranium and plutonium, which can either be recycled as fresh nuclear fuel for its use in a nuclear reactor that is adapted to this type of fuel or sold as raw material.

Hence, taking this into consideration, the front end main activities are mostly the same for both strategies: natural uranium extraction (mining); uranium conversion; enrichment, and; fuel fabrication. The exact processes may vary according to the type of fuel and the technologies used for each step, but overall the activities can be summarized in this way.

Nevertheless, back end activities are highly dependent on the nuclear fuel strategy that is implemented. Therefore, all back end stages will be explained below for each of the fuel cycle strategies.

2.1. Once-through Cycle

The once-through cycle comprises two main back end stages: interim storage and final disposal. In between those stages there is also an encapsulation step and transportations.

First, SNF is extracted from the reactor and cooled inside the reactor pools for a period of at least five to 10 years [20], though in some cases it was considered three years [21]. This stage is mandatory for every nuclear fuel cycle strategy.

After this period of initial cooling, nuclear fuel can be transferred into a dry interim storage, which can be either at the NPP site or at a centralized location that stores SNF from more than one NPP. However, in some cases, such as Spain, SNF is stored at the reactor pools as interim storage and transferred into a dry storage when its capacity is full [17,22,23].

Finally, after a minimum period of 50 to 100 years of interim storage, SNF must be transferred to a final disposal facility. Currently, the preferred option is a deep geological repository (DGR), which is an underground emplacement in stable geological formations. However, currently, there are only a few under construction in some countries, such as Finland and Sweden, but none of them are yet commissioned or operating [20].

2.2. Twice-through Cycle

The twice-through cycle is a little more complicated than the once-through cycle and, thus, involves more stages both in the back end and in the front end.

After the initial cooling at the reactor pools, SNF is transported to the reprocessing facility, where the U and the Pu are separated from the minor actinides (MA) and fission products (FP), by means of the PUREX (Plutonium and Uranium Redox Extraction) process [24], which is currently the only commercially available technology.

Once they have been separated, MAs and FPs are vitrified in a glass matrix and stored as HLW. Eventually, they will be transferred into a final disposal facility, most probably a DGR. However, since SNF is composed of more than 96% of U and Pu, the final HLW volume is reduced about 80%, its radiotoxicity decreases about 90% and its decay heat is also reduced compared to the once-through cycle [25,26]. Thus, the cost of a DGR for vitrified waste would be about 25% of the cost of a DGR for SNF [27,28].

Meanwhile, the extracted plutonium can be recycled into MOX (mixed oxide) fuel after being transferred to a MOX fuel fabrication plant. This type of fuel can be used in about 10% of the world reactors, but currently the number of Pu recycles is limited to two or three, due to technological limitations. Thus, irradiated MOX has to be stored until its final disposition or until advanced reprocessing technologies are available [20].

Additionally, uranium can also be reused after re-enrichment and fabrication at a dedicated plant. However, recovered uranium is more radioactive than natural uranium. Thus, the cost of the processes of enrichment and fuel fabrication are higher for recovered uranium than for natural uranium, because it requires dedicated facilities to avoid natural uranium contamination. Hence, this practice is not very common at this time and, for this reason, reprocessed uranium is usually stored [20].

2.3. Advantages and Disadvantages of the Current Nuclear Fuel Cycle Strategies

The advantages of reprocessing compared to the direct disposal option come from the reduction of natural uranium requirements and the decrease in volume, radiotoxicity and decay heat of the final HLW, as long as irradiated MOX is treated separately. Consequently, the final repository volume is reduced, which also decreases its cost. Furthermore, HLW compaction and vitrification facilitates the handling of the final waste [19,29,30].

However, there are some disadvantages associated with reprocessing due to the complexity of the nuclear fuel cycle, which creates more stages and fuel transportations. Moreover, reprocessing requires the extraction of the nuclear fuel, which increases the exposure risk, as well as LILW (low and intermediate lived waste) volumes. In addition, the separation of pure plutonium increases the proliferation risk. However, there are some options to solve this issue such as the recycling of Pu in MOX fuel, which reduces the Pu inventory, but also some advanced reprocessing techniques that jointly extract Pu and U, avoiding pure Pu separation, which will be described in Section 4.2, and advanced nuclear reactors technologies, such as Generation IV, which will be further explained in Section 2.4.

In relation to the cost, numerous international studies have estimated the costs of both strategies, under different hypothesis. The cost of the twice-through cycle is usually higher than for the once-through cycle. However, the magnitude of the difference between these costs depends on the methodology and the adopted hypothesis and assumptions [20,27,28,31–40].

It can be observed that the once-through cycle has always been more economical than the reprocessing option. Furthermore, the trend lines of both costs (once-through and twice-through cycles) have a declining slope, which means that these costs are decreasing with time due to the maturity of technology. However, this slope is slightly more pronounced for the twice-through cycle, which means that, eventually, reprocessing might become as economically viable as the once-through cycle, or even more. Figure 1 shows the evolution of the nuclear fuel cycle strategies costs with time, in mill/kWh, for different international studies [27,28,30–33,38,40], which have been updated to 2016. One of these costs [30] is much lower, so it has been excluded from the trend lines.

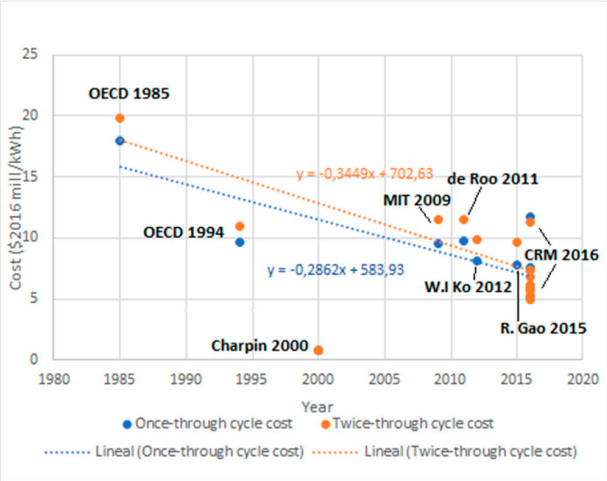

Figure 1. Evolution of nuclear fuel cycle costs.

Additionally, these trend lines and the difference in their slopes can be explained in Figure 2, which shows the evolution of the estimated DGR costs for the once-through cycle and the costs of reprocessing with time.

As can be seen in Figure 2, the reprocessing costs have been decreasing with time due to the maturity of the technology. However, the opposite happens when it comes to the DGR costs, because it is a new, untested technology. As reported [27,28], the DGR cost dominates the back-end costs

for the direct disposal option and reprocessing is the most important cost for the back end of the twice-through cycle. Therefore, this explains why the twice-through cycle costs are decreasing more rapidly than the once-through cycle costs.

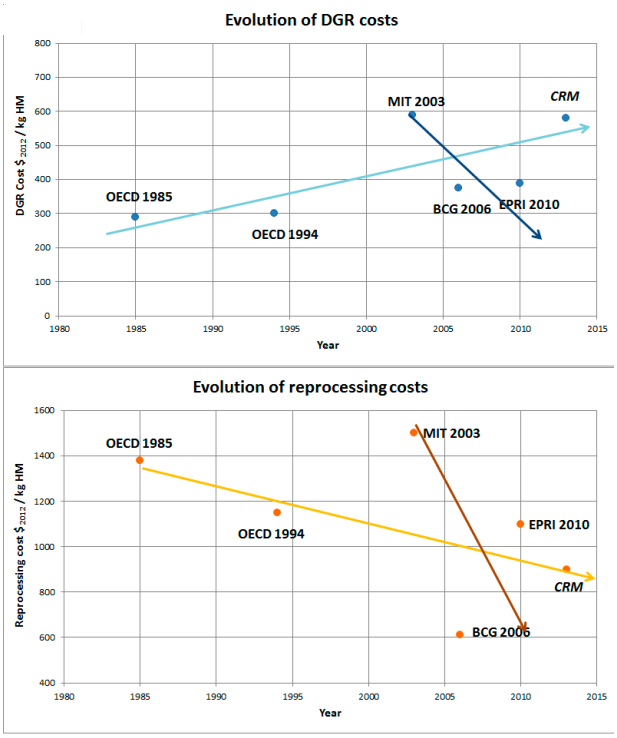

Figure 2. Evolution of deep geological repository (DGR) and reprocessing costs.

2.4. Advanced Cycles

There are currently different types of advanced cycles under research, whose purpose is to achieve nuclear power sustainability by further reducing the volume and radiotoxicity of the final waste, as well as improving resource utilization, while maintaining its economic viability [25,41]. Most of them have something in common: they are based on partitioning and transmutation (P&T) technologies, which seek to shorten the half-lives of the long-lived isotopes that compose SNF into short-lived stable isotopes [22].

Thus, there are two main areas of research: nuclear reactors and reprocessing technologies. The latter will be further explained in Section 4.

The most widely spread advanced nuclear reactor technologies are Generation IV fast breeder reactors (FBR) and accelerator driven systems (ADS) [19,22,23]. However, there are also other research areas that have recently been the focus of some studies, such as fusion-fission hybrids, which seek to start introducing nuclear fusion in a more feasible manner and producing nuclear fuel for fission reactors, avoiding the need of reprocessing and, consequently, the proliferation risk [42–44].

2.4.1. Gen. IV Reactors

Fast reactors have been developed since the 1950s [22] and there are prototypes around the world, such as in the USA, France, India, and so forth [45]. However, they are still not commercially available.

These reactors use uranium completely, both fissile U-235 and fertile U-238, as well as plutonium and MAs, which makes them capable of extracting more energy from nuclear fuel and reducing the volume of the final HLW and its radiotoxicity [22].

Generally, Gen. IV reactors are defined as breeders, which means that they can produce more fissile material than they consume. Currently, there are different designs of FRs around the world: Gas-Cooled

Fast Reactor (GFR); Lead-Cooled Fast Reactor (LFR); Molten Salt Reactor (MSR); Sodium-Cooled Fast Reactor (SFR); Supercritical-Water Cooled Reactor (SCWR), and; Very High Temperature Reactor (VHTR) [19].

All these designs have their own advantages and disadvantages. However, worldwide, the greatest concerns about fast reactors are their economic cost, the effective time necessary for their commercial development and that, like any other reactor, they are critical reactors and are supercritical for the short time in which they increase the power.

2.4.2. ADS

ADS consist of a subcritical reactor connected to a particle accelerator. They are of great international interest because they possess greater flexibility and they allow burning not only Pu and MAs, but also FPs, even when the U content is nearly zero, which reduces HLW radiotoxicity by a factor of 100 [19,22,46].

Moreover, they can also be used as fuel breeders and, because they are subcritical reactors, their safety is improved compared to FBRs, which allows them to maintain subcritical conditions in the shutdown [23]. Additionally, ADS have less limitations regarding fuel composition [25].

2.5. Advantages of the Advanced Cycles

Due to their improved sustainability, advanced cycles present numerous advantages compared to the current nuclear fuel cycle strategies. Some of these advantages, which depend on the type of advanced fuel cycle, are listed below [25,41].

- Heavy metal content can be reduced in more than three orders of magnitude in final disposal.
- FP mass (per kWh) and, hence, the HLW volume can be decreased about 30% in an all-FR (Fast Reactor) strategy, due to the higher efficiency of fast reactors.
- Fully closed cycles can achieve a 100-fold TRU (Transuranics) content reduction.
- Natural uranium needs vary from 10% to about two orders of magnitude lower than the once-through cycle.
- Decay heat can be decreased by several orders of magnitude.
- FPs separation and management can reduce decay heat from 17 to 30 times, and vitrified HLW containers from 25% to 40%.

However, the advanced cycle costs are higher than for the current strategies and the trend line in Figure 3 has a slightly increasing slope, due to the lack of maturity of these new technologies.

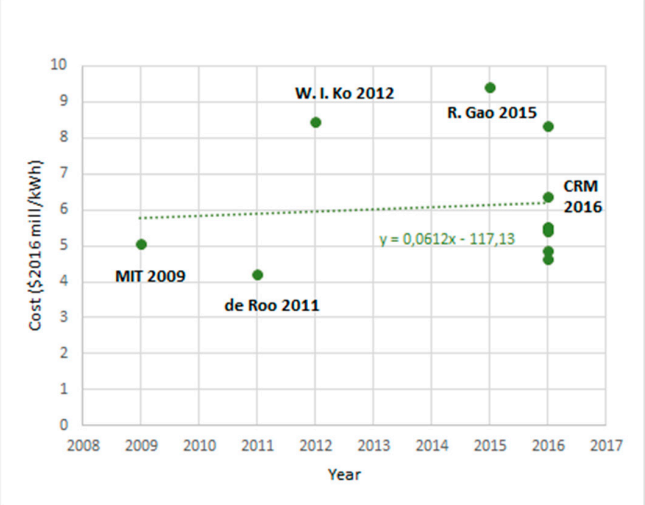

Figure 3. Evolution of advanced cycles cost.

Nonetheless, Monte Carlo simulations show that advanced cycles might eventually be more economically viable than direct disposal [25].

3. Interim Storage

Interim storage is the first step for back end of the once-through cycle and its purpose is to safely store and monitor SNF while its activity decays and enough heat is removed to proceed to the final disposal.

There are different types of interim storage and its emplacement may vary. Thus, it can be located at the NPP site, whether at the reactor or away from the reactor (AFR), or at a centralized location. The main advantage of centralized storages is that monitoring is simpler, because all containers are located at the same emplacement. However, the main disadvantage is that SNF transportations increase.

Regarding technology, there are two main types of interim storage: wet and dry. The former is usually the reactor pools and it is not frequently used at AFR locations. The latter, however, is the most common and expanding technology for both AFR and centralized storages [47].

The reason behind the increasing interest in dry interim storage is their passive cooling systems, their low O&M (operation and maintenance) requirements and its subcriticality. Thus, even though water has the function of heat dissipation and shielding, it is also an excellent moderator, so there is the possibility of criticality [48,49]. Moreover, since the Fukushima Daiichi accident, the loss of coolant in pools is a major concern, which R&D efforts are trying to solve, as in the case of the upper-pool of a Mark-III containment [50].

Therefore, the actual trend is to transfer SNF to dry storage as soon as possible, which is the safest method to reduce the danger of fires when there is lack of coolant [51].

Dry Storage Technologies

Even though interest in dry storage has been increasing over the past few years, its origins are not new: their research began in Canada in 1960 and in 1970 the first dry storage was built in the UK. However, no large storage facility was constructed until 1987 [47,48].

Since then, dry storage has improved and different technologies have been developed around the world. At first, they were single-purpose systems (storage), but nowadays double-purpose (transportation and storage) and multi-purpose systems (transportation, storage, and final disposal) have been developed [47].

Currently, there are two basic types of dry storage technologies: vaults and container systems, which can be subdivided into silos, metal casks and concrete casks. These are described in [47].

The type of technology used varies from one country to another. However, since Germany ceased reprocessing and, especially, since they decided to abandon nuclear energy in 2011, they have become leaders on the construction of dry storage systems and they have planned to fabricate 800 double-purpose containers over the next 15 years [52].

4. Reprocessing

As seen in Section 2, SNF is composed of 96–97% of recyclable materials, which means that it still contains most of its energy potential. Thus, the purpose of reprocessing is to chemically separate the components of SNF in order to exploit its full potential [19]. At this time, U and Pu are extracted from SNF, but future trends lead to MA and FP separation as well, with the purpose of burning them in advanced reactors or store them separately.

Reprocessing started in the 1940s with military purposes and different technologies were tested, but they were replaced for solvent extraction technologies and PUREX process was developed [19,20]. However, commercial reprocessing started in the 1960s [26] with the purpose of reducing natural uranium requirements and interest started growing especially due to the unprecedented rise in uranium prices in the 1970s, although they moderated shortly afterwards [20].

Over the years, many countries have reprocessed their SNF, but there are only a few countries that have built their own reprocessing facilities: France, the UK, India and Russia [19,24]. The USA is a special case, because they have built three different reprocessing facilities, but none of them have ever operated. Also, Japan has been planning and building its own reprocessing facility for two decades in Rokkasho, but it is still not operational [53–55]. Finally, China's ambitious plans for nuclear energy, due to the environmental problems related to the massive use of coal [13,14], include the construction of a reprocessing plant [56].

4.1. PUREX Reprocessing

PUREX is a mature technology and, currently, the only one that is commercially available. The process starts with the dissolution of SNF in nitric acid and then it is subjected to a process of solvent extraction using tri-n-butyl phosphate (TBP), where uranium and plutonium are separated (with a 99.9% efficiency) and then purified [26]. MAs and FPs remain in the acid solution and are vitrified in a glass matrix and packaged into a universal canister (UC), which will be disposed in a DGR [19,57]. Finally, the U and Pu extracted from SNF can be recycled into fresh nuclear fuel: MOX fuel or reprocessed uranium fuel.

In the past years, several modifications to the PUREX process have been researched to improve its performance. For example, in Japan they have co-precipitated plutonium and uranium in order to avoid pure Pu separation and the subsequent proliferation risk [19,20,25].

4.1.1. MOX

Pu-239 and Pu-241 are fissile isotopes, so they can be used as fuel in a thermal reactor instead of U-235 [33]. Thus, MOX is composed of about 3% [58] to 5% or 8% [39] of extracted Pu and uranium, which can either be low-enriched uranium, depleted uranium from enrichment plants [27] or even separated uranium [19,25].

MOX is a mature technology and is used in about 10% of the world reactors (40 NPPs), but its use is mostly found in France [20]. Its main purpose is to reduce natural uranium needs, which with current technologies are estimated to decrease from about 8–12% [20,25] to 20–25% [29,34]. Moreover, an additional consequence is the reduction of pure Pu inventory and, thus, the proliferation risk.

However, due to technological limitations, spent MOX can only be reprocessed and recycled once or twice. Nonetheless, advanced technologies may be able to use spent MOX and, therefore, allow Pu storage without proliferation risk [20].

4.1.2. Reprocessed Uranium Oxide (REPUOX)

Separated uranium can be recycled into reprocessed uranium oxide (REPUOX) after re-enrichment. However, this is used on a limited scale around the world because reprocessed U is more radioactive than natural uranium and, thus, it requires dedicated facilities to avoid natural uranium contamination. Consequently, as this increases the cost, most of the recovered U is stored for future use [20].

4.2. Advanced Reprocessing Technologies

Nowadays, there are several reprocessing technologies being researched around the world, whose purpose is to improve reprocessing efficiency, avoid proliferation risk, reduce the final waste volume (both HLW and LILW), and so forth.

4.2.1. Wet Reprocessing Technologies

There are two different lines of research for wet processes: (i) technologies based on the solvent extraction of the PUREX process with advanced separation of different components, and; (ii) variations on the chemistry of the solvent extraction process in order to only separate uranium from SNF, while keeping Pu, MAs and FPs in the waste solution for further separation. The purpose of these processes

is to extract not only U and Pu, but also Np or other Mas and some FPs in order to reduce the radiotoxicity and heat load in the final HLW. Also, in some cases, pure Pu separation is avoided to reduce proliferation risk.

There are different processes being researched around the world using various types of extractants and solvents. Some of these processes are explained below.

- UREX (Uranium Extraction): Uranium extraction separates U and Tc from SNF. It is mostly the same procedure as PUREX with the addition of AHA (CH_3C=ONHOH), which prevents the separation of Pu and Np and reduces solid waste volume. There are different modifications of this process which include other processes for further separation [19,26,44,59].
    - UREX+: it consists of a series of five consecutive solvent-extraction processes that separate SNF into several fractions: (i) UREX; (ii) Cs and Sr recovery (Chlorinated Cobalt Dicarbollide and Poly-Ethylene Glycol (CCD-PEG)); (iii) NPEX (Pu and Np recovery); (iv) TRUEX (recovery of Am, Cm and rare-earth FPs), and; (v) separation of Am and Cm from the rare-earth FPs [25,26].
    - UREX+1: it is a modification of the UREX process which further separates Pu and Np in order to recycle those materials in MOX fuel [19].
    - UREX+2: it uses ion exchange instead of concentrated nitic acid to separate Tc from U [19,25].
- COEX (Combined Extraction): it is a simplification of the PUREX process which jointly extracts Pu and U and, thus, avoids proliferation risk and the complex steps of separation of the PUREX process [19,60].
- DIAMEX (Diamide Extraction): it uses a solvent based on amides as alternate to phosphorous reagent in order to extract MAs and lanthanides from HLW. As this solvent is totally combustible, this process generates minimum organic waste. There is a similar process in Japan known as TODGA, which uses tetra-octyl-diglycol-amide [26].
- TRUEX (Transuranic elements Extraction): this process extracts different transuranic elements, such as Am and Cm, from the HLW solution by using Carbamoyl Methyl Phosphine Oxide (CMPO) together with TBP [19,26].
- SANEX-N and SANEX-S (Selective Actinide Extraction process): these processes separates actinides from lanthanides using either N-bearing extractants (Bis-triazinyl-pyridines, BTPs) or S-bearing extractants (such as a synergistic mixture of Cyanex-301 with 2.2-bipyridyl) [26].
- SESAME (Selective Extraction and Separation of Americium by Means of Electrolysis): it separates Am from Cm by oxidation of Am to Am (VI) and subsequent extraction with TBP for separation from Cm [26].
- CSEX (Cs Extraction): it separates Cs from HLW using Calix-crown extractants [26].
- SREX (Sr Extraction): it separates Sr from HLW using dicyclohexano 18-crown-6 ether [26].
- CCD-PEG (Chlorinated Cobalt Dicarbollide and Poly-Ethylene Glycol): it extracts Cs and Sr from UREX raffinate [26,61].
- GANEX (Group ActiNide Extraction): it consists of uranium extraction followed by group recovery of all actinides and subsequent DIAMEX/SANEX [26,46].
- Supercritical CO_2: it is a solvent extraction process which uses supercritical CO_2 with TBP. It has similar characteristics to PUREX and it extracts UO_2 and U_3O_8 [19].

Besides these modifications on the PUREX process, there is also another technology which has raised interest: the OREOX process (Oxidation and Reduction of Oxide Fuel). This process is designed to fabricate CANDU (CANada Deuterium Uranium) fuel from PWR (Pressurized Water Reactor) spent fuel and takes advantage of both types of reactors fuel characteristics, as the fissile content of a PWR spent fuel is similar to the requirements of a CANDU reactor. This nuclear fuel cycle is known as DUPIC (Direct Use of spent PWR fuel In CANDU) cycle [18,25,33].

4.2.2. Non-Aqueous Processes

The most important non-aqueous reprocessing technology is pyroprocessing, which usually focuses on advanced nuclear fuel cycles, FBRs and ADS systems, because SNF that arises from these cycles usually has very high decay heat which current technologies cannot handle [20,25]. It was first investigated in the 1950s as an alternative to PUREX to increase the radiation resistance and stability of material used in extraction processes [26].

The pyrochemical process does not involve dissolving SNF in an acid solution but its techniques are based on using metals and salts at high temperatures: melt-refining, volatilisation, gas-solid reaction, electro-deposition, electro-refining, electro-winning, and so forth. This technology has a lower separation factor; however, it takes advantage of the ability of fast reactors to use fuel with more impurities than thermal reactors. Therefore, it allows transuranic elements recycling and fully exploits nuclear fuel energy potential, which, consequently, considerably reduces the final waste volume and natural uranium requirements, up to two orders of magnitude [25,26].

This technology has other advantages, such as very high proliferation resistance, but it is still under research in order to improve its performance and determine the specific characteristics, such as the most thermodynamic stable configurations [19,26,62].

Additionally, there are other advanced technologies being investigated which combine several aqueous and non-aqueous processes. Some examples are Fluoride Volatility and FLUOREX (Fluoride volatility and solvent Extraction). The former is a process of physical separation which uses the property of volatility of uranium, plutonium and neptunium hexafluoride to separate them from other materials and is more effective when used in fuel with small amounts of Pu and Np. The latter comprises various processes including AIROX (Atomics International Reduction Oxidation) process (holes are drilled in the fuel cladding and it is subjected to cycles of oxidation and reduction), Fluoride volatility and PUREX [19].

5. Final Disposal

The final stage of the back end of the nuclear fuel cycle is unavoidable and common for all the strategies, despite of the reduction in waste volume and radiotoxicity with current or future reprocessing techniques. This last step is the final disposal of the waste and whether it is untreated SNF or vitrified HLW arising from reprocessing, it is still necessary to safely store them for the long-term until its radioactivity reaches safe levels. However, the period of time they require safe storage is highly dependent on the reprocessing technologies.

Over the years, many final disposal methods have been studied, some of which were discarded due to safety or political issues, such as disposal under the sea bed or in geological subduction zones. Nonetheless, the concept of disposal in stable geological formations has been debated and approved in many forums by international organisations and has achieved a broad consensus around the world [41]. Therefore, the preferred option continues to be the underground emplacement in a deep geological repository (DGR) [20,63].

5.1. DGR

A DGR performance is based on a multi-barrier concept, which helps to safely contain radioactivity for thousands of years. There are two types of barriers: the geological barrier and the engineered barrier. The former is the host rock, which provides stability and isolation to HLW, and its characteristics are unlikely to significantly change over the relevant time scales [20,25,41].

The latter is formed by a series of protection layers specially designed to protect HLW over the years from external agents (water filtrations, corrosion, etc.). These layers are usually a metallic container, a buffer surrounding the container to fill voids and fractures, a backfill to fill transport and access galleries and, finally, seals and plugs [25].

These different layers provide robustness and safety functions to the repository [25,41,64]:

- Physical containment: a watertight barrier isolates the radioactive waste from groundwater and no release of radionuclides can occur (at least 1000 years).
- Slow release: after container failure, various physicochemical processes strongly limit radionuclide releases.
- Retardation: the radionuclides dissolved in the groundwater start to migrate through the buffer and the host formation. However, many of them will be sorbed onto minerals.
- Dispersion and dilution: radionuclides are released into overlying or surrounding aquifers, but dispersion and dilution processes reduce their concentration.

5.2. Types of DGR

There are different types of DGRs according to the type of host rock, which are being studied around the world: hard rock formations, such as granite (Canada, Finland, Spain, Sweden, etc.); argillaceous formations, such as clay (Belgium, France, the Netherlands, etc.); salt formations (Germany and the Netherlands), and; volcanic formations, such as tuff (USA, Yucca Mountain) [25,41,65].

Additionally, there are two main types of DGR configurations: galleries, where waste is placed along the axis of a gallery; and boreholes, where waste is placed in horizontal or vertical boreholes drilled from a gallery.

Therefore, over the years many countries have executed geological studies of their bedrock formations and, thus, they have developed different designs for their DGRs. One country may have different suitable locations for its DGR, so the final selection of the site may involve political or strategic reasons.

5.3. DGR and Advanced Cycles

As seen in Section 2.5, there are many advantages from the advanced cycles, which can be extended to the final DGR design and its size. This is not only due to the direct volume reduction of the vitrified HLW and the number of containers, but, to a greater extent, to the significant decay heat reduction of the final waste, because the major constraint to the DGR size is thermal limitations: the less thermal load, the greatest repository total size reduction [25].

Some of these effects on the repository size have been published on the NEA (Nuclear Energy Agency) report titled "Advanced Nuclear Fuel Cycles and Radioactive Waste Management" [25], and several examples are listed below:

- DGR in argillaceous formations (Belgium): DGR galleries length is reduced in a factor of 3.5 and up to 9.4 with Cs and Sr separation.
- DGR in granite (Spain): final repository size is considerably smaller for advanced cycles. The length of the disposal galleries for advanced cycles has a reduction ratio from 0.844 to 0.238 compared to the direct disposal option due to the lower decay heat of the advanced cycles final waste.
- DGR in volcanic formations (USA): Pu and Am removal increases drift loading a factor of 4.3 to 5.4. Additional separation of Cs and Sr allows a greater increase up to a factor of 42.7. Subsequent studies have estimated an increase up to a factor of 100 [41].

Moreover, as seen in Section 2.5, advanced cycles can reduce the inventory and radiotoxicity of the waste by a factor of 100 to 200 and, therefore, the required time scales to reach safe radioactivity levels would be reduced from over 100,000 years to between 1000 and 5000 years. Finally, the peak dose was estimated to be reduced by a factor up to eight in advanced cycles, although any nuclear fuel cycle studied maintains the peak dose well below the regulatory limits [25,41].

These advantages, provided by the advanced technologies that are currently under research, are very important. However, they are not, at present, commercially available. Therefore, it is essential to design DGRs with retrievability as an option [66–68], in order to be able to recover SNF in the future and reduce its radiotoxicity and volume when these technologies are available.

5.4. Alternatives to the DGR

Even though the DGR is the most widely accepted option for final disposal some lines of investigation have recently raised new interest in some countries. This is the case of the space disposal system for the nuclear waste (mostly FPs), which had been studied by NASA but was discarded due to safety issues and the high cost. Nevertheless, space disposal has been redesigned and improved in a new study as an alternative to a DGR in Korea, because, due to its geographic situation and high density of population, a DGR would be difficult to implement [41,69].

There are other options which have been considered for countries in the same situation as Korea, whose geographical situation is complicated or lack suitable stable geological formations, such as the construction of a joint DGR. This DGR would store HLW from several countries, its location would be the most strategically favourable, taking into consideration that the final geological formation must be suitable. Nonetheless, this practise is currently forbidden and sanctioned, so, in order to proceed with this option, legislations would have to be modified.

6. Spanish Case and Future Challenges

As seen in Section 1, the world is living an energy transition period and the future of nuclear power is uncertain. In this context, some of the Spanish political parties are also questioning its future and proposing the closure of all NPPs at the end of their initially designed lifetime (40 years). Furthermore, in Spain the uncertainty surrounding nuclear power is also occurring regarding SNF management, because of the delays and controversy surrounding some of the proposed plans.

In 2006, the company which manages nuclear waste in Spain, ENRESA, planned the construction of a centralized interim storage facility (CTS) and was considered a priority in the sixth general plan of radioactive waste [17], which was in line with the proposal of following a direct disposal strategy. This CTS was planned to start operating in 2010, but due to political reasons, its construction has not started yet, even though the location has been selected and approved.

Moreover, the vitrified waste which came from reprocessing in France and the UK had to return to Spain in 2015, but due to these important delays in the CTS construction, which was the facility that was going to receive and storage this waste, they have not returned yet. This is going to cost millions of euros to Spain. Thus, it is a priority for Spain is to solve this issue as soon as possible.

Therefore, the current efforts surrounding nuclear waste management are concentrating on solving this interim storage problem and there are no real projects for the construction of a deep geological repository, which will be necessary sometime in the future. However, Spanish research centers, such as CIEMAT (Center for Energy, Environmental and Technological Research), have studied several DGR host rock formations in Spain and different configurations. These R&D efforts and these studies have served as an international reference for other DGR projects [25] and have laid the ground for future DGR plans in Spain.

Furthermore, despite not being currently on the spotlight, CIEMAT, as well as other research centers and some companies are working and researching on different nuclear waste subjects. Some of these are advanced reprocessing technologies, which are very interesting and important regarding the future of SNF and HLW, as it could lead to very significant volume and radioactivity reductions and, also, to an important decrease of the final waste isotope's half-lives.

Despite having considered a direct disposal option as a SNF management strategy in Spain, due to all the uncertainty regarding nuclear waste management, reprocessing could still be reconsidered in Spain, taking advantage of the benefits that this entails, such as volume reduction. Additionally, in 2012 the Taxation Measures Law on Environmental and Energy Sustainability Matters [70] was introduced in Spain, which penalizes nuclear waste generation. This law is very important, as it can lead to the closure of the Spanish NPPs, because it levies the production of electricity generated by nuclear power in such way that it becomes economically not profitable.

Therefore, this law has a major impact in SNF management [36] and it could benefit the reprocessing option if managed properly. In this scenario, SNF should not be considered nuclear waste

but an energy source and, thus, NPPs should not be levied for SNF production. For this purpose, a consensus would have to be reached between the electric companies, which have been damaged with this law; the government, which is involved in the decision-making process; and the compromise towards future generations.

7. Conclusions

Nuclear power has been in constant research and development since its beginnings. Current technologies have thoroughly improved and they provide solutions for the short-term. However, advanced technologies that are currently under research have the potential of solving one of the key issues regarding nuclear power: waste management.

Therefore, it is important to continue R&D in nuclear energy and, while these new technologies are not commercially available, address the nuclear waste management issue as conveniently as possible for each country and its particular context, taking into account several factors such as economic viability, environmental impact, material flows, and resource utilization.

Additionally, there is an important factor to consider: retrievability. This is essential for the DGRs that are currently under construction, because it provides the possibility of recovering SNF and nuclear waste in the future, when advanced technologies are commercially available and, thus, eventually reducing the volume, radiotoxicity and half-life of the waste. Thus, all of the current DGR designs must include this feature in their design.

Furthermore, governments, electricity companies and the nuclear sector around the world should increase measures to improve social acceptability of nuclear power, by means of educating the public, demonstrating its advantages and implementing effective strategies that enhance key issues such as nuclear waste management.

Finally, for the Spanish case, the taxation system should be reconsidered and, instead of paying taxes as a disincentive for waste management, those earnings should be reinvested in improving its sustainability and safety, with the purpose of avoiding leaving the problem to future generations.

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