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Review

Trends and Perspectives on Nuclear Waste Management: Recovering, Recycling, and Reusing

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Abstract: This paper focuses on the highly radioactive, long-lasting nuclear waste produced by the currently operating fission reactors and on the sensitive issue of spent fuel reprocessing. Also included is a short description of the fission process and a detailed analysis of the more hazardous radioisotopes produced either by secondary reactions occurring in the nuclear installations or by decay of the fission fragments. The review provides an overview of the strategies presently adopted to minimize the harmfulness of the nuclear waste to be disposed, with a focus on the development and implementation of methodologies for the spent fuel treatments. The partitioning-conditioning and partitioning-transmutation options are analyzed as possible solutions to decrease the presence of long-lived highly radioactive isotopes. Also discussed are the chemical/physical approaches proposed for the recycling of the spent fuel and for the reusing of some technologically relevant isotopes in industrial and pharmaceutical areas. A brief indication is given of the opportunities offered by innovative types of reactors and/or of new fuel cycles to solve the issues presently associated with radioactive waste.

Keywords: nuclear waste; spent fuel; nuclear installations; fission products; actinides; transmutation chains; radioisotope recovery

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1. Introduction

The technology of nuclear fission is undergoing a paradigm shift from a mere electricity producer to a provider of sustainable energy for a variety of applications, from water desalination to hydrogen production. Moreover nuclear-renewable hybrid energy systems to produce electricity and heat are now seen as a promising way to decrease greenhouse gas emissions and effectively address the issue of climate changes [1].

The more recent IAEA/PRIS information on nuclear reactors designed to generate electricity for a power grid indicates a total of 419 nuclear power reactors with operational status in 32 countries, providing ~16% of the electricity in the world [2]. Due to the conjunction of the increased power demand expected in the near future and of the necessary energy transition, 63 nuclear reactors are under construction in 16 countries and another 128 are planned in 19 countries. Another 30 countries have expressed their intention to adopt, for the first time, nuclear energy in their energy mix [3].

However, one of the most criticized characteristics of nuclear energy is the waste produced as a by-product of the fission process and this raises a real problem when nuclear reactors or fuel processing plants are refueled or decommissioned [4]. The development of solutions for radioactive waste management faces many different challenges that span from the technical issues to a series of complex social/political factors and to unavoidable economic influences [5]. Moreover, different approaches must be used depending on the kind of waste. What is known as "nuclear waste" encompasses, indeed, the spent fuel coming from the reactor's core but also the radioactive materials coming from medical or industrial applications and from research facilities [6]. It is noteworthy that, even if the IAEA offers legal instruments to address the issue of spent fuel and radioactive waste management on a global scale [7,8], there are differences in the way each country tackles the topic of waste categorization and of subsequent waste disposal

The indications of the US-NRC, followed by several countries, establish the processing and handling of the nuclear waste according to three levels of radioactivity, namely, high, intermediate, and low [9].

The high-level radioactive waste is made of “spent fuel”, mainly coming from nuclear power reactors, but also includes fuel cladding. This waste represents only 3% (vol) of the total but accounts broadly for 95% of the radioactive hazard [10] and requires specific treatments and immobilization operations in “high level waste facilities” where it is stored in pools to cool for many years and then moved to above-ground concrete casks [11]. Such temporary storage solutions, designed to safely store the waste for a limited period, require the presence of technical staff to monitor the chemical/physical conditions of the disposal site and of security agents to prevent terroristic acts. Containers for final longer-term unmanned storage are designed to isolate the waste for about 100,000 years in deep geological repositories, either retrievable by future generations or permanently sealed. The disposal of the high-level radioactive waste is typically perceived by the public as an extremely difficult problem that requires high financial investments to solve [5].

The radioactive isotopes employed for medical, industrial, agricultural, and research purposes constitute a low-level waste. Another kind of low-level waste is produced by the milling of ores from which Uranium or Thorium are extracted [9]. The radioactive content of all these materials, which contain low concentrations of radionuclides with long half-lives, is typically about 1%. The low-level waste, which represents about 90% (in volume) of the radioactive materials to be disposed of, is typically stored in near-surface appropriate facilities until it turns into a harmless waste. Some additional precautions are adopted in the case of the waste with radioactive content up to 4%, consisting mainly of lightly contaminated items, like tools and work clothing from power plant operators.

It must be noted that in today’s more common power reactors, the “nuclear fuel” is ^{nat}U or ^{nat}U enriched with 3–5% ^{235}U isotope, contained in rods generally assembled in square grids. Exceptions are the Russian Light Water Reactors (LWRs), characterized by a hexagonal assembly.

The timing of fuel replacement depends on the position of the assemblies in the reactor. In 25–33% of the reactors, the fuel is removed every 12–24 months; in other cases, the fuel can operate for 3–7 years. Also, the fuel that remains in the reactor core can be removed. As a whole, all the removed fuel constitutes the so-called “spent fuel”, a waste to be managed following two different nuclear fuel cycle concepts, namely, the open and the closed ones. In the open cycle, the irradiated fuel makes a once-through passage from the removal at the end of its use to the disposal in appropriate repositories. In the closed fuel cycle, the fissile material is recovered from the irradiated fuel, reprocessed, and re-utilized for the fabrication of new fuel [10].

However, to consider the used fuel assemblies as “spent fuel” is rather misleading. Indeed, as has been suggested, it would be better to replace this term with the more precise “slightly used fuel” because, independently of the kind of reactor, the fuel assemblies extracted from the reactor vessel still contain large amounts of U, Pu, fission products, and minor actinides [12].

As an example, the fuel extracted from a typical LWR (see Section 2) still contains fission products, Pu isotopes, and minor actinides, in addition to ~96% of the pristine ^{238}U and ~0.9% of the ^{235}U [13]. Moreover, the decay of the primary radioisotopes produced by the fission process gives rise to a variety of other radionuclides [10,14].

It is therefore evident that a huge amount of radionuclides are still present in spent fuel, which is to be considered a precious resource rather than a hazardous waste to be disposed of [15]. Following the more recent industrial-scale technologies, almost 96% of the spent fuel extracted from the reactors can be recycled. Reprocessing the used nuclear materials enables the production of new fuels and offers the additional advantage of reducing the volume of waste to be stored, at the same time avoiding the consumption of raw materials, sometimes critical for geopolitical reasons.

In this view, some countries, in *primis* France, Russia, China, Japan, and India, are carrying out the option to reprocess their used fuels, recovering fissile nuclides, which can be supplemented to the fresh fuel of nuclear power plants [9]. Sustainability, safety, and cost benefits have been the general objectives that the present initiatives wish to reach, even if the many differences in the parameters of existing and envisaged installations make it impossible to have a fully shared roadmap [10].

The main ways in which the nuclear waste can be disposed are schematically illustrated in the following table, Table 1.

Table 1. Disposal procedures for the main classes of nuclear waste.

SPENT FUEL AND MATERIALS	DISPOSAL
Fuel cladding, filters, components	Storage as low/intermediate-level waste
Reprocessed U and Pu	Reuse in thermal or fast reactors
Long-lived minor actinides	Partitioning and transmutation before storage
Long-lived fission products	Storage as high-level waste
Selected radionuclides	Extraction and recycling in industry and medicine

This paper aims to analyze the current status of the strategies adopted to minimize both the volume and threat of the nuclear waste to be disposed of whilst highlighting the potential options and the most promising perspectives for the recycling and reusing of spent fuel.

2. A Look at the Fission Technology

The majority of the worldwide working nuclear reactors employ Uranium-based fuels and convert in power the energy released by the fission of the fissile isotopes ^{235}U, ^{233}U, and ^{239}Pu.

The more common reactors are the so-called "Slow Neutron Reactors" (SNRs) where fission is induced by neutrons of energy at about 0.025 eV. These thermal neutrons have indeed the right "cross section" for neutron capture by such fissile isotopes and are therefore able to sustain the nuclear lifecycle [16]. In this context, a moderation performed by light atoms (H, D, C) is needed to reduce the high energy (1–2 MeV) of the prompt neutrons emitted when the nucleus of a fissile isotope is split into two fragments. The slow neutron reactors can be broadly classified into two main classes, Light Water Reactors (LWRs) and Heavy Water Reactors (HWRs), depending on the kind of water (H_2O or D_2O) used as a neutron moderator. A third class of SNR is represented by the graphite-moderated gas-cooled reactors (GCRs) and high-temperature gas-cooled reactors (HTGRs) that, in the past, operated in France, Japan, Italy, Germany, the United Kingdom, and the United States and that, in their last configuration—Advanced Gas-cooled Reactors (AGRs)—are still operating in the UK. Regarding the HTGRs, presently one is up and running in Japan and three in China.

In the SNRs, the nuclear fuel is typically UO_2 (either ^{nat}U or 3–5% ^{235}U-enriched ^{nat}U) in the form of small ceramic pellets encased into metal rods that are bundled together generally into square assemblies (Figure 1, Left).

To operate efficiently, the various kind of reactors require different kinds of fuel. To go critical, the PLWRs must use a U-fuel enriched (3–5%) in ^{235}U, whereas the PHWRs can run using ^{nat}U, which contains about 0.72% of the fissile ^{235}U isotope. The AGRs, which employ graphite for neutron moderation, have been designed to operate with 3.5%-enriched U.

Each fission event produces, on average, two and a half neutrons, so if all the neutrons were allowed to initiate a new fission, a branching process rather than a linear chain would occur. To maintain the reactor under critical working conditions, it is essential that at least the same number of neutrons are produced from one generation to the next or, in other words, to have $k \geq 1$, where k is the neutron multiplication factor. If this condition is no longer achieved, the reactor becomes subcritical and shuts down. The strategy to prevent this is the use of control rods that enable a one-for-one reaction and assure the maintenance of a linear fission chain.

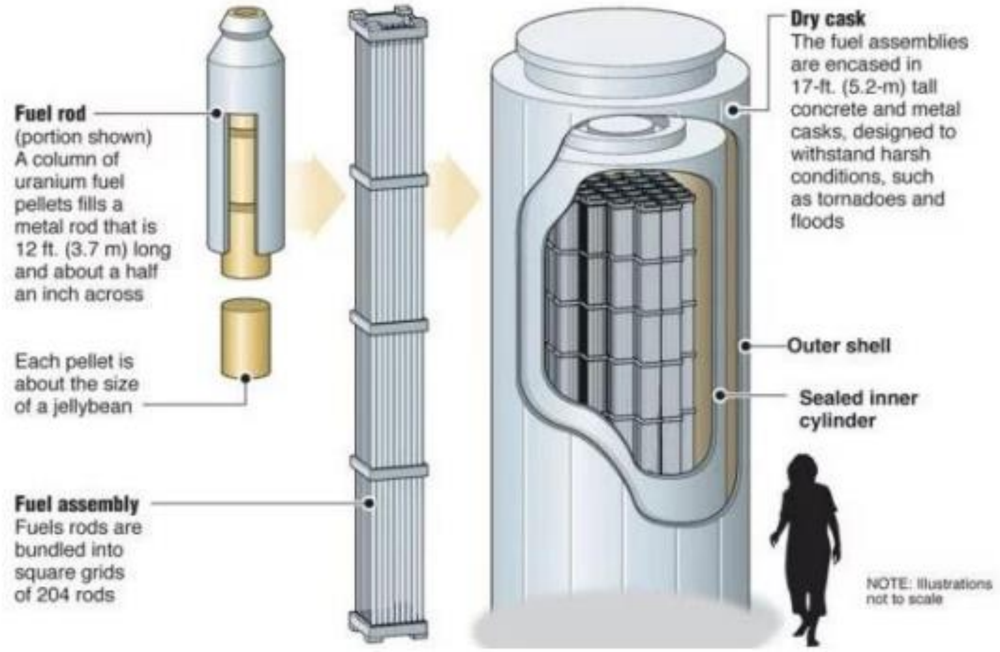

Figure 1. Left: Schematic view of fuel rods and of their assembles in square grids; Right: Scheme of the dry cask used for the storage of spent fuel [Source: U.S. Nuclear Regulatory Commission—NUREG].

Of the three fissile nuclides, ^{235}U, ^{233}U, and ^{239}Pu, only ^{235}U is naturally found (about 0.72% in ^{nat}U); the two others must be produced by nuclear reactions.

Small amounts of ^{239}Pu are obtained as a by-product of neutron capture by the fertile ^{238}U in SNRs through the reaction chain:

$$^{238}\mathrm{U} + \mathrm{n_{th}} \rightarrow {}^{239}\mathrm{U}(\beta^-) \rightarrow {}^{239}\mathrm{Np}(\beta^-) \rightarrow {}^{239}\mathrm{Pu}.$$

However, the yield in ^{239}Pu can be strongly increased by an improved neutron economy in the Fast Neutron Reactors (FNRs), a much less common class of reactors where fission of Uranium isotopes is induced by “fast” neutrons (energies > 1 MeV). This peculiarity opens reaction paths rather different from those occurring in slow neutron reactors and enables the utilization of U-based fuels about 60 times more efficiently than a common reactor [17]. Some of the ~20 FNRs that have been working since the 1950s are the so-called “fast breeder reactors”, specifically designed to generate more fissile nuclear fuel than they consume [18].

Using appropriate installations, it is also possible to produce the fissile ^{233}U from the fertile ^{232}Th isotope:

$$^{232}\mathrm{Th} + \mathrm{n_{th}} \rightarrow {}^{233}\mathrm{Th}(\beta^-) \rightarrow {}^{233}\mathrm{Pa}(\beta^-) \rightarrow {}^{233}\mathrm{U}.$$

This last reaction could also efficiently take place in a cutting-edge Accelerator Driven System (ADS), designed to combine a Th-based reactor with a spallation neutron source. As demonstrated by Rubbia et al., the fission of ^{233}U derived from the conversion of ^{232}Th in an ADS reactor is a clean and inherently safe fission process because an ADS can only run when neutrons are supplied to it [19,20].

An in-depth analysis of the current state of art and of international ongoing developments regarding fast reactors can be found in [21]. Given the limited number of the currently operating FNRs, from there onwards, the present paper will cover mainly the topic of the spent fuel from SNRs.

The composition of the spent fuel at the end of the operational cycle of a SNR is determined by two different processes, namely, (i) fission of fissile nuclides and (ii) thermal-neutron capture followed by sequential β^--decay.

The fission induced by thermal neutrons in a fissile nucleus of atomic number Z produces a bimodal mass distribution of fragments, with the light-fragment peak at around ~0.41 Z and the heavy one at around ~0.59 Z. The ^{235}U isotope (Z = 92) can split into two Z = 46 nuclides, following the less frequent symmetric splitting mode, or in a Z = 40 plus a Z = 52, a Z = 39 plus Z = 53, and so on. Next, the unstable neutron-rich primary fission fragments decay to more stable states emitting a sequence of highly energetic β^--particles, giving rise to a wide range of nuclides. An example is shown in Figure 2, which illustrates the case of fission-producing ^{94}Sr (Z = 38) and ^{140}Xe (Z = 54) nuclides and shows the decay chains starting from such fission fragments.

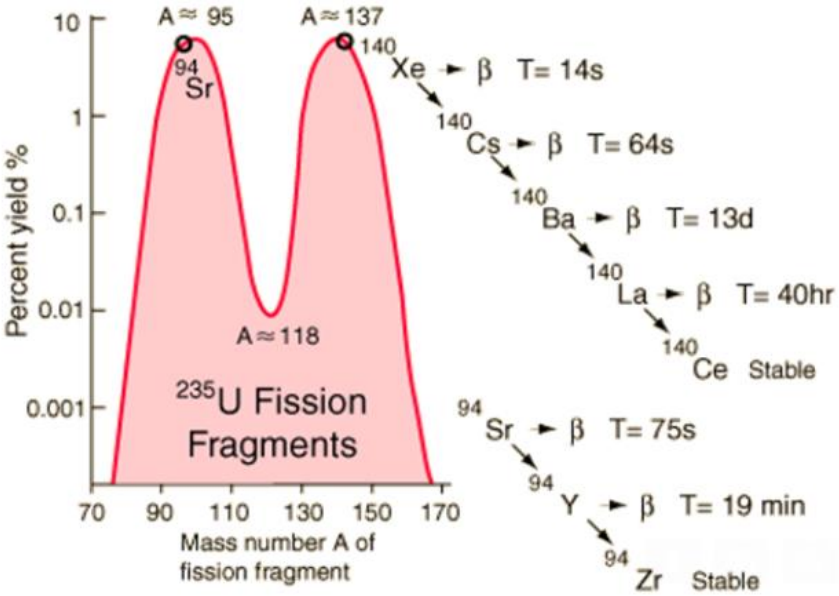

Figure 2. A scheme of ^{235}U fission producing the ^{94}Sr and ^{140}Xe fission fragments [Reproduced by courtesy of Rod Nave, Georgia State University].

It is to be noted that the fission of ^{235}U can occur following more than fifty different modes, giving rise to many hundreds of either radioactive or stable nuclides [22]. Collectively, the fission fragments and the isotopes produced by the secondary processes are called fission products.

Whereas the pristine ^{235}U isotope is quite completely split into fission fragments, other ^{235}U is generated by secondary reactions and this explains why, in reactors running with ^{nat}U, the removed fuel is found slightly enriched in ^{235}U. In these reactors, only a small fraction of ^{238}U present in the fuel undergoes fission, whereas this fertile isotope can capture neutrons emitted by the fission process giving rise to ^{239}U and to a series of actinides, mainly Np and Pu isotopes. The Plutonium concentration in the fuel increases with time [23] and the fissile ^{239}Pu generated from ^{238}U supplies up to 1/3 of the energy provided by a typical LWR [4]. The final composition of the fuel discharged from a LWR, at normal burn-up levels, depends not only on the type, chemical composition, and degree of ^{235}U enrichment of the pristine fuel but also on the neutron energy spectrum.

As an example, what happens in a conventional PLWR nuclear reactor scaled to an electricity power output of 1 GW_e after one year of uninterrupted operation? To accomplish such a task, the initial amount of required fuel is 27.228 tonnes of Uranium Dioxide ($^eUO_2$) enriched with 3.54% of ^{235}U. This material contains 23.16 tonnes of ^{238}U plus 0.84 tons of

^{235}U, totaling 24.0 tonnes of ^{e}U [24]. After one year of reactor operation, the consumed amount of the pristine ^{e}U is 1649 kg, of which there is 1018 kg of ^{238}U and 631 kg of ^{235}U. These quantities constitute the "nuclear ashes", the small fraction of the fission products and actinides resulting from the burning of the initial fuel. It is to be remarked that 1.06 kg of the initial pristine fuel produces 8.76 TWh of electricity for external use, with a process efficiency of ~33% (a burning of ~46 GWd/ton of ^{e}U).

The spent fuel contains 472.3 kg of actinides, responsible for ~1.5 × 10^5 TBq of total activity, plus another 1176.6 kg of fission products (~50 × 10^5 TBq). The composition and activity of the waste produced after 1 year of operation by a typical LWR of 1-GW$_e$ output are reported in Table 2.

Table 2. Composition of the total spent fuel for a conventional PWR of 1-GW$_e$ electricity output after one year of uninterrupted operation: (i) unused pristine fuel (upper part); (ii) actinides (middle part); (iii) fission products (bottom part) [24].

Radionuclide	Half-Life [a]	Mass [kg]	Activity [TBq]
^{235}U	7.04×10^8	209.0	1.67×10^{-2}
^{238}U	4.46×10^9	22,141	0.28
Total	**------**	**22,350**	**0.30**
^{234}U	2.45×10^5	6.2	1.43
^{236}U	2.34×10^7	137.5	0.33
^{238}Pu	87.7	5.6	3.55×10^3
^{239}Pu	2.41×10^4	177.2	4.07×10^2
^{240}Pu	6.56×10^3	69.1	5.81×10^2
^{241}Pu	14.3	37.2	1.43×10^5
^{242}Pu	3.75×10^5	15.3	2.23
^{237}Np	2.14×10^6	13.5	0.352
^{241}Am	4.33×10^2	6.8	0.86×10^3
others	-------	3.9	------
Total	**-------**	**472.3**	$\mathbf{1.48 \times 10^5}$
^{89}Sr	0.138	0.1	1.08×10^5
^{90}Sr	28.9	18.0	0.91×10^5
^{106}Ru	1.02	6.7	0.82×10^6
^{134}Cs	2.05	35.1	1.69×10^6
^{137}Cs	30.0	42.9	1.38×10^5
^{144}Ce	0.78	13.8	1.62×10^6
^{147}Pm	2.62	12.0	4.12×10^5
^{154}Eu	8.59	1.2	1.20×10^4
Others	-------	136.3	0.85×10^5
Stables	-------	910.5	-------
Total	**-------**	**1176.6**	$\mathbf{49.8 \times 10^5}$
Grand Total	**-------**	**23,9989**	$\mathbf{51.3 \times 10^5}$

The 22.350 tonnes of unburned fuel contain 0.209 tonnes of ^{235}U; therefore, such a big fraction of the "nuclear waste" is still considerably enriched (~0.94%) with fissile ^{235}U. The unused fuel can be totally or partially prepared by separating it from the spent fuel to obtain a new fuel for further reactor operation.

In the spent fuel, Pu isotopes are the most abundant ones among the produced actinides, whereas ^{134,137}Cs and ^{90}Sr are the predominant ones in the fission products group. The ^{239}Pu can be separated from the reprocessed fuel to be mixed with U to produce a mixture of U and Pu oxides (MOX), which can serve as a new fuel. Among the actinides contained in the spent fuel, the ^{241}Pu isotope is the most active and the ^{236}U is the least active. Around 270 kg of active fission products present in the spent fuel provide a total activity ~34 times greater than the ~470 kg of all the actinides and ~17 million times greater than the 22.35 tonnes of the unused fuel (see Table 2).

3. Used Fuel Processing

The methodologies used in processing spent fuel strongly depend on the reactor where the recycled material is going to be reused. Moreover, different methodologies must be applied if the reuse of nuclides from spent fuel is meant for purposes other than energy production. These last cases regard the mining of selected radionuclides for medical or industrial applications but also initiatives aimed at reducing the amount of high-level radioactive waste.

The reprocessing of spent fuel for use as fresh fuel in reactors is carried out by means of two main methodologies, namely, the aqueous and the non-aqueous ones. The last involves an initial pyroprocessing step that employs molten salts to dissolve the fuel rods and subsequent electroplating processes aimed at separating the various elements. This separation method was proposed some decades ago [25] but pyroprocessing technologies are still at the prototype stage and are not yet commercialized [26].

Today the more commonly used method is aqueous recycling. All the commercial reprocessing plants use the hydrometallurgical process PUREX (Plutonium Uranium Reduction Extraction), based on the dissolution of the used fuel rods in concentrated nitric acid, followed by a series of solvent extraction steps to separate the different components. The spent fuel contains a large spectrum of mostly radioactive nuclides with varying valency states and this makes its reprocessing very complex. The PUREX method enables an efficient separation of the rod materials into three main groups, namely, U, Pu, and a remaining highly radioactive component.

The composition of the used U recovered from the aqueous acid phase by the PUREX process depends on the enrichment degree and on the time the fuel has worked [27]. The fuel extracted from a LWR is valuable for its fertile potential. It is slightly enriched in ^{235}U and contains small amounts of ^{236}U, as well as traces of ^{232}U; the rest consists of the largely unused ^{238}U. The ^{232}U, produced by the α-decay of ^{236}Pu, generates daughter nuclides that emit strong γ-rays. Whereas this makes rather difficult the handling of the reprocessed material, such a Uranium isotope does not represent a trouble for the operating reactor because it easily captures a neutron becoming a fissile ^{233}U. Conversely, the 0.5% of ^{236}U, also a strong neutron absorber, reduces the number of neutrons available for fission processes and makes it necessary to further enrich ^{235}U the reprocessed fuel before the reuse in a reactor. Other U isotopes present in traces, such as the fertile ^{234}U and the short half-life β^--emitter ^{237}U, do not affect, at all, the performances of reprocessed fuel.

The current approach for the reuse of the extracted U, if necessary returned to a plant for a re-enrichment, is to mix its oxide (UO_2) with PuO_2. The mixed oxide fuel (MOX) produced may be suitable for use in water-cooled reactors. It is noteworthy that, while in the past, such reactors could be only partially fueled by MOX, the last generation is able to operate with a 100% MOX core. However, the economy of the process limits the recycling to only once or possibly twice.

Modifications of the PUREX process have led to several other separation methodologies, all operating by a solvation mechanism [28]. The most significant is the UREX process, a modified method of U extraction that is set up to avoid the issues associated with the production of Pu. Beyond preventing Pu extraction, the UREX process enables the efficient recovery of some fission products, such as ^{129}I (by 95%) and ^{99}Tc (by 90%), as well as smaller amounts of ^{135}Cs and ^{90}Sr [29].

Whereas for use in today's LWRs, the reprocessed U needs to be enriched, the Pu obtained from the PUREX process can be directly used for the fabrication of MOX fuel. As an average, the Pu component (1–2% of the discharged fuel) consists of ^{239}Pu (~0.5%), ^{240}Pu (~0.9%), ^{241}Pu (~0.1%), and ^{242}Pu (~0.05%). The isotopic concentration of the separated Pu phase, and therefore its value for refueling, is determined by the fuel burn-up level. High levels of burn-up increase, indeed, the percentage of non-fissile Pu-isotopes and of minor actinides.

The ^{239}Pu coming from the extraction processes is immediately sent to MOX plants, in order to avoid any potential security risk associated with a Pu storage [30]. The threats that the building up of Pu stockpiles would pose are a concern that led the USA, in the frame of a non-proliferation policy, to rule out any civilian reprocessing able to completely separate the fissile isotope. In particular, the separation of ^{239}Pu from ^{240}Pu is prevented because the contamination by ^{240}Pu inherently limits the possible use of ^{239}Pu in a nuclear bomb. In effect, beyond the α-decay mode leading to ^{236}U, the ^{240}Pu isotope also undergoes spontaneous fission, releasing a neutron flux that makes it difficult to reach the conditions for an implosion [31]. Small amounts of recovered Pu are instead retained in secure storage facilities to produce, by β^--decay of ^{241}Pu ($T_{½}$ = 14 yr), the isotope ^{241}Am, a very suitable source for the powering of nuclear batteries [32].

The waste remaining after the U and Pu removal by the PUREX process and follow-up treatments is, in general, conditioned/stabilized and incorporated into insoluble borosilicate glass materials for storage in a repository. However, this high-level radioactive liquid phase still contains valuable amounts of fission products and minor actinides.

Regarding the fission products, these represent about 3% of the treated fuel. Although most of the radioisotopes are derived from the fission decay within days or weeks, the remaining fission products account for most of the heat generated by the nuclear waste.

Among the minor actinides, some very hazardous isotopes, such as ^{237}Np, ^{241}Am, ^{243}Am, and ^{244}Cm, are still present (about 1%) in the liquid waste and are responsible for the short- and long-time high radiotoxicity.

4. Further Steps: Conditioning, Partitioning, Transmutation

The PUREX process enables the recovery of U and Pu isotopes from spent fuel and the manufacture of a MOX material suitable for reuse. This conventional reprocessing not only saves about 20% of U raw material but also reduces, by a factor of about 3.5, the total volume of the high-level waste to be treated; conditioned; dispersed in a glass, ceramic, cement, or bitumen matrix; and finally stored in a suitable repository [33]. However, this complex and expensive procedure does not solve the problem of nuclear waste that remains highly radioactive for thousands of years due to the presence of minor actinides and some fission products [34]. The contributions of the actinides and fission products to the total residual radioactivity of spent fuel produced by the burn-up of 1 tonne of U is shown in Figure 3.

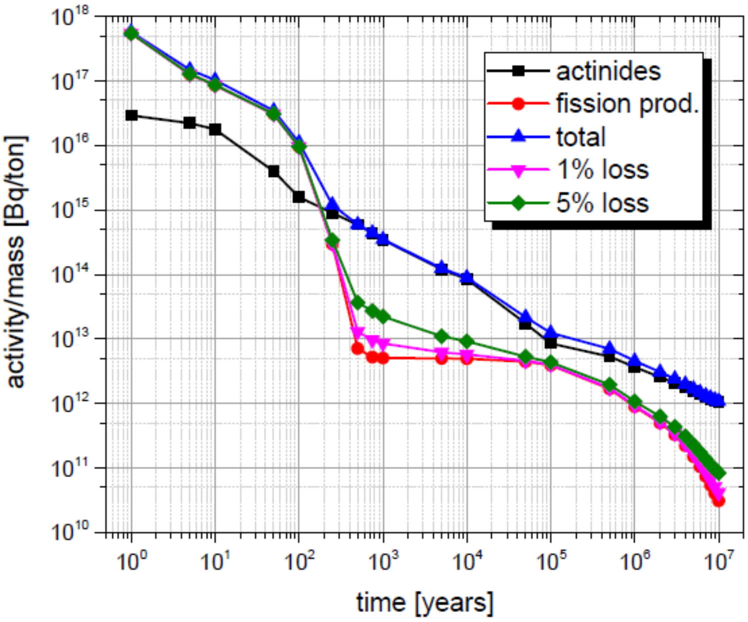

Figure 3. Trend of total radioactivity vs. time of the spent fuel produced by the burn-up of 1 tonne U (blu points), with an indication of the contribution of actinides (black points) and of fission products (red points). The effect of actinides losses during reprocessing is also indicated: 1% loss (pink points), 5% loss (green points) [Reproduced from Ref. [35] (open access)].

A more effective management of spent fuel requires methodologies able to remove from the liquid phase not only U and Pu but also the other nuclides that drive the main long-term hazard associated with radioactive waste [36]. In this view, a series of chemical procedures have been established with the objective of separating long-lived radionuclides from the short-lived ones. This chemical step, known as partitioning, is very often carried out before the conditioning and the final disposal of the nuclear waste.

However, a reprocessing approach more advanced with respect to the "partitioning-conditioning" (P&C) one is the "partitioning-transmutation" (P&T) strategy, proposed in the 80s to reduce the inventory of radioactive wastes to be stored [37]. The term transmutation refers to nuclear processes induced by neutron bombardment, so the neutron energy spectra and the cross-sections for neutron absorption drive the competition between fission and neutron capture. The coupling of chemical separation processes (partitioning) with transmutation processes has been conceived to transform highly radioactive and long-lived isotopes present in the waste into less dangerous or significantly shorter-lived isotopes [38–40].

The strategies for the management of used fuels pursue two main goals, short- and long-term ones. The short-term objective is to reduce the radiotoxicity of the fuel just removed from the reactor, radiotoxicity that is mainly due to the presence of Pu and minor actinides. The second goal is to reduce the residual radioactivity, minimizing the long-term risk of hazardous leaks from the repository into the biosphere.

The solubility and mobility of the waste components, which are not only in solid form but also in gas and liquid phases, represent indeed a further risk associated with the treated spent fuels [41]. To avoid any release of volatile radioactive compounds and any migration of metallic particles in liquid effluents, complex and expensive engineering solutions for underground repositories must be taken [42].

The P&T methodology has been proposed as a valid approach to decrease the amounts of long-lived highly radioactive isotopes and to mitigate their thermal effects, reducing the precautions required for the handling of used fuels and downsizing the areas of the disposal facilities [43,44].

The complexity of the nuclear processes occurring during the running of a fission reactor and, thereafter, during the storage of the spent fuel makes it necessary to consider separately the issues of short-term and long-term hazards due to accumulation of the more significant isotopes.

Regarding the actinides, studies of decay data and parent/daughter relations have suggested that, beyond the U and Pu isotopes, ^{237}Np, ^{241}Am, ^{243}Am, and ^{244}Cm are the ones more meant for transmutation [45]. In the initial period of storage, the most hazardous among the minor actinides is the α-emitting ^{244}Cm ($T_{½}$ = 18.11 a), which decays to ^{240}Pu. The ^{244}Cm isotope is characterized by a large cross-section for thermal neutron capture and, therefore, can be easily transmuted in ^{245}Cm. While a fraction of the ^{245}Cm isotopes undergo fission, small amounts of the higher isotopes, $^{246–248}$Cm, are produced by successive capture of neutrons. Further β-decay processes also create some isotopes of Bk, Cf, Es, and Fm.

Conversely, in the time range of 100–100,000 a after being removed from the reactor, the radioactivity of the fuel is determined by the Pu and Am isotopes [46]. In particular, after about 100 a of storage, more than 90% of the total radiotoxicity due to actinides comes from ^{241}Am. This isotope, produced in low-enriched power reactors through the successive formation of a series of Np (including the ^{237}Np) and Pu isotopes, continues to be generated in storage facilities by the β-decay of ^{241}Pu ($T_{½}$ = 14.3 a). The long-term storage of ^{241}Am leads, through the ^{242}Am and ^{242}Cm nuclides, to the formation of the ^{238}Pu. Also, the ^{243}Am is mainly obtained from a member of the Pu family, in this case from the short-lived ^{242}Pu through the ^{243}Pu and partially from ^{242}Am through the metastable isomer ^{242m}Am. After that, neutron capture by ^{243}Am generates the ^{244}Am ($T_{½}$ = 10 h) and the dangerous ^{244}Cm. This last isotope also comes partially from ^{243}Cm, in turn, a product of neutron capture by ^{242}Cm.

After 100,000 a, the radioactivity is produced rather exclusively by ^{237}Np ($T_{½}$ = 2.144 × 10^6 a), an α-emitter which, through the short-lived ^{233}Pa (27 d), the ^{233}U (1.6 × 10^5 a) and the ^{229}Th (0.793 × 10^4 a), finally generates the very long-lived ^{209}Bi ($T_{½}$ = 2.0 × 10^{19} a).

Figure 4 shows the isotopes typically produced from a low-enriched U fuel and highlights the all of the actinide transmutation chains. As one can see, the complex scheme of nuclear reactions and parent/daughter radioactive decays that starts from ^{238}U and ^{235}U leads to the formation of a number of Pu and minor actinides isotopes.

All these actinides, not present in the unirradiated nuclear fuel, strongly demand the use of transmutation approaches to transform highly radioactive nuclides into less hazardous ones [47].

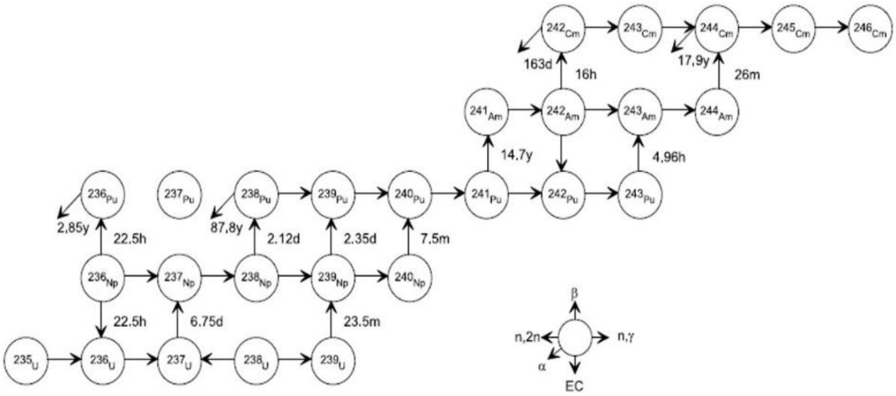

Figure 4. Scheme of actinide transmutation chains. The principal production pathways of the various isotopes, i.e., the nuclear reactions (n,2n) and (n,γ) and the radioactive α, β, and EC decays, are indicated at the right bottom (Reproduced with permission from Salvatores and Palmiotti, Ref. [48], copyright Elsevier, 2011).

An efficient transmutation of actinides critically depends on the efficiency of the initial partitioning step. High-performance procedures for the separation of Np, Am, and Cm, either on a collective or individual basis, are needed to produce high-purity targets and avoid residuals that can interfere with the transmutation processes.

In particular, it is critical to achieve the most complete removal of the fertile ^{238}U isotope that would capture neutrons generating further transuranic isotopes and of the lanthanides, also present in the remaining liquid waste [48]. The lanthanides have indeed a high cross-section for neutron absorption; therefore, it is necessary to eliminate them in order to create conditions favorable for the transmutation of actinides [27]. However, due to the quite similar chemical behavior of actinides and lanthanides, an effective separation and complete removal of lanthanides is a very difficult task [49,50].

A further observation is that the actinide's transmutation of itself may have a strong impact on both fuel handling and repository performance. An example is the unavoidable generation of ^{252}Cf, which undergoes spontaneous fission releasing an extremely high number of neutrons (~10^{12} n g^{-1} s^{-1}). Depending on the type of reactor, different solutions have been suggested to mitigate the troubles while continuing to assure better repository performances and compliance with non-proliferation regulations. The various options include the partial recycling of actinides, the reduction in the number of cycles, and the extension of the cooling steps between successive fuel loading [51,52].

By the partitioning-transmutation approach, also, the fission products, either the pristine fragments or the nuclides produced by secondary reactions and decay chains, could, in principle, be separated from the waste and transmuted into less dangerous isotopes [33]. Among the fission products, ^{79}Se, ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, and ^{135}Cs are the ones that deserve more attention because they strongly contribute to the high radioactivity of the conditioned nuclear waste during the first 100 a after its removal from the reactor. In particular, ^{99}Tc and ^{129}I account for over 90% of the activity of the long-lived fission products and their removal can significantly reduce the activity of the waste products [38]. In any case, just after the reactor shutdown, a complex interplay between the build-up and decay of fission-derived radioactive species modifies the chemistry of the spent fuel, as has been highlighted in the case of Iodine [53]. A capture process can no longer occur, whereas the β-decay of the parent ^{135}I ($T_{½}$ = 6.7 h) still generates further ^{135}Xe [54].

It is to be noted that, whereas a huge amount of data is presently available for the transmutation of Np, Am, and Cm, only a small number of studies have been performed on the transmutation of the fission products. The first technical reason for this is that the transmutation of fission products requires larger amounts of neutrons with respect to that of actinides [55,56]. An example is given by the transmutation of ^{99}Tc to the stable ^{100}Ru, after neutron capture and the decay of ^{100}Tc (β^--decay, $T_{½}$ = 16 s), or of ^{129}I to the stable ^{130}Xe, after neutron capture and the decay of ^{130}I (β^--decay, $T_{½}$ = 12.4 h). The (n,γ) reactions that would enable the elimination of the hazard arising from ^{99}Tc and ^{129}I require intense neutron fluxes that cannot be generated in conventional reactors but need different types of installations [56,57].

A second point is that studies about the radiotoxicity of highly active fission products have evidenced how the long-term threat due to such nuclides is lesser than that of the actinides [47]. As reported in [48], after 100 a from the discharge, the radiotoxicity of the fission products derived from 1 ton of enriched U fuel would be of about 1.4×10^7 Sv. However, due to the relatively rapid decay of some of the most significant fission products, such as ^{90}Sr and ^{137}Cs ($T_{½}$ about 30 a), the radiotoxicity after 1000 a is expected to be only ~870 Sv.

As a whole, the transmutation approach is not considered the best option to lessen the negative effects of fission products and the low interest in reducing the radioactivity of the fission products has led the scientific community to no longer investigate their possible transmutations. Instead, more efforts have been focused on the development of technologies able to assure an efficient and safe containment of waste in dry casks that could withstand strong radiations for at least 100 a.

5. Major Issues, New Opportunities

Whereas the transmutation of fission products is of very little interest, emerging reasons are pushing toward the settling of even more efficient procedures for their extraction from nuclear waste. This is due to the fact that the mitigation of hazards is not the only reason that drives experimenting with new methodologies for the selective extraction of components from used fuels. The value of some radioisotopes that are generated in the reactors strongly impacts programs and strategies for waste management and is making successful the "separate-extract-reuse" concept. The great demand for some radioisotopes that find huge applications in medical and industrial fields makes their recovery from used fuel attractive.

For industrial applications, separation methods enable the extraction of high-purity individual isotopes, such as the ^{241}Am widely employed in nuclear batteries [58]. From the spent fuel some other radioisotopes qualified to power nuclear batteries can be recovered, such as the fission products ^{85}Kr, ^{90}Sr, ^{144}Ce, and the actinides ^{277}Ac and ^{244}Cm [32]. Very recently, as high-power source for nuclear batteries has been proposed also the ^{232}U, generated by nuclear reaction chains in fuels containing ^{234}U, ^{235}U, ^{238}U, and traces of ^{232}Th [59,60].

Of particular economic relevance is the recovery of some isotopes of the Platinum Group, as the rather rare and high-priced Ru, Rh, and Pd metals are used as catalysts in diverse industrial areas, including petrochemical plants. In this view, emphasis has been put on the recovery of the Pd components from nuclear waste by a synergistic binding strategy [61]. A non-contact photoreduction method has proved itself able to selectively recover Pd (99.8%), Rh (99.9%), and Ru (90.0%) from high-level radioactive liquid waste [62]. A scheme of the chemical/physical multi-step process designed to achieve the efficient separation of each metal is shown in Figure 5.

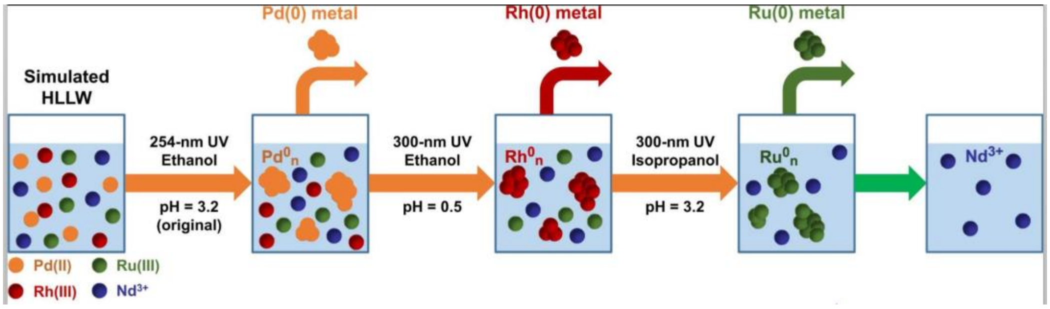

Figure 5. A scheme of the photo reduction multi-step process for recovery of Pd, Rh, and Ru from a simulated high-level liquid waste [Reproduced with permission from Weng et al., Ref. [62], copyright Elsevier, 2023].

A sequence of chemical separation from the liquid waste, even/odd isotope separation, and nuclear transmutation allows the collection of ^{105}Pd and ^{107}Pd for commercial uses [13,63]. The efficient chemical/physical separation processes recently settled for Pd are expected to additionally enable the recovery of selected Ru and Rh isotopes, thereby overcoming the drawback related to the handling and reuse of materials with radioactivity exceeding the limits permitted for industrial applications [15].

The recovery of radionuclides for medical therapies and diagnostics deserves a special mention. The mass production of such isotopes is indeed a global problem that the extraction from spent fuel may help to mitigate. The more commonly employed isotopes, namely, ^{125}I for prostate cancer therapy; ^{90}Sr for cardiovascular therapy; ^{99}Tc for diagnostic imaging and radiopharmacy; as well as the ^{188}W/^{188}Re, ^{133}Xe, and ^{32}P radiosources, are all present in the radioactive waste of nuclear reactors from which they can be obtained [64].

The implementation of waste partitioning procedures is of great importance, other than in preserving the earth's assets by reducing the mining of critical elements, in the case of elements that do not exist in nature and need to be produced by expensive technologies. The most interesting example is given by the above-cited ^{99}Tc ($T_{½}$ = 2.11 × 10^5 a), an isotope that represents over 90% of the activity of the long-lived fission products and is a very significant component of nuclear waste from about 10^4 to 10^6 years after its production.

In this view, the storage and disposal of ^{99}Tc-containing waste need to sequester/immobilize it in materials able to address the challenges posed by such a hazardous radionuclide [65]. The ^{99}Tc comes from the fission of ^{236}U, in turn, produced from ^{235}U by neutron capture in research reactors that use highly enriched U targets. The precursor of ^{99}Tc is the neutron-rich light fission fragment ^{99}Mo that decays by β^--emission ($T_{½}$ = 66 h) to ^{99}Tc through the metastable nuclear isomer $^{99\text{m}}$Tc [66]. This precious isotope, the most used one in nuclear medicine all over the world, decays to ^{99}Tc emitting γ-rays of 140 keV with a half-life of 6 h. Due to its peculiar chemical/physical/biological characteristics, $^{99\text{m}}$Tc plays a fundamental role in radiopharmacy and is widely applied as a tracer and imaging agent in diagnostic procedures, such as SPECT (Single Photon Emission Computed Tomography) [67].

A quick retrieval of the fission product ^{99}Mo from the spent fuel could ensure a twofold benefit. The first one is to prevent the ^{99}Tc generation and the second is to obtain $^{99\text{m}}$Tc by a path alternative to the fission processes carried out in some dedicated reactors [68,69] or in cyclotrons [70].

A further medical radioisotope that can be mined from nuclear waste is the α-emitter ^{225}Ac ($T_{½}$ = 10 d), which is being proposed for radio-immunotherapy [71]. This isotope is produced using various techniques, including proton cyclotrons and linear accelerators. However, for now, the largest source of high-purity ^{225}Ac comes from ^{229}Th ($T_{½}$ = 7.9 × 10^3 a) recovered from the ^{233}U used in Th cycles [72,73].

Presently, none of the 27 molecules labeled with ^{225}Ac has yet been approved by the U.S. Food and Drug Administration but it is expected that Ac-based drugs might reach the market by 2028 [74]. In this case, the rising demand for ^{225}Ac will require a significant increase in the production capacity and, therefore, the development of multiple production routes, including infrastructures for ^{225}Ac extraction from nuclear waste. Moreover, this recovery will meet the US strategy to eliminate the ^{233}U that originally was intended for use in nuclear reactors but presently represents a security concern due to the high radioactivity of this isotope, which is also a potential weapon material.

In Russia, as part of the non-proliferation policies and strategies for nuclear weapons recycling, the ^{225}Ac that is being proposed for the therapy of brain tumors, bladder cancer, neuroendocrine tumors, and leukemia, is routinely obtained by the separation of ^{229}Th from the ^{233}U originally produced for weapons applications [64]. Additionally, Degueldre et al. [75] very recently analyzed the feasibility of radiopharmaceutical applications for a series of short-life fission products extracted from molten salt reactors.

The development of reprocessing approaches also shows a strong potential for the pre-conditioning of used fuels that are not meant to be recycled and need, therefore, to be stored in a repository. Aiming to decrease the repository sizes, the objective is to obtain a nuclear waste containing residuals of long-lived radionuclides as low as possible, reducing in such a way not only radiotoxicity but also the heat produced by nuclear decays.

6. Technological Limits and Developments

The multiple objectives that the technology of waste management aims to achieve include sustainability, reliability, safety, cost advantages, and prevention of nuclear weapons proliferation. As has been illustrated in the previous sections, the current P&T strategies offer interesting options complementary to the long-term storage of highly radioactive nuclides. The partial elimination of some hazardous components and their threatening effects has the potential to somewhat reduce the waste volume and simplify the requirements for disposal facilities. An example is represented by Pu. The extraction by PUREX methodologies from the spent fuel (and its reuse in power plants) helps to overcome the concerns about the risk of possible stealing from the storage facility for misuse in nuclear weapons.

Speaking about non-proliferation objectives, it is of note that the strategies settled on to process spent fuel offer the exciting opportunity to convert the warhead-grade deadly U and Pu into fuel for nuclear power plants. In the framework of the 20-year program "From Megatons to Megawatts" initiated in 1993, the highly enriched U extracted from 20,008 nuclear warheads was converted in Russia to nearly 15,000 tonnes of LEU (low-enriched Uranium) used in US civil power reactors [76].

However, none of the presently used approaches can be considered a solution to the big problem of used fuel disposal. To fully achieve all the settled objectives, it will be indeed necessary to rely on approaches that, initially designed to assure an ultra-efficient use of the U and Pu resources, are now also considered a way to lessen the risks associated with the long-term storage of nuclear waste. This means the implementation of installations able to consume recycled fuel to run, to significantly decrease the inventory of both minor actinides and long-lived fission products, and, in short, to switch from the concept of the SNR to that of FNR, or of the innovative ADS [6,77].

Fast neutron reactors have special capabilities and offer many important advantages with respect to water-cooled reactors that, even adopting the more advanced fuel treatments, do not allow the extraction of much more than ~1% of the nuclear value stored in the waste. Compared with today's traditional SNRs, a FNR can utilize the ^{238}U about 60 times more efficiently, converting such an otherwise useless isotope into the fissile ^{239}Pu. [78]. Moreover, the capabilities of FNRs enable them to run consuming fuel that has been recycled many times. Even if some nuclides, such as ^{239}Pu and the long-lived fission products ^{99}Tc and ^{129}I, could also be transmuted in principle by the thermal neutrons produced in LNRs, under the physical conditions of such reactors, to complete the process would take several decades.

The advantage of destroying actinides using fast neutron reactors can be appreciated if one compares the probability of their fission in a fast neutron energy spectrum with that in a thermal neutron spectrum. The ratios of the fission probabilities are 1.2 for ^{243}Cm, 23 for ^{243}Am, 53 for ^{242}Pu, and 55 for ^{240}Pu [10]. In this context, a smart strategy carried out by some countries is the extraction of highly active and long-lived isotopes of Pu and of minor actinides from the waste and their burning in one of the six presently working fast neutron reactors, where such radionuclides can be successfully destroyed by neutron-induced fission reactions. Such a P&T approach, accompanied by the removal from the waste of the longer-lived fission products, has proven to be a valid one in terms of minimizing the amount of hazardous waste [10].

The transmutation processes will be instead an integral part of the closed fuel cycles in specifically designed fast neutron reactors or in the accelerator-driven systems (ADSs), where fluxes of high-energy neutrons are produced by spallation [47,48,77–79].

With respect to the currently adopted P&T approaches, a much more winning strategy will be that of the closed fuel cycle in Generation IV reactors. In such integrated fuel reprocessing, an enhanced utilization of the resources occurs and continuous power is generated inside such innovative reactors by the cyclic reuse of Pu and of all the actinides.

A different consideration should be made for transmutation by an ADS, where neutron beams produced by the accelerator hit an assembly formed by the waste and by a small amount of fissionable fuel. Here, the capture of neutrons by the isotopes contained in the waste provokes their fission and produces energy. It is noteworthy that, due to the peculiar design of these installations, the ADSs could also be used to generate nuclear power from non-fissile nuclides, such as ^{232}Th [47]. A further potentiality envisaged for ADSs is the feasibility of disposing of weapons-grade Plutonium more efficiently than burning it as MOX in conventional reactors [80].

To define priorities in planning future transmutation installations, important R&D programs have been launched in some of the countries, such as France, India, Japan, China, Russia, and the UK, that carry out nuclear fuel reprocessing [13,57]. In this context, the Riken Nishina Center (Japan) has designed a new type of high-power accelerator for industrial-level transmutations. This accelerator, expected to deliver a 1A deuteron beam, will utilize the high-energy neutrons produced by the reaction of deuteron break-up at a liquid Li target [62]. The goal that all these innovative installations aim to achieve is to have waste that becomes radiologically harmless in only a few hundred years, making possible the use of less demanding waste repositories. It is interesting to cite the recently emerging tendency to consider reactor, fuel cycle, and waste as parts of the same integrated system [81]. Within this approach, the design of a new reactor should no longer be separated from the prediction of the type and amount of nuclear waste produced by that reactor and from the development of strategies for spent fuel disposal [35].

In the frame of disrupting technological innovations, an important place is occupied by the Small Modular Reactors (SMRs) [82], largely proposed for their inherent safety and the reduction in the various burdens associated with the traditional large nuclear plants, in primis the issue of waste management [83].

However, studies addressed to compare the spent fuel produced by a 1100-MW$_e$ PLWR with that produced by some types of SMRs, namely, the water, molten/salt, and sodium-cooled ones, demonstrated that these last designs could generate a larger volume of highly radioactive waste [84]. These effects, ascribed to processes of neutron leakage that result enhanced in small reactor cores, make it clear that the challenge is to develop spent fuel management concepts and disposal technologies specific to each reactor design [85].

Figure 6, taken from a paper by Poinssot et al. [86], allows one to take a look at the evolution of the nuclear systems, which include reactors, fuel cycles, and spent fuel treatments, from the 1960s to the distant future.

As outlined in Ref. [86], the projections indicated in the scheme are not intended to predict future developments but rather to indicate the directions along which to move for improved sustainability of the nuclear systems in terms of natural resources consumption, the efficiency of fuel cycles, the safety of waste repositories, reactor security, and nuclear proliferation contrast.

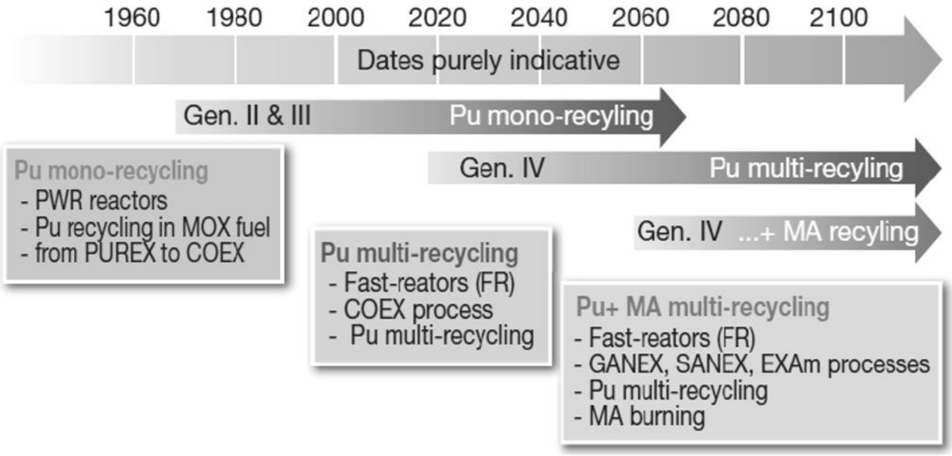

Figure 6. Evolution of nuclear systems and indications of strategies for improved sustainability of nuclear energy. [Reproduced from Ref. [86] (open access)].

7. Some Final Thoughts

Whereas the urgent need to develop a global energy policy is nowadays exploiting the potential of fission reactors, there are some key issues, such as safety, cost competitiveness, waste management, and proliferation resistance, that are still causing concern. It is noteworthy that such issues are mostly entangled in each one. This is the case for the high costs, due mainly to the need to satisfy stringent safety demands during the reactor runs but also when the fuel is disposed of during the refueling or decommissioning phases.

On the other hand, the issue of resistance against terrorist crimes or nuclear weapons proliferation is closely linked to the right management of nuclear waste. In particular, the implementation of effective partitioning and transmutation methodologies and a judicious choice of fuel cycles can help to reduce the storage volume of the high-level radioactive wastes, preventing, at the same time, the formation of inventories of weapon-grade fissile materials. Waiting for the foreseen transition from low-neutron to fast-neutron reactor technology, the problem of waste disposal is a legacy issue with which existing and future nuclear technologies must contend. The analysis of successes and failures in managing the spent fuel inventories shows where and how progress has been made, as well as what more is needed.

In this context, the presently tested, as well as the envisaged cutting-edge fuel cycles could be a key enabling an expansion of fission-based nuclear power. However, the roadmap for their efficient utilization still needs a lot of research because such advanced cycles require materials and procedures that can be very different from those used in traditional fuel cycles.

It should be considered that a change of perspective is now leading researchers to look at the spent fuel as a possible valuable resource rather than as a mere trouble and, therefore, to consider waste reprocessing a route complementary to its storage. The economic benefits provided by the recovery and reuse of some nuclides are now attracting the attention of governments and also of private investors, who are planning to invest in advanced waste management programs.

It is hoped that the new initiatives and the achievements obtained in reducing the hazard of nuclear waste could contribute to the creation of a favorable image of nuclear power, an essential, indispensable player in the dynamic scenario of global energy policy.

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Abbreviations

ADS	Accelerator Driven System
AGR	Advanced Gas-cooled Reactor
COEX	Co-Extraction of U and Pu
DOE-US	Department of Energy (US)
EXAm	Extraction of Am
FNR	Fast Neutron Reactor
GANEX	Grouped Actinides Extraction
GCR	Gas-cooled reactors
HTGR	High-temperature gas-cooled reactors
HWR	Heavy Water Reactor
IAEA	International Atomic Energy Agency
LEU	Low Enriched Uranium
LWR	Light Water Reactor
MA	Minor Actinides
MOX	Mixed Oxides
NEA	Nuclear Energy Agency
NRC	Nuclear Regulatory Commission
OECD	Organization for Economic Co-operation and Development
P&C	Partitioning-Conditioning
P&T	Partitioning-Transmutation
PHWR	Pressurized Heavy Water Reactor
PLWR	Pressurized Light Water Reactor
PRIS	Power Reactor Information System (IAEA)
PUREX	Plutonium Uranium Reduction Extraction
SANEX	Selective Actinides Extraction
SMR	Small Modular Reactor
SNR	Slow Neutron Reactor
SPECT	Single Photon Emission Computed Tomography
^{e}U	Enriched Uranium
UREX	Uranium Reduction Extraction
W_e	Electrical power
WNA	World Nuclear Association

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