Review

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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 way to deal with the sensitive issue of spent fuel reprocessing. The review includes 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. Also outlined are the strategies presently adopted to minimize the harmfulness of the nuclear waste to be disposed and discussed advanced options and possible solutions proposed for the recycling of the used fuel and for the reusing of some technologicaly relevant radioisotopes.

**Keywords:** nuclear waste management; spent fuel processing; nuclear installations; fission products; actinides; transmutation chains; recovery of radioisotopes

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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 is now seen as a promising way to decrease greenhouse gas emissions and effectively address the issue of climate changes [1].

The more recent WNA report indicates that a total of 440 nuclear power reactors (second and third generation) are presently operating in 32 countries , providing 16% of the world's electricity [2]. Due to the conjunction of the increased power demand expected in the near future and of the necessary energy transition, 61 nuclear plants are under construction in 16 countries , 110 are planned and 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 characteristic 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 decommisioned [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 economical 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 reactors core but also the radioactive materials coming from medical or industrial applications and from research facilities [6] . It is noteworthy that, even if IAEA offers legal instruments to address the issue of spent fuel and radioactive waste management on global scale [7], there are differences in the way each country tackles the topic of waste categorization and of subsequent waste disposal .

The indications of US-NRC, followed by several countries, establish the processing and handling of the nuclear waste according to three level of radioactivity , namely high, intermediate and low [8].

The high-level radioactive waste is made of “spent fuel”, mainly coming from nuclear power reactors, but includes also fuel cladding .This waste represents only 3% (vol) of the total, but accounts broadly for 95% of the radioactive hazard [9] 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 [10]. Such temporary storage solutions, designed to safely store the waste for a limited period, require the presence of a 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 opinion as an extremely difficult problem that requires high financial investments to solve it [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 [8]. The radioactive content of all these materials, that contain low concentrations of radionuclides with long half-lives, is typically about 1% .The low-level waste, that represents about the 90 % (in volume) of the radioactive materials to be disposed of, is typically stored in near-surface appropriate facilities until it turns 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 (LWR), characterized by a hexagonal assembly .

The timing of fuel replacement depends on the position of the assemblies in the reactor . In the 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 fabrication of new fuel . [9].

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, ìndependently of the kind of reactor, the fuel assemblies extracted from the reactor vessel still contains large amounts of U, Pu, fission products, and minor actinides [11].

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 $^{235}$U [12]. Moreover, the decay of the primary radioisotopes produced by the fission process give rise to a variety of other radionuclides [9,13].

It is therefore evident that a huge amount of radionuclides are still present in spent fuel, that is to be considered a precious resource rather than a hazardous waste to be disposed of [14]. Following the more recent industrial-scale technologies, almost 96% of the spent fuel extracted from the reactors can be recycled. Reprocessing of the used nuclear materials enables to produce new fuels and offers the additional advantage to reduce the volume of waste to be stored, at the same time avoiding 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 fresh fuel of nuclear power plants [8]. 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 [9].

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

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

| SPENT FUEL & MATERIALS | DISPOSAL |
|---|---|
| Fuel cladding, filters … | Storage as low/intermediate level waste |
| Reprocessed U and Pu | Reuse in thermal or fast reactors |
| Long-lived minor actinides | Partitioning & 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 analyse the current status of the strategies adopted to minimize both volume and threat of the nuclear waste to be disposed, whilst highlighting the potential options and the most promising perspectives for recycling and reusing of spent fuel.

## 2. A Look at the Fission Technology

The majority of the worlwide 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" (SNR) where fission is induced by neutrons of energy 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 [15]. 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 in two main classes, Light Water Reactors (LWR) and Heavy Water Reactors (HWR), depending on the kind of water ( $H_2O$ or $D_2O$) used as neutron moderator. A third class of SNR is represented by the graphite-moderated reactors that in the past operated in France, Japan, Italy and that, in their last configuration- Advanced Gas-cooled Reactors (AGR) - are still operating in UK.

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

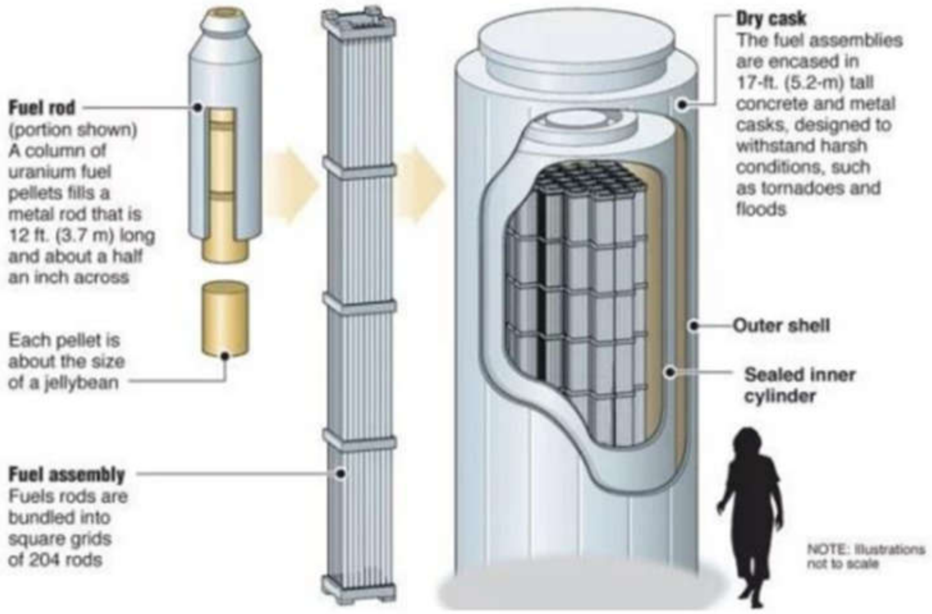

**Figure 1.** Left: Schematic view of fuel rods and of their assembles in square grids; Right: Scheme of the dry cask used for the storage of spent fuel [Source : US-NRC].

To efficiently operate, the different 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, that contains about 0.72% of the fissile $^{235}$U isotope. The AGRs, that employ graphite for neutron moderation, have been designed to operate with a 3.5% enriched U.

Each fission event produces on average two and 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 mantain 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, in other words to have $k \geq 1$ , where $k$ is the neutron multiplication factor. If this condition is no more 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.

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 :

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

However, the yield in $^{239}$Pu can be strongly increased by an improved neutron economy in the Fast Neutron Reactors (FNR), a much less common class of reactors where fission of Uranium isotopes is induced by “fast” neutrons (energies > 1MeV). This peculiarity opens reaction paths rather different from those occurring in slow neutron reactors, and enables to utilize U-based fuels about 60 times more efficiently than a common reactor [16]. Some of the ~ 20 FNR that have been working since the 1950s are the so-called “fast breeder reactors”, specifically designed to generate more fissile nuclear fuel that they consume [17].

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

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

This last reaction could efficiently take place also 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 a ADS reactor is a clean and inherently safe fission process, because an ADS can only run when neutrons are supplied to it [18,19].

An in-depth analysis of the current state-of-art and of international ongoing developments regarding fast reactors can be found in [20]. 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 reactor 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 around ~ 0.41$Z$, and the heavy-one around ~ 0.59Z. 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⁻-particle, giving rise to a wide range of nuclides. An example is shown in Figure 2, that illustrates the case of a fission producing $^{94}$Sr (Z=38) and $^{140}$Xe (Z=54) nuclides, and shows the decay chains starting from such fission fragments.

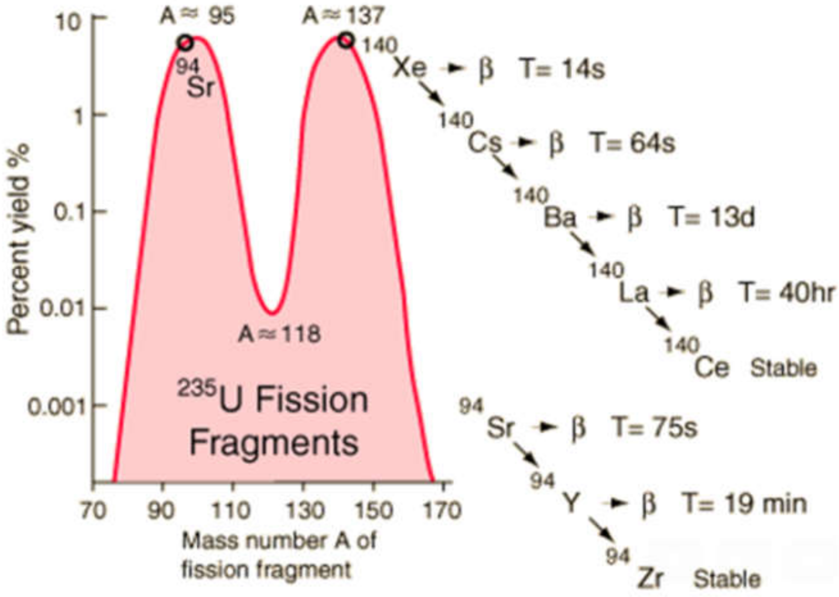

**Figure 2.** A scheme of $^{235}$U fission producing the $^{94}$Sr and $^{140}$Xe fission fragments [Reproduced by courtesy of Rod Nave, Georgia State University ].

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 [21]. 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 [22], 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 ($^{e}$UO$_2$) enriched with 3.54 % of $^{235}$U. This material contains 23.16 tonnes of $^{238}$U plus 0.84 tons of $^{235}$U, totalling 24.0 tonnes of $^{e}$U [23].After one year of reactor operation the consumed amount of the pristine $^{e}$U is 1,649 kg, of which 1,018 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 other 1176.6 kg of fission products (~ 50 × 10$^{5}$ TBq). 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 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) [23].

Radionuclide Half-life [a] Mass [kg] Activity [TBq]

$^{238}$U 4.46 × $10^{9}$ 22,1410.28
$^{235}$U 7.04 × $10^{8}$ 209.0 1.67 × $10^{-2}$

Total ------ 22,350 0.30___

$^{236}$U2.34 × $10^{7}$ 137.5 0.33
$^{234}$U 2.45 × $10^{5}$6.21.43
$^{238}$Pu 87.75.6 3.55 × $10^{3}$
$^{239}$Pu 2.41 × $10^{4}$177.2 4.07 × $10^{2}$
$^{240}$Pu 6.56 × $10^{3}$ 69.15.81 × $10^{2}$
$^{241}$Pu 14.337.21.43 × $10^{5}$
$^{242}$Pu 3.75 × $10^{5}$15.3 2.23
$^{237}$Np 2.14 × $10^{6}$13.5 0.352
$^{241}$Am 4.33× $10^{2}$ 6.80.86 × $10^{3}$
others ------- 3.9------

Total -------472.31.48 × $10^{5}$

$^{89}$Sr0.138 0.1 1.08 × $10^{5}$
$^{90}$Sr28.9 18.0 0.91 × $10^{5}$
$^{106}$Ru 1.02 6.7 0.82 × $10^{6}$
$^{134}$Cs 2.05 35.11.69 × $10^{6}$
$^{137}$Cs 30.0 42.91.38 × $10^{5}$
$^{144}$Ce 0.78 13.81.62 × $10^{6}$
$^{147}$Pm 2.62 12.04.12 × $10^{5}$
$^{154}$Eu 8.59 1.2 1.20 × $10^{4}$
Others-------136.30.85 × $10^{5}$
Stables-------910.5 -------

Total ------- 1176.649.8 × $10^{5}$

**Grand Total ------- 23,9989 51.3 × $10^{5}$**

The 22.350 tonnes of unburned fuel contain 0.209 tonne of $^{235}$U, therefore such 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 , $^{241}$Pu isotope is the most active, $^{236}$U the least one. 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 above).

## 3. Used Fuel Processing

The methodologies used in processing spent fuel strongly depend on the reactor where the recycled material is going to be re-used. Moreover, different methodologies must be applied if the re-use 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 to reduce the amount of high-level radioactive waste.

The reprocessing of spent fuel for using as fresh fuel in reactors is carried out by means of two main methodologies, namely the aqueous and the non-aqueous ones. These last involves a first pyroprocessing step that employ molten salts to dissolve the fuel rods and subsequent electroplating processes aimed at separating the various elements. This separation method has been proposed some decades ago [24], but pyroprocessing technologies are still at the prototype stage and are not yet commercialized [25].

Today the more commonly used method is the 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 in 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 [26]. 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 $\alpha$-decay of $^{236}$Pu, generates daughter nuclides which emit strong $\gamma$-rays. Whereas this makes rather difficult the handling of the reprocessed material, such 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, as the fertile $^{234}$U and the short half-life $\beta^-$-emitter $^{237}$U, do not affect at all the performances of reprocessed fuel.

The current approach for the re-use 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 devise several other separation methodologies , all operating by a solvation mechanism [27] . 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 also to efficiently recover some fission products, such $^{129}$I (by 95%) and $^{99}$Tc (by 90%), as well as smaller amounts of $^{135}$Cs and $^{90}$Sr [28].

Whereas for the use in today's LWRs the reprocessed U needs to be enriched, the Pu obtained from the PUREX process can be directly used for 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 re-fueling, 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 [29]. The threats that the building up of Pu stockpiles would pose is a concern that led 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 $\alpha$-decay mode leading to $^{236}$U, the $^{240}$Pu isotope undergoes also spontaneous fission, releasing a neutron flux that makes difficult to reach the conditions for an implosion [30]. Small amounts of recovered Pu are instead retained in secure storage facilities to produce, by $\beta^-$-decay of $^{241}$Pu ($T_{½}$ = 14 yr) , the isotope $^{241}$Am, a very suitable source for the powering of nuclear batteries [31].

The waste remaining after the U and Pu removal by 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 of minor actinides.

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

Among the minor actinides, some very hazardous isotopes, 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 to recover U and Pu isotopes from spent fuel and to manufacture 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 [32]. However this complex and expensive procedure does not solve the problem of a nuclear waste that remains highly radioactive for thousands of years due to the presence of minor actinides and some fission products [33]. The contributions of the actinides and fission products to the total residual radioactivity of a spent fuel produced by the burn-up of 1tonne U is shown in Figure 3.

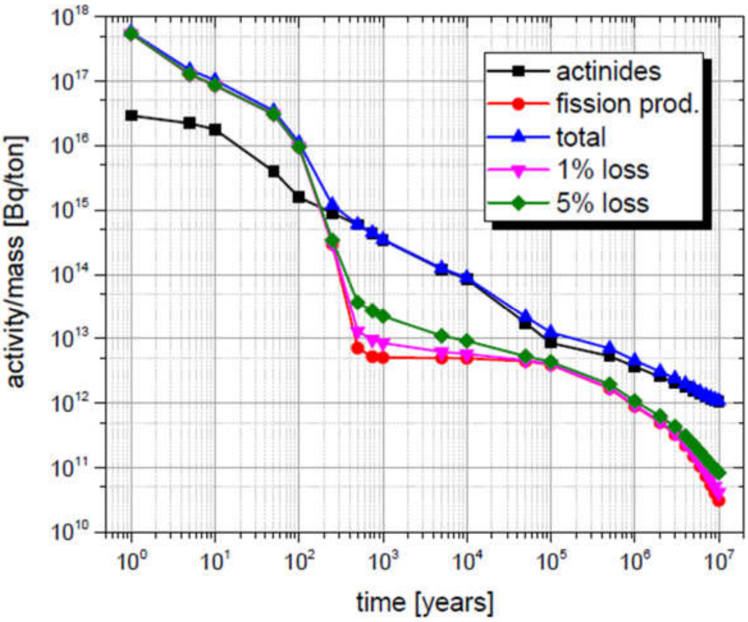

**Figure 3.** Trend of total radioactivity *vs* time of the spent fuel produced by the burn-up of 1 tonne U (blu points), with indication of the contribution of actinides (black points) and of fission products (red points). The effect of actinides losses during reprocessing is also indicated:1% loss (pink points), 5% loss (green points) [Reproduced from Ref.34 ].

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 to radioactive waste [35]. In this view a series of chemical procedures have been established with the objective to separate 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 80's to reduce the inventory of radioactive wastes to be stored [36]. 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 [37–39].

The strategies for the management of used fuels pursue two main goals, short- and long-term ones. The short time objective is to reduce the radiotoxicity of the fuel just removed from the reactor, radiotoxicity 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, that are not only in solid form but also in gas and liquid phases, represent indeed a further risk associated with the treated spent fuels [40]. 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 [41].

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 [42,43].

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 hazard due to accumulation of the more significant isotopes.

As regards 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 [44]. In the initial period of the storage the most hazardous among the minor actinides is the $\alpha$-emitting $^{244}$Cm ($T_{½}$ = 18.11 a), that decays to $^{240}$Pu. The $^{244}$Cm isotope is characterized by a large cross section for thermal neutrons capture, and therefore can be easily transmuted in $^{245}$Cm. While a fraction of the $^{245}$Cm isotopes undergoes fission, small amounts of the higher isotopes $^{246-248}$Cm are produced by successive capture of neutrons. Further β-decay processes create also some isotopes of Bk, Cf, Es and Fm .

Conversely, in the time range 100–100,000 a after being removed from the reactor, the radioactivity of the fuel is determined by the Pu and Am isotopes [45] . 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 comes also 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 ($T_{½} = 2.144 \times 10^6$ a) an $\alpha$-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 ($T_{½} = 2.0 \times 10^{19}$ a).

All these actinides, not present in the unirradiated nuclear fuel, strongly demand the use of transmutation approaches to transform highly radioactive nuclides in less hazardous ones [46]. A transmutation chain of actinides typically produced from a low-enriched Uranium fuel is schematically shown in Figure 4 .

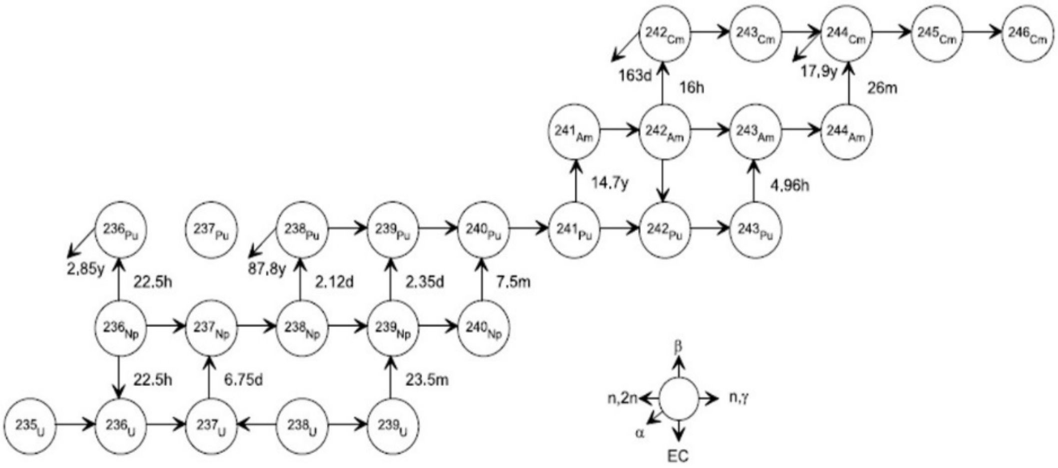

**Figure 4.** Scheme of Actinide transmutation chains [Reproduced with permission from Ref. 47]. .

An efficient transmutation of actinides critically depends on the efficiency of the initial partitioning step. High-performance procedures for 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 [47]. The lanthanides have indeed a high cross section for neutron absorption, therefore it is necessary to eliminate them in order to create conditions favourable for the transmutation of actinides [26]. However, due to the quite similar chemical behaviour of actinides and lanthanides, an effective separation and a complete removal of lanthanides is a very difficult task [48,49].

A further observation is that the actinides transmutation of itself may have a strong impact on both fuel handling and repository performance. An example is the unavoidable generation of $^{252}$Cf, that undergoes spontaneous fission releasing an extremely high number of neutrons (~10$^{12}$ n/g.s). 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 of the number of cycles, and the extension of the cooling steps between a successive fuel loading [50,51].

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 [32]. 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 [37]. In any case, just after the reactor shutdown, a complex interplay between 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 [52]. 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 [53].

It is to be noted that, whereas a huge amount of data is presently available for transmutation of Np, Am and Cm, only a small number of studies have been performed on the transmutation of the fission products. A first technical reason of this is that transmutation of fission products requires larger amounts of neutrons with respect to that of actinides [54,55]. An example is given by the transmutation of $^{99}$Tc to the stable $^{100}$Ru, after neutron capture and decay of $^{100}$Tc ($\beta^-$-decay, $T_{½}$ = 16 s ), or of $^{129}$I to the stable $^{130}$Xe, after neutron capture and decay of $^{130}$I ($\beta^-$-decay, $T_{½}$ = 12.4 h). The (n,$\gamma$ ) 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 type of installations [55,56].

A second point is that studies about the radiotoxicity of the highly active fission products have evidenced how the long-term threat due to such nuclides is lesser than that of the actinides [46]. As reported in [47], after 100 a from the discharge the radiotoxicity of the fission products derived from 1 ton of enriched U fuel would be of about $1.4 \times 10^7$ 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 more investigate their possible transmutations. Instead, more efforts have been focused on developments 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. Big Questions, New Opportunities

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

For industrial applications, separation methods enable to extract high-purity individual isotopes, as the $^{241}$Am widely employed in nuclear batteries [57]. From the spent fuel can be recovered also some other radioisotopes qualified to power nuclear batteries, such as the fission products $^{85}$Kr, $^{90}$Sr, $^{144}$Ce and the actinides $^{277}$Ac and $^{244}$Cm [31]. 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 [58,59].

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 used as catalysts in diverse industrial areas, including the petrochemical plants. In this view emphasis has been put on the recovery of the Pd components from nuclear waste by synergistic binding strategy [60]. A sequence of chemical separation from the liquid waste, even/odd isotope separation and nuclear transmutation allows to obtain $^{105}$Pd and $^{107}$Pd for commercial uses [12,61]. The efficient chemical/physical separation processes recently settled for Pd are expected to enable also the recovery of selected Ru and Rh isotopes, thereby overcoming the drawback related to handling and re-use of materials with a radioactivity exceeding the limits permitted for industrial applications [14].

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 [62].

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 ($T_{½} = 2.11\times 10^5$ 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 [63]. 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 $\beta^-$-emission ($T_{½}$ = 66 h) to $^{99}$Tc through the metastable nuclear isomer $^{99m}$Tc [64]. This precious isotope, the most used one in nuclear medicine all over the world, decays to $^{99}$Tc emitting $\gamma$-rays of 140 keV with an half-life of 6 h. Due to its peculiar chemical/physical/biological characteristics, $^{99m}$Tc plays a fundamental role in radiopharmacy and is widely applied as tracer and imaging agent in diagnostic procedures, as the SPECT (Single Photon Emission Computed Tomography) [65].

A quick retrieval of the fission product $^{99}$Mo from the spent fuel could ensure a two-fold benefit. The first one to prevent the $^{99}$Tc generation, the second to obtain $^{99m}$Tc by a path alternative to the fission processes carried out in some dedicated reactors [66,67] or in cyclotrons [68].

A further medical radioisotope that can be mined from nuclear waste is the $\alpha$-emitter $^{225}$Ac ($T_{½}$ = 10 d), which is being proposed for radio-immunotherapy [69]. 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 ($T_{½} = 7.9\times 10^3$ a) recovered from the $^{233}$U used in Th cycles [70,71].

Presently no one 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 [72]. In this case, the rising demand for $^{225}$Ac will require a significant increase of 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 that 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 [62]. Additionally, Degueldre et al. [73] very recently analysed the feasibility of radiopharmaceutical applications for a series of short-life fission products extracted from molten salt reactors.

The development of reprocessing approaches shows a strong potential also 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 residual 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 a long-term storage of highly radioactive nuclides. The partial elimination of some hazardous components and of their threatening effects has the potential to somewhat reduce the waste volume and to simplify the requirements for disposal facilities. An example is represented by Pu. The extraction by PUREX methodologies from the spent fuel (and its re-use in power plants) helps to overcome the concerns about the risk of a possible stealing from the storage facility for misuse in nuclear weapons.

Speaking about non-proliferation objectives, it is to note that the strategies settled 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 [74].

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 considered also a way to lessen the risks associated to 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 SNR to that of FNR or of the innovative ADS [6,75].

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 to extract much more of ~1% of the nuclear value stored in the waste. Compared with a today’s traditional SNR , a FNR can utilise the $^{238}$U about 60 times more efficiently, converting such otherwise useless isotope into the fissile $^{239}$Pu. [76]. 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 be transmuted in principle also by the thermal neutrons produced in LNRs, under the physical conditions of such reactors to complete the process would take several decades.

The advantage to destroy 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 [9]. 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 reactor where such radionuclides can be successfully destroyed by neutron-induced fission reactions. Such P&T approach, accompanied by the removal from the waste of the longer-lived fission products, has proven to be a valid one in view of minimizing the amount of hazardous waste [9].

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 (ADS), where fluxes of high-energy neutrons are produced by spallation [46,47,75–77].

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 a 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 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 notewhorty that, due the peculiar design of these installations, the ADSs could be used to generate nuclear power also from non-fissile nuclides, as $^{232}$Th [46]. A further potentiality envisaged for ADSs is the feasibility to dispose of weapons-grade Plutonium more efficiently than to burn it as MOX fuel in conventional reactors [78].

To define priorities in planning future trasmutation installations, important R&D programs have been launched in some of the countries, as France, India, Japan, China, Russia, UK, that carry out nuclear fuel reprocessing [12,56]. 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 [61]. The goal that all these innovative installations aim to achieve is to have wastes which become radiologically harmless in only a few hundred years, making it 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 a the same integrated system [79]. Within this approach the design of a new reactor should be no more separated from the prediction of type and amount of nuclear waste produced by that reactor and from the development of strategies for spent fuel disposal [34].

In the frame of disrupting technological innovations an important place is occupied by the Small Modular Reactors (SMR) [80], largely proposed for their inherent safety and the reduction of the various burdens associated to the traditional large nuclear plants, *in primis* the issue of waste management [81].

However, studies addressed to compare the spent fuel produced by a 1,100-MW$_e$ with that produced by some types of SMR, namely the water-, molten salt- and sodium-cooled ones, demonstrated that these last designs could generate a larger volume of highly radioactive waste [82]. 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 for each reactor design [83].

## 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, as safety, cost competitiveness, waste management and proliferation resistance, that are still causing concern. It is noteworthy that such issues are mostly entangled each one. This is the case of the high costs, due mainly to the need to satisfy stringent safety demands during the reactor runs but also when the fuel is disposed at the refueling or decommissioning phases.

On the other hand, the issue of resistance against terrorist crimes or nuclear weapons proliferation is closely linked to a right management of nuclear waste. In particular the implementation of effective partitioning and transmutation methodologies and an 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 show where and how progresses have being 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 the fission-based nuclear power. However the road map for a 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 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 re-use of some nuclides are attracting nowadays the attention, besides of governments, also of private investors, who are planning to invest in advanced waste management programmes.

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 the nuclear power, an essential, indispensable player in the dynamic scenario of global energy policy.

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## List of Acronyms

ADS - Accelerator Driven System

AGR -Advanced Gas-cooled Reactor

DOE-US -Department of Energy (US)

FNR – Fast Neutron Reactor

HWR – Heavy Water Reactor

IAEA-International Atomic Energy Agency

LEU- Low Enriched Uranium

LWR- Light Water Reactor

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

PUREX-Plutonium Uranium Reduction 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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