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Transmutation: reducing the storage time of spent fuel

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Abstract

Transmutation can reduce the storage time of spent fuel. Efficient transmutation requires a high flux of neutrons and can therefore be done only in nuclear reactors. The article shows the concepts of different solutions of transmutation in nuclear reactors. Knowledge of transmutation is supplemented by information on spent fuel and its radiotoxicity.

Keywords: Transmutation, Nuclear waste, Accelerator drives systems, Fast reactors, Radiotoxicity, Nuclear spent fuel

1. Introduction

One of the most common arguments used by the opponents of nuclear power is the issue of nuclear waste, and in particular the storage of spent nuclear fuel and the potential risk to the public. The question of how nuclear waste from existing nuclear reactors affects our generation and the next generation is also one of the first questions asked by people from outside the nuclear community. Even non-nuclear technical and engineering professionals raise concerns about the safety of nuclear waste storage.

In debates about nuclear power, opponents often highlight risks associated with the radiotoxicity contained in the spent fuel and related consequences for future generations. Methods of disposal proposed by most countries rely on insulating the waste fuel deep underground in geological repositories. The project closest to realization is to enclose the spent fuel rods in copper capsules and cement them deep underground. Such projects are in progress in Sweden and Finland.

In other countries, such as France, Belgium and Switzerland, spent fuel reprocessing was selected. This process uses plutonium contained in the spent fuel to create MOX (mixed oxide) fuel which is subsequently used in light water reactors. The leftover remaining after separation of the uranium that has not been burned in the reactor (approximately 95% by weight of fuel) is reused, thus reducing the volume destined for spent fuel storage. Fission products and minor actinides (without uranium and plutonium) are encapsulated in metal containers and ready to be stored deep in geological repositories.

In both cases, the geological depository is designed to store spent fuel over a period of about 100–300 thousand years (after this period, the radiotoxicity of the spent fuel will be equal to the radiotoxicity of natural uranium, meaning it really would be safe). Research shows that such depositories are able to store spent fuel for the required period. However, the question is how to inform future generations performing excavation works in the areas of geologi-

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cal repositories about the potential risks connected to spent fuel, for instance in 50 000 years' time.

An alternative solution to the two processes described involves the use of transmutation, which was proposed by Claiborne in 1972. Transmutation involves fissioning the most radiotoxic nuclides (actinides) or replacing them with less radiotoxic substances. Originally, the idea of Claiborne (transmutation) [1] was that the plutonium and actinides would be fully used up in pressurized water reactors. The remaining waste would reach radiotoxicity equal to the radiotoxicity of natural uranium after a period of 1 000 years. Such a reduction in storage time reduces the risk of future generations chancing on the geological depository.

2. Spent fuel and its radiotoxicity

To be able to deal with issues of spent fuel or transmutation of radioactive waste, it is imperative to know which radioactive nuclides are produced and how radiotoxic they are. It is also necessary to know which radioactive nuclides need to be removed in order to considerably reduce radiotoxicity in short term (up to 400 years), medium term (up to 20 000 years) and long term (up to 300 000 years) storage.

3. Production of radioactive nuclides in nuclear power reactors

The main fuel in nuclear power reactors is uranium. In light water reactors, natural uranium is enriched with ^{235}U to 3–5% mass concentration in fuel, the remaining part is mainly ^{238}U. Most of the energy (60%) produced in power reactors comes from the fission reaction of ^{235}U with the rest coming mainly from the fission of plutonium nuclides.

Fission products have atomic weights of 60 to 170 u and most of them are unstable and subsequently decay. The half life of certain elements varies from a few hours to several days. They are the main source of radiotoxicity in the short term. The main nuclides responsible for radiotoxicity in the short term (up to 400 years) are listed in Table 1.

Nuclear fission is not the only reaction that takes place in the reactor and requires the capture of neutrons. Neutron absorption is accompanied by other

Table 1: Nuclides responsible for short term radiotoxicity (up to 400 years)

Nuclide	Half life
^{131}I	8 days
^{85}Kr	10.8 years
^{90}Sr	28.8 years
^{137}Cs	30.1 years

reactions that result in the creation of transuranic nuclides, which are primarily responsible for radiotoxicity in the fuel. An exemplary reaction is shown below:

$$n + {}^{238}U \rightarrow {}^{239}U^{*} \rightarrow {}^{239}Np + \beta \rightarrow {}^{239}Pu + \beta$$

The above reaction (double β decay) is responsible for the creation of plutonium (^{239}Pu). Other heavier isotopes of plutonium are the result of further neutron absorption reactions of plutonium isotopes. Americium (Am) is produced in the reactors in two ways: by the β decay of ^{241}Pu

$$^{241}Pu \rightarrow {}^{241}Am + \beta$$

or by the neutron absorption of ^{242}Pu:

$$n + {}^{242}Pu \rightarrow {}^{243}Pu \rightarrow {}^{243}Am + \beta$$

Curium (Cm) is produced by the reaction of neutron absorption of americium. The amounts of plutonium, americium and curium depend on the length of the fuel cycle, the initial composition and the neutron flux impinging on fuel. *Plutonium, americium and curium* are primarily responsible for radiotoxicity in the long term.

Table 2: Initial fuel composition, enrichment of 4.2%

Nuclide	Mass concentration
^{235}U	4.2
^{238}U	95.8

In modern light water reactors fuel burnup is approximately 5%. Table 2 shows an initial composition of fuel for light water reactors enriched to 4.2%

Table 3: Fuel composition after full fuel cycle of burnup equal to approximately to 50 GWd/t [2]

Nuclide	Mass concentration
^{235}U	0.767
^{236}U	0.552
^{238}U	92.186
^{237}Np	0.072
^{238}Pu	0.042
^{239}Pu	0.623
^{240}Pu	0.286
^{241}Pu	0.155
^{242}Pu	0.095
^{241}Am	0.038
^{243}Am	0.028
^{244}Cm	0.010
^{245}Cm	0.001
Fission products	5.145
Transuranic elements	1.350
Actinides without U and Pu	0.149

(mass enrichment). Table 3 shows the mass composition of the fuel after a whole cycle, with the burnup of fuel equal to approximately 50 GWd/t.

It can be observed that:

- 1/6 of the initial amount of ^{235}U was converted to ^{236}U and ^{237}Np by neutron absorption.
- 3.8% of the initial amount of ^{238}U was either fissioned or transformed into plutonium by neutron capture. The probability of fission of ^{238}U in the neutron spectrum in light water reactors is approximately 8%., i.e. approximately 0.3% of the initial amount of ^{238}U was fissioned and the remaining 3.5% was transformed into plutonium by neutron capture.
- As shown in Table 3, the concentration of plutonium and heavier nuclides is about 1.3% in the fuel after a full fuel cycle. That means that approximately 60% of the plutonium was fissioned. In light water reactors 40% of the energy is produced through plutonium fissioning.

4. Radiotoxicity

Spent fuel is highly radioactive and poses a threat to living organisms. Radioactivity itself is not a good measure of risk for living organisms. Alpha, beta, gamma or neutron radiation affect living cells differently. The location of the radiation, i.e. whether it acts from the outside or from the inside, needs to be taken into account.

The dose absorbed is expressed by the following formula:

$$h = \sum \epsilon_j A_j$$

where A_j—activity of absorbed substance [Bq], ϵ_j—coefficients depending on the manner in which the radiation was absorbed. The International Commission on Radiological Protection publishes regular values of coefficients ϵ.

Table 4: Coefficients of ϵ in case of absorption by an average person [3]

Nuclide	[nSv/Bq]
^{235}U	47
^{236}U	47
^{238}U	44
^{237}Np	110
^{238}Pu	230
^{239}Pu	250
^{240}Pu	250
^{241}Pu	5
^{242}Pu	240
^{241}Am	200
^{243}Am	200
^{244}Cm	120
^{245}Cm	210
^{90}Sr	28
^{137}Cs	13
^{129}I	110
^{131}I	22

Table 4 shows the set of coefficients ϵ for an average person in case of ingestion of radioactive substances.

Table 4 shows which elements are the most dangerous. From looking at the table, it would seem that plutonium, americium and curium are only four times more dangerous than natural uranium. That is

Table 5: Absorbed dose by an average person in case of 1 mg ingestion

Nuclide	
Natural uranium	0.018 mSv
^{239}Pu	0.57 Sv
^{243}Am	1.48 Sv
^{245}Cm	1.35 Sv

illusory. The absorbed dose depends on the radiation coefficient and activity. Americium, plutonium and curium are much more active than natural uranium. Table 5 shows the absorbed dose in case of ingestion of 1 mg by an average person.

The average annual dose of radiation in Poland is 3–4 mSv. A dose of 1–2 Sv causes acute radiation syndrome (ARS), but it is possible to make a complete recovery after a few weeks. A dose of 4 Sv is lethal to every second person and few will survive an absorbed dose of over 6 Sv.

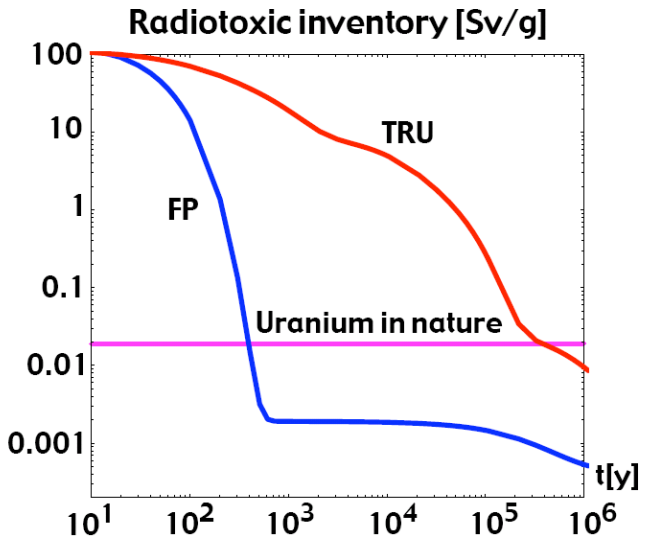

Figure 1: Comparison of radiotoxicity of fission products (FP) and transuranic nuclides (TRU) [4]

The radiotoxicity of 1 g of spent fuel of the composition given in Table 3 is depicted in Figures 1 and 2.

After 400 years, the radiotoxicity coming from fission products will be less than the radiotoxicity of uranium found in nature, as a result of radioactive decay mostly of ^{85}Cr, ^{90}Sr and ^{137}Cs, whose half life

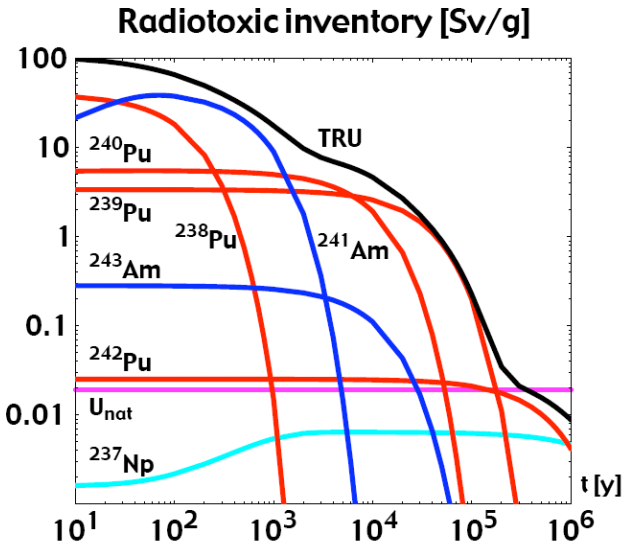

Figure 2: Radiotoxicity of selected minor actinides [4]

is about 30 years old, as shown in Figure 1.

Transuranic nuclides, plutonium, americium and curium have the greatest impact on the length of spent fuel storage. If spent fuel is taken directly from a nuclear power station to a geological repository, it should stay deep under ground for a period of 100 000–300 000 years (Figure 1). Transmutation of plutonium, americium and curium is one solution to reduce the storage period.

5. Transmutation

Theoretically, it is now known what needs to be done to reduce the quantity and storage period of radioactive waste: Transmutation of elements is followed by reaction with neutrons (absorption, fission). To make it efficient, there is a need for a high flux of neutrons, which can be found in nuclear reactors.

5.1. Transmutation in Light Water Reactors

Through appropriate reprocessing methods it is possible to separate approximately 99% of plutonium from spent nuclear fuel and use it in the production of new nuclear fuel—Mixed Oxide (MOX) fuel. During the production of MOX nuclear fuel, a mixture of plutonium isotopes is mixed with depleted or natural uranium (or even enriched uranium). During production it is possible to use Reactor Grade

Plutonium (RGP) from spent nuclear fuel or even Weapon Grade Plutonium (WGP) containing lighter plutonium isotopes [5]. The next stage of the process is to place MOX fuel in the existing reactor fleet without power, operation time or safety penalties.

Out of safety cocnerns current light water reactor designs allow the use of reactor cores with no more than 30% MOX fuel assemblies [6]. After design modifications it is possible to add more MOX into the core and theoretically achieve an upper limit of 100%.

Nevertheless, standard MOX fuel does not enable the plant to operate safely with reprocessed MOX. To recycle fuel more than once it is necessary to use plutonium multi-recycling schemes with new fuel designs. Reprocessing standard MOX and using its plutonium content more than once leads to a positive coolant void value and makes reactor fuel unsafe, which is unacceptable [4, 6, 7]. Additionally, the use of standard MOX fuel in water reactors has only a minor impact on the total plutonium inventory in the fuel cycle. In order to reduce overall plutonium radiotoxicity other methods must be applied [8].

There are various plutonium multi-recycling concepts and options. For instance, there is a special design called CORAIL, with MOX fuel rods and partly enriched UOX fuel rods in a special proportion. This design allows one to equalize plutonium breed and burn, and hence stabilize its inventory in the fuel cycle. Computations show that it demands about 80% of the reactor park with this fuel [4, 9]. Another option is a MOX-UE (MIX) fuel with a mix of plutonium isotopes (about 12%) and enriched uranium (with 3–4% enrichment). In this case we need only 30% of the whole reactor fleet, using this fuel to stabilize the plutonium inventory [4, 7]. Interstingly, MOX-UE fuel allows for the addition of americium with a maximum concentration of 1%. For fleet with 40% of such fuel, it could enable simulations to be made for the stabilization of plutonium and americium inventory.

Elimination by partitioning and transmutation of all plutonium in the UOX fuel could reduce the safe storage time to about 20 000 years. In the case of typical MOX fuel the disappearance of all of the plutonium isotopes would reduce the storage time to about 50 000 years [10].

In order to further reduce the time required to store spent nuclear fuel, it is crucial to burn actinides with a mass number above plutonium (minor actinides). The principal minor actinide is americium and its transmutation is crucial for further reduction. Utilization with special emphasis on americium is possible in light water reactor, but it is highly technically demanding, expensive and unsafe.

Total elimination of most of the plutonium and americium could lead to radical reductions in storage time to a few hundred years. To reduce total storage time it would be necessary to transmute curium isotopes. For a typical PWR spent UOX fuel curium radiotoxicity is higher than uranium ore radiotoxicity for about 200 years [10]. In the case of transmutation using MOX fuel, total elimination of plutonium and americium will reduce the necessary storage time to somewhere in excess of 20 000 years due to the high production of long lived curium isotopes. Additionally, in MOX fuel high radiotoxicity is generated by neptunium and thorium, which is negligible in the case of UOX [10].

Curium irradiation in the thermal reactor neutron spectrum is unsuitable because it leads to the production of californium (for example ^{245}Cm is transmuted into ^{252}Cf) which has a very high spontaneous fission cross-section and it is a very strong neutron emitter. Fuel fabrication with the addition of curium isotopes is extremely difficult because some of it generates huge decay heat (for example: isotope ^{244}Cm generates about 2.8 W/g). Due to an array of issues with light water reactor transmutation technology, it is more suitable to use fast reactors or dedicated transmutation facilities with Accelerator Driven Systems to transmutate and solve the problem of nuclear waste [4].

Another problem is the transmutation of fission products, which cause high radiotoxicity for the first few hundred to a thousand years. Without eliminating it, reducing storage time to below one thousand years is not feasible.

5.2. Transmutation in Fast Spectrum Reactors

Initially, fast reactors were created to breed plutonium and hugely increase the available energy density of nuclear fuel. In the early days of nuclear power, overall forecasts of energy consump-

tion and available resources were worse than now and a potential lack of energy sources was a real problem. Later, fossil fuel reserves were adjusted sharply upwards and even potential uranium reserves are higher. Expensive fast breeder reactor technology was not irreplaceable.

Currently, within the framework of the Generation IV Forum and the general concept of Gen-IV installations, fast reactors constitute a tool dedicated to withstand sustainable development and as high a possible utilization of nuclear fuel [11]. Gen-IV fast reactor designs are mainly based on liquid metal technology (sodium, lead or lead-bismuth) and gas technology (helium, carbon dioxide).

Current plans and designs do not assume the breeding of plutonium. Instead, it is viewed as preferable to burn the plutonium stockpile and reduce its long term radiotoxicity in the whole fuel cycle. Fast reactors are more useful than light water reactors in terms of transmutation strategy. Importantly, the development of burner fast reactors preserves technology which could be relatively easily used to return to the breeding concept in the whole fuel cycle should it become necessary and economical.

Since plutonium does not generate severe problems when used in fast reactors, it could be simply maintained at a constant inventory in the cycle and even effectively removed from the cycle [4].

Under the CAPRA project, which was dedicated to plutonium burning in the European Fast Reactor (EFR), it was calculated that in order to burn produced plutonium in nuclear reactors, 24% of the total number of reactors would have to be fast reactors [12].

The minor actinides present a problem, especially americium. Addition of those elements into the fuel in large quantities has an adverse impact on the safety parameters of fast reactors [13]. Nevertheless there are no problems with fuel fabrication, as there were light water reactors transmutation dedicated fuel since the production of californium is much lower. For example, the upper limit for addition of americium into existing fast reactors is about 3% and with new designs it is about 5%. A nuclear fleet with 50% being fast reactors operating with such fuel could keep its minor actinides inventory on a constant level [4, 13].

5.3. Transmutation in Accelerator Driven Systems

An alternative option to critical fast reactors are sub-critical fast reactors called Accelerator Driven Systems (ADS). ADS is based on spallation - where a large number of neutrons are emitted from heavy nuclides due to being hit by high energy charged particles—like protons. The charged particles are accelerated in a linear accelerator to energies in the range of 1 GeV and they hit the spallation target. Cascades of neutrons (up to 50 neutrons) interact with surrounding fission material and create new neutrons, which causes fission and generates further generations of neutrons. They could also interact with fission products or actinides and transmute them. An ADS reactor works like an energy-amplifier: energy is inputted in the form of a particle current and due to fission reaction 20–50x times more energy is created. Neutron flux in the ADS reactor is fast, allowing plutonium and minor actinides to be safely reduced in huge quantities. ADS installations have much smaller obstructions in terms of safety than critical fast reactors and about ten times more minor actinides can be added—up to as much as 50% of the fuel.

This type of installation is purely dedicated to the transmutation of nuclear waste due to heat removal limitations in the spallation target [4, 5, 8, 10].

Studies show that ADS systems can be used for higher actinides, in particular, the transmutation of minor actinides. If transmutation ADS systems were used, they would constitute 10% of all reactors and at the same time would practically enable full transmutation to be achieved [4].

By eliminating the plutonium, americium and curium from spent fuel, the time required for storing radioactive waste can be reduced to less than 1 000 years.

6. Summary

Transmutation in light water reactors is possible, but only reduces the required storage time of spent fuel to 20 000 years. To reduce the toxicity of spent fuel to the level of natural uranium in a timeframe of

1 000 years, trecourse to fast reactors or ADS systems appears inevitable at this stage [4]. Two solutions to achieve this end are shown in Figure 3.

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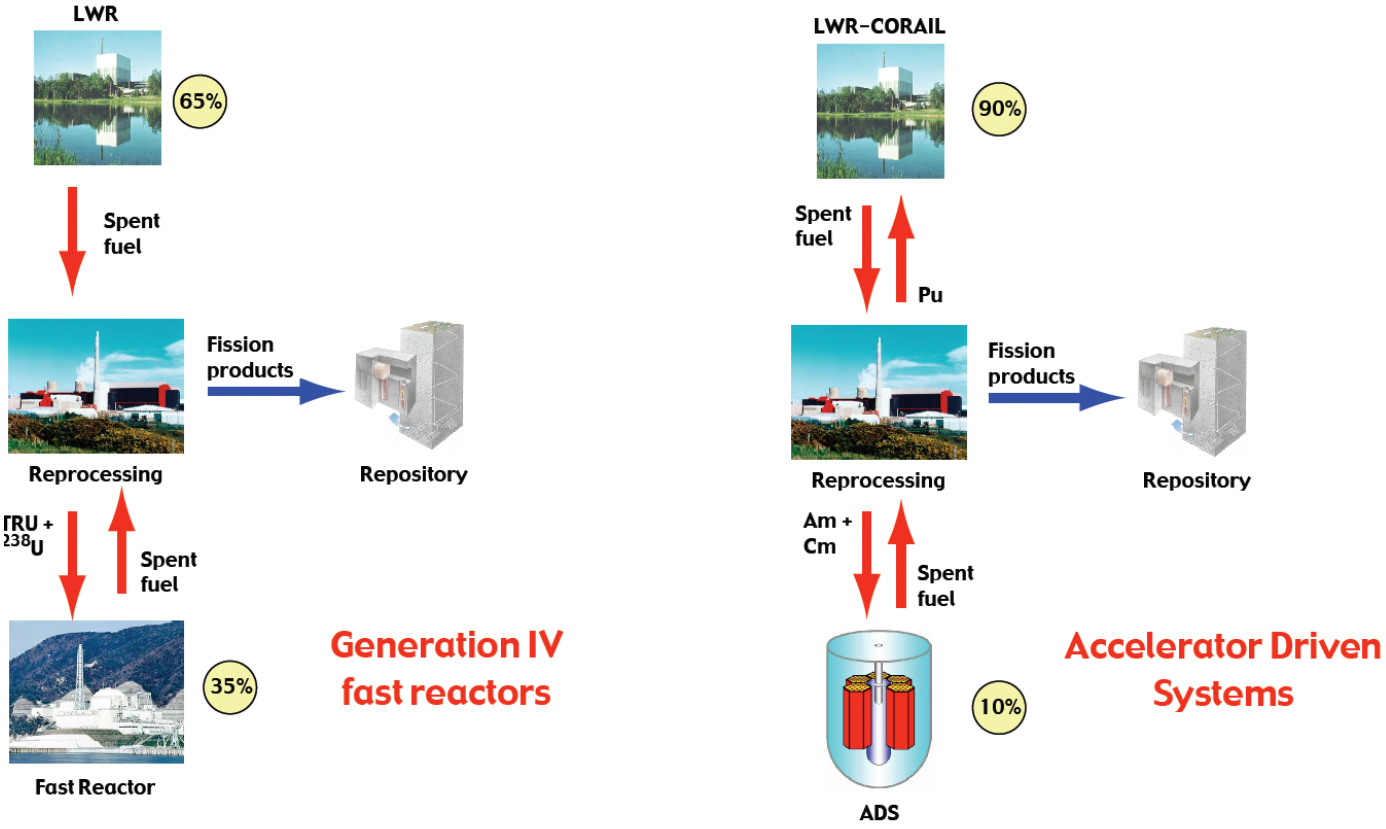

Figure 3: Possible solution to reduce the radiotoxicity of spent fuel to 1 000 years [4]