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WASTE MANAGEMENT IN FUTURE PARTITIONING AND TRANSMUTATION (P&T)

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ABSTRACT

Current research and development (R&D) in radioactive waste management is mainly associated with the quantities and toxicity of high level waste and spent fuel. One of the solutions that already exists, but has not yet reached scientific and technological maturity, is the process of partitioning and transmutation (P&T). Partitioning is the selective separation of radiotoxic isotopes from reprocessing streams. After the successive partitioning has been done, the long-lived radionuclides are converted into shorter-lived or stable nuclides by process called transmutation. P&T can reduce the radiotoxic inventory of spent fuel by a factor of 100 to 1000 and can achieve the reduction of time needed to reach the radioactivity level of the uranium ore from 100,000 to 5000 years. To achieve this, the separation of plutonium, minor actinides and long-lived fission products has to be implemented as early as possible in the fuel cycle strategy. Currently, P&T is still at the research and development stage and it needs to be scaled up, before it can be introduced on an industrial scale, therefore the paper will present the current status of the development of P&T and plans for the future.

1 INTRODUCTION

The management and disposal of radioactive waste from the nuclear fuel cycle is one of the most difficult problems currently facing the nuclear power industry. Today, more than forty years after the first commercial nuclear power plant entered service, no country has yet succeeded in disposing of the high-level, long-lived, highly radioactive, and most technologically challenging of the waste streams generated by the nuclear industry.

In most countries, the preferred technological approach is to dispose waste into the deep geological repositories constructed in rock formations hundreds of metres below the earth's surface [1]. Although several experimental and pilot facilities have been built, there are no operating high-level waste repositories, and all countries have encountered difficulties with their programs. The perceived lack of progress towards successful waste disposal clearly stands as one of the primary obstacles to the expansion of nuclear power around the world.

The problem is mainly associated with spent fuel that is discharged from nuclear reactors and so will remain highly radioactive for many thousands of years. The contents of spent fuel from the reactors can be roughly divided into 4 categories: 1) uranium; 2) plutonium; 3) fission products; 4) minor actinides (neptunium, americium and curium). Annual production in EU (15 members) [2] is 2500 tons of fuel assuming that the burnup is 37.2MWd/kg. Among 2500 tons of spent fuel the estimated amount of plutonium is 23 tons,

minor actinides 4 tons and some selected fission products: technetium-99 is 2.3 tons; iodine-129 is 0.8 tons; caesium-137 is 2.3 tons and strontium-90 is 1.0.

Some fission products (^{99}Tc, ^{129}I), plutonium and minor actinides, although present at relatively low concentrations in the spent fuel, are very long-lived and highly radiotoxic, so they present a hazard to life. Therefore their disposal requires isolation in stable deep geological repositories for more than 100,000 years.

Radiotoxicity is the measure of the toxicity to health of a radionuclide. Some general features of the radiotoxicity of spent fuel are shown in figure 1 [3]. A reference point is the radiotoxicity associated with uranium ore and its daughters. The radiotoxicity of the fission products dominates the total radiotoxicity during the first 100 years. Thereafter, their radiotoxicity decreases and reaches the reference level after about 300 years. The long-term radiotoxicity is dominated by the actinides, mainly by the plutonium and americium isotopes. The reference radiotoxicity level is reached by spent nuclear fuel only after periods of more than 100,000 years.

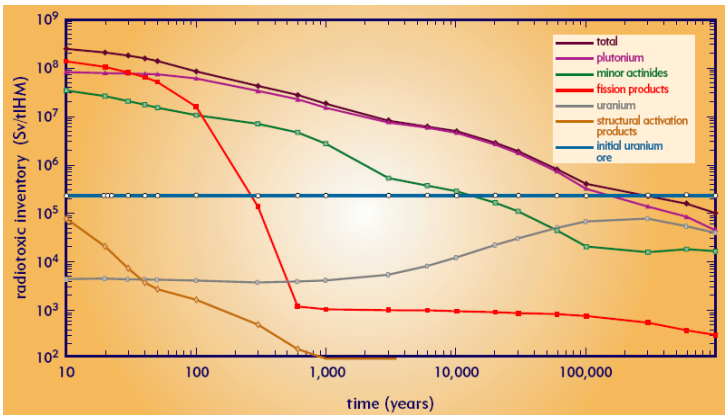

Figure 1: Changes in the radiotoxic inventory over time, expressed in Sielverts per metric ton of initial heavy metal (Sv/tiHM), of a spent fuel (uranium oxide enriched with 3.7% of uranium-235)

The objective of current research in waste management is to reduce the quantities and toxicity of waste with the use of the P&T process. Briefly the main goals of the P&T process should be:

- Reduction of the hazard associated with spent fuel at medium and long times (>300 years) by significant reduction of the inventory of plutonium and minor actinides.
- Reduction of the time interval required to reach a reference level of radiotoxicity by recycling plutonium and minor actinides.
- Decrease of the spent fuel volume by separation of uranium to enable more efficient storage or disposal. This should result in an increase of the effective capacity of a final repository.

However at each step of the fuel cycle new wastes would be created, which means that partitioning and transmutation will not eliminate the need for a deep geological repository for certain long-lived radioactive wastes from spent fuel. Therefore a highly efficient system of separation of the radionuclides of choice would be needed to make the technique feasible.

2 PARTITIONING

A prerequisite for transmutation by irradiation with neutrons is that the nuclides to be transmuted are separated (partitioned) from the other nuclides in the spent fuel. Since 95% of the spent fuel is constituted of slightly enriched uranium, the elimination of uranium as a fertile breeder material for plutonium is the first step to be made unless one wishes to produce more plutonium and other actinides. Separation of the various elements can at least in principle be achieved by mechanical and chemical processes.

Currently there are some large scale facilities for separation of uranium and plutonium from the spent fuel. This selective partitioning, carried out using the PUREX process, can efficiently separate uranium and plutonium, however it can not separate the minor actinides and fission products from high level waste. The concept of separation can be extended to these highly radiotoxic, long-lived radionuclides found in waste. Once the separation of these radionuclides is successful preformed, the remaining waste would lose its radiotoxicity much more quickly. The separated products would then need to go through another process, such as transmutation or, in some cases, specific conditioning.

This advanced separation process is mostly applied for separation of neptunium, americium, and curium. After plutonium, they represent the highest radiotoxic inventory in spent fuel. At a later stage, attempts could be made to extract other long-lived fission products such as iodine-129, technetium-99, and cesium-135. These three elements produced in significant quantities in the fission process are soluble and can migrate relatively quickly through ground water. The research program strategy for separating these long-lived radionuclides consists in adding extra processes to existing industrial spent-fuel reprocessing methods, and exploiting the full potential of the PUREX process. R&D has defined the frameworks of an advanced separation process based on new chemical processes using highly selective molecules. Studies have shown that neptunium can be separated out, just like iodine and technetium, by adjusting the PUREX process. Other processes have been also demonstrated for separation of actinides and lanthanides, but none of them have yet reached the industrial level.

Traditionally, partitioning techniques used in the nuclear industry are dominated by liquid-liquid extraction based on a combination of organic and aqueous solutions. One of the main problems of these techniques is the degradation and destruction of organic molecules by high-energy radiation, which complicates the design of processes and increases operating costs. Hence, numerous attempts have been made over the years to develop non-aqueous partitioning alternatives, which would have much smaller sensitivity to radiation. This section briefly describes aqueous and non-aqueous separation techniques currently used on industrial scale and research activities in the field of new separation methods for more effective separation of minor actinides and fission products.

2.1 Aqueous partitioning techniques

The main aqueous technique considered for partitioning of actinides, lanthanides and long-lived fission products is the PUREX process, which is universally employed in the irradiated fuel reprocessing industry. PUREX is a wet chemical process based on the use of TBP (tri butyl phosphate), solvent containing phosphorus. Uranium and plutonium are co-extracted by TBP and thus separated from the bulk of the fission products which remain in the aqueous phase. The behaviour of the minor actinides and long-lived fission products in the PUREX process can be divided into three categories:

- elements already partially separated by the PUREX process: Np, Tc and I. For these elements, the R&D objective involves process extensions to achieve the desired separation performance.
- elements separable by TBP, for which a complementary step to the present PUREX process can be developed. This applies to Zr.
- elements that cannot be separated by the PUREX process:
    - Am and Cm,
    - Cs, Sr and probably the other fission products.

To separate these elements, it is necessary to develop new classes of extractants, or to resort to different separation methods. Several processes have been studied and tested in hot facilities; among the most important are: TRUEX (USA, Japan, Russia, Italy, India), TRPO (China, India), DIAMEX (France, Italy, Europe, Japan, USA, India) for actinide-lanthanide (An-Ln) group separation, coupled to recent CYANEX 301™ (China, Germany, India), SANEX, ALINA (Germany) and BTP processes which allow the An and Ln separation. For separation of Am/Cm the SESAME (France, Japan) process was developed.

2.2 Non-aqueous methods

What is generally understood by non-aqueous partition techniques is chemical processing at elevated temperatures. Thus, such processes are also often called pyrochemical. The major attraction of these processes lies in the fact that metals, oxides, or salts are used, which in general are much more resistant to damage by ionizing radiation than molecular liquids like water or organics. Another favourable property is that much higher concentrations can be handled, decreasing the size of the plant. A third good property is the assumption that pyrochemical processing of spent nuclear fuel can be designed in such a way that separation and production of pure plutonium can be avoided, made very difficult, or perhaps nearly impossible to achieve. Non-aqueous methods are in general more proliferation resistant than the typical PUREX process.

The pyrochemical process is particularly suited for processing high-burnup fast reactor (FR) fuels and irradiated targets in order to shorten the cooling times in the processing cycles. The separation ability is presently limited to groups of elements, and in order to achieve higher separation factors and element separation, multi-stage separation will have to be developed. The development of pyrochemical partitioning processes requires also the development of equipment that is suitable for the industrial application of such processes. Material and corrosion problems must be solved as well as technical and radiological safety issues. The road to industrial application is probably longer for the pyrochemical processes than for the aqueous based processes.

The separation efficiency between different processes is shown in figure 2 [4]:

Process	PUREX	Advanced Aqueous Reprocessing(*)	Pyrochemistry
Status	Industry	Laboratory	Laboratory
U	99,9 %	99,9 %	99,9 % (Prototype)
Pu	99,8 %	99,9 %	99,9 % (Prototype)
Np	95 %	99,9 %	99,9 %
Am	-	99,9 %	99,9 %
Cm	-	99,3 %	?
Ln in MA	-	≤ 5 %	<10 %
Cs-135,137(**)	-	99,9 %	-
Tc-99	-	~80 %	-
I-129	98 %	99,9 %	-

(*) PUREX + DIAMEX + SANEX
(**) CALIXARENES

Figure 2: Separation efficiency for various actinides and fission products in different chemical processes

After the successful partitioning has been done by aqueous or non-aqueous technique the radionuclides selected for transmutation then need to be fabricated into some sort of target and put into a device where they can be bombarded with neutrons to convert them into more stable or short-lived radionuclides.

3 TRANSMUTATION

As mentioned in the introduction, the objective of transmutation is to change one nuclide into another as a result of nuclear reaction – in radioactive waste management, the aim is to produce shorter-lived or more stable nuclides. The main nuclides of interest for transmutation are the Pu and minor actinides (Am, Np, Cm) which can be most effectively transmuted by nuclear fission. Capture reactions will only lead to transmutation from one long-lived nuclide to another via decay of some interim short-lived nuclides. Many of those elements are only fissionable in fast neutron spectra. This means that for an effective transmutation a strong fast neutron flux is needed. Two types of facilities are considered for creating such a strong neutron flux:

- a fast reactor with a self-sustaining chain reaction
- accelerator-driven subcritical reactor system (ADS)

The properties of the fast reactor are well known, since they have been in operation for many years. Only a few of these systems however are still in operation, due to the problems in cooling of liquid sodium. Concerning ADS, two so-called roadmaps for development of the ADS were published in USA 1999 [5] and in Europe 2001 [6]. Both these studies emphasise that the first major step towards the implementation of an ADS should be the construction of a demonstration facility of some reasonable minimum size.

3.1 Accelerator-driven systems

In contrast to conventional nuclear reactors, in which there are enough neutrons to sustain a chain reaction, sub-critical systems used in accelerator driven systems need an external source of neutrons to sustain the chain reaction. These extra neutrons are provided by the accelerator. The accelerator produces high-energy protons which produce neutrons via a spallation source. Such a sub-critical system has more control and flexibility in the design and operation of the critical reactor. This is required when the reactor is being used to transmute large amounts of nuclear waste in the form of minor actinides (MAs) due to their high intensity of radiation.

An ADS accelerator must deliver an intense proton beam. It should do so in a highly controllable way which involves flexibility and minimal beam losses. Although the accelerator technology developed for the needs of basic science already meets most of these requirements, no ADS accelerator has yet been built. Progress still has to be made to reach "reactor-quality" reliability [7]. The essential points are the constancy of the beam output (over a period of the order of seconds) and the number of beam interruptions (typically between a few tens and a few hundreds per year). These were not essential issues in basic nuclear and particle research.

Today it appears that ADS has great potential for waste transmutation and that such systems may go a long way towards reducing the amounts of waste and thereby reducing the burden on underground repositories. Such facilities must meet very high standards of safety and radiation protection, and be environment friendly. They must be economically viable and have good proliferation resistance. The large amount of energy released in the transmutation process should be used in a proper way. In other words the processes and facilities must be acceptable to society.

Of course the strategy for P&T depends on the national nuclear energy program. The essential differences between the various systems are evaluated with the help of representative fuel cycle schemes.

4 FUEL CYCLES

4.1 Once-through cycle

The once-through cycle (OTC) scenario is the main alternative in Slovenia as in the countries like Canada, Finland, Spain, Sweden, USA and many others. This is due to the very low uranium prices; thereby OTC provides the lowest cost nuclear energy production. However, it implies that the residual fissile material content (~1 % Pu and 0.8 % ^{235}U) as well as the remaining fertile material (^{238}U) of the spent fuel will not be recovered and becomes a waste material.

The long-term radiological impact of the OTC can be controlled by a man-made system and natural barriers which should provide protection for as long as the life-time of the radiological source term they confine. The long time periods involved require a careful analysis of the confinement technology and of the long-term consequences for conceivable scenarios.

At the present time, there is no world-wide agreement on the time intervals for confinement of high level radioactive wastes in a geologic repository. Periods of 1000, 10000, 100000 years or even longer have been considered but no internationally accepted confinement period has been established [8]. For example: the licensing period for the proposed U.S. Yucca Mountain repository is 10000 years [4].

4.2 Reprocessing fuel cycle

Since natural uranium contains only 0.72% of fissile ^{235}U isotope, the recycling of U and Pu from spent fuel through the reprocessing fuel cycle scenario has been from the beginning of the nuclear era the standard scenario of nuclear energy production. There has been reduced support for this approach in many countries in recent years owing to economic factors and particularly because of proliferation concerns. By processing according to this RFC scenario the major fraction (~99.9 %) of the U and Pu streams is extracted and only a very minor fraction of minor actinides is transferred to the liquid HLW (and consequently to the vitrified HLW) and eventually to the geologic repository.

However, if the public and/or political acceptance of very long term disposal of HLW could not be obtained, the removal of minor actinides from spent fuel would be a technical solution which might reduce the residual radiotoxicity of the HLW. Moreover, with increasing burn-up, the generation of minor actinides becomes more and more important. The addition of a minor actinides partitioning process to the standard reprocessing plant would, in such a case, be the most obvious change to the current RFC. Countries with a reprocessing infrastructure (France, UK, Japan, India, Russia and China) and their associated partners could in the medium term realize a partial partitioning scenario by which the HLW would be practically free from long-lived transuranium elements.

From the radiotoxic point of view, the overall gain is rather limited since only ~25 % of the recycled Pu is consumed and about 10 % is transformed into a long term radiotoxic minor actinides source term. Recycling of spent LWR fuel as MOX provides an overall mass reduction with a factor of about 5, but does not significantly reduce the total radiotoxicity. Double, or perhaps at the limit, triple recycling of LWR-MOX is theoretically possible in LWRs if fresh plutonium is available, but in the short term the resulting radiotoxicity drastically increases throughout the subsequent recycling campaigns because of ^{244}Cm build-up. This avoids the reprocessing or the fabrication of new fuels.

If it were required to further reduce the global radiotoxic inventory it would be necessary to transfer the plutonium and minor actinides of spent LWR-MOX fuel into a FR-MOX fuel cycle scenario which becomes a part of the advanced fuel cycle (AFC).

4.3 Advanced fuel cycle

The advanced fuel cycle (AFC) with P&T incorporated is the most comprehensive approach. The AFC is a series of chemical, metallurgical and nuclear operations by which the plutonium and the minor actinides (Am, Cm, Np) and some selected fission products (^{99}Tc, ^{129}I) are separated from the main stream and recycled as targets into dedicated nuclear reactors and/or ADS to obtain a significant reduction of their radiotoxic inventory. AFC requires reactor facilities, which go far beyond the current nuclear technology. In particular, the transmutation approach calls for the development of FR-burners and/or ADS facilities, which may take 20 to 30 years to become industrially available. AFC is a very complex fuel cycle which offers a final solution of efficient radwaste management and can not be discussed briefly, so the detailed report of AFC is described in reference [8].

5 P&T IN RADIOACTIVE WASTE MANAGMENT

As pointed out in the introduction, the objective of partitioning and transmutation is to eliminate or at least substantially reduce the amount of long-lived radionuclides that has to go to a repository for final disposal. This implies that a successful outcome of the ongoing research and development on P&T may have a considerable impact on, in particular, future waste management programmes but also on the general development of nuclear power and nuclear energy. To investigate the effects of separation on the radiotoxicity reduction, three cases have been considered. The resulting radiotoxicity curves are shown in figure 3 [9], in which the cross-over point indicates the time at which the radiotoxicity of the waste reaches the reference level.

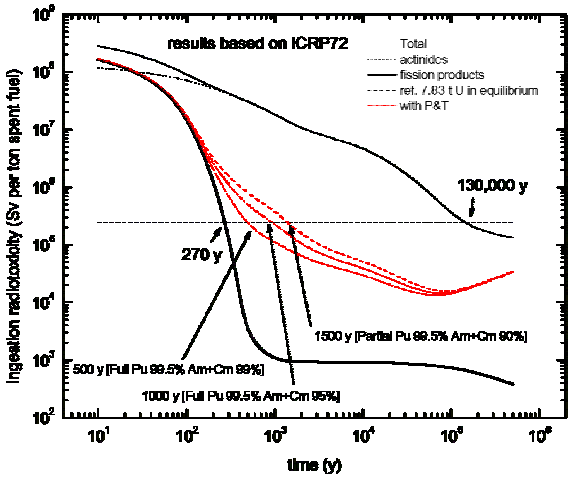

Figure 3: Ingestion radiotoxicity of one ton used nuclear fuel based on ICRP72 effective dose coefficients

The following observations have been made:

1. The open cycle: the spent fuel is directly sent to the long-term storage with no P&T. It takes 130,000 years before the radiotoxicity reaches the reference level.
2. The full multi-recycling of Pu as well as Am and Cm with high overall efficiency of P&T processes (99.5 % for Pu and 99 % for Am + Cm). The crossover point is 500 years. If the Cm is left in the waste, this time is extended to 1,000 years.
3. The full multi-recycling of Pu as well as Am and Cm with less overall efficiency of P&T processes (99.5 % for Pu and 95 % for Am + Cm). The crossover point is 1,000 years.
4. The partial multi-recycling: multi-recycling of the Pu (99.5 % of P&T efficiency), and one single recycling of the Am and Cm. In this case the Am and Cm are transmuted in targets in a fast reactor, and then 90 % of P&T overall efficiency is foreseen. Thus the cross over point is around 1,500 years. In this strategy, we can also consider leaving the Cm in the waste, and then 3,000 years are required.

Based on these results it can be concluded that P&T can help to reduce the time from 130,000 years to between 500 years to 1500 years. The fission products radiotoxicity curve gives the theoretical limit to the total radiotoxicity reduction in the case that all the actinides are partitioned and transmuted - no losses. This time is about 270 years. If the separation of Pu is 99.5% and the separation of Am and Cm is 99% then the reference point is reached in 500 years, but if the separation of Am and Cm is slightly less efficient (95%) the time for radiotoxity to reach the reference time is extended by 500 years. It can be concluded that for an efficient P&T process it is very important to have a very efficient separation.

6 CONCLUSIONS

It has been shown that today the problem of waste management, especially the concern of people about the hazard associated with high level waste and spent fuel, can be extenuated by the use of P&T technology. This means that today the choice of the flow of nuclear energy could have serious consequences for the future.

It should be indicated that the development of the P&T system will require several decades. Even the deployment and operation of the necessary industrial facilities will take another 100 years or more before the stocks of long-lived radionuclides from currently existing nuclear power reactors have been transmuted.

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