Challenges and potential benefits of partitioning and transmutation (P&T)

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Most of the radioactive hazard from irradiated nuclear fuel originates from only a few chemical elements – plutonium, neptunium, americium, curium and some long-lived fission products such as iodine and technetium. These radioactive by-products, although present at very low concentrations in the irradiated fuel, are a hazard to life forms when released into the environment. As such, their disposal requires isolation from the biosphere in stable, deep geological formations for long periods of time.

Partitioning and transmutation (P&T) is considered a means of reducing the burden on a geological repository. As plutonium and the minor actinides are mainly responsible for the long-term radiotoxicity, when these nuclides are first removed from the irradiated fuel (partitioning) and then fragmented by fission (transmutation), the remaining waste loses most of its long-term radiotoxicity.

It can be shown that the radiotoxicity inventory can be reduced by as much as a factor of 10 if all plutonium is recycled in reactors. Reduction factors higher than 100 can be obtained if, in addition, the minor actinides are burned. A prerequisite for these reduction figures is nearly complete actinide elimination by fission, for which multi-recycling is required. Moreover, the P&T strategy allows, in principle, a combined reduction of the radionuclide masses to be stored and their associated residual heat, and, as a potential consequence, the volume and the cost of the repository. To achieve this, however, there are still a number of outstanding challenges to be met, especially in the fields of separation and fuel development.

Recycling in LWRs and fast reactors

All P&T scenarios imply fuel reprocessing and recycling of actinides and possibly fission products. Plutonium recycling is a necessary first step. At present, this strategy is an industrial reality and has been implemented in several countries, using standard light water reactors (LWRs). Several studies have been performed to evaluate the recycling of plutonium and minor actinides in critical reactors. The comparison of detailed characteristics, including tables of plutonium, minor actinide and fission product inventories, of the three following fuel cycle strategies can be found for example in reference 1:

- multi-recycling of plutonium in LWRs;
- multi-recycling of plutonium and minor actinides in LWRs;
- recycling of plutonium and plutonium plus minor actinides in fast reactors.

A major finding is that the most promising approach to plutonium and minor actinide multi-recycling is based on the use of fast reactors. In fact, the multi-recycling of all minor actinides in LWRs has a very significant impact on the fuel cycle (e.g. at fuel fabrication, due to an increase of neutron doses of a factor of ~10 000), which makes this strategy impracticable.

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The recycling of plutonium and minor actinides in fast neutron spectrum reactors can be performed either in a homogeneous or a heterogeneous way. The homogeneous recycling mode consists of a system capable of recycling plutonium and minor actinides together (avoiding a separation of plutonium and minor actinides), stabilising both plutonium and minor actinide mass flows, and sending only a small fraction of the radiotoxic actinides (losses at reprocessing) into the wastes. In fact, if the losses at reprocessing are assumed to be of the order of 0.1%, homogeneous recycling allows one to obtain a reduction of the potential radiotoxicity by a factor of almost 200 with respect to the open cycle scenario, and this over the entire timescale of 100 to 1 000 000 years. This reduction is such that the radiotoxicity in deep geological storage becomes comparable to that of the initial uranium ore after less than a thousand years. The main advantages of homogeneous recycling are that the concept is designed to produce energy, allowing for an optimised use of resources and can, in principle, accommodate several options in terms of reactor size and fuel, reactor coolant and waste forms, among others.

Heterogeneous recycling consists of performing the transmutation of minor actinides in the form of targets to be loaded in specific subassemblies of critical cores of a "standard" type. The potential advantage of heterogeneous recycling is to concentrate the handling of a reduced inventory of minor actinides (separated from plutonium) in a specific fuel cycle. A potential limitation of this approach is the very high irradiation times needed to fission a significant amount of minor actinides. In fact, for heterogeneous recycling the limiting factor is the fission rate value which can be reached under realistic conditions, while for homogeneous recycling the limiting factor is the separation chemistry performance. Another potential draw-back of this approach is the impact on the reactor characteristics (e.g. on power distributions) due to the presence of target-loaded subassemblies in the core. Overall, most studies indicate that the transmutation of long-lived fission products (such as technetium-99 and iodine-129) is rather impracticable and its impact questionable.

Dedicated accelerator-driven systems (ADS)

Another approach is to separate the minor actinides fuel cycle and the transmutation technology from the electricity production. This would be feasible by using dedicated fast neutron cores, where the plutonium-based fuel is heavily loaded with minor actinides. Possible drawbacks with such critical dedicated cores are the difficulties related to the degradation of safety parameters, such as a very low delayed neutron fraction and a reduced Doppler effect. These disadvantages have helped promote the concept of accelerator-driven, fast neutron, sub-critical systems (ADS) and the so-called "double-strata" fuel cycle concept described later in this article.

To develop an idea of the characteristics of a typical ADS (600 MeV proton accelerator coupled via a spallation target to a fast neutron sub-critical reactor core), a rather simplified calculation shows that the accelerator beam must be of the order of 5 mA (3 MW_{th} in the particle beam) for a sub-critical core of 0.99 and about 25 mA (15 MW_{th} in the beam) for a sub-criticality of 0.95. This indicates that the choice of the sub-criticality level is crucial and that it is probably difficult to envisage a very sub-critical core (e.g. $k < 0.95$), in view of the demanding characteristics of the required accelerator (>15 MW_{th} in the beam), the stringent requirements in terms of accelerator reliability and the cost of the energy to feed it. The demonstration of the ADS concept components (i.e. high power proton accelerator, spallation target, sub-critical core) and of its behaviour during operation (e.g. the continuous and effective monitoring of the sub-criticality with appropriate experimental techniques) is a significant R&D challenge.

P&T scenarios based on fast neutron spectrum cores

This section describes three of the most commonly discussed partitioning and transmutation scenarios. All three go beyond the strategy of the "once-through" ("open") fuel cycle (whereby fuel is irradiated only once before being placed in final storage) and imply fuel reprocessing. Their specific characteristics are outlined below.

Development of nuclear energy with waste minimisation

This scenario can be implemented in Generation IV fast reactors, with homogeneous recycling of plutonium (Pu) and minor actinides (MA) together (2-5% MA in the fuel). It allows a drastic minimisation of the radioactive waste in terms of volume, radiotoxicity and heat load. It preserves resources (Pu is an essential resource) and provides enhanced resistance to proliferation (Pu and MA are kept together).

A variant can also be envisaged, using the heterogeneous recycling mode as described above.

Targets of minor actinides (for example on a uranium support) would then be loaded at the periphery of a Generation IV fast reactor.

The "double-strata" fuel cycle

The double-strata fuel cycle would make use of commercial reactors burning plutonium using mixed-oxide fuel and separate minor actinide management, typically through an ADS. The minor actinides would be managed in a dedicated transmuter system, which could either be a low-conversion ratio-critical fast reactor, or a sub-critical accelerator-driven system (ADS) loaded with uranium-free fuel.

The main interest in this scenario is the possibility of keeping the management of minor actinides independent from the commercial fuel cycle. The expected reduction of radiotoxicity is similar to that expected in scenario 1 above, if the separation performance (e.g. losses during reprocessing, or transuranics recovery rate) is approximately the same in the two scenarios.

The reduction of transuranic (TRU) stockpiles

This scenario, which relies on multi-recycling of plutonium and minor actinides in dedicated transmuters, offers a potential means of reducing stockpiles of these elements in spent fuel, for use for example in the case of the phase-out of nuclear power plants. However, if implemented by a country in isolation, this scenario implies a substantial deployment of new installations (such as fuel reprocessing and fabrication facilities and accelerator-driven systems). Moreover, it would take approximately 100 years to eliminate 80% of the initial TRU inventory.

Potential benefits of P&T

Partitioning and transmutation offers significant potential benefits to the fuel cycle, such as:

- Reduction of waste volume and heat load for deep geological storage, meaning that a larger amount of radioactive waste can be stored in the same repository.
- Reduction of the radiotoxicity in the deep geological repository (which is important in the case of an "intrusion" scenario).
- If the transuranic elements are not separated (through homogeneous recycling in a fast neutron reactor for example), improved proliferation resistance is achievable.

The loading capacity of a typical radioactive waste repository of the Yucca Mountain type can be increased substantially if some of the actinides and fission products are removed from the waste before being despatched to the repository. Assuming a separation rate of 99.9%, it can be shown that a removal of plutonium and americium will enable an increase in the repository loading factor of about 6. A further separation of curium, cæsium and strontium would allow increased loading factors of about 50 and higher.

The expected heat load reduction in a repository is shown in Figure 1. The multiple plutonium recycling and minor actinide disposal have limited benefits (below a factor of 2). The multiple plutonium and americium recycling associated with curium disposal has a more favourable impact (by a factor of 5 or 6 at 1 000 years after disposal). If curium is stored and not disposed, the theoretical heat load reduction is comparable to what is achievable when all transuranic elements are fully recycled in a fast reactor.

Figure 1. Heat load in a repository

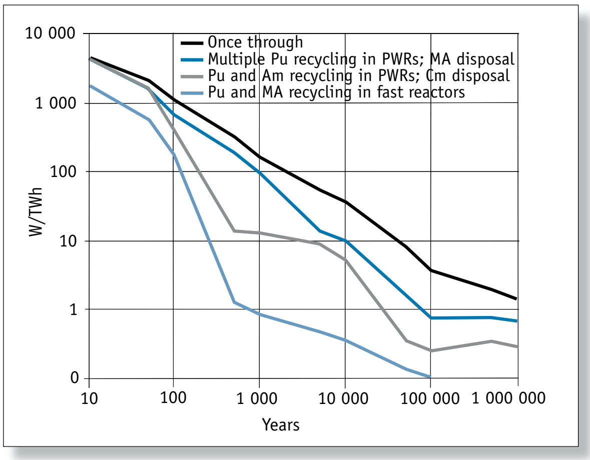
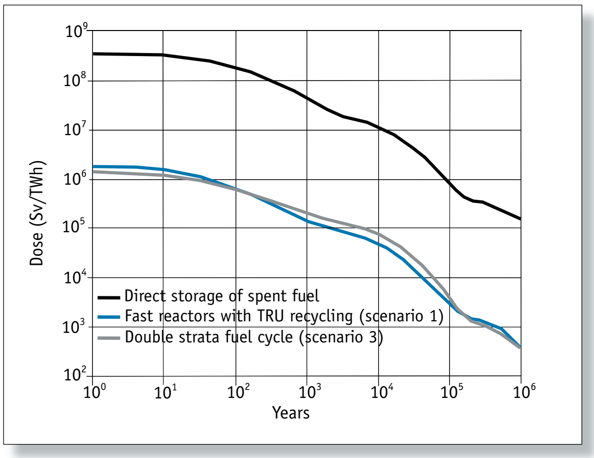


Figure 2 shows the reduction in radiotoxicity for different scenarios. It can be noted that the same reduction is obtained for homogeneous recycling as for the double-strata fuel cycle scenario, assuming the same chemical separation performance. The reduction is such that, at equilibrium, the potential radiotoxicity of the radioactive waste sent to a repository is reduced to the level of the radiotoxicity of the initial uranium ore after less than 1 000 years.

Some outstanding challenges associated with P&T

In general it can be stated that the physics of transmutation is well understood. Experiments have been performed irradiating pure transuranic isotope samples in power reactors, and transmutation rates

Figure 2. Radiotoxicity reduction



have been compared successfully to calculations. The main challenges are thus mainly in the areas of actinide separation and fuel development. A few examples are:

- The chemistry of actinides is complex, for example the separation of americium and curium from lanthanides is a challenging task. Significant results have been obtained, in particular with aqueous processes (see for example reference 3), but the industrial implementation of the processes developed at laboratory scale is still a major challenge.
- The development and processing of transmutation fuels, and in particular of the uranium-free fuels foreseen in ADS or of the targets of a heterogeneous recycle, are still under investigation.
- Dry (pyrochemical) processes (potentially more appropriate for U-free fuels) still need significant development efforts. Production and management of secondary wastes is also a concern.
- Large decay heat and high neutron emission of several higher-mass transuranic elements present new problems with respect to standard fuel manufacturing.

In addition, scenarios including ADS require the validation of new concepts such as highly reliable intense proton accelerators with 5-20 MW in the beam, spallation targets with solid or liquid metal, and a full coupling of the different ADS components and validation of the dynamic behaviour of a sub-critical system in the presence of an external source.

Finally, no P&T strategy can be implemented without a careful cost/benefit analysis. A first analysis has been performed (see reference 4), that attempts to quantify the impact on all of the installations of the fuel cycle (including different types of geological environments), and that gives preliminary cost estimations.

Conclusions

Based on the above and the supporting studies in reference, the following conclusions can be drawn:

- P&T technologies offer the potential for significant radioactive waste minimisation.
- P&T does not eliminate the need for deep geological storage, whatever the strategy, but enables an increase in its capacity, a drastic reduction in the burden and a potential improvement in public acceptance.
- P&T can be applied to widely different fuel cycle strategies.
- Critical fast reactors offer the most adapted and flexible tool in order to implement P&T. The use of ADS can be seen as an option or a potential back-up solution.
- Demonstration of P&T implies experimental demonstration beyond the laboratory scale of all of the "building blocks" of the strategy: adapted fuels, adapted reprocessing techniques, and reactor behaviour when loaded with significant quantities of MA.
- The implementation of P&T could benefit from a "regional" approach to the fuel cycle.
- The transmutation of long-lived fission products is questionable. However, an appropriate management of cæsium-137 and strontium-90 could have a significant impact on geological repository performance. ■

References

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