STATUS AND ASSESSMENT REPORT ON ACTINIDE AND FISSION PRODUCT PARTITIONING AND TRANSMUTATION

On behalf of the OECD/NEA Working group on P&T

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Abstract

A working group composed of 30 scientists representing 12 countries and three international organisations have prepared a comprehensive status and assessment report on the Partitioning and Transmutation (P&T) of actinides and fission products in the frame work of an advanced fuel cycle option and as an alternative or complementary waste management option.

The report is subdivided into two main parts: the first part gives a general overview of P&T with a critical evaluation and general conclusions; the second part covers a technical discussion and a systems analysis of partitioning, transmutation and risk analysis within an advanced fuel cycle and in a long term waste management option. Separate chapters are covering the Japanese and French R&D programmes which are the leading programmes within the OECD countries.

Introduction

For about 10 years the Partitioning and Transmutation (P&T) option has been revived due to the OMEGA initiative taken by the Japanese government in 1988. In 1990 the French government decided to launch a similar programme called SPIN (*Séparation - Incinération*) which was requested by law to investigate the alternative high-level waste (HLW) treatment by P&T in parallel with the development of the geological disposal option.

In the mean time important R&D efforts have been accomplished throughout the world to investigate means to reduce the long-term radiotoxic inventory resulting from the nuclear power generation.

Partitioning is a complex series of chemical and/or metallurgical operations, intended to separate selected radiotoxic nuclides or groups of nuclides occluded in the spent fuel elements. The separated fractions can be stored as such or transformed into new fuel elements or irradiation targets.

Transmutation is the general term covering as well elementary nuclear transmutation as a result of single neutron capture or more complex phenomena as fission of heavy nuclei, spallation and other nuclear reactions involving neutrons produced in nuclear reactor cores or in subcritical facilities connected to high energy proton accelerators. The ultimate aim of the transmutation is to reduce the radiotoxic inventory by transforming the initial nuclide either into short-lived radionuclides or stable isotopes.

Fuel cycle options

Fuel cycles

At the industrial level there are two major fuel cycle options:

- The "Once-through-cycle" (OTC) or direct disposal option of spent fuel.
- The "Reprocessing fuel cycle" (RFC) with MOX recycle and disposal of HLW.

The OTC is an option taken by the US, Sweden, Spain, Canada, Korea and some other countries. The RFC is the traditional nuclear fuel cycle which limits the recycling to uranium and plutonium and leaves the minor actinides (MA: neptunium, americium and curium) in the HLW mixture, this option is industrially available in France, United Kingdom and Japan. Some other countries: Russia, China and India have important activities in this field.

The P&T option is an extension of the RFC. Partitioning requires additional separation operations to extract the MAs and some selected Long-Lived Fission Products (LLFP: ^{99}Tc, ^{129}I, ^{79}Se, ^{93}Zr and ^{135}Cs) from HLW. In order to recycle and burn the MAs and to transmute LLFPs, dedicated nuclear facilities have to be developed. The P&T option is part of a so called "Advanced Fuel Cycle" (AFC) which could be realised as an extension of the present fuel cycle industry and the nuclear power plant (NPP) park.

Alternatively the AFC could be accomplished by separate operation of, on the one hand, the electricity producing reactors and their associated fuel cycle facilities and, on the other hand, the recycling and burning facilities of MAs and LLFPs operated independently from the major industrial nuclear-power activity. This type of AFC option is called "the double strata fuel cycle" and was pioneered by Japanese nuclear research organisations.

Fuel cycle issues

The OTC is undoubtedly the cheapest nuclear energy production but it leads to the accumulation of large inventories of spent fuel containing long lived actinides and fission products. The long-term impact has to be controlled by man-made systems and natural barriers which should provide protection for as long as the life of the radiological source term they confine.

The RFC option results in a partial recycling of major actinides (U and Pu) and produces in terms of residual radiotoxicity essentially vitrified high level waste (HLW) which has to be disposed of in suitable repositories. However, these glasses contain MAs and all the fission products. The radiotoxic inventory of these wastes is significantly lower than spent fuel but their toxic lifetime is determined by the presence of some MAs and LLFPs. The MAs have half-lives which range from several hundreds to million years and some LLFPs have half-lives of hundred thousand to several million years. Recycled U and Pu in fresh LWR-MOX fuel constitutes, in comparison with the OTC option, a reduction of the Pu-containing spent fuel volume but does not decrease the overall long-term radiotoxicity.

The AFC comprises all the fuel cycle and reactor sequences of the RFC but involves additionally a complex set of chemical separations to remove the MAs and some LLFPs from HLW before vitrification. Apart from the use of LWRs for energy production the AFC option requires the industrial deployment of Fast Reactors (FRs) in order to reduce the inventory and the radiotoxic half-life of separated MAs by transmutation or "incineration". The LLFPs could, in principle, be transmuted to inactive or shorter-lived isotopes in dedicated LWRs or in accelerator-driven systems (ADS).

The combined use of LWRs and FRs would stabilise the actinide inventory and reduce the MA content in the vitrified HLW. However, the gradual increase of the Pu+MA inventory in the reactors and fuel cycle facilities calls for additional actinide destruction tools such as ADS in order to eliminate the accumulated actinides and possibly to transmute some LLFPs.

Whatever option taken, it will always be necessary to build and operate geological repositories in order to isolate the HLW, the α bearing medium level waste and ultimately residual reactor cores from dedicated reactor systems.

Advanced reprocessing and partitioning

Improved aqueous reprocessing

Industrially aqueous reprocessing is carried out according to PUREX process by which spent fuel is dissolved in HNO_3 and treated by liquid extraction with tributylphosphate (TBP). Uranium and plutonium enter into the TBP phase and the other components remain in aqueous solution which is concentrated into HLW. This highly active waste stream contains the MAs and most of fission products (FP). Eventually this waste stream is vitrified and stored until disposal.

The additional operations to realise an AFC process can partially be carried out, on line, within the current reprocessing facilities and partially, off line, from the stored liquid HLW stream before vitrification.

The easiest adaptation of the conventional reprocessing process is by addition of the neptunium separation to the first cycle of the PUREX process. The neptunium separation process could be included in the current large reprocessing plants if some additional modifications of the separation flowsheet by REDOX reactions would be performed.

Some LLFPs constitute the very long-term risk for the disposal of HLW and are, or could be separated, during the PUREX process. The most important nuclides are: ^{129}I and ^{99}Tc.

The current reprocessing process involves the separation of ^{129}I ($t_{1/2}=15.7\times10^6$) from dissolver off-gases and its discharge into the ocean as medium-level liquid waste. The iodine scrubbing could be complemented by a specific conditioning operation in order to obtain a stable matrix (as insoluble as possible) in aqueous media.

Separation of ^{99}Tc ($t_{1/2} = 213\ 000$ years) is a more complex issue since it occurs as insoluble residue from the dissolver process and as soluble Tc in HNO_3. By scrubbing the TBP stream from the first extraction cycle with high-acidic NO_3, ^{99}Tc could quantitatively be eliminated from the HLW and treated separately, if necessary, in order to combine it with the insoluble fraction and to condition this in a stable metallic or oxide form.

Separation of minor actinides (MA) from HLW

Whereas Np could be separated during conventional aqueous reprocessing, this is not the case for the two main radiotoxic MAs: Am and Cm. Special off-line separation techniques from liquid HLW are necessary. This separation is difficult because rare earths (lanthanides) are generated during the fission process with ten times lager mass than MA and because the chemical behaviour of lanthanides is very similar to that of the MAs. Very high separation factors have to be reached in order to get an acceptable chemical purity of the separated MA fractions.

For about 10 years R&D on this subject has been performed in the US-DOE laboratories (ANL, SRP, LANL), in Japan (JAERI, PNC), France (CEA-FAR & Marcoule), Russia (Saint-Petersburg,) China and within the national nuclear laboratories in Europe and the laboratories of the European Commission (TUI).

TRUEX process

At ANL (USA) a process was developed using a very powerful extractant, CMPO, which allows to extract the MAs together with the lanthanides from strong acid solutions. The method was tested on real HLW solutions at PNC (Japan) and showed to be very efficient. There are some technical problems to be solved but as a whole this technique is a possible route to decrease the radiotoxicity of HLW. The TRUEX process is compatible with the PUREX technology.

The main unresolved question is the separation of lanthanides from MAs and the associated production of secondary waste.

DIDPA process and talspeak process

At JAERI, a new process was developed using DIDPA as extracting agent, this process has been very thoroughly investigated but suffers from the drawback that concentrated liquid HLW is much too acidic and that denitration has to be performed prior to the extraction. This leads to losses of insoluble actinides (Pu+MA) and reduced separation yields. One of the advantages of the DIDPA process is the incorporation of Np into the Am-Cm separation.

The separation of the Am-Cm fraction from the lanthanides is proposed by a so called "TALSPEAK" process used industrially for the separation of individual lanthanides. This process, if slightly adapted, could advantageously be used on the direct extraction of MAs from the more dilute first aqueous-raffinate of the PUREX process.

TRPO Process and cyanex-301 process

Recently a new process was developed in China using TRPO as extractant for the mixture of actinides and lanthanides from 2M HNO_3 waste stream. A straight-forward extraction of Ac-Ln fraction followed by stripping stages to remove impurities has been demonstrated with slightly diluted liquid HLW at the TUI of Karlsruhe.

Very promising performances in the direct separation of Am-Cm from lanthanides have been demonstrated with a special reagent "CYANEX-301". Although it is applied to low-acidic solution, this development is the most important one in this field for many years.

DIAMEX-SESAME Process

New, very effective extraction reagents for MAs, called "DIAMIDES", which are fully incinerable, have been developed by CEA-FAR and Marcoule and tested in hot-cell conditions. The reagent DMDBTDMA is almost as powerful as CMPO and can be used in very acidic media. Additional development work will be necessary to make this process applicable in industrial conditions in order to obtain acceptable purity of An in the product. The electrochemical oxidation process called "SESAME" process is the only one in the world which can, in principle, separate Am/Cm in one process step. The combination of DIAMEX with SESAME offers perspectives to separate the An from Ln and prepare pure chemical fractions of Am.

Conclusions and recommendations

From the above overview it is clear that great progress has been accomplished in the group separation of Ln+An from liquid HLW but that purification and separation of each MA by liquid extraction are still in the exploratory phase. Most of these methods are still on the bench scale or at hot-cell laboratory level. Table 1 shows a synthetic overview of the development state of each method.

In order to be useful on industrial scale, the above listed methods have to be **upscaled in terms of radioactivity level and throughput** of the liquid extraction rigs. Moreover very much attention will have to be devoted to the **secondary waste** issue which may become the bottleneck of their applicability in industrial facilities.

Table 1. **Status of R&D on aqueous separation techniques**

	Phase 1	Phase 2	Phase 3	Remarks
U and Pu separation (PUREX)	-	-	-	achieved industrially
Np separation (PUREX)			○	95% separation
(PUREX)		○		> 95% separation
(DIDPA)		○		
(HDEHP)		○		
(TRUEX)		○		
Am + Cm separation:				
* based on An/Ln co-extraction				
(TALSPEAK)			○	
(DIDPA)		○		
(TRUEX)		○		
(TRPO)		○		
(DIAMEX)		○		
* based on An selective extraction				
(TPTZ)	○			
(Picolinamides)	○			
(CYANEX 301)	○			SF=5 900
* based on precipitation				
(Ferricyanide)	○			
Am separation in the oxidised state				
(SESAME)		○		Am/Cm separation
Tc separation (PUREX)			○	soluble Tc
(PUREX)	○			insoluble Tc
Tc - PGM separation				
(Denitration precipitation)		○		
(Active carbon adsorption)		○		
I separation (PUREX)			○	95% separation
Zr separation (PUREX)		○		
Cs separation (Calixarenes)		○		
(Zeolite)		○		
Sr separation (Titanic acid)		○		
Cs and Sr separation (Dicarbollides)			○	
Pd (PGM), Se, Ru separation				
(Electrolytic extraction)	○			soluble Pd, Se, etc.

However it is also important that the more fundamental type of R&D, on the **synthesis of new selective extractants**, on their analytical characteristics and on their radiation stability, would be continued.

Dry reprocessing

In the future, nuclear fuel will reach higher burnups and recycling of spent fuel will have to be performed after short cooling times to be economically and ecologically attractive. Dry reprocessing using pyrochemical methods received for a number of decades the attention of some research institutes: the RIAR (Dimitrovgrad) in Russia; ANL (Argonne (Idaho)) in the USA; CRIEPI in Japan, and some older studies in France, Belgium etc.

Dry reprocessing is a pyro-electrochemical extraction of actinides from chlorinated spent oxide fuel which involves the following high temperature (650°C) processes: chlorination of oxide fuel, pyro-electrolysis of UO_2, selective precipitation, and additional electrolysis for the TRU nuclides. Recycling of Li-KCl, Cd and/or Bi reagents, conditioning of the salt and metal waste forms.

The proposed processes are very complex and necessitate the use of highly controlled atmospheres to avoid hydrolysis and precipitation reactions. However these processes have the inherent advantage in criticality safety considerations and in the compactness of the plant

Except for the pilot-scale demonstration of the pyro-electrolysis at Argonne-West and RIAR, all the other studies are at the laboratory scale. If other fuel types, e.g. nitride fuel or metallic fuels are to be used in multi-recycling processes, pyro-electrochemical methods might become necessary to implement this option.

Very much R&D work will be required in order to upgrade the pyro-electrochemical reprocessing process to the level of the present industrial aqueous reprocessing. But in order to shorten the cooling times which determine the recycle period of highly irradiated fuel (>150 GWd/tHM) the development of alternative recycling processes to the present PUREX process will become mandatory.

Possible scenarios for separated MAs

There are several scenarios which can be considered in the management of separated minor actinides:

- Neptunium resulting either directly from the reprocessing process or extracted from HLW could be separated easily from the other nuclides and recycled into MOX fuel or kept in separated form until advanced transmutation-incineration processes (FR, ADS) become industrially available. Alternatively, the separated neptunium could be conditioned in a thermodynamically stable phase and kept in a retrievable long-term storage. This matrix could be designed to serve either as irradiation matrix or as final disposal waste form.

- The Americium-curium fraction separated from HLW could be treated as a single fraction both in a transmutation strategy and in a disposal option. Both elements behave chemically so similar that very special radiochemical and/or electrochemical techniques have to be used to achieve the americium/curium separation. In a group separation option the americium-curium fraction could be stored as such or conditioned as a ceramic type of matrix and kept in engineered storage until new transmutation techniques become available.

- Americium could ultimately be separated as an individual element and recycled into special LWR-MOX or FR-fuel to be partially transmuted or conditioned in a ceramic form for extended transmutation-incineration processes by multi-recycling and ADS processes.

- The management of separated curium has not yet received convincing answers not only because the americium-curium separation is difficult but also because this very radioactive nuclide (heat and neutron source) cannot be recycled in LWR- or FR-MOX fuel. Storage of the separated nuclide for a century would alleviate the processing problems and make recycling in an advanced fuel cycle possible.

Transmutation of radiotoxic and long-lived nuclides

Since transmutation is the only method to reduce the half life of the separated nuclides, the separated MA and LLFP fractions have to be transformed into irradiation targets and submitted to neutron fluxes. Two options are open; irradiation with thermal neutrons or with fast neutrons.

Irradiation of MAs and LLFPs by thermal neutrons

The most obvious route to be followed is apparently to try to destroy the radiotoxic nuclides in the existing facilities in which they were produced. The existing NPPs could for example be specialised to carry out such job.

Transmutation of Neptunium

Among the MAs, Np is the easiest nuclide to be separated during reprocessing, and without major problems, it can be mixed with MOX fuel and irradiated. The transmutation yield (Table 2) in a single pass varies from 40 to 50% but the main product of the transmutation is ^{238}Pu with 87 years half life. The gain resulting from this operation is the disappearance of ^{237}Np with $2x10^6$ years half-life but its replacement by a highly toxic nuclide which would interfere with multi-recycling operations by aqueous reprocessing. Mixing of Np with MOX fuel will increase the radiotoxic inventory of the spent LWR-MOX assemblies, but it will reduce a very long-term risk. The Np reduction factor by such single step irradiation varies from two to three. Transmutation of Americium

Mixing of a pure ^{241,243}Am fraction with MOX fuel requires already an adaptation of the fuel fabrication equipment and technology. The gamma emission of ^{241}Am increases the radiation field around the fabrication units which have to be shielded and remotely operated. But if this adaptation is made ^{241,243}Am fraction can be recycled into fresh LWR-MOX fuel.

During irradiation, ^{241}Am undergoes transmutation into ^{242}Am which partly decays into ^{238}Pu; and ^{243}Am is mainly transmuted into ^{244}Cm, a highly radioactive nuclide which decays into long-lived ^{240}Pu. The annual depletion of Am ranges from 40 to 80% (Table 2). However the main end products of a single-pass irradiation are ^{238}Pu and ^{240}Pu.

Table 2. **Mass balances for homogeneous recycling in thermal reactors**

		PWR-UOX* moderation ratio 2	PWR-MOX** moderation ratio 2	PWR-MOX*** moderation ratio 3
Burn-up (*GWd/t*)		47.5	47.5	47.5
Initial minor actinide content (%) (mass of heavy metal)		1	1	1
Np	Initial mass (*kg*)	1 104	1 104	1 104
	Specific consumption (*kg/TWhe*)	12	10	11
	Depletion (% of initial mass)	53	43	45
Am	Initial mass (*kg*)	1 104	1 104	1 104
	Specific consumption (*kg/TWhe*)	19	5	10
	Depletion (% of initial mass)	83	20	42
* U-235 enrichment of 5.5% to recycle Np, or Am (standard enrichment 4.5%) ** Pu enrichment of 12% to recycle Np and 12.5% for Am (standard enrichment 9%) *** Pu enrichment of 7.7% to recycle Np, 8.2% for Am (standard enrichment 5.7%)				

Transmutation of Curium

Recycling of the ^{243,244,245}Cm fraction into fresh LWR-MOX fuel is a very difficult task which cannot be realised unless dedicated fuel fabrication plants with extensive neutron and gamma shielding are being developed. The irradiation of the Cm fraction yields a range of long-lived nuclides, e.g. ^{246,247}Cm, and decay products ^{239,240}Pu, and ^{241}Am. There is no gain in radiotoxicity and a long term reduction of the Cm inventory would require multi-recycling by very special reprocessing processes, e.g. pyro-electrochemical techniques.

Presently the preferred strategy would be to store separated Cm for ~100 years and to re-inject the resulting Pu nuclides into MOX fuel.

Transmutation of long-lived fission products

Iodine-129

Iodine being separated from dissolver off gases can be transformed into a single compound, e.g. insoluble AgI, PbIO$_4$ or kept as soluble NaI. Transmutation of ^{129}I is a very slow process since the capture cross section is very small for practical transmutation purposes.

Whatever the final product, it is necessary to develop a vented capsule or pin, since ^{129}I transforms during irradiation into gaseous ^{130}Xe. The thermal stability of the target material during irradiation is a questionable issue dedicated reactors would in any case be necessary because of the high iodine inventory.

Technetium-99

If the soluble and insoluble Tc fraction can be combined into a single metallic target, irradiation in LWRs is possible since the transmutation reaction yields a metallic Ru-100 which is inactive. The half-life of this irradiation its inventory reduction with a factor two is about 40 years in LWR-UO$_2$ and about 80 years in LWR-MOX.

Other LLFP

The other significant LLFP are: ^{93}Zr, ^{135}Cs.

Due to the relatively low mass fraction of the radionuclide (20% ^{93}Zr, 10% ^{135}Cs) in the total fission yield the presence of other isotopes would interfere in the transmutation reaction. Further advance in this field can only be achieved if isotopic separation becomes an economically feasible intermediate step.

Conclusions for R&D activities on transmutation in thermal neutron spectra

- There is a need for more precise determinations of the **MA generation** in spent LWR-MOX fuel by a more elaborate verification of the yields in high burnup LWR-UO$_2$ and LWR-MOX assemblies.

- Study of the impact of **multi-recycling of MAs** in LWR-MOX as a function of cooling time.

- The **delayed transmutation of Cm** after 100 years storage would have to carefully examined in order to establish the potential benefits of recycling.

- Among the LLFP **only ^{99}Tc** could technically be considered for transmutation in thermal reactors. The core management of this option needs to be carefully established: in core inventory, transmutation yields, residence times etc. as a function of a preset inventory reduction.

Transmutation and incineration in fast reactors

The prime advantage of irradiating nuclides particularly MAs in fast neutron fluxes is the increase of the fission contribution in the total transmutation yield. A secondary effect is the very high fast flux in FRs, which can be partially thermalised if necessary, e.g. by installing solid neutron-moderator materials in the peripheral section of the FR core.

Irradiation of MAs in FRs

There are two recycling modes to be considered: homogeneous and heterogeneous recycling. The advantages of heterogeneous recycling are apparent for the fuel cycle operations (reprocessing, fuel refabrication waste management) and the total transmutation capability is almost identical for homogeneous and heterogeneous recycling. However in standard core calculations homogeneous recycling is used as standard method.

Either specially fabricated fuel assemblies with high MA content (25 to 50%) or pure Np, Am, Cm targets have to be introduced in the FR core. But for reasons of reactor safety the total MA content in a sodium cooled core may never exceed 2.5% because of the positive sodium void coefficient.

By using lead cooling higher MA contents can be used, but the negative reactivity of the MA has to be compensated at BOC by an increase in the fissile isotope (^{239,241}Pu or ^{235}U) content. Due to the formation of fissile nuclides (^{239}Pu, ^{242}Am, ^{245}Cm) from the "fertile" isotopes (^{237}Np, ^{241}Am, ^{244}Cm) the burnup-reactivity swing becomes smaller than in a conventional core loading.

According to computations (Table 3) the depletion yield for ^{237}Np and ^{241,243}Am targets/fuels in FR-MOX reactors amounts to 60% composed of 23% fission and 37% capture. The deletion rate of ^{241,243}Am amounts to 45% partly fissioned and partly transmuted into other actinides.

Table 3. **Mass balances for homogeneous recycling in fast reactors**

		FBR*
Burn-up (*GWd/t*)		120
Initial minor actinide content (%) (mass of heavy metal)		2.5
Np	Initial mass (*kg*)	1 010
	Specific consumption (*kg/TWhe*)	10
	Depletion (% of initial mass)	60
	Fissioned fraction (% of initial mass)	27
	Fissioned mass (*kg*)	273
Am	Initial mass (*kg*)	1 174
	Specific consumption (*kg/TWhe*)	9
	Depletion (% of initial mass)	45
	Fissioned fraction (% of initial mass)	18
	Fissioned mass (*kg*)	211
*EFR type reactor with three enrichment zones (15.32, 18.18 and 22.08%)		

The mixed MA loading method, a combination of the homogeneous and heterogeneous methods was examined as an alternative; Np is uniformly loaded in the core region and a small number of subassemblies containing Am, Cm and RE nuclides is loaded into the blanket region. Parameters were surveyed systematically to investigate the basic characteristics of MA transmutation in a 1 000 *MWe*-class FR core with mixed oxide fuel.

The mixed MA-loading method can transmute a large amount of MAs without serious drawbacks in terms of core performance. The transmuted mass of MAs is ~530 *kg/cycle*, which is almost 16 times the mass produced by an LWR of the same power output.

It was found that a combination of homogeneous and heterogeneous loading methods, has the potential to achieve the maximum transmutation of MAs with no special design consideration.

Transmutation of MAs and LLFPs in Dedicated Systems

Dedicated transmutation systems are being studied at JAERI and CEA and based on a strategy named the double strata fuel cycle concept (or multi-component concept). The double strata concept is to consider a P&T fuel cycle (second stratum) separated completely from the conventional fuel cycle for commercial power reactors.

The P&T cycle could be made very compact by co-locating the entire facilities. This would minimise the transportation of nuclides that are troublesome with respect to waste management, and confine them effectively in the P&T fuel cycle.

Both critical reactors (burner reactors) and sub-critical reactors (hybrid systems or accelerator-driven systems) are potential candidates as dedicated transmutation systems used in the second stratum.

In dedicated "burner" reactors studied by JAERI: lead-cooled MA-burner reactor and He-cooled MA burner reactors with coated particle nitride fuel the transmutation is much higher, but the price to be paid is an initial enrichment of the MA-fuel with 35% ^{235}U.

In this case burner fuel containing 65% MAs has been considered. The major characteristic of this type of reactor core is the very high mean neutron energy which amounts to 720 keV which is close to the fission threshold of MAs. The MA fission in the ABRs ranges from 11 to 13% per cycle and is 190 to 200 Kg/GWth-year.

Accelerator driven transmutation systems

Accelerator-driven systems (ADS, frequently called hybrid systems) combine high-intensity proton accelerators with spallation targets and a subcritical core with or without blanket. The proton accelerator will be either a linear accelerator (linac) or a circular accelerator (cyclotron). The high-intensity continuous-wave (CW) proton beam with an energy around 1 *GeV* and a current of several tens *mA* are injected into a target of heavy metal. This results in spallation reaction that emits neutrons, which enter the subcritical core to induce further nuclear reactions. The subcritical core can, in principal, be operated with either a thermal or a fast neutron spectrum.

ADSs have unique features to burn MAs and possibly FPs, preferably in the double strata option. They operate in a subcritical mode and can more easily address safety issues associated with criticality than in critical systems. They also offer substantial flexibility in overall operation. ADSs can provide more excess neutrons compared to critical reactors. The excess neutrons may be utilised for transmutation, conversion, and breeding purposes. These features may be exploitable to prepare a safe and efficient means of transmuting nuclear waste. Both homogenous and heterogeneous fuel recycling is possible.

Various concepts of ADS have been proposed with different goals and approaches. Relevant R&D programmes are being pursued at CEA, JAERI, LANL, CERN, etc. In recent years, all the system concepts proposed by these groups have converged on a fast neutron spectrum because of its large neutronic advantage over the thermal one, and the reduced production of higher actinides.

Technical issues

High intensity proton accelerators are operational at Los Alamos National Laboratories (USA) and at the Paul Scherrer Institute in Switzerland. The LANSCE is a 1 MW linear accelerator while the PSI accelerator is a 1 MW cyclotron.

In the high-intensity accelerator, beam loss should be minimised to avoid resultant damage and activation of accelerator hardware. For industrial applications, primary issues are the stability, efficiency, reliability, operability and maintainability of the accelerator. Recent reviews of the operation reliability of accelerators have shown that beam trips and proton source failures are very frequent. Substantial improvement is required and R&D priority should therefore be given to that essential part of ADS. Further R&D and operational experience will increase the dependability. Reducing the cost and size of the accelerator should also be included in the design priorities.

Lead and lead-bismuth spallation targets are presently being considered in design studies. At present most of the available data and codes are sufficient for approximate evaluations. But uncertainties in the prediction of spallation products and neutron multiplication factors should be reduced in order to allow detailed technical design studies.

Trips and fluctuations of the incident proton beam are inevitable, causing thermal shocks in the core components. Therefore the design must take this into consideration: power distribution, effective neutron multiplication factors, the neutron flux shape transient response and the size of the system.

The accelerator-driven subcritical system has clear safety advantages for severe reactivity accidents. It can cope with fast ramp rate accidents which could occur too rapidly for scram systems in critical reactors. A margin to accommodate fast reactivity insertions is important to avoid supercriticality accidents.

The consequences of cooling failure for ADSs are similar to critical reactors. A reliable beam shut-off system is, therefore, required for an ADS, just as a reliable scram system is required for a critical reactor. A reliable emergency decay heat removal system is required for both.

Performance assessment studies

Over the past few years a number of different ADS concepts have been developed. The most attractive are those where a fast neutron flux is being used. Two studies are commented in the OECD-NEA report; one by FZK, Karlsruhe and one by JAERI, Tokai.

In the FZK study the sodium cooled, fast spectrum ADS employs a very high power accelerator of up to about 200 *mA* of proton beam with an energy of 1.6 *GeV*. The spallation target is MAO_2 or PuO_2 fuel itself. The proton beam is spread over the entire surface of the target to avoid the hot-spot problem. In this scenario, three different cores are required; the first one is for the incineration of MA from 14 units of LWRs, the second core is loaded with ^{99}Tc and ^{129}I and plutonium produced in 19 LWRs. ^{129}I eventually left over from this core is transmuted in the third core, fuelled by a fraction of Pu coming out of the second core. It was shown that the considerable power swings between the BOC (k_{eff} = 0.95) and the EOC (keff = 0.80) stages could be smoothed out by adjusting the beam current. A technical difficulty of this concept is the use of a spallation target of MAO_2 or PuO_2 and the use of spread proton beam.

JAERI designed two-types of subcritical cores of 820 *MWt* with nitride fuels. One type is a Na-cooled core with a solid tungsten (W) target at the core centre and the other is lead-bismuth target and coolant system. The spallation targets are bombarded by high energy and power protons of 1.5 *GeV* and 45 *mA*. The parametric studies were conducted to obtain the optimal neutronic characteristics of the subcritical core to maximise the MA transmutation rate and to minimise the burnup reactivity swing during irradiation by adjusting the MA and Pu fraction of nitride fuels with ZrN inert matrix as thermal diluent. The coolant void reactivity is negative in the lead-bismuth cooled core, though it is positive in the Na-cooled one. The MA transmutations in both cores are 250 *kg/y* and this amount corresponds to the annual production of MA in about 10 LWRs.

Transmutation of long-lived fission products (LLFP)

The transmutation capacity for long-lived fission products in conventional reactors is very limited, and these neutron-absorbing substances tend to poison the core. The reactor neutron balance makes it conceivable to recycle some but certainly not all of them. The transmutation yield is very low because the cross sections are low, but the presence of other, e.g. inactive isotopes in the target may result in the production of LLFP and interfere with the depletion reactions.

Only the irradiation of ^{99}Tc and ^{129}I in standard PWRs has been considered in calculations with these fission products located in special target pins and inserted into the guide tubes of the PWR assembly. ^{99}Tc was assumed in metallic form, iodine was considered to be in the form of cerium iodide (CeI_3) or sodium-iodide (NaI). The stability of the iodine target during the irradiation is the most important bottle-neck in the search for high transmutation yields and conversely shorter irradiation half-lives.

Transmutation of ^{99}Tc in fast reactors may be accomplished in several ways: in a special moderated sub-assembly loaded at the periphery of the core or in the inner core, or in a non-moderated subassembly loaded in the core. Moderation could be realised with a material like CaH_2. Table 4 gives for ^{99}Tc some calculated annual transmutation rates in LWRs and FRs, together with the radionuclide inventories and the irradiation half-lives.

As a general conclusion it can be postulated that **transmutation of ^{99}Tc or ^{129}I in present reactors is not encouraging** because of the long transmutation half-lives and the huge inventories of fission products required. Special-purpose high flux reactors or ADS systems may perhaps improve the prospects.

Table 4. **Ranking of reactors with respect to ^{99}Tc transmutation capability**

Reactor	Configuration	Inventory ^{99}Tc (kg)	Transmutation ^{99}Tc (kg/y)	Transmutation ^{99}Tc (kg/MWe y)	Irradiation Half-life(*) (y)
FR	Moderated S/A in inner core	2 741	122	0.11	15
FR	Non-moderated S/A in inner core	2 662	101	0.09	18
LWR	Pin in guide tube UO$_2$ fuel	3 633	64	0.07	39
LWR	Pin in guide tube MOX fuel	1 907	17	0.02	77

(*) Irradiation half life $T_{1/2} = \ln 2 / \sigma \varphi$

Impact of P&T on risk assessment and waste management

Radiological risk assessment

The radiological risk associated with the long-term waste management is a very complex issue which involves the radiotoxic inventory of the waste or the spent fuel, the confinement conditions of the radionuclides during the disposal period and the expected collective dose to man resulting from the release into the biosphere over a very long time interval.

It is recommended to abandon the term 'potential radiotoxicity', in speaking of nuclear wastes, because it gives the illusion of a management scenario for these wastes, whereas it is merely an inventory. It is preferable to express for each radionuclide the inventories in *Bq/t* of fuel (or of heavy metal it contains) or in *Bq/TWhe*.

Radiotoxic inventory of wastes

The general strategy of introducing P&T as an additional waste management option is based on the radiological benefit which is expected from such an option. The ranking of the actinides and long lived fission products can be made on the comparison of their intrinsic hazard (effective dose coefficients, *Sv/Bq*) coupled to their radioactive concentrations in spent fuel or HLW (*Bq/tHM*), as a function of time.

In the case of the OTC all radionuclides contribute to the source term and the long term radiotoxic inventory is mostly due to Pu, MAs and some LLFPs. However, the conditioning operations can provide artificial barriers which are potentially capable of confining the radionuclides within their package for thousands of years. After this time interval nothing can be predicted except that the solubility of the actinides (except Np) is generally very low whereas the long-lived fission products, particularly ^{135}Cs, ^{129}I and in some cases ^{99}Tc, display high mobilities in the geosphere.

In the case of the RFC, the reprocessing operation and recycling of Pu as LWR-MOX significantly reduces the radiotoxic inventory of the HLW. In this case the long-term radiotoxicity is essentially determined by the minor actinides (Np, Am and Cm,) and the long-lived fission products ^{99}Tc, ^{126}Sn, ^{79}Se and ^{135}Cs. An important long-lived fission product ^{129}I does not appear in this list since it is discharged into the ocean as a consequence of the reprocessing operations. Between 100 and 10 000 years the most important radiotoxicity contribution in the HLW comes from Am and Cm isotopes. Beyond that period both isotopes have significantly decayed to ^{239,240}Pu (daughters of ^{243,244}Cm) and ^{237}Np (daughter of ^{241}Am) which becomes predominant.

The radiotoxic inventory of spent MOX fuel is much higher since the Pu content of seven spent UO_2 assemblies is "concentrated" in one MOX fuel element, the "actinide" radiotoxic inventory of a spent MOX fuel element is about eight times that of spent UO_2 fuel elements.

In a long-term perspective of waste management the disposal of spent MOX fuel is a major factor in the overall assessment of the radiotoxic inventory in the RFC fuel cycle. Reprocessing of spent MOX fuel and recycling of recovered plutonium are therefore important issues which determine the long term radiotoxicity.

The main impact of the AFC strategy is a significant reduction in the radiotoxic inventory of the vitrified wastes with conversely a transfer of the long-lived actinides to the fuel cycle facilities and reactor core inventories. This long-term benefit has to be weighed against the short term doses to workers, and the production of additional contaminated wastes, due to the increased complexity of the fuel cycle

P&T facilities for TRU reduction

In order to reduce the overall radiotoxic inventory of a 100 GWe reactor park it is important to emphasise the additional nuclear facilities which have to be made available.

Fuel cycle facilities

If the P&T strategy intends to improve the radiotoxic inventory situation on world-wide scale, additional industrial reprocessing plants totalling 3500 *tHM/y* should be built and operated.

However, in a first phase only the necessary chemical facilities for separation of MAs from HLLW have to be designed and constructed around existing reprocessing plants.

A 100 *GWe* park will discharge annually 1.54 *t* Np, 1.56 *t* Am, and last but not least 94-180 *kg* of Cm. The corresponding fuel fabrication outputs are: 60 *tHM* U-Np fuel and 60 *tHM* U-Pu-Am fuel with 2.5% MA concentration. Provisionally a storage capacity of 3 to 5 *t* Cm would be needed to cover, e.g. a 30 year production period. All these facilities would preferably be constructed on the reprocessing site in order to minimise transportation of these highly active concentrates. After conditioning and encapsulation a transfer to a dedicated reactor site for transmutation should in principle be possible. However, if multi-recycling of the targets is required, either the irradiation facilities should be installed near the existing processing units or dedicated (e.g. pyrochemical) reprocessing facilities should be erected near the reactor buildings.

Recycling scenarios in a power reactor grid

Research conducted in recent years has helped to clarify the feasibility of recycling actinides (mainly Am and Np) in the different types of reactor (FR, PWR) and in different recycle modes (homogeneous or heterogeneous).

Plutonium recycling and multi-recycling have been thoroughly studied. However, to reduce the overall radiotoxic inventory, it has been emphasised that any Pu multi-recycling strategy must consider the essential goal of not increasing the production of minor actinides. Theoretically, the advantage of FRs seems obvious.

According to present views and insights, a scenario with simultaneous management of plutonium and MAs seems possible in order to achieve equilibrium between production and consumption of Pu, Am, Np and Cm within the reactors. The radiotoxic inventory of the wastes generated by advanced reprocessing of these reactor fuels would significantly be reduced. The overall losses in the different reprocessing and partitioning operations would represent the minimum radiotoxic inventory of the waste resulting from the nuclear electricity generation. Several reactor park compositions with balanced reactor types can be considered.

The following reactor park compositions have been proposed in this study:

- THERM   100% PWRs, some of which are used for the multi-recycling of Pu and MAs.

- MIX 1   70% PWR-UO$_2$ and 30% FR capacity.

- MIX 2   70% PWR-UO$_2$, 10% PWR-MOX, 20% FR capacity.

- FAST   100% FRs of the EFR type.

Since these reactor systems all have the same installed capacity, they are said to be in equilibrium in two respects.

1. The characteristics of these systems (proportions of the different reactors, actinide contents etc.) are determined so that equilibrium exists between actinide production and consumption. In the reactors, the actinide inventory remains unchanged. Only the losses in the different cycle operations (fabrication, reprocessing) represent the ultimate wastes intended for disposal.

2. However, since the PWR MOX and FBuR incinerator reactors cannot consume all the actinides loaded in a single passage, this obviously implies a change in the isotopic composition of the fuel during multi-recycling.

Isotopic equilibrium is reached after about five to six cycles, and considering the inter-cycle times (cooling, reprocessing, fabrication), this corresponds to at least 50-70 years depending on the intercycle cooling time. Nevertheless, it must be emphasised that mass equilibrium between actinide production and consumption is always achieved in the first five cycles and before isotopic equilibrium is reached.

The reactor system consisting exclusively of PWRs (THERM) has the following features:

- The recycling of MAs must be dissociated from that of Pu, with some of the PWR reactors recycling Pu in MOX and others dedicated to recycling the MAs.

The characteristics of the other types of reactor system considered are as follows:

- All the PWRs and FRs have an electrical capacity of 1 450 *MWe*. The burnup of PWR UO$_2$ is 55 GWd/tHM.

- For the MIX 1 and MIX 2 scenarios, the FR cores correspond to the CAPRA reference oxide, with initial enrichment of 45% and core residence time of 855 EFPD.

- For the FAST scenario, the cores correspond to the reference EFR with zero regeneration gain (configuration with two axial blankets of 5 *cm* and one ring of radial fertile assemblies), with initial enrichment of about 20% and core residence time of 1 700 EFPD.

Neptunium is recycled homogeneously and mixed with the fuel in the core. Americium is recycled heterogeneously in the form of targets placed in the first peripheral ring of the core.

Curium is assumed to be stored for a century to allow the ^{243}Cm and ^{244}Cm to decay to Pu. This Pu is then recycled with the standard Pu flux. The residual curium (essentially ^{245}Cm) is recycled with americium in the targets.

Only the losses during reprocessing and fuel cycle operations are considered with assumptions of 0.1% for Pu and 1% for MAs.

Results

The radiotoxic inventory, expressed in *Sv/TWhe*, going to the wastes, and their changes over time are graphically shown for the different scenarios, and are compared with the radiotoxic inventory resulting from a reactor system with the same capacity consisting exclusively of PWR UO_2 managed in open cycle, i.e. with all the irradiated fuel discharged in the wastes. The reduction of radiotoxic inventory in the wastes are found to be similar for all the scenarios and close to a factor of 100.

Table 5**. Radiotoxic inventory reduction factor due to MA recycling as a function of disposal times in comparison OTC option**

Time (years)	100	1 000	10 000	100 000	1 000 000
MIX 1	74	72	77	130	82
MIX 2	71	75	84	140	80
FAST	110	100	100	150	150

If Curium is not recycled, which is the present proposal, the reduction factors become much lower and range between 7 and 24 during the first 10 000 years. Beyond that period, the daughter nuclide of ^{244}Cm, ^{240}Pu has decayed and the reduction factors become more favourable.

One important result of all the scenarios examined is that the Pu+MA inventory in the cycle can be stabilised. but the actinide mass inventory in the reactors increases steeply from 580 kg/GWe for the THERM case to 3184 kg/GWe for MIX1, 3 289 kg/GWe for MIX2 to reach 6721 kg/GWe for the FAST case.

Risk and hazard assessment over time

While the radiotoxic inventory is a physical-biological concept intrinsically based on the laws of radioactive decay and the radiological damage due to a quantity of radioactivity incorporated in the human body, the risk and hazard concepts on the contrary rely on the extent of conditioning and packaging of waste streams, on the long-term behaviour of waste packages in geological media and on the routes which could be followed by radioactive releases on their return to the biosphere and to mankind.

- In a first phase one has to assess the direct radiological impact of additional fuel cycle operations.

- The second step is the radiological assessment of the waste types created by the AFC operations.

- Finally the hazard assessment over time is closely related to the repository design and location.

Direct radiological impact of nuclear power production

The statistical data on the collective dose to the workers suggest that compared with the RFC, the AFC with P&T would only induce a moderate increase of the dose to man in the fuel cycle activities, and particularly to those in fuel and target fabrication. However, appropriate measures must be taken to reinforce shielding, especially against neutrons, throughout the entire recycling facilities, and this will significantly increase the overall investment cost.

In the OTC scenario, uranium mining and milling constitutes 90% of the total regional dose. In the RFC scenario, the impact of reprocessing on the regional dose amounts to 10% in the UNSCEAR study and 28% in the Cogéma study. If the AFC should become an industrial reality in those countries having a reprocessing infrastructure, the contribution of the MAs and FPs separation and conditioning operations would slightly increase the overall radiological impact on the environment but not modify the overall picture.

Impact of P&T on waste management decisions

The potential impact of P&T is an improvement of the long-term hazard but it requires additional actinide handling facilities and does not eliminate the necessity of geological disposal.

- Without reprocessing, P&T cannot be implemented.

- Partitioning of MAs from HLLW is possibly a first step in the gradual decrease of the radiotoxic inventory of vitrified HLW.

- Long-term storage of partitioned MAs waste fractions and long lived fission products will be necessary since special reactors have to be developed for transmutation.

- Partially "incinerated" or "deactivated" separated actinide or fission product targets will need special preparation and conditioning before disposal.

- The risk of contaminating the geosphere will be decreased if the conditioning of the toxic radionuclides is improved (e.g. by using ceramic matrices or improved glass compositions for the separated MAs).

- The fraction of radionuclides involved in fuel cycle and waste management activities will shift from mainly disposed materials in the OTC to mostly stored inventories as shown in Table 6. Compared to these reactor and facility inventories, the weight of waste discharged annually by a reactor park of 100 GWe-0.8 year (= 700 TWhe) would be drastically decreased in comparison with the OTC as shown in Table 7.

Table 6. **Mass of transuranic elements in reactor park (tHM/100 GWe)**

Elements	UO_2	MIX1	MIX2	FAST
Np	2.9	7.3	7.7	3.3
Pu	52.2	249.2	257	620
Am	2.3	54	56.1	41.6
Cm	0.6	7.9	8.1	6.8
Total	58	318.4	328.9	671.7

Table 7. **Annual discharge of TRU wastes (tHM/100 GWe-0.8 Year)**

Elements	UO_2	MIX 1	MIX 2	FAST
Np	1.155	0.021	0.022	0.0056
Pu	22.78	0.076	0.056	0.111
Am	1.152	0.075	0.0798	0.0354
Cm	0.126	0.042	0.040	0.018
Total/year	25.21	0.214	0.198	0.17
Total/30 year	756	6.42	5.94	5.1

Impact of P&T on waste characteristics and management

In the OTC option with a continuous nuclear power production of 320 *GWe*, additional large repositories will be necessary for a single type of "waste" form. If the maximum content of a repository is 70 000 *tHM* spent fuel like that of Yucca Mountain in USA, then without any reprocessing, every 13 years a new repository of that size or equivalent will have to be constructed. With partial reprocessing (3 900 *tHM/y*), the interval is increased from 13 to 25 years and vitrified HLW together with Medium Level Waste forms will have to be disposed of. The repositories will have to be adapted to the nature of the waste types and the size of the national or the regional nuclear power capacity.

In an AFC strategy the separation of Pu, MAs and LLFP will create a multiplicity of waste forms which could be conditioned according to the required chemical or metallurgical properties. A better conditioning will decrease the long-term risk and will allow in the mean time the development of transmutation technologies which are the only route to long term hazard reduction.

In the MIX 1, MIX 2 and FAST options, the equilibrium TRU inventory in the reactor and nuclear facilities amounts to 98% of the total amount of actinides involved in the fuel cycle, only one percent is discharged as waste. However, this strategic option implies the replacement of 20% or more of the present LWRs by FRs, and a complete recycle of recovered plutonium. Apart from the construction of dedicated transmutation and incineration facilities, this option also implies the construction of specialised fuel fabrication facilities capable of processing 1 490 *tHM* UO_2 fuel, 236 *tHM* LWR-MOX fuel, and last but not least the construction of 20 FRs with an actinide throughput of about 100 *tHM* per year.

Conclusions

- Fundamental R&D for the implementation of P&T needs long lead times and requires large investments in dedicated fast neutron spectrum devices (FRs, ADS which may complement or substitute for operating LWRs), extension of reprocessing plants and construction of remotely manipulated fuel and target fabrication plants.

- Partitioning facilities for MAs and some long-lived fission products could be designed and constructed as extensions to the existing reprocessing plants.

- Partitioning methods for long-lived radiotoxic elements have been developed on a laboratory scale but much work is still to be done on improving and scaling up the partitioning technology in order to make it compatible with industrial reprocessing practices.

- The short-term impact of partitioning would be to reduce long-term radiotoxic inventory of the resulting HLW at the expense of an increase of the operational requirements for the nuclear facilities concerned.

- Fast Neutron-spectrum devices (dedicated FR or ADS facilities) are more efficient than current LWRs for recycling and transmuting long-lived radionuclides. The ADS might play an important role in "incinerating" the actinides and some FPs from the spent LWR-MOX or FR-MOX fuel in order to reduce the long-term radiotoxic inventory of the wastes.

- Recycling of Pu+MAs in a composite reactor park could stabilise the TRU inventory of an electro-nuclear park. Multiple recycling of TRUs is a long-term venture which may take decades to reach equilibrium TRU inventories.

- Conditioning of separated long-lived nuclides in appropriate matrices which are much less soluble than glass in geological media, or which could serve as irradiation matrix in a delayed transmutation option, is a possible outcome for the next decades.

- P&T will not replace the need for appropriate geological disposal of HLW, irradiated TRU concentrates and residual spent fuel loads from a composite reactor park.

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