

ROLE AND INFLUENCE OF PARTITIONING AND TRANSMUTATION ON THE MANAGEMENT OF NUCLEAR WASTE STREAMS

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ABSTRACT

The report provides a global analysis of the present situation in the backend of the fuel cycle and emphasizes the expansion of the reprocessing capacity as a prerequisite for P&T. The radioactive source term with long term implications is discussed on the basis of the isotopic hazard indexes and their evolution with time. The Pu-241, Am-241, Np-237 is the sequence with greatest radiological impact.

The feasibility of the partitioning is analyzed on the basis of present day technology of aqueous and dry reprocessing methods. Rerouting of Np and separation of Am are the first objectives to be reached. The long lived fission products I-129 and Tc-99 determine to a great extent the long term dose to man and have to be examined together with the actinides.

The last and most challenging aspect of P&T is transmutation which covers two distinct concepts: transmutation to a short lived nuclide or incineration of actinides into fission products. The technical feasibility and limitations of each of the transmutation devices (reactors, accelerators) are discussed.

INTRODUCTION

The discussion on the validity of P&T as an alternative waste management option is based on a more extensive report (1) prepared for the Nuclear Development Division of OECD-NEA.

The present analysis starts from the current situation in the fuel cycle as practised by the major nuclear countries in the world. Worldwide, nuclear electric capacity amounts to 340 GWe and produces a total spent fuel output of roughly 9000 THM/year.

The fate of the spent fuel depends to a great extent on the national fuel cycle policy. For one half of the worlds' capacity of nuclear power plants, the back-end of the fuel cycle relies on reprocessing, plutonium recovery, vitrification of residues and disposal of wastes.

Among the countries following this course are France, UK, Japan, Germany, Belgium, Switzerland, the CIS and the countries of its former alliance.

The other half of the worlds' nuclear capacity produces spent fuel as a waste product. The so-called "once-through-cycle" is being pursued in the US, Canada, Sweden, Spain and some other countries. The long term above ground storage is presently the main trend as well for spent fuel as for HLW.

Storage is to be followed by disposal in a suitable geologic formation.

Since P&T is a strategy to reduce the very long term impact of long lived actinides and fission products, it implies that the total output of spent fuel (and the backlog) ought to be reprocessed in order to provide a technical basis for partitioning the most radiotoxic and long-lived nuclides.

Table I shows the existing or planned reprocessing capacity of 3800 THM and the increment required to provide a worldwide P&T technology basis. Since nearly all the spent fuel discharged from LWR's is made of UO₂, the currently available PUREX reprocessing process is the only one capable of implementing this gap of 4200 THM per year. Only if this additional reprocessing capacity is made available, the nuclear community will be ready for a comprehensive P&T strategy.

Dry reprocessing techniques will play a role in a later phase of P&T when recycling of e.g. metal fuel in fast reactors can be realized.

If this option is not entirely adopted by all countries for whatever political or public acceptance reasons, the benefit of P&T will be curtailed. In that case, the P&T option cannot fulfil its promises for the very long lived fission products (I-129, Tc-99, ...) and Np-237, which have a global circulation

TABLE I
Fuel Cycle Options

Option	Group	GWe	THM/y (*)	Repro. Capacity (**)	Increment For P & T
Reprocessing	OECD	134	3620	3800	-
HLW Disposal	NON OECD	47	1270	(1000)	270
Once through	OECD	137	3700	-	3700
Direct disposal	NON OECD	18	486	-	486
	WORLD	337	9076	4800	4276

(*) 27 THM/GWe - 0.75 y

(**) UP.2, UP.3, THORP, ROKKASHO, TOKAI (RUSSIA)

pattern independent of the technical lifetime of a geologic repository.

However, on local and regional scale, P&T might decrease the nuclide inventory of actinides and fission products and decrease radiological impact on the biosphere or the time interval during which the source term decays to lower activity levels. Whatever the future fuel cycle policy, it is not reasonable to expect an overall inventory reduction exceeding a factor of 25, although locally a reduction with a factor of 100 may be pursued.

HAZARD RANKING OF ACTINIDES AND F.P.

The selection of actinides and long-lived fission products, which are beneficial to eliminate by transmutation, can be made on the comparison of their hazard factors. The volume of drinking water (or groundwater), necessary to reduce the radioactivity content of one tonne heavy metal to drinking water limits, is a measure of the radiotoxicity. By using the ratio between the radioactivity content of HLW or spent fuel, and the Annual Limits of Intake (ALI's) $\times 10^{-4}$, a ranking is obtained which differs according to the ALI's proposed by ICRP 30 or ICRP 61.

Table II shows the data for actinides and fission products.

During the first thousand years, Am-241 is the dominant radiotoxic nuclide followed by Am-243 in HLW and Pu-240-239-242 in spent fuel.

During the period from 1000 to 200.000 years, the mixture of Pu isotopes is the dominant radiotoxic source term.

Beyond that period, Np-237 and long-lived fission products are determining the radiologic impact on the global biosphere.

Depending on the local hydrogeologic conditions, I-129 or Tc-99 are the main radiotoxic fission products to be taken into consideration.

C-14, Sn-126, Cs-135 and Zr-93 are nuclides which have to be assessed for each hydrogeologic situation individually.

During the first 600 years, the radiological impact is dominated by the main fission products Cs-137 and Sr-90 and by Am-241 as most important actinide. From technological point of view, both Cs-137 and Sr-90 are the most important nuclides since they determine nearly the total heat output of HLW and spent fuel.

Transmutation of these fission products does not seem feasible according to the present state of the art. Total confinement of HLW and spent fuel during the first millennium must therefore be postulated as the only viable waste management option.

TECHNICAL PARTITIONING TARGETS AND THEIR IMPLEMENTATION

In order to set a scientific frame for the targets to be achieved, there are a number of approaches which can be followed:

- From environmental point of view reduce the Minor Actinides (M.A.) and Fission Product (F.P.) concentrations in the HLW residues in order to reach a concentration level equivalent to HLW with a global M.A. concentration of 100 nCi/g after 1000 years.

Under these hypothesis, the amount of radioactivity stored, does not constitute a nuclear hazard beyond that period of human control.

Table III shows the data per nuclide.

TABLE II
Hazard Ranking of Actinides and Fission Products in HLW

Actinides					
	ICRP-30	I/THM		ICRP-61	I/THM
1	Am-241	2 - 7 10^{13}	1	Am-241	2.3 - 12.3 10^{13}
2	Am-243	1.1 10^{12}	2	Am-243	1.8 - 2 10^{12}
3	Np-237	4.7 - 6.4 10^{11}	3 (1 %)	Pu-240	7.6 - 8 10^{11}
4 (1 %)	Pu-240	1.5 10^{11}	4 (1 %)	Pu-239	3.2 - 3.5 10^{11}
5 (1 %)	Pu-239	6.4 - 6.9 10^{10}	5	Np-237	4.7 - 6.4 10^{10}
6	Cm-246	1.4 - 1.6 10^{10}	6	Cm-246	2.4 - 2.7 10^{10}
Fission Products					
		Nuclide			I/THM
	1	Sr-90			3.9 10^{12} - 2.13 10^4
	2	Cs-137			3.8 10^{12} - 3.66 10^4
(1)	3	Tc-99			1.6 10^9
(2)	4	Sn-126			6.6 10^8
(3)	5	I-129			5.8 10^8
(4)	6	Cs-135			1.3 10^8

TABLE III
Actinide Concentrations and Target DF's for HLW after 1.000 and 10.000 years

Nuclide	Actinide concentration Ci/THM		Decontamination factors based on 10 nCi/g	
	1.000 y	10.000 y	1.000 y	10.000 y
U-234	0.0198	0.0195	5	5
Np-237	0.524	0.578	130	144
Np-239	15.2	6.54	$3.8 \cdot 10^3$	$1.6 \cdot 10^3$
Pu-239	3.77	5.12	$9.42 \cdot 10^2$	$1.28 \cdot 10^3$
Pu-240	8.30	3.20	2.10^3	8.10^2
Pu-241	0.128	0.0612	32	15.3
Pu-242	0.0192	0.0206	4.8	5.1
Am-241	273.0	0.0614	$6.8 \cdot 10^4$	15.3
Am-242	0.04	-	10	-
Am-243	15.2	6.54	$3.8 \cdot 10^3$	$1.6 \cdot 10^3$
Cm-242	$3.34 \cdot 10^{-2}$	-	8.3	-
Cm-245	0.127	0.0611	31.7	15.27
Cm-236	0.194	0.052	48.5	13.0
TOTAL	318	21		

If geological disposal is accepted for a period up to 10,000 years, the same approach leads to partitioning targets which are somewhat more relaxed.

- From radiochemical point of view, we have to take into account that the radioactive "series" contain interconnected nuclides which have to be considered globally in order to get a meaningful result e.g. Pu-241 - Am-241 - Np-237; Am-243 - Pu-239, ...
- From nuclear technology point of view, some minor actinides with a short half life (especially Cm-244, $t_{1/2} = 18$ y) have important consequences for the industrial development of a partition strategy due to its intense heat and neutron emission, particularly during the fuel fabrication activities. The non-removal of Cm isotopes has a direct consequence on the long term target DF's values to aim at.

The technical implementation of partitioning involves a series of activities which have to be improved in the present reprocessing process:

- Due to the decrease of the Pu ALI's (ICRP 61), it would be advisable to reduce the residual Pu concentration in HLLW with a factor of 10 to 100. This can be done by increasing the extraction coefficient during the conventional reprocessing or by installation of an additional Pu separation rig. A further decrease in Pu concentration cannot be justified taking into account the Pu content of MLW and fuel fabrication residues.
- The rerouting of Np is a second step which could seriously improve the management of M.A. in the nuclear fuel cycle. By modifying slightly the U-Pu extraction process, it should be possible to quantitatively reroute the dispersed Np streams into one single product stream:

- the extraction raffinate from the 1st extraction column, or
- the Pu product stream.

The raffinate from the first extraction column of the PUREX process is the starting point of the HLLW stream, which can serve as a source term for new partitioning techniques.

- The extraction of Tc-99 from the U-Pu-TBP extractant phase of the first extraction column and its recycling to the insoluble residue fraction in the head-end step of reprocessing, would be beneficial to reduce the subsequent Tc-99 contamination of the U/Pu product streams. Tc-99 interferes with the normal enrichment steps.

Partitioning of M.A. and F.P. requires additional separation technologies to be installed in or around the fuel reprocessing plants.

- The HLLW stored in cooled tanks constitutes the source for M.A. and most F.P. The recently developed TRUEX process, combined with the HDEHP process are suited for extraction of Pu and M.A. from 4 M HNO₃ HLLW solutions. See Fig. 1. This M.A. stream does not contain interfering Rare Earths amounts and produces separate Pu, Np and Am-Cm fractions. If Cm has to be separated from Am, an additional separation process has to be added.
- Iodine is presently removed from the fuel processing streams but discharged into the ocean. In order to perform a real partitioning process, this element ought to be precipitated from the MLW streams and transformed into a chemical form suitable for intermediate storage and/or into a transmutation target.

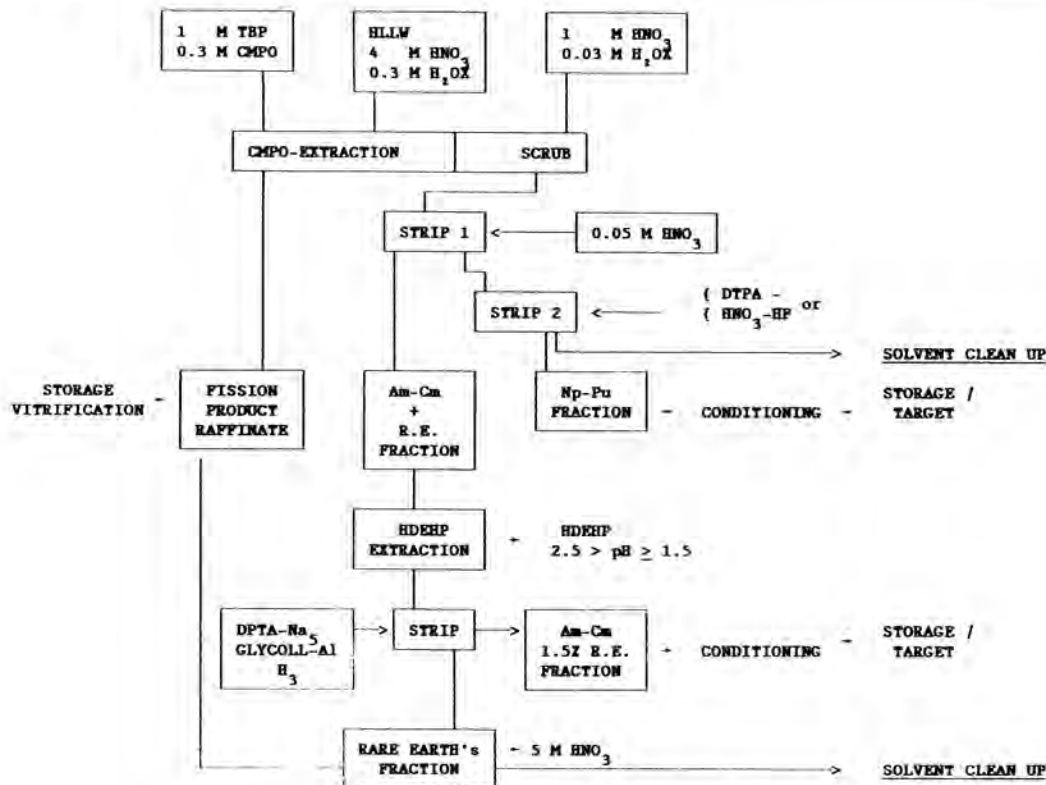


Fig. 1. Combined TRUEX-HDEHP-Process.

- Technetium is for 50 % found in the insoluble residues and for 50 % in the reprocessing feed solution. By rerouting Tc from these two separate streams into one "platinum metal" fraction, a separate treatment can be developed after a prolonged storage. Indeed, the Ru-Rh-Pd isotopes decay within 50 years to a very weakly active resource from which it is possible to extract economically attractive nuclides from otherwise lost resources which constitute 30 % of the known world reserves in platinum elements. This new development is preferable to Tc incorporation into HLW glass for final storage unless the geological repository can provide a perfect confinement for millions of years. Moreover, large concentrations of platinum metals are detrimental to the long term stability of waste glass.
- The radiological significance of miscellaneous long lived fission products e.g. C-14, Se-79, Zr-93 and Cs-135 has to be evaluated before concrete steps are undertaken to remove them from waste streams. In some cases e.g. Zr-93 and Cs-135, it is almost impossible to achieve this goal since it would involve isotopic separation from other isotopes.

The development of a partition technology is obviously the condition-sine-qua-non for P&T but it is also very useful for the future developments in waste management. The conditioning of waste forms is at present limited to vitrification for HLW and embedment into concrete or bitumen for MLW. Glass is an excellent matrix for fission products and is stable for one or a few millennia, a time interval which is more than sufficient for most F.P. to decay. By partitioning the HLW into F.P. and actinides a more suitable matrix might become avail-

able for the long term storage of actinides in case transmutation would show to be too difficult or too expensive.

RECYCLING OF M.A. FOR TRANSMUTATION

In order to assess the magnitude of the M.A. conditioning operations including : chemical purification, conversion to oxide, encapsulation and incorporation into fuel elements or irradiation targets, it is necessary to quantify the total inventory of M.A. produced by reprocessing. A total production of 1700 kg Np and 1500 kg Am-Cm would be expected from advanced reprocessing.

Table IV shows the production capability of M.A. in reprocessing plants as constructed or planned in the OECD countries. It has to be kept in mind that this capacity is roughly 40 % of the total output of M.A. in spent fuel worldwide.

Based on these macroscopic quantities, we can derive the required fuel fabrication or target production capacity to produce the recycled fuel. But it depends on the reactor or transmutation device how the M.A. will be recycled and at what concentration level in the reactor materials.

- Homogeneous LWR fuel

Recycling of M.A. in LWR's is technically possible for Np in MOX fuel. A fuel fabrication capacity of 70 THM ought to be developed for that purpose but the transformation of Np into Pu-238 and the negligible net transmutation yield of Np make this option less attractive from radiological and waste management point of view.

Homogeneous recycle of Am-Cm involves the quantitative separation of rare earths, the construction of a 70 THM/year fuel fabrication plant with full remote handling and remote maintenance equipment. The

TABLE IV

Production Capability of Minor Actinides in Reprocessing Plants as Constructed or Planned in OECD Countries

Nuclide	pecific yields	M.A. Annual production in kg/year		
	g/THM	La Hague UP2 + UP3	Sellafield THORP	Tokai (Rokkasho)
Np-237	437	700	524	131. (349)
Am (241 + 243)	380	608	456	114. (304)
Cm (242 - 247)	1.5	2.4	1.8	0.45 (1.2)
Resid. Pu (0.7 %)	65	104	78	19.5 (52)

presence of Cm-244 requires additional neutron shielding.

The transmutation of Am-Cm leads essentially to heavier nuclides with the same radiotoxic potential.

- Heterogeneous Recycle in LWR

Heterogeneous recycle in LWR's is an option which has not yet been investigated thoroughly because of the very stringent specifications imposed on nuclear fuel and the potential regulatory problems associated with these fuel targets in a conventional power plant.

- Fast Reactor Fuel

Recycle in fast reactors received from the beginning most of the attention because of the fissioning potential of even and uneven nuclides in a fast neutron spectrum. In principle, a fast reactor can use large amounts of M.A. and is not sensitive to Rare Earth poisoning. MOX fuel fabrication facilities for fast reactor fuel are standard industrial practice for non gamma emitting nuclides.

Homogeneous recycle of Np is in principle concentration-wise not limited but the residual concentration of Pu-238, present in the spent fuel, is the limiting factor because of the heat emission of Pu-238 (0.5 W/g) and its interference in the subsequent aqueous recycling processes.

Homogeneous recycle of Am-Cm with a non negligible RE contamination (e.g. 10 - 20 %) requires the industrial development of new fuel fabrication techniques. Sol Gel techniques were investigated in the past on laboratory scale and are not suited for mass production of fuel. However a combination of Sol Gel and Vibrocompaction may become a viable route which still needs extensive development in order to reach a throughput of 60 - 80 THM/y.

- The feasibility of heterogeneous recycling of Np and Am-Cm, in MOX fuel types has been demonstrated on the scale of individual fuel pins prepared at the TUI. Fuel containing 45 % Np and a mixture of 21 % Np + 18 % Am was prepared by Sol-Gel, followed by pellet pressing. One of the major advantages of heterogeneous recycling of M.A. is the possibility to produce fuel pins along a specially designed fuel fabrication process in tailor made, relatively low throughput, fuel fabrication facilities.

In order to handle the total M.A. output of Np resp. Am-Cm from the OECD reprocessing plants, an

annual fuel fabrication capacity of 4 T (Np + inert diluent) resp. 8 T (Np + Am(Cm) fuel + inert diluent) is sufficient to transform all the M.A. into fast fuel assemblies.

This option would of course be much more preferable to the dilution of the M.A. throughout the entire reactor fuel load.

- The latest development in M.A. fuel fabrication consists of the preparation of metallic fuel according to a pyro metallurgical method followed by direct injection casting. The use of large amounts of Cd metal and salts, to process the spent metal fuel as well as the production of a mixed radiotoxic-chemotoxic waste salt is a heavily mortgaged process to start with. It is premature to consider this option as proven but it is, up to now, the only one producing metal fuel which is a necessary substrate for special reactor systems such as the IFR and MABR reactor concepts which have a much higher transmutation potential than MOX fuelled fast reactors. Very much R&D will be required to bring this option up to the level of knowledge and confidence of the MOX fuel preparation.

M.A. TRANSMUTATION IN NUCLEAR REACTORS

The generic term "transmutation" covers two distinct processes: the transformation of a nuclide into a heavier species by neutron capture and the "incineration" which produces neutron induced fission products.

In thermal neutron spectra, the transmutation by neutron capture of M.A. is by far the predominant process which transforms the M.A. nuclides into fissionable species only after one or two neutron captures e.g. Np-237 into Pu-239, Am-241 into Am-242, Cm-244 into Cm-245 etc. . .

In a fast reactor spectrum all M.A. nuclides are fissionable but the capture/fission ratio is different for each element and dependent on the neutron energy. Fast reactors, with a metal fuel core, produce the highest yield of fission products. Nonetheless the capture process is still very important and not negligible, particularly for Np-237.

- Transmutation in LWR's is not advisable since the overall balance between the depletion and intrinsic formation of each nuclide is almost at equilibrium. The radiotoxic potential remains roughly constant but the only positive result for Np is the shortening of the half life from 2 M years for Np-237 to 80 years for Pu-238.

Transmutation of Np targets in High Flux reactors is an option to be pursued.

- Transmutation of M.A. in a F(B)R - MOX reactor is a much more attractive venture since 1 GWe fast reactor can transmute the output of 6 LWR's. However, the transmutation is also in this spectrum made up of 80 % capture and 20 % fission for Np-237 and Am-241 with a resulting buildup of Pu-238.

The main advantage of a F(B)R is its ability to accumulate within the reactor core substantial quantities of M.A. (720 kg Np and 710 kg Am) which correspond to the total annual output of a large reprocessing plant or 60 GWe LWR's.

The annual "incineration" capacity of a MOX F(B)R being low, a large installed capacity of 22 - 25 GWe would be required to eliminate the M.A. output from all the OECD reprocessing plants.

With a mean burnup of 12%, eight cycles of irradiation, cooling, reprocessing and fuel refabrication have to be realized in order to achieve this ambitious goal within a time span of 20 to 25 years depending on the duration of each step. The overall actinide depletion amounts to 96% if 0.5% of loss is to be expected at each recycle step. Theoretically a M.A. - D.F. of 25 is obtained in this way.

- Transmutation of M.A. in metal fuelled fast reactors, IFR's and MABR's, constitutes the most advanced reactor option available up to now. Due to the very high neutron energy (780 keV) beyond the fission threshold, fission becomes the main transmutation process without or with minimal production of transuranic elements from the core fuel.

The M.A. inventory of a 400 MWe - ABR is 2670 kg M.A. which corresponds to the annual output of 100 GWe - LWR's. In order to destroy this M.A. output a M-ABR capacity of 6.4 GWe should be constructed. This is 4 times less than with F(B)R - MOX reactors. However, this reactor type is only at the conceptual design stage and very much RD&D will be required to prove the validity of this option, particularly the pyrometallurgical reprocessing and the thermo-mechanical behaviour of the fuel in reactor conditions.

M.A. TRANSMUTATION IN ACCELERATOR DRIVEN SYSTEMS

When a beam of high energy protons (1.6 GeV) impinges on a heavy metal core (W, Pb, U, Actinides) neutrons, fission products, and spallation nuclides are formed. With beams ranging from 25 to 250 mA, a very high number of neutrons can be generated (up to 10^{20} n/s) and produce locally a very high neutron flux ($\leq 10^{16}$ n/cm² s.).

Three proton accelerator projects are presently under investigation:

- | | |
|---------------------|--|
| JAERI project: | 30 mA beam subcritical metal core - Na cooling. |
| PHOENIX project: | 100 mA beam subcritical MOX core - Na cooling. |
| LOS ALAMOS project: | 250 mA beam thermal neutron flux, D ₂ O moderated facility. |

The combination of a high energy physics machine, operating under extreme vacuum and a subcritical irradiation facility is undoubtedly a formidable technical challenge.

The construction and continuous operation of such high energy facilities is a first objective in itself. The connection between the accelerator and the spallation target is the most critical component of the system.

Many features of the subcritical assembly are identical to the fast reactor technology : fuel loading, Ar gas blanket, sodium loops etc... The burnup of the "spent" fuel before recycling should be driven at much higher levels than in an equivalent fast reactor to become competitive with a nuclear reactor concept since the investment costs will be much higher. Recycling of partially transmuted metal fuel requires the same pyrochemical reprocessing attachment as the IFR or MABR's.

The implementation of the PHOENIX project involves advanced reprocessing of spent LWR fuel, MOX fuel fabrication, specialized recycling facilities for MOX fuel. The outspoken potential of 2.6 THM M.A. per year is by any standards very large.

The LOS ALAMOS project is, apart from his size, the most complex system including the 250 mA LINAC, a molten lead spallation target, a molten salt loop for heat transfer and nuclide transport and a heavy water moderator.

It looks premature to consider this option before a few decades taking into account its fundamentally new character which relies on the most advanced high energy physics concepts, a molten salt reactor and a super high flux reactor. Such a facility is capable of transmuted any radionuclide of the table of isotopes from heavy actinides to fission products independent almost of the cross section. However even in this case the necessity occurred to incorporate also isotopic separation as a preparatory step to insure full section.

CONCLUSIONS

- P&T is according to this analysis not "the third option" for the backend of the fuel cycle, but is a potential improvement of the reference fuel cycle with reprocessing and disposal of HLW.
- A systematic analysis of P&T would be very useful to make a comparison of the three options : direct disposal, reprocessing & disposal, P&T; on the basis of a comprehensive data base, flowsheet analysis, fuel fabrication and transmutation scenario's in order to quantitatively assess the possible impact of P&T and its economic implication.
- P&T should in the first place aim at providing new answers for those nuclides which are not or not sufficiently confined by geologic disposal particularly the very long lived nuclides with global circulation.
- Partitioning as an advanced form of reprocessing (possibly delayed) could be useful to improve the conditioning of the waste packages for disposal or to serve as irradiation matrix for future transmutation or to build up a strategic resource for the future (noble metals).
- Transmutation in LWR's does not decrease the radiotoxic potential of actinides. Transmutation in F(B)R's is theoretically possible but requires a very high investment cost and will be subject to the same

public acceptance questions as any other fuel cycle activity.

- Transmutation in accelerator driven systems is a bright futuristic option which might become interesting for specific isotopic targets containing nuclides with very low cross section.

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