The Impact of Partitioning and Transmutation on the Risk Assesment of a Spent Nuclear Fuel

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I. INTRODUCTION

Nuclear power produces steadily a mass of spent fuel which contains a part from the short lived fission products, a significant amount of actinides and fission products with height toxicity and long half-lives. These nuclides constitute the long-term radiotoxic inventory which remains as a hazard far beyond human perception.

The reprocessing recycles most of the major actinides (Uranium and Plutonium), while the Minor Actinides (MA) (mainly Neptunium: Np, Americium: Am and Curium: Cm) with half lives up to 2 million years remain with the fission products which are vitrified before being buried in deep repositories. Partitioning of the minor actinides and some of the fission products is an efficient method to reduce the long term radiotoxicity of the residual waste components with a factor proportional to the separation yield. The improved minor actinide nuclides would be recycled into a fuel cycle activities and returned to the reactor inventory of fissile and fertile material for transmutation to short lived isotopes. Progressively the MAs and some long-lived fission product (LLFP) could be burn up out. This option would reduce the long term contamination hazard in the high-level waste and shorten the time interval necessary to keep the actinides containing wastes confined in a deep geologic repository. Partitioning and Transmutation (P&T) is, in principal, capable of reducing the radiotoxicity period, while a number of practical difficult remain to be surmounted.

II. RADIOTOXIC INVENTARY OF WASTE

The general strategy of introducing P&T as further 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 hazards (effective dose coefficients, Sv/Bq) joined with their radioactive concentration in spent fuel (Bq/tHM). Based on this criterion, the long term radiotoxic inventory depends on the source term which is determined by the type fuel (LWR-UO_2, LWR-MOX, FR-MOX), the burn-up and the storage time (up to a million years). These fuels contain the actinides and the long lived fission products as major radiotoxic constituents.

The effective dose coefficients (FD_{RN}) for the most important actinides are given in Table II.1 [1]

Element	Nuclide	Sv/Bq
Uranium	^{235}U	4.6×10^{-8}
	^{238}U	4.4×10^{-7}
Neptunium	^{237}Np	1.1×10^{-7}
Plutonium	^{238}Pu	2.3×10^{-7}
	^{239}Pu, ^{240}Pu	2.5×10^{-7}
Americium	^{241}Am, ^{243}Am	2.0×10^{-7}
Curium	^{243}Cm	1.5×10^{-7}
	^{244}Cm	1.2×10^{-7}
	^{245}Cm, ^{246}Cm	2.1×10^{-7}

Table II.1. **Effective dose coefficients of actinides FD_{RN}**

The short-term radiotoxic inventory of some fission products is comparable to that of the actinides within a time horizon up to 100 years. After 300 years only the long-lived fission products remain radioactive (^{99}Tc, ^{93}Zr and ^{135}Cs) and constitute a radiotoxic inventory which is roughly 1000 time smaller than that of actinides [2]. ^{129}I in term of effective dose coefficients (Sv/Bq) comparable with the actinides but its radiochemical concentration in the spent fuels, expressed in Becquerel by metric ton of Heavy Metal (Bq/tHM), is much lower. The effective dose coefficients (FD_{RN}) for the most important actinides are given in Table II.2 [3].

Element	Nuclide	Sv/Bq
Strontium	^{90}Sr	3.4×10^{-10}
Zirconium	^{93}Zr	8.6×10^{-10}
Technetium	^{99}Tc	6.4×10^{-10}
Iodine	^{129}I	1.1×10^{-7}
Cesium	^{135}Cs	2.0×10^{-9}
	^{137}Cs	1.3×10^{-8}

Table II.2. **Effective dose coefficients of fission products FD_{RN}**

In the case of Once Throw Cycle (OTC) all radionuclides contribute to the source term and the long – term radiotoxic inventory is mostly due to Pu, MA and some LLFP. The conditioning operation can present artificial barriers which are potentially capable of confining the radionuclides within their package of thousand of years. After this time interval nothing can be predicted that the solubility of the actinides (except Np) is generally low whereas the long lived fission product, particularly ^{135}Cs, ^{129}I and in some case ^{99}Tc, display high mobility in the geosphere.

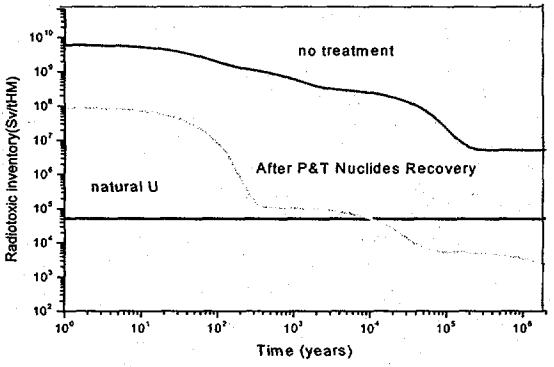
The waste management issues in case of an Advanced Fuel Cycle (AFC) with partitioning and transmutation scenarios are very different from the previous option. The impact of advanced reprocessing on the radiotoxic inventory of the High Level Waste in solidified form (HLW) is quite remarkable, since the actinides (U, Pu, Np, Am and Cm) are ideally removed from the HLW with a high separation factor. During the first 200 years the fission products are dominant and the radiotoxicity of the actinides is of the same degree as the Pu losses (~0.1%).

The main Impact of the AFC strategy is a significant reduction in the radiotoxic inventory of the vitrified waste with conversely a transfer of the long lived actinides to the fuel facilities and reactor core inventories. Comparing the radiotoxicity balances of the OTC and the AFC scenarios shows that the glass compositions become much more favorable. The radioactivity, although similar during the first few years, decreases by factor of 10 and 50 after 300 to 1000 years, respectively. The residual heat of the HLW becomes 3, 250 and 350 times lower after 100, 1000 and 10 000 years. This is the main potential contribution of the AFC to the waste management [4].

III. The benefit of Partitioning and transmutation operations on waste management:

Nuclear power produce a considerably a mount of spent fuel which contains, a part of short lived fission products and a significant amount of actinides and fission products with high toxicity and very long half-lives. These nuclides constitute the long term radiotoxic inventory which remains as a hazard far beyond human perception.

Partitioning of the minor actinides (Np, Am and Cm) and some selected fission products is a method which would reduce the long term radiotoxic of the residual waste components with a factor proportional to the separation yield Fig.1. The recovered minor actinides would be recycled into the fuel cycle activities and returned to the reactor inventory of fissile and fertile material for transmutation to short lived isotopes.



Recovery Rate ()	
U	99.9
Np	99.9
Pu	99.99
Am	99.99
Tc	99.9
I	99
Cs	99

Fig1. **The radiotoxicity after P&T nuclides recovery**

Gradually the minor actinides and some long-lived fission products would be burned out. This technique must reduce the long term contamination hazard in the high-level waste and shorten the time interval necessary to keep the actinides containing waste confined in the deep geologic repository. P&T is the most important option capable to decrease the radiotoxic period, although the technical and practical difficulties remain to be surmounted.

IV. Risk and hazard assessment over time:

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

The first positive Impact is the decrease in uranium mining requirements. It may be estimated that recycling Pu in LWR-MOX reduces the uranium needs by 20%. If the MAs were also recycled a maximum benefit if 25% could be expected [5].

Reprocessing process is a key technology for P&T strategy. Without reprocessing, a total amount of 50 t 60 t of plutonium and 7 to 8t of MAs have to be disposed of annually world-wide. From a long term radiotoxic point of view, reprocessing of spent nuclear fuel is a preferable strategy since it decrease the Pu amount in height level waste HLW. In the AFC scenario, the contribution of MAs and FPs separation and conditioning operations would slightly increase the overall radiological impact on the environment [6].

This impact can be limited if the appropriate separation plan are installed on the same sites as the large plants of the Light Water Rector (LWR) fuel reprocessing. The partitioning of MAs from height level liquid waste is the first step in the gradual decrease of the radiotoxic inventory of vitrified HLW. The long-term storage of partitioning MAs and long lived FPs will be necessary since special reactors have to be developed for transmutation.

V. Conclusion

Partitioning and transmutation of radioactive and long lived component from the highly radioactive waste stream in order to reduce or probably eliminate their radiotoxic inventory was the important option for the nuclear waste management. The principal radionuclides contribute to the long term radiotoxic inventory is mostly due to Pu, MA and some LLFP.

The conditioning operation can present artificial barriers which are potentially capable of confining the radionuclides within their package of thousand of years. After this time interval nothing can be predicted that the solubility of the actinides (except Np) is generally low whereas the long lived fission product, particularly ^{135}Cs, ^{129}I and in some case ^{99}Tc, display high mobility in the geosphere. Conditioning of separated long-lived nuclides in appropriate matrix which could serve as irradiation matrix in a delayed transmutation option is possible outcome for the next decades.

The general strategy of introducing Partitioning and Transmutation as an additional waste management option is based on the radiological benefit which is expected from such an option. 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 (FR or ADS facilities) are more efficient than current LWRs for recycling and transmuting long – lived radionuclides.

References:

[1] ICRP Publication 68, *Dose Coefficients for Intake of Radionuclides by Workers*, Vol. 20, N°4, 1994.

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[4] *Status and assessment report on actinides and fission product Partitioning and Transmutation.* NEA/PTS/DOC (98). OECD-NEA Paris November 1998. *Final report of Expert Group on Actinides and Fission Product Partitioning and Transmutation.*

[5] L.H. Baetslé, T. Wakabayashi, S. Sakurai, *Status and assessment report on actinides and fission product Partitioning and Transmutation.*, Proceeding of the fifth International Information Exchange Meeting , Mol, Belgium. 25-27 November 1998

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