Neutronics, Reactor Systems and Fuels for Transmutation

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The purpose of transmutation is to reduce the mass and the radiotoxicity inventories of Minor Actinides and Long-Lived Fission Products of nuclear waste. In France, the law voted in 1991 for waste management has requested in particular the study of solutions and processes on the subject of transmutation. This review gives conclusions, at the end of the 15 year research period defined by the law, on the scientific and technical feasibility of transmutation based on neutronic aspects, reactor systems and possible fuels, from the CEA point of view.

Important results are now available concerning the possibility of significantly reducing the quantity and the radiotoxicity of long-lived waste in association with a sustainable development of nuclear energy. As France has confirmed its long-term approach to nuclear energy, the most effective implementation of (recycling-)transmutation of Minor Actinides other than Plutonium and Uranium depends on the fast neutron GEN IV systems which are designed to recycle and manage their own actinides. The perspective to deploy a first series of such systems around 2040 supports the idea that progress is being made: the nuclear long-term waste would only be made up of Fission Products, whose radio toxicity considerably drops within a few hundred years. Future work will deal with pre industrial demonstrations of transmutation.

KEYWORDS: Transmutation, Minor Actinides, Neutronics, Fuels, Scenarios, GEN IV reactors, ADS

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I. Introduction

The production of nuclear energy in France has been associated, since its inception, with the optimization of radioactive waste management, including the partitioning and the recycling of recoverable energetic materials.
The public's concern regarding its long-term management made the French Government in 1990 to delay the implementation of the geological disposal and to prepare a law, passed on December 30, 1991, requesting in particular the study of solutions and processes for:

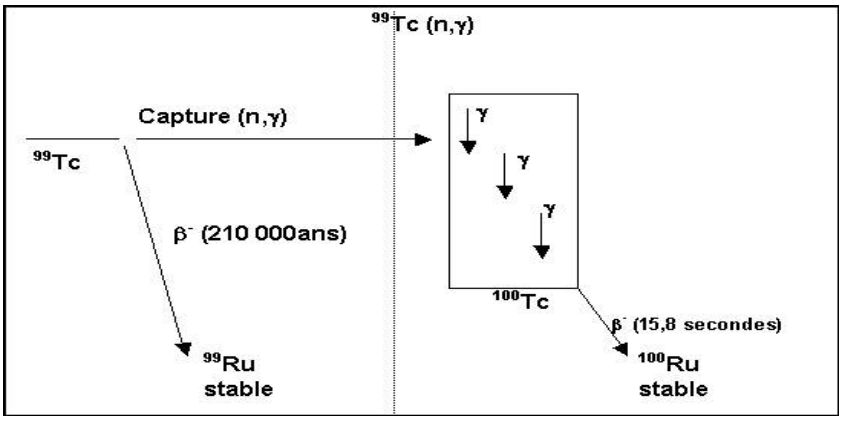
- minimizing the quantity and the hazardousness of waste, via partitioning and transmutation,
- either reversibly or irreversibly disposing the waste in deep geological formations,
- waste conditioning and long-term interim storage.

Studies on transmutation aim at changing the most radiotoxic long-lived elements present in the waste (Minor Actinides Americium, Curium and Neptunium and Long-Lived Fission Products LLFP Technetium 99, Iodine 129 and Cesium 135) through recycling in nuclear reactors into non-radioactive or shorter-lived elements. The transmutation of partitioned minor actinides MAs (americium, curium and neptunium) would reduce to a few hundred years the time necessary for the radiotoxicity of the vitrified waste to become similar to that contained in the natural uranium ore originally used.
Studies on transmutation, which were initiated before the 1991 Law, rapidly led to concluding that MAs transmutation was feasible in particular in fast neutron spectra. Results obtained confirm that the feasibility of transmutation is demonstrated, both in pressurized-water reactors (recycling and transmutation of plutonium, optionally but with more difficulty of americium and neptunium) and in advanced systems of nuclear-energy production (GEN IV fast-spectrum reactors, with recycling and transmutation of all heavy nuclides, uranium, plutonium, the minor actinides) or in dedicated incinerator reactors, either critical or sub critical.
Work on transmutation has been also focused on technical elements necessary for the demonstration of its technological feasibility. Irradiations for fuel transmutation studies have been carried out namely in the Phénix reactor in a very sustained way from 1995 and will continue until the shut down of the reactor in 2009.
We present in this paper an overlook of the French program and an update on the progress made by the research conducted on transmutation, in terms of neutronics, reactor systems and fuels.

2. Neutronics aspects

The interaction neutron-nucleus leads mainly to two types of reactions, the reaction of neutron capture by the target nucleus and the reaction of fission.
For the fission products, the capture in general makes it possible to generate, after successive transformations, a stable element. The typical example is the Technetium 99 (210000 years of period) which by neutron capture is transformed into technetium 100 of very short radioactive half-life 15,8 seconds) ending up by beta disintegration at stable ruthenium 100 (Fig.1).

Figure 1: Technetium 99 decay vs (n,γ) reaction



For Minor Actinides, the capture reaction is to be avoided because it results mainly in generating of other radioactive actinides, which generally does not achieve a decrease of radiotoxicity. For example, in, case of Am 241, the reaction of capture gives Am242 and mainly Cm242 (by beta disintegration). The half-live of this isotope is only of 163 days, but in fact the Cm 242 decay on Pu238 and so on up to stable Lead. The comparison of the radiotoxicity induced from Am241 and Cm242 (and their sons) shows that the overall result is not positive in terms of reduction of radiotoxicity.

It is obviously the fission which it is necessary to support for the destruction of Minor Actinides, because on the one hand fission leads to residues (Fission Products) with short life then stable, less radiotoxic in the long term than destroyed actinide, and on the other hand fission produces additional neutrons usable to destroy other waste or to take part in feeding reaction chain while producing energy. So the capacity to obtain a process of effective Minor Actinides transmutation depends on the competition between the two reactions of fission and neutron capture. The probability of occurrence of each reaction is characterized by the cross section of the isotope considered. These cross sections are quantified in Tab.1 where the values of capture to fission ratio of average cross sections integrated on the spectra of definite neutrons are indicated, representative as the distribution in energy of the neutron population present in the nuclear reactor: a "thermal" PWR spectrum with a UOX fuel, an "epithermal" PWR spectrum with a MOX fuel and a Na-cooled Fast Reactor spectrum.

Table 1: Capture to fission ratio of MA average cross sections

Isotope	PWR UOX	PWR MOX	FR
	$\alpha = \sigma c / \sigma f$	$\alpha = \sigma c / \sigma f$	$\alpha = \sigma c / \sigma f$
^{237}Np	**63**	**30**	**5,3**
^{241}Am	**100**	**45**	**7,4**
^{243}Am	**111**	**63**	**8,6**
^{244}Cm	**16**	**13**	**1,4**
^{245}Cm	0,15	0,2	0,18

Clearly, there is a great advantage for fast spectrum by comparison to thermal/epithermal neutron spectrums.

The evaluation of the cross sections alone, although a good indicator, is not sufficient. It is also necessary to consider what happened under irradiation taking into account others isotopes produced. For a standard irradiation time in PWR type and RNR reactors, the disappearance rates of the considered elements, expressed in percent of the mass presents initially, are indicated in Tab.2. The rates of fission, integrated over the irradiation time, are also indicated. These fission rates integrate the contributions to fission, not only of the father isotope present initially, but also those of the daughter isotopes produced during the irradiation.

Table 2: MA transmutation (T) and fission (F) rates for PWR and FR

Reactor	PWR		FR	
Burn Up	60 GWd/t		140 GWd/t	
Flux level	2.5 10^{14} n/cm^2/s		3.4 10^{15} n/cm^2/s	
Irrad. time	1500 EFPD		1700 EFPD	
Fission (F) and Transmutation (T) rates	T (%)	F (%)	T (%)	F (%)
Np237	46	4	63	24
Am241	70	10	69	24
Am243	65	6	63	15
Cm244	44	16	50	27

From the analysis of these results, it arises the principal points:
- under the standard conditions of reactor's irradiation, it is not possible to reach a complete disappearance of the elements considered with the only once through irradiation and multi-recycling is necessary,
- the fission rates obtained in FR are higher, in a ratio 2 to 6 with those obtained in PWR,
- in PWR, the transmutation of the considered elements being done primarily by successive captures, the production of higher elements will be very clearly enhanced.

This last point is of main importance. In fast spectrum, production of higher isotopes is quite limited by comparison with thermal spectrum for which production of isotopes higher than curium is much larger. Even if mass production is quite small, due to very high spontaneous fission yield mainly for Cm246 and Cm248 but also upper element produced like Californium, this is quite a large drawback for the fuel cycle operations.

Concerning transmutation of LLFP, the isotopes whose possibilities of transmutation have been assessed are ^{99}Tc, ^{135}Cs and ^{129}I. These isotopes are pure neutron capturers. In PWRs, their transmutation proves very difficult and would require the fuel's overbreeding. The FRs can be used with specific assemblies in which the neutron spectrum is changed locally to benefit from large thermal neutron cross sections. Cesium transmutation is not realistically foreseeable due to the two reasons that ^{135}Cs capture cross section is very low, about 1 barn in thermal spectrum, and that a prior isotopic partitioning to isolate ^{135}Cs from others Cs isotopes is necessary before transmutation. The destruction of technetium and iodine is possible in FRs by using targets placed in a moderated neutron spectrum. Nevertheless, calculations show a low transmutation performance, even in optimal neutronic conditions. For example, for technetium's most favourable case, 20 to 30 years of irradiation are needed to reduce the quantity of technetium by half. Technetium and iodine transmutation is therefore hardly realistic when the goal is to reduce their radiotoxicity.

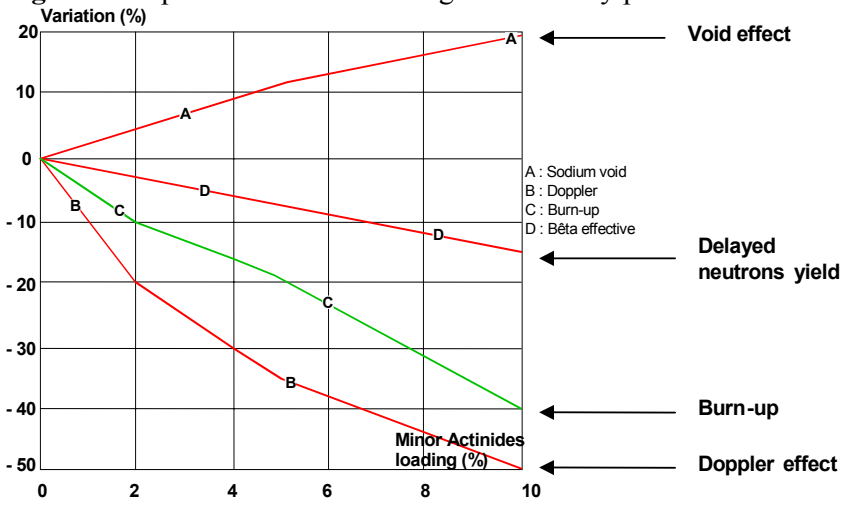
3. Reactor systems

3.1 Critical PWR and FR reactors

The main restriction to introducing minor actinides into critical reactors is linked to their impact on the core's reactivity and kinetic parameters. This would produce:
- a drop in the fuel temperature coefficients (Doppler effect),
- an increase on reactivity effect linked to the coolant voiding,
- a reduction on delayed neutron yield.

For instance, these impacts in function of Minor Actinides content loading are given for the case of a large Fast Reactor in Fig. 2. The Minor Actinides content limit is around 3% of total heavy nuclides for large sodium cooled reactors. The Gas Cooled Fast Reactor allows a upper loading in MA than liquid Metal Fast Reactor due to the disappearance void effect constraint, 5 % of MA seems acceptable even for a large core.

Figure 2: Impact of the MAs loading on the safety parameters for LMFR



The void effect is also actually the most restricting criteria of MA loading in PWR for the physic and safety impact point of view. Without the coolant, the neutron spectrum moves on higher energy and the minor actinides contributions of the thermal and epithermal resonances vanish. The Minor Actinide content must then be limited to about 1% of total heavy nuclides.
For standard reactors, the final conclusions are:
- the admissible quantities of minor actinides in the core have to be kept low (about 1% for PWR type and 3% to 5% for FR type),
- the maximum MA's fission rates are about 5 to 10 % in PWRs and from 15 to 30 % in FRs. In both cases, a multi recycling is necessary in order to reach satisfactory overall performances,
- in PWRs, Curium recycling is to be avoided as it produces by itself and by producing Californium, an intense source of neutrons. In FRs, Curium recycling undoubtedly produces upper elements; however they stabilize at a far lower level than in the PWRs and therefore do not pose any specific problems.

3.2 Accelerator Driven Systems

Systems dedicated to MA transmutation comprise a subcritical reactor coupled to an external neutron source supplied by a proton accelerator. Such dedicated ADS (Accelerator Driven System), operating in a sub critical mode, offers:

- a safety guard against accidental reactivity increase which allows considering large MA loads whose reactivity feed back (Doppler effect) and proportion of delayed neutrons would be prohibitive with critical cores,
- an acceptance of a greater variation in the fuel's isotopic composition, resulting from transmutation during the cycle, as the system is no longer constrained by self-criticality during the whole fuel time in reactor.

The complexity of these systems means that they cannot be considered as electricity generating reactors. Their development involves the design of highly technical elements, such as a reliable power accelerator which has to supply an intense high energy proton beam for long period of time without beam interruption, a spallation target which has to produce high energy neutrons under the effect of the acelerator's proton using the lead bismuth eutectic (LBE) and a window separating the accelerator void from the LBEf, and a reactor core operating in sub critical and fast spectrum mode. For the first time ever, the main components have been successfully assembled for studies at zero power in the CEA Cadarache Masurca reactor, as part of the Muse program. The results of this program have allowed developing validated sub criticality level measurement techniques. Nevertheless, several technological constraints still remain and must be overcome before judging the viability of a powerful ADS; studies are on going on these subjects as part of the European EUROTRANS project dealing with ADS designs (both experimental and industrial), neutronics, fuels, LBE technology and specific nuclear data.

4. Fuels and targets for transmutation

A massive research programme on transmutation fuels and targets, including in particular an experimental irradiation programme, has been launched since 1995. This experimental irradiation programme has been based, for a large part, on tests in the European HFR in Petten and in the CEA Phénix fast reactor which power restart has been effective in late 2003. Flux conditions of Phénix are well suited to allow the studies of irradiation damages of fuels and targets for transmutation under representative conditions of fast and partly moderated flux, which are considered to be the most efficient for transmutation of MA and some LLFP. A first phase of experiments has been conducted in Phénix:

- Ecrix B and H targets, made of pellets containing americium oxide microdispersed in an inert magnesia matrix, under fast (B) and partly moderated (H) neutron flux (Ecrix H has been unloaded in 2005),
- 3 Metaphix sub assemblies, containing experimental pins with metallic UPuZr fuel and dispersed MA, inside the framework of an agreement with the Japanese CRIEPI (Metaphix 1 has been unloaded in 2004),
- Anticorp 1, consisting of 3 pins containing pure metallic ^{99}Tc,
- Profil R for the measurement of capture sections of FP, lanthanides, actinide isotopes under fast flux neutrons.

A second phase, which will integrate the knowledge gained with the experimental irradiations carried earlier in Phénix, in the thermal French Siloe reactor and the European HFR and the R and D results about fabrication process (macrodispersion and new matrix materials), has started in 2006; the important joint CEA-DOE-ITU-JAERI Futurix-FTA experiment (oxide, nitride and metallic ADS fuels) is also actively prepared for a start of irradiation planned beginning in 2007 up to the final shutdown of Phénix in 2009.

Americium oxide is today the reference compound for the transmutation targets of this radionuclide. The experimental feasibility of Americium transmutation has been shown in several irradiation experiments, one of which in HFR has resulted in a transmutation rate higher than 99% and a fission rate of 52%. For the inert matrix associated with the MA compound to form the target, magnesium oxide, spinel, yttriated zirconium oxide and metallic molybdenum are under investigation. Results of the ongoing irradiations should permit the selection of the most suitable material once the tests are completed.

V. Scenarios studies

To assess through simulation the nuclear reactor's transmutation capacities, calculation software programs which reproduce the physical phenomena occurring in the core are used. These neutron calculation programs rely on three large subsets whose quality determines the level of uncertainty of the calculations' results: data libraries, programs or calculation codes and qualification experiments. Developments and additions have been made to each subset to cater for the specificities of waste-transmuting reactors. These models and data make it now possible to perform preliminary calculations, whose results and tendencies are relevant and credible for feasibility studies.

Assessing a waste management solution can only be achieved through an overall view of the cycle (fuel, reactors, cycle plants). Therefore, the scenario studies which consider the cycle's hypothetical developments over time are essential for assessing the interest of and the possibilities to implement (partitioning)/transmutation.

The scenario studies provide these overall views of the cycle and waste produced at various times, from the current situation to balanced situations, which could be reached after several decades if feasible. The transitional scenarios must therefore also be assessed and so must their feasibility on the basis of the existing reactor fleet. The CEA uses the calculation COSI code to simulate how the fuel cycle functions (reactors and cycle facilities), so as to provide, according to time, the development of fluxes and of nuclear material inventories which are present in the cycle and the characteristics of the waste produced.

Different major scenario families have been considered. Since plutonium is both a recyclable energetic material and the main contributor to potential long-term radio toxicity, all scenario studies considered must be consistent with the policies contemplated for the long-term management of plutonium. A first appraisal of the scenarios with current technology reactors (PWRs and FRs) have indicated that the multi recycling of plutonium offers a first advantage by reducing the potential radiotoxicity of ultimate waste by a factor of 3 to 10, compared to once-through cycle, and that partitioning and transmutation of minor actinides would bring a further advantage reflected by a global reduction factor in the order of 100, depending on the partitioning/transmutation modes considered.

As early indicated, transmutation physics favors the fast neutron spectrum. Neptunium transmutation, homogeneously related to plutonium recycling, does not pose any specific difficulties in terms of feasibility. However, this operation does not make a significant contribution to waste management. For the recycling of all of the minor actinides in PWRs, the appearance of californium (due to curium recycling) is prohibitive; associated with curium, it would especially create significant difficulties during the recycling and manufacturing of the fuel, given the presence of curium and californium (high thermal release, intense neutron emission). Americium recycling on its own in PWRs is hardly realistic. Introducing americium into the PWR fuel requires, for reactor operating safety reasons, additional uranium 235 enriching and would therefore only make use of a part of the plutonium's energy capacity in comparison to the recycling of plutonium on its own.

Furthermore, the americium inventory would not be stabilized by 2100 and would require significant modifications to the existing cycle plants.
Finally, the scenario studies have shown that the optimal solution for the implementation of partitioning/transmutation is connected to the deployment of fast neutron spectrum systems like those of the fourth generation systems. The introduction of partitioning/transmutation options which use fast neutron spectrum reactor technology, implemented during the 21st century to renew the French current fleet, has been the subject of an assessment of fleets transitional scenarios from the current situation to balanced situations. Those scenarios based on fast neutron critical reactor systems mean that the aim to minimize minor actinides is reached through the full actinide recycling operation which is possible in these systems; these reactors can reprocess their waste and the minor actinides produced by the current PWR fleet.

VI. Conclusion

Important results are now available concerning the possibility of significantly reducing the quantity and the radiotoxicity of long-lived waste in association with a sustainable development of nuclear energy. As France has confirmed its long-term approach to nuclear energy, the most effective implementation of (recycling-)transmutation of actinides other than Plutonium and Uranium depends on the fast neutron GEN IV systems which are designed to recycle and manage their own actinides. The perspective to deploy a first series of such systems around 2040 supports the idea that progress is being made: the long-term waste would only be made up of fission products, whose radio toxicity considerably drops within a few hundred years. Future work will deal with pre industrial demonstrations of transmutation. A new waste management Law has been passed by French Parliament in 2006, demanding that P and T research will continue in strong connection to GEN IV systems development and allowing a detailed cost/benefit analysis by 2012, in order to decide or not the industrial implementation of P and T, if proved of interest with regards to the consequent simplification of the conditions of the necessary geological disposal.

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