

## **SESSION II**

# **THE NUCLEAR FUEL CYCLE AND P&T**

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## RECENT TOPICS IN R&D FOR THE OMEGA PROGRAMME IN JAERI

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### Abstract

The R&D for partitioning and transmutation (P&T) technology has been carried out in Japan since 1988 under the OMEGA (Options for Making Extra Gains from Actinides and fission products) programme. In this programme JAERI has proposed the double-strata fuel cycle concept as a partitioning and transmutation system for long lived radioactive nuclides. The system consists of three technical areas or processes of the partitioning, nuclear transmutation and fuel processes. This paper summarises the JAERI's activities on these topics, focusing on the recent technical achievements in each process.

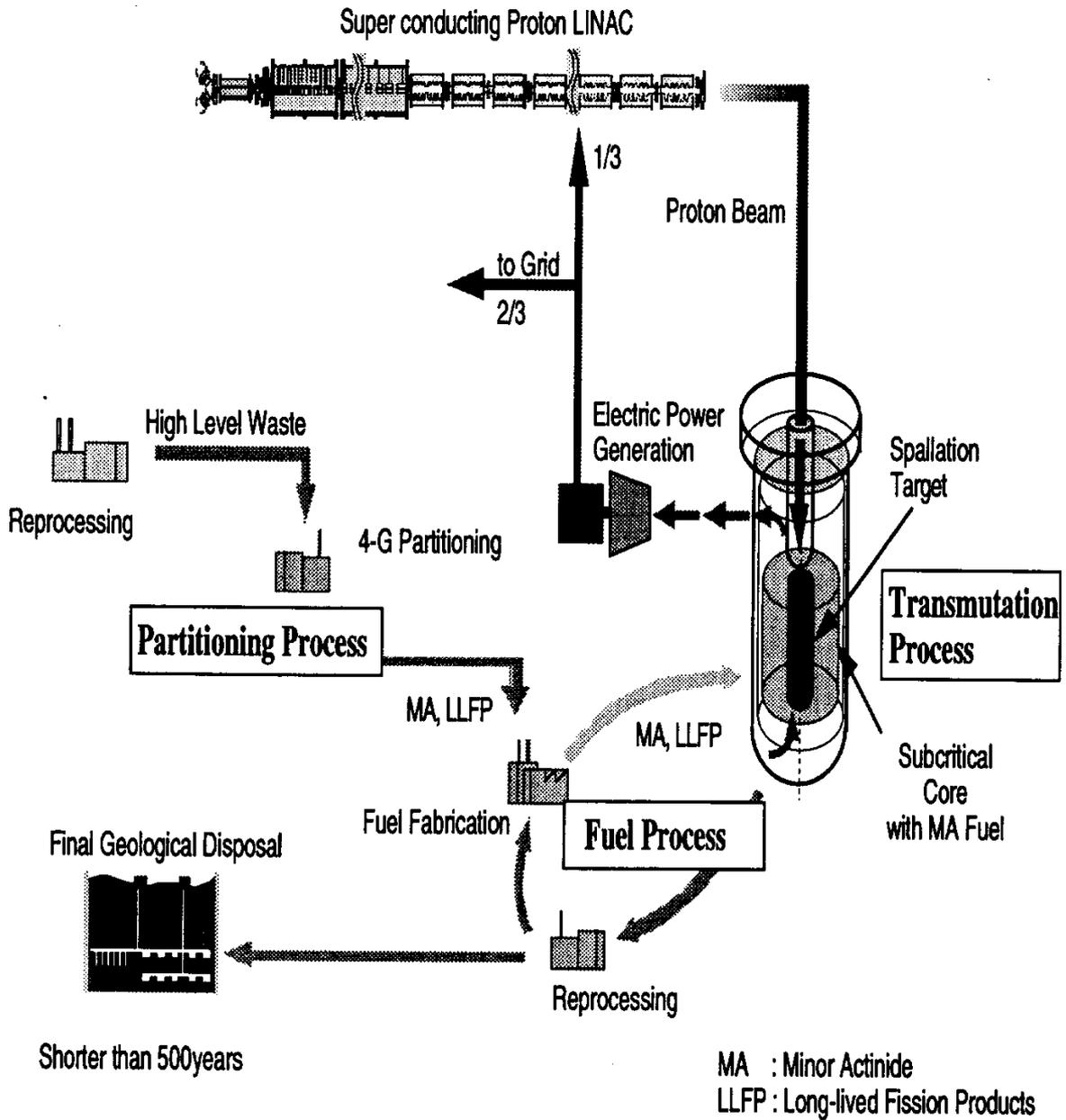
## 1. Introduction

The double-strata fuel cycle concept has been proposed by JAERI as a partitioning and transmutation system for long lived radioactive nuclides. Mukaiyama *et al.*, reviewed the activities in JAERI for research and development of this concept [1]. The system consists of the following three technical areas or processes, the partitioning, nuclear transmutation and fuel processes.

- **Partitioning process:** The four-group partitioning process (4-GPP) has been developed to separate the elements in high-level liquid wastes (HLLW) into transuranium elements (TRU), Tc and Platinum group metals (PGM), Sr-Cs group, and others. TRU are separated by extraction with diisodecylphosphoric acid (DIDPA), Tc and PGM by precipitation through denitration, Sr and Cs by adsorption with inorganic ion exchangers (titanic acid and zeolite). A hot verification test was performed using concentrated real HLLW. As a modification effort of the present 4-GPP, a more powerful ligand, tridentate diglycolamide (DGA), has been studied to extract actinides directly from HLLW. From fundamental studies, tetraoctyl 3-oxapentandiamide (TODGA) was selected as the most proper DGA extractant.
- **Transmutation process:** A lead-bismuth cooled accelerator driven system (ADS) with nitride fuel has been proposed as a dedicated transmutation system to be deployed into the second stratum. The 800 MWt plant has a pool-type configuration and a power conversion system operated on a saturated steam cycle.
- **Fuel process:** Nitride is suitable for the fuel material for MA transmutation from the viewpoint of supporting hard neutron spectrum and heat conduction ability. In addition, actinide mononitride with NaCl-type structure will have a mutual solubility leading to the flexibility of fuel composition. Pyrochemical processing has several advantages over wet process in case of treating MAs concentratedly with large decay heat and fast neutron emission. One of the drawbacks of nitride fuel is that nitride with  $^{15}\text{N}$  enriched nitrogen must be used to minimise the  $^{14}\text{C}$  production. But the pyrochemical process has the practical feasibility of recovering expensive  $^{15}\text{N}$ .

In this paper, the recent JAERI's activities in the P&T technology development are reviewed by focusing on the major technical achievements in each process shown in Figure 1.

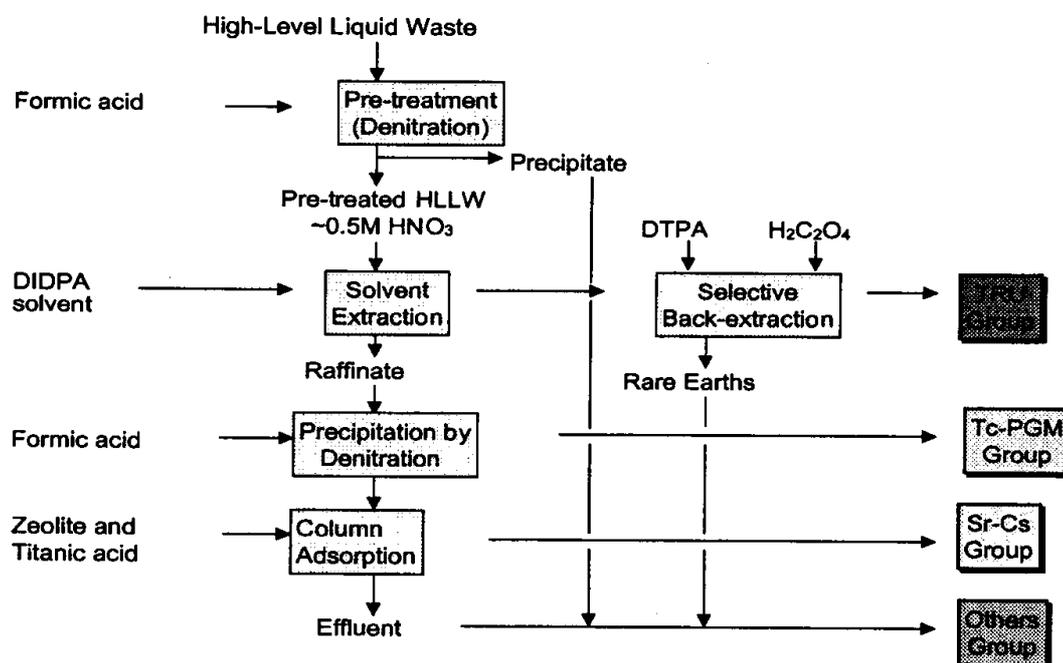
Figure 1. Partitioning and transmutation system for long-lived radioactive nuclides at JAERI



## 2. Partitioning process

A hot verification test of the 4-GPP with concentrated real HLLW was carried out in the Partitioning Test Facility in the hot cell [2]. For the preparation of the concentrated HLLW, about 14L (11 TBq) of the raffinate from the co-decontamination cycle of Purex Process were first denitrated and then concentrated to about 2.5L. The raffinate was obtained by two reprocessing tests with about 1 kg of spent fuel burned up to 8 000 MWd/t and with about 1.5 kg of spent fuel burned up to 31 300 MWd/t. The flow sheet is shown in Figure 2.

Figure 2. Flow-sheet of 4-group partitioning process



Results of the present test well agreed with the either result of previous tests using the unconcentrated real HLLW and the simulated HLLW added with a small amount of real HLLW. Table 1 summarises the fractional distribution of each element at the 1st mixer-settler. More than 99.998% of Am were extracted from the HLLW with the organic solvent containing 0.5M DIDPA – 0.1M TBP, and 99.986% of Am were back-extracted with 4M nitric acid. Cm showed the same behaviour as Am. Np and Pu were extracted simultaneously in a high yield, and more than 99.9% of them were back-extracted with oxalic acid. In the denitration step for the separation of Tc and PGM, pH of the solution was increased to 2.8 after the denitration, and then more than 90% of Rh and more than 97% of Pd were precipitated. About half of Ru were remained in the denitrated solution, but the remaining Ru were quantitatively precipitated after neutralization of the denitrated solution to pH 6.4, which was performed for the preparation of the feed solution to the adsorption step with the inorganic ion exchangers. In the adsorption step, both Sr and Cs were separated effectively. Decontamination factor for Cs was more than  $10^6$  in all the effluent samples.

Table 1. Fractional distribution (%) of each element at the 1st mixer-settler (from [2])

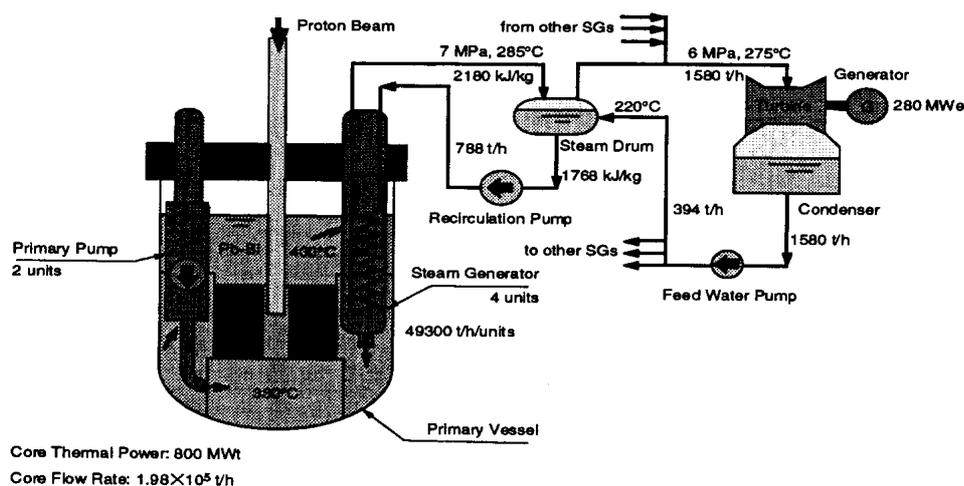
Element	Raffinate	Stripped with 4M HNO <sub>3</sub>	Solvent	Mass balance
Am	<0.002	99.986	0.012	77
Cm	<0.001	99.984	0.015	79
Eu	<0.0004	98.7	1.3	79
Np	1.8	0.3	97.9	110
Cs	99.95	0.05	0.001	106

The present 4-GPP necessitates a pre-treatment step, i.e. denitration-filtration, to reduce the acidity of an aqueous feed in harmony with the capability of diisodecylphosphoric acid (DIDPA). Thus as a modification effort of the 4-GPP, to eliminate the above step, more powerful ligand to extract actinides directly from high-level liquid waste (HLLW) has been developed in line with the CHON principle [3]. We found that a tridentate diglycolamide (DGA) with an ether oxygen at a centre of diamide molecule forms more stable complex with trivalent actinides and lanthanides than that of bidentate malonamides. Aiming at a high solubility of the extracted metal-DGA complex in n-dodecane solvent, to avoid the third-phase formation, and a high extractability toward all actinides, we have examined the effect of chain length of alkyl groups attached to the two amidic nitrogen atoms, and selected a ligand, tetraoctyl 3-oxapentandiamide (called TODGA hereafter) as the most proper DGA-extractant. To facilitate the development of TODGA-partitioning process, fundamental studies on i) the extraction behaviour of TODGA for various valency states of actinides, trivalent lanthanides, and some fission products ii) radiolytic degradation of TODGA by  $^{60}\text{Co}$  gamma rays have been carried out. The results of above investigation revealed that the TODGA is a satisfactory extractant to be applied to the process of separation of actinides and lanthanides (III) in the 4-GPP.

### 3. Transmutation process

Preliminary design of an 800-MWt lead-bismuth cooled accelerator-driven system has been developed as a dedicated transmutation system to be deployed into the second stratum in the double strata fuel cycle [4]. The system employs lead-bismuth for the target and primary coolant material. The plant has a pool type configuration and a power conversion system operated on a saturated steam cycle. An analysis of beam trip transient was made for this type of accelerator-driven transmutation plant [5]. Transients of the primary coolant temperature, the water/steam temperature, the water/steam pressure, the turbine flow rate, and the electric output were calculated using a simple network model based on a simplified flow diagram of ADS plant shown in Figure 3. The plant and turbine trips were required at 380 s after beam trip to prevent from overcooling. The maximum temperature swing was 185°C in lead-bismuth, and 82°C for in water/steam for the case when beam recovered at 370 s.

Figure 3. Simplified flow model of ADS plant



Mass balance in the proposed double strata transmutation system was analysed for cases where the type of power reactors in the first stratum is  $\text{UO}_2$ -LWR, MOX-LWR and FBR. The analysis shows that the transmutation rate for (Pu, MA) composition from a MOX-LWR becomes one half than that

from an  $\text{UO}_2$ -LWR. The number of transmutation systems and the amount of transmuted minor actinide are estimated for several possible scenarios of the future nuclear power development, assuming the deployment of transmutation systems starts in 2030. It was concluded that the introduction of ADS could play a significant role as “transmuter” in the back-end of fuel cycle [6].

A code system “ATRAS” was developed for the neutronics design of ADS [7]. The code system consists of the nucleon-meson transport code, Sn code and burn-up analysis code. In order to obtain the nuclear data required for the development of ADS, the Actinide File and the High Energy File were developed along with the JENDL General Purpose File.

## 4. Fuel process

Fabrication of MA nitride, irradiation tests of nitride fuel and the development of pyrochemical process for nitride fuel have been carried out [8].

### 4.1 Fuel fabrication process

Fabrication of Pu and MA-bearing nitrides and preparation of the thermodynamic database have been carried out besides the irradiation tests of (U,Pu)N fuel up to 4.6 at%. High-purity AmN and (Pu,Cm)N were fabricated by carbothermic reduction of the dioxides by use of  $^{243}\text{Am}$  and  $^{244}\text{Cm}$  nuclides. X-ray diffraction patterns showed almost the single phase of NaCl-type structure. On the other hand, PuN pellets containing inert matrix nitrides such as ZrN and TiN were fabricated and characterised. Vapour pressure of Np(g) over NpN, (U,Np)N and (Np,Pu)N was measured by high-temperature mass spectrometry to clarify thermodynamic properties of the solid nitride phase. Thermodynamic property of Np(C,N), which is an intermediate product of carbothermic reduction for fabricating NpN from  $\text{NpO}_2$ , was also evaluated by both experiments and calculation. The results suggested that Np (C,N) could be treated as ideal solid solution as is the case of Pu (C,N). Measurements of heat capacity and thermal expansion of NpN and PuN are underway by use of the sintered sample for preventing oxidation. The irradiation of two  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{N}$  fuel pins at fast test reactor JOYO was completed in 1999 under the joint research with JNC. The non-destructive post irradiation examinations are underway and any failure of fuel pins was not observed. The destructive examinations will start in the latter half of this year.

### 4.2 Fuel reprocessing

As for pyrochemical process, the electrochemical dissolution behaviour of NpN and PuN were measured by cyclic voltammetry and the equilibrium potentials of the nitrides in LiCl-KCl eutectic melt were determined. On the other hand, the electrochemical deposition behaviour of Pu at liquid Cd cathode was investigated. In this case the potential of deposition and dissolution shifted positively compared with the case of solid cathode in correspondence with a thermodynamic stabilisation by formation of intermetallic compound. Indeed, the formation of  $\text{PuCd}_6$  phase was observed at the cathode by microprobe analysis. By adjusting electrochemical parameters such as current density during electrolysis, ten-gram scale of Pu was recovered at liquid Cd cathode with high Pu concentration. In addition, the electrochemical deposition behaviour of Np at liquid Cd cathode and the phase relationship of Am-Cd binary system were experimentally studied. Nitrogen releasing behaviour from NpN and PuN at an anode, and the results of distillation and nitridation of the Cd cathode after the electrolysis were examined. It was proved that the pyrochemical process is fundamentally suitable for recovery of expensive  $^{15}\text{N}$ -enriched nitrogen gas compared with the wet process.

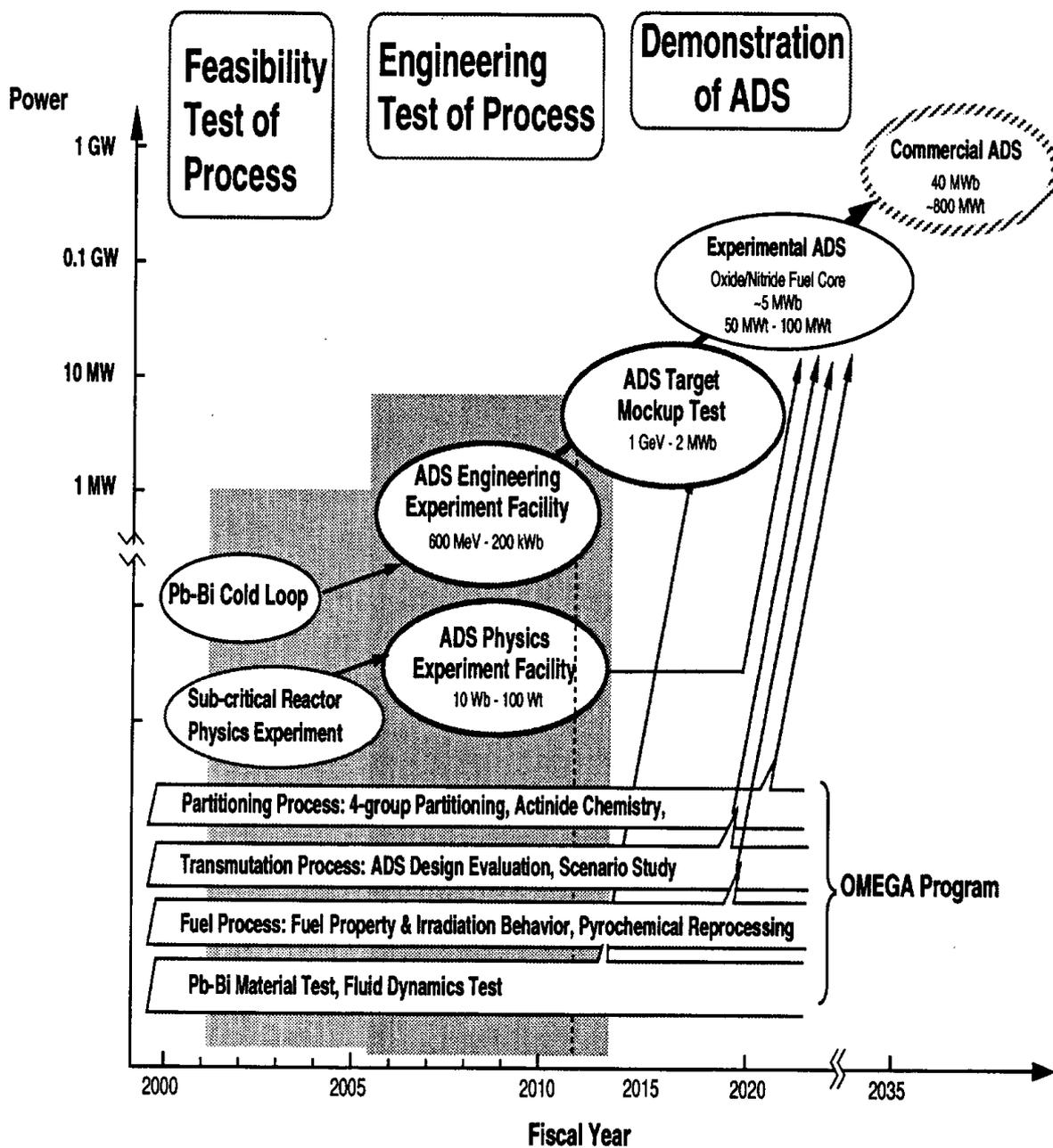
## 5. Concluding remarks

In 1999, the Atomic Energy Commission of Japan (AEC) established the Advisory Committee on Nuclear Fuel Cycle Back-end Policy to conduct the Check-and-Review of the outcome of the OMEGA programme. The Committee concluded, in the report issued in March 2000, that the present status of the programme would be the level of basic studies and tests, and that various concepts of the P&T system were evaluated and required technologies were developed. They also concluded that the future R&D should be proceeded in order to convert the high level waste into useful resources and to reduce the environmental impact associated with its disposal. Their recommended processes are as follows; to study the P&T implementation scenario taking account the situation of nuclear fuel cycle in Japan, to carry out basic experiments to demonstrate the feasibility of the process, and to conduct engineering scale experiments to obtain safety data of these systems.

After getting the results of the above mentioned C&R by AEC, JAERI will proceed the R&D in each process of the P&T system from the basic experimental step to the engineering mock-up step including the following areas Figure 4:

- **Pb/Bi material test:** One of the technical issues for ADS developments is the corrosion/erosion of material in liquid-bismuth coolant. The beam window represents a major technical challenge in ensuring the structural integrity as it suffers a high differential pressure load as well as thermal stress and radiation damage.
- **Am and Cm characteristics:** A “high temperature chemical cell” is to be constructed in Nuclear Fuel Cycle Safety Engineering Facility of JAERI for gram-scale experiments of Am and Cm.
- **ADS experimental facility:** Another issues to be tested or to be demonstrated are sub-critical reactor physics, and system operation and control. The experimental programmes to solve these technical issues for ADS developments have been planed within the framework of the JAERI-KEK Joint Project for High-Intensity Proton Accelerators.

Figure 4. Scenario for development of ADS



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## **TRANSURANICS TRANSMUTATION ON FERTILE AND INERT MATRIX LEAD-BISMUTH COOLED ADS**

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### **Abstract**

Different strategies for the back-end of the nuclear waste are explored, including different strategies of ADS application to nuclear waste transmutation. In this paper the results of the detailed simulation studies of ADS systems, both with fertile (Th) and inert (Zr compounds) matrix fuels, but always with lead-bismuth coolant will be presented. In addition, several options are considered for the plutonium isotopes: direct burning in ADS together with the minor actinides, a separate partial burning in MOX LWR before its load to the ADS and intermediate solutions. Depending on the case, the studies are performed from two perspectives: the situation of the equilibrium of the fuel cycle and the approach to the equilibrium from the actual LWR discharge composition.

## 1. Introduction

CIEMAT is actively working on the evaluation of the possible roles of ADS systems on the nuclear waste management within a collaboration agreement with ENRESA, the Spanish enterprise responsible for the radioactive waste management. Different strategies for the back-end of the nuclear waste are explored, from direct disposal to different strategies of ADS application to nuclear waste transmutation.

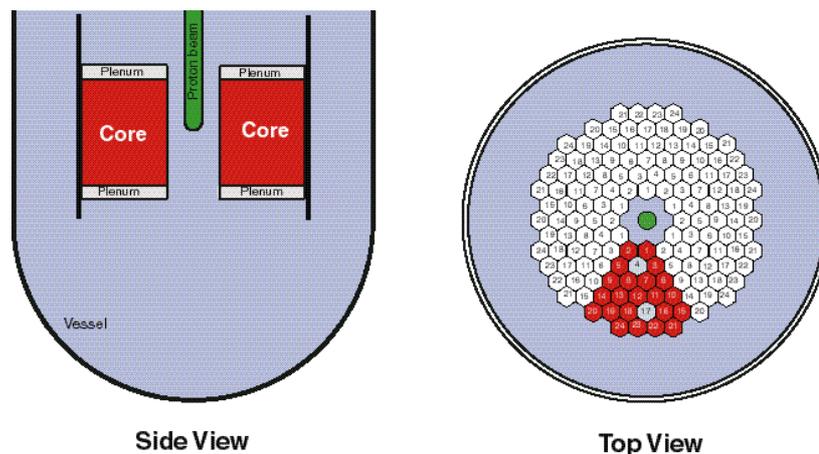
In this paper the results of the detailed simulation studies of ADS systems, both with fertile (Th) and Inert (Zr compounds) matrix fuels, but always with lead-bismuth coolant will be presented. In addition, several options are considered for the plutonium isotopes: direct burning in ADS together with the minor actinides, a separate partial burning in MOX LWR before its load to the ADS and intermediate solutions. Depending on the case, the studies are performed from two perspectives: the situation of the equilibrium of the fuel cycle and the approach to the equilibrium from the actual LWR discharge composition.

The studies are grouped in two wide groups. The first one is based on an ADS with a MOX fuel based on a  $\text{ThO}_2$  matrix and the second one for the inert matrix cases is based on ZrN plus AcN. The ADS systems are similar for the two cases but not exactly the same, on the other hand the methodology of the detailed simulations is in both cases the same, and always based on the EVOLCODE system [1].

## 2. ADS systems main characteristics and simulation methodology

The ADS concept used in all the studies includes a fast core with an hexagonal arrangement of fuel elements cooled by lead (fertile matrix) or lead bismuth eutectic (inert matrix) in forced convection, and operates at constant thermal power close to  $800 \text{ MW}_{\text{th}}$ . The external neutrons are produced in a windowed spallation target, of the same material that the main coolant, by the action of a 1 GeV proton beam. The mass and composition of the fuel depends on the case.

Figure 1. Side and top view of the ADS core concept used for the inert matrix simulations



The ADS used for the fertile fuel case had already been presented in several papers and conferences [2,3]. In the inert matrix cases, the core geometry has been slightly modified, including a total of 132 fuel assemblies, to introduce 12 special rod positions (see Figure 1). These positions are

reserved for control bars, shutdown bars, sample irradiation channels, special instrumentation and others, however in the present studies they have been considered as filled with coolant. Table 1 gives additional details on the inert matrix ADS concept.

Table 1. **Inert matrix ADS parameters**

<b>Hexagonal fuel subassemblies</b>		<b>Proton beam and spallation target</b>	
Flat to flat	210.96 mm	Kinetic energy	1 000 MeV
Total height	150 cm	Beam pipe material	HT9
Active length	120 cm	Beam window	Steel
Subassembly wall thickness	5 mm	Vacuum beam pipe thickness	3 mm
		Vacuum beam pipe external diameter	200 mm
<b>Power + Primary circuit</b>		<b>Fuel pins</b>	
Nominal power	800 MW <sub>th</sub>	Number of pins per subassembly	Var. 169 – 331
Coolant/Convection type	Pb/Bi E./Forced	Pitch (mm)	Var. 15 – 10.7
Inlet temperature	300°C	External radius of fuel pins	4.1 mm
Outlet temperature	450°C	Cladding thickness	0.35 mm
<b>Core</b>		Void thickness	0.1 mm
Fuel	(Zr,TRU)N	External radius of fuel pellets	3.65 mm
TRU elements	Pu, Np, Am, Cm	Internal radius of fuel pellets	0.55 mm
Coolant and moderator	Pb/Bi		
Cladding material	Steel HT9		
Configuration	Hexagonal		
Number of fuel assemblies	132		
Number of special rod positions	12		

The simulation of the ADS systems, their  $k_{\text{eff}}$  values, power distributions and isotopic composition evolution during burn-up has been performed using the EVOLCODE system. The system is based on the combination of: LAHET [4] for the simulation of the neutron spallation in lead produced by the proton beam, and the transport of these neutrons down to 20 MeV; MCNP4B [5] for the complete neutron transport by Monte Carlo for energies below 20 MeV, and to calculate the neutron multiplication, the neutron flux energy spectra at different positions inside the core, the neutron flux intensity magnitude and distribution, the specific power distributions and the energy release by fission; and ORIGEN2.1 [6] with ad-hoc libraries for the burnup calculations. Further details on EVOLCODE can be found in [1]. For the purpose of the simulations of material evolution with burn-up, each fuel assembly is logically subdivided in 10 longitudinal zones.

### 3. Transmutation based on fertile or inert matrix ADS

Two approaches are considered in the CIEMAT transmutation studies. The first one uses a Th matrix (ThO<sub>2</sub>) for the fuel. The matrix provides chemical, mechanical and thermal characteristics very similar to the well known MOX fuels, and in addition, the breeding required to achieve very long burn-ups of the fuel (1 500 days). On the other hand, at the end of the transmutation process a substantial amount of <sup>233</sup>U has been bred from the Th matrix. This fuel cycle concept will make sense if the <sup>233</sup>U is used in the LWR substituting the <sup>235</sup>U or if the U-Pu cycle was to be replaced by the Th-U cycle. This second option will provide a much smaller production of transuranics and finally the

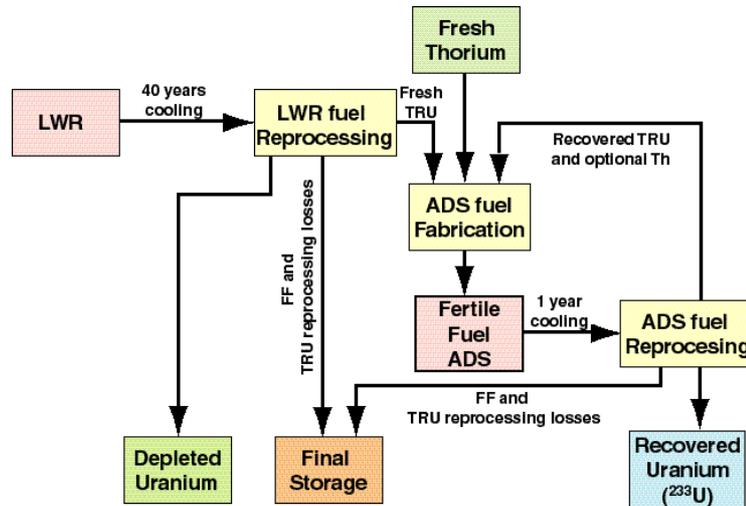
radiotoxicity to be managed could also be reduced. In this last approach the use of TRU from the LWR in the ADS will be a transitory operation where most probably the equilibrium of the cycle will not be reached before the TRU are exhausted (depending on the different countries strategies). The study will concentrate on the first cycles of the TRU burning on ADS.

The most recent studies are devoted to evaluate the inert matrix option both in the mixed oxides or mixed nitrides versions. This option has the advantage of not introducing new isotopes in the fuel cycles, although the enrichment on some of the higher actinides becomes in certain phases unusually high. In the hypothesis of stability of the fraction of energy generation from the fission process, either the present LWR or new reactor types should provide most of energy and the ADS will only contribute with a small fraction of the total produced energy. In this circumstances the transmutation ADS has to handle a continuous amount of TRU regularly being produced at the same rate that they are eliminated. As in most strategies of transmutation on ADS, fuel recycling inside the ADS is required to obtain high elimination levels. It can be easily demonstrated that these two conditions are sufficient to progressively approach in the ADS to an equilibrium fuel composition. The behaviour of the system after reaching equilibrium decides the final TRU elimination efficiency of the system. For these reasons the studies of inert matrix concentrate on the fuel cycle after the equilibrium has been reached.

#### **4. Fertile matrix (Th based) fuel option for TRU elimination**

The concept explored in this study has been to close the LWR fuel cycle operation by introducing all the transuranic isotopes, TRU, contained in the LWR nuclear wastes, after 40 years cooling time, homogeneously in a fuel based on a thorium matrix. This fuel in the MOX chemical form is used in a fast ADS using lead as coolant. The ADS is then operated for a total of 1 500 days at a mean power of 800 MW<sub>th</sub> reaching a burn-up of 146 GWd/THM (the burn-up for TRU reaches 238 GWd/T). The right choice of TRU/Th allows to obtain these very long burn-ups without interruption of the ADS operation, by the precise compensation of fissile isotopes consumption and breeding (mainly from the Th matrix). The ADS fuel is reprocessed after discharge, assuming an uniform 99.9% efficiency for all actinides. The fission and activation products and the reprocessing losses are stored in an appropriated repository. The uranium recovered, mainly <sup>233</sup>U, is available to be used in the operation of other reactors or ADS systems devoted to energy production. The recovered TRUs are mixed with fresh thorium and new TRUs from the LWR nuclear wastes to produce the new cycle fuel. Figure 2 and [2,3] provide more details of the global fuel cycle.

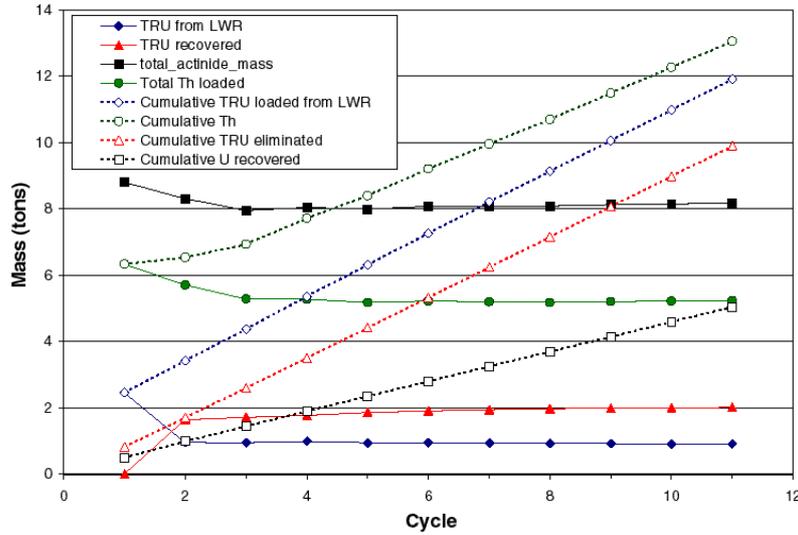
Figure 2. Fuel cycle assumed in the fertile fuel ADS TRU transmutation studies



The first eleven cycles of operation of such an ADS system had been carefully studied in detail. This number of cycles might be sufficient to exhaust most of the TRU contained in the nuclear wastes produced by one reactor generation (from beginning of nuclear reactors till the end of life of the presently installed LWR) of a country with moderate nuclear energy production (10 GWe) in a small number of ADS systems. The fuel composition of each reload is carefully tuned in order to maintain a sufficiently stable neutron multiplication (between 0.96 and 0.98) and to optimise the transmutation efficiency. The respect of the thermomechanical limits of the fuel during the burn-up is also verified.

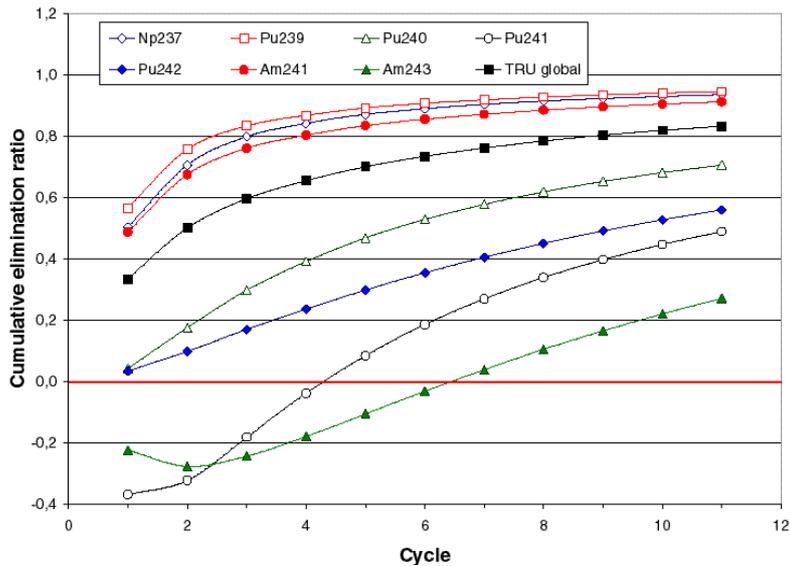
Figure 3 shows the masses of the different components of the fuel for each reload: the TRU recovered from the previous cycle, the new TRUs from the LWR wastes, the total Th and the total actinide mass. In addition, the figure also shows the accumulated TRU from LWR, accumulated Th entered in the system, the eliminated TRU and the recovered U after each cycle. It can be observed that these last four quantities increase linearly with the cycle number after the 4th cycle. The amount of TRU remaining in the ADS after each cycle decreases progressively approaching a constant value, as a consequence that as more and more cycles are performed and equilibrium is approached, the TRU transmutation efficiency increases reaching at the latest cycles very high values. The total TRU mass loaded in the eleven cycles was 11.9 tons while at the discharge of the 11th cycle the total remaining TRU is close to 2.0 tons. This means a global cumulative elimination ratio close to 83.1%. When reprocessing losses are taking into account the global cumulative TRU elimination efficiency is 82.8% in 11 cycles.

Figure 3. Mass composition of the different ADS reloads and evolution of cycle parameters



In addition to the mass reduction, the isotopic composition changes with the transmutation cycles. This is a consequence of the difference of the cumulative elimination efficiencies for the different isotopes. The evolution of this parameter for the most abundant TRU isotopes is presented in Figure 4. For all the main components of the LWR TRUs the cumulative elimination efficiency increases with the cycle number, reaching at the 11<sup>th</sup> cycle values as high as 94% for <sup>239</sup>Pu, 93% for <sup>237</sup>Np, 91% for <sup>241</sup>Am, 70% for <sup>240</sup>Pu, 49% for <sup>241</sup>Pu, 56% for <sup>242</sup>Pu and 27% for <sup>243</sup>Am. <sup>238</sup>Pu is not produced nor eliminated and the curium isotopes are continuously produced in the system, but in any cases the final masses of these isotopes represent only 7% of the TRUs in the ADS discharge after the 11<sup>th</sup> cycle.

Figure 4. Evolution of the cumulative elimination efficiencies for the most abundant TRU



The theoretical limits of this system with very large number of cycles had been computed assuming that the 11<sup>th</sup> cycle is a good representative for the behaviour of the system at equilibrium.

Reprocessing losses both from the LWR and the ADS reprocessing had been taken into account assuming extraction efficiencies of 99.9% for all TRUs. The asymptotic limits of these curves had been computed giving as result that the 0.34% of the LWR TRU will end on the final storage, from reprocessing losses.

## 5. Inert matrix fuel options for TRU elimination

A parametric study of the characteristics of different inert matrix fuels with a ZrN matrix and different Pu-MA fractions [7] (from a cycle with  $UO_2$  LWR and a single pass for all the Pu in LWR MOX fuel and in an ADS with 20 tons of nitride fuel), showed that the evolution of the ADS neutronic multiplication varies from a rapidly falling neutron multiplication to a continuous breeding to configurations close to critical as the MA fraction increases (see Figure 5). Of particular relevance is the existence of Pu-MA mixtures that allow achieving very long burn-ups with minimum variation of the neutron multiplication during the ADS operation. It is also important to note that the fraction Pu/TRU in these stable mixtures is approximately 40%, half the fractions produced in  $UO_2$  LWR or in similar cycles.

The detailed studies on inert matrix fast ADS applications to nuclear wastes elimination had been performed on the scope of the cycle described in Figure 6. The  $UO_2$  fuel is consumed in LWR. The resulting spent fuel is reprocessed separating four streams: the recovered depleted U, the Pu, the minor actinides (MA), and the fission fragments, activation products and reprocessing losses. The Pu is used to produce MOX and then this fuel is used once in LWR. The Pu and MA in the spent MOX and the MA from the LWR are send to the ADS described in Figure 1 and Table 1. The spent fuel of the ADS is then continuously recycled after reprocessing and addition of more Pu and MA from the spent  $UO_2$  and MOX from the LWR.

Figure 5. Evolution of the  $k_{eff}$  for different inert matrix Pu-MA fuel mixtures in a fast Pb-Bi cooled ADS

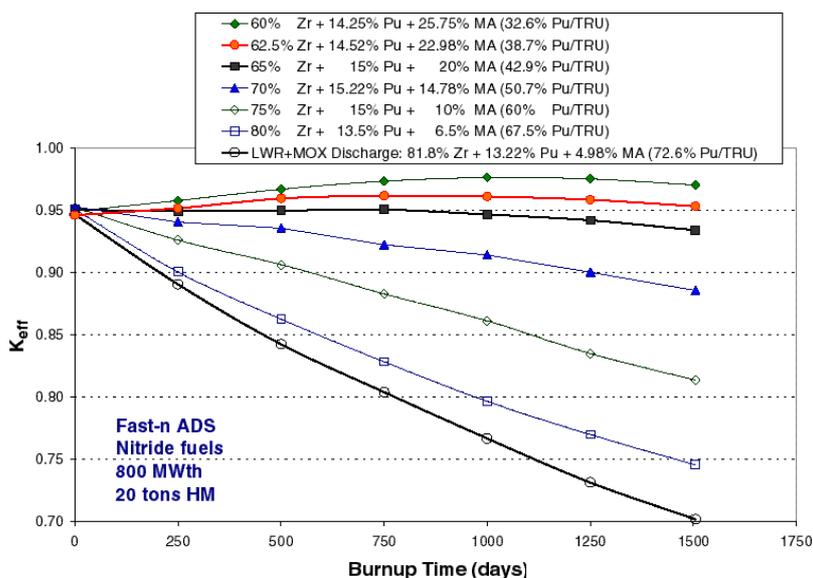
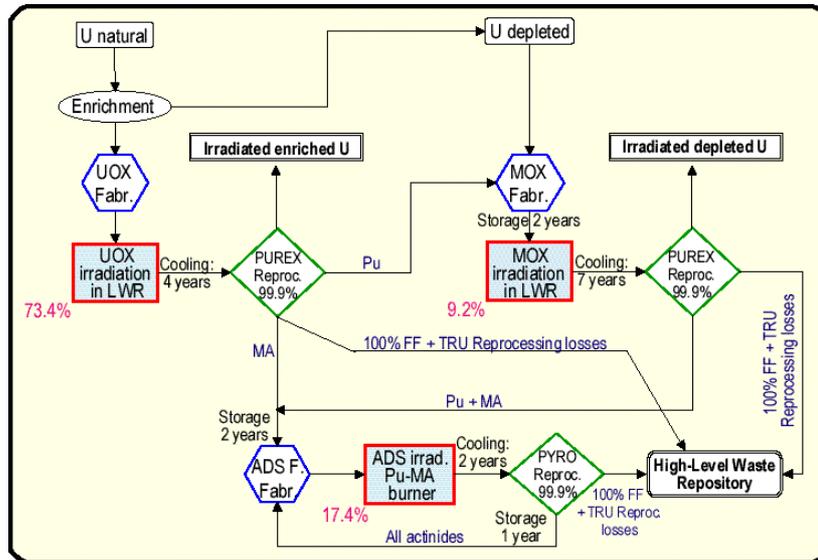
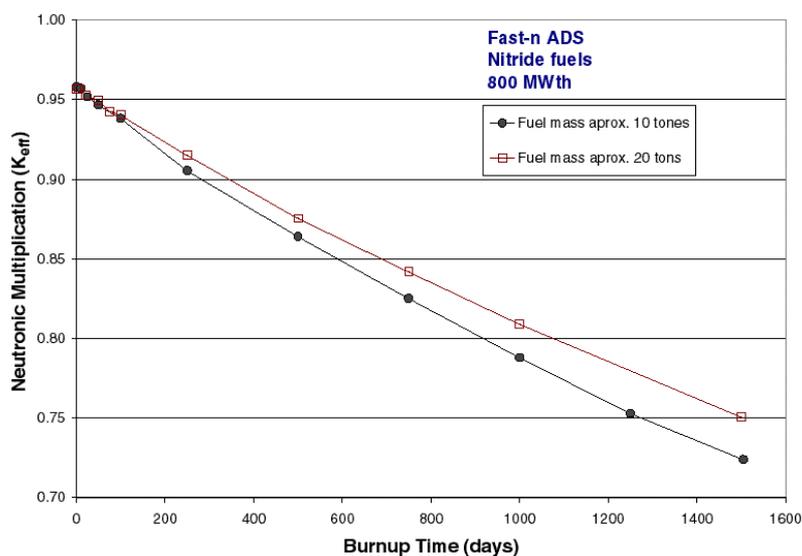


Figure 6. Fuel cycle assumed in the inert matrix ADS TRU transmutation studies



Because the transformation of the actinide isotopic composition vector along the ADS cycle (ADS burn-up, cooling, reprocessing and storage) is contractive, the fuel composition at the beginning of irradiation will progressively approach to an equilibrium value from cycle to cycle as far as the feed from the LWR reprocessing is kept constant. For the long-term consideration and in the hypothesis of maintenance of the present level of energy production from fission, the relevant information is the performance of the ADS cycle after this equilibrium has been reached. The isotopic composition of this equilibrium ADS fuel depend on the isotope vector coming from the ADS, the ADS characteristics and the burn-up per ADS cycle. This last dependence is however small and variations in the burn-up from 600 to 1 500 days introduce corrections smaller than 15% in the main isotopes. On the other hand the ratio between the LWR (UO<sub>2</sub> and MOX) TRUs and the total fuel mass in the ADS fuel depends strongly on this burn-up. The ADS equilibrium fuel composition was computed for the cycle of Figure 6. This fuel includes 2.5% U, 3.8% Np, 72.8% Pu, 13.1% Am and 7.8% Cm. The operativity of the ADS loaded with this fuel in nitride form distributed in a ZrN matrix was studied. Figure 7 shows the evolution of the neutron multiplication with the burn-up, for two different fuel masses of the ADS, 10 and 20 tons. The figure shows that it will be very difficult and expensive (from the accelerator point of view) to maintain the operation more than 150 days.

Figure 7: Evolution of  $k_{\text{eff}}$  of an inert matrix fast ADS loaded with the equilibrium fuel of the cycle of Figure 6



This peculiarity of the ADS loaded with this fuel will be a serious difficulty. On one hand, it will require frequent interruptions of the ADS that will reduce its energy production cost competitiveness, and on the other hand, it will mean many reprocessing passes for the TRU before it is significantly reduced. Many possibilities can be envisaged to mitigate this difficulty in the ADS application to the transmutation of TRUs in equilibrium with simple LWR energy producing cycles. One type of possibilities already proposed by other authors are: the continuous or quasi-continuous fresh fuel supply by means of liquid fuels (e.g. molten salts), particle fuels (e.g. pebble-bed fuels), sliding fuel assemblies or designs of cores that allows to move fresh and spent fuel assemblies between the ADS core and a region neutronically decoupled inside the main vessel. A different possibility is the use of burnable absorbers or control rods in order to maintain a stable sub-criticality level. A third option, implicitly included in the double strata concept, consist in changing the isotopic composition of the equilibrium fuel. What is need is to severely reduce the Pu content on the equilibrium fuel. This can be achieved by reducing the Pu from the LWR reprocessing. This plutonium can not be simply stored, the natural option should be to use its potential as energy producing fuel by continuously reprocessing it on (critical or sub-critical/LWR or fast) reactors devoted to energy production. In this paper two additional options are discussed: the use of the equilibrium fuel in several batches and the use of a partially fertile matrix for the ADS.

### 5.1 Inert matrix fuel ADS for TRU elimination: batches with equilibrium fuel

One possible method to extend the burn-up of the fuel in the case of equilibrium fuel in an inert matrix ADS configuration is to irradiate the fuel in batches. Figure 8, shows a sketch of the possible batch refueling scheme studied in this paper. The approach is OUT-IN, with the fresh fuel coming to the ADS periphery there the fuel is irradiated for a period of time (166 days). When the neutron multiplication has fall below the accelerator possibilities, the ADS stops and the fuel elements move inward, extracting the inner most batch and introducing again fresh fuel in the periphery. Figure 9 shows the variation of neutron multiplication constant  $k_{\text{eff}}$  during one refueling bath. An equilibrium load has been computed that allows to charge exactly the same amount of fuel per 166 days batch in

the 4 batches scheme, allowing to achieve a fuel burn-up at discharge of 140 GWd/THM, with a fluctuation on the  $k_{\text{eff}}$  from 0.956 to 0.936 during each batch. The fresh fuel composition introduced at the periphery has 75.3% Zr, and 24.7% of TRUs, with their equilibrium composition. This OUT-IN scheme allows also reducing the picking ratio of the power distribution inside the ADS.

Figure 8. OUT-IN refueling scheme studied for the inert matrix ADS with equilibrium fuel

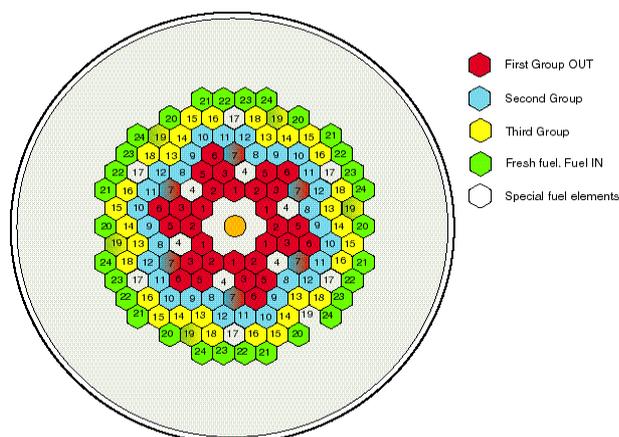
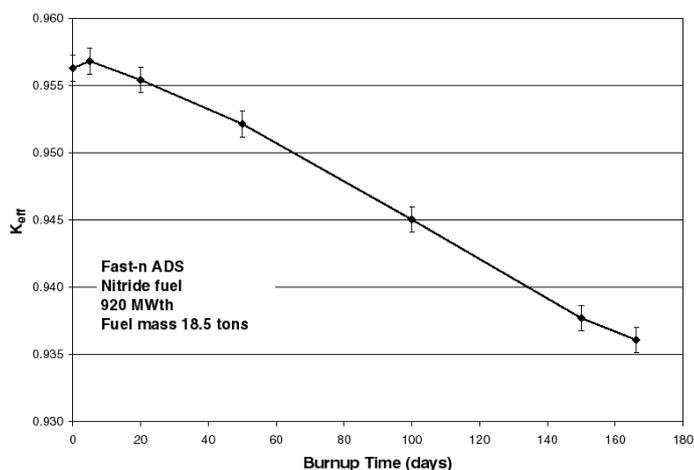


Figure 9.  $k_{\text{eff}}$  evolution of an inert matrix ADS with equilibrium fuel during one of the four refueling batches



## 5.2 Partially fertile matrix fuel ADS for TRU elimination with equilibrium fuel

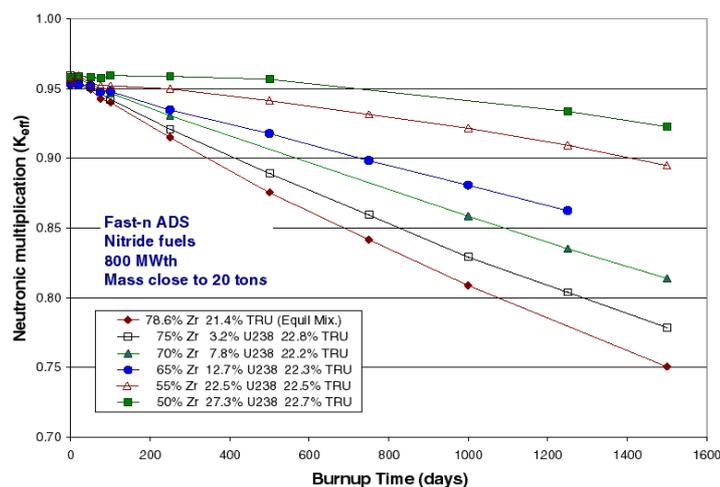
Another option to extend the burn-up of the fuel per ADS cycle is the use of partially fertile matrix adding either  $^{238}\text{U}$  or  $^{232}\text{Th}$ . The peculiarities of fertile fuel with Th had been described in the previous section. The natural choice for the breeding material should be  $^{238}\text{U}$ . This isotope will introduce no new isotope in the fuel cycle and the main effect would be the reduction of transmutation efficiency per cycle. Figure 10 shows that a fuel with 65% Zr, 12.7% U and 22.3% TRU allows to extend the operativity of the ADS loaded with 20 tons of nitride fuel for more than 500 days. Configurations with 55% Zr, 22.5% U and 22.5% TRU and 21.7 tons of nitride fuel

allow to operate for more than 1 200 days. Mixtures of Th with 70% Zr, 7% U and 23% TRU and 18 tons of fuel also allow to obtain operation for more than 500 days. These configurations improve the achievable burn-up per cycle of the ADS but reduce the transmutation efficiency of TRU. Figure 11 shows the change in TRU elimination from pure inert matrix to the 65% Zr, 13% U and 22% TRU fuel after 500 days of irradiation. The eliminated TRU mass is 72% of what could be transmuted in a pure inert matrix ADS if it could be operated for 500 days. The main effect of this reduction is to increase in the complementary proportion the number of reprocessing passes and the corresponding reprocessing losses, as well as increasing the time required for TRU elimination. Both inconveniences are easily acceptable and well compensated for the extension of the single pass burn-up.

### 5.3 Reprocessing losses estimation

To estimate the TRU fraction finally going to the nuclear waste storage, from the reprocessing losses, in the inert matrix scenario, the 4 batches refueling concept with 660 total irradiation time and an average burn-up of 140 GWd/THM, will be used, as the simpler solution for a realistic operation of an inert matrix TRU transmuter ADS. For these parameters and assuming that the reprocessing efficiencies are 99.9% for all the TRUs in the reprocessing of the LWR, the MOX and the ADS spent fuels, simple arithmetic allows to estimate the fraction of TRUs going to the repository between 0.7 and 0.8% of the originally produced. The value obtained from the detailed simulation is 0.707%.

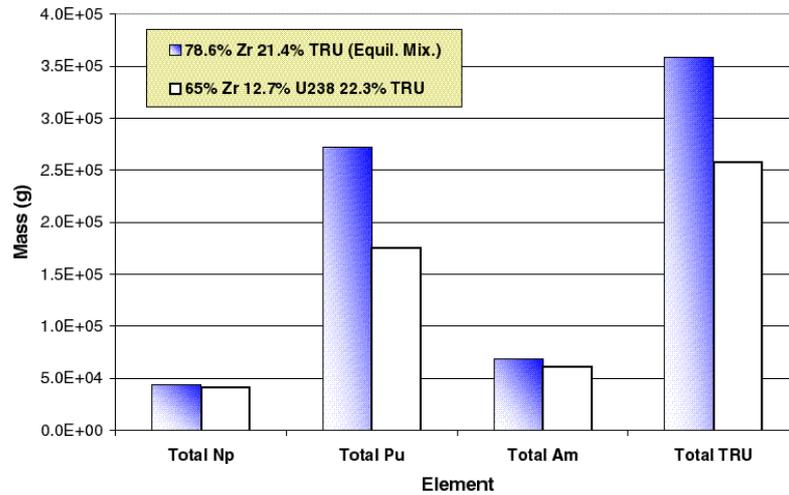
Figure 10.  $k_{\text{eff}}$  evolution of an partially fertile (Zr-<sup>238</sup>U) matrix ADS with equilibrium fuel



## 6. Conclusions

The previous exercises have shown that both the inert matrix and fertile matrix allow to reduce the amount of TRUs to be stored in the final nuclear waste repository by a factor larger than 100 if the cycle is maintained sufficiently long. The inert matrix choice is the solution of minimum perturbation of the present fuel cycle but it has the difficulty of short burn-up per cycle. Several solutions are possible for this problem, again the minimum deviation from the present cycle would be the use of refueling batches or partially fertile (Zr-<sup>238</sup>U) matrixes. A more advance solution is the introduction of Pu recycling in the energy production strata, although this probably will require the use of new types of reactors. Finally the use of Th based matrix ADS will be more justified as a transition from the U-Pu fuel cycle to the Th-U fuel cycle for energy production, although intermediate solutions are also possible.

Figure 11. Transmutation efficiency per 500 days batch in inert and partially fertile matrix ADS



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## **ACTINIDE AND FISSION PRODUCT BURNING IN FAST REACTORS WITH A MODERATOR**

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### **Abstract**

Calculations have been carried out with respect to the transmutation of long-lived wastes (minor actinides and fission products) in fast reactors using special devices based on large amounts of moderator material. Such devices can replace both radial and axial blanket sub-assemblies. It has been shown that the implementation of these devices will allow the achievement of long-lived waste burn-up up to a level of 90-95%, decreasing essentially the radiotoxicity of wastes to be buried.

## 1. Introduction

Spent fuels of modern nuclear reactors contain a rather large quantity of long-lived high-active wastes (plutonium, minor actinides (MA) and fission products (FP)). This amount of waste increases with the electricity production. If plutonium can be used as a fuel for reactors (both thermal and fast), ways should be found for decrease the amount of minor actinides and fission products. Therefore, the major challenge for the future nuclear energy system is the decrease of possible contamination of the environment by such wastes.

Nevertheless, the existing reactors are able to solve these type of problems before development and operation of new promising nuclear systems. However, the implementation of thermal reactors for solving these problems is not efficient due to essential limitations of core physics. The use of traditional fast reactors allows solving the task partially. A homogeneous waste addition to the fuel makes it possible to utilise annually only 15% of all produced MAs in this period and less than 10% of long-lived FP. However, a noticeable degradation of neutronic parameters (increase in sodium void reactivity effect (SVRE) and decrease in Doppler-effect) requires a search for other decisions, more acceptable from a safety perspective.

One of the possibilities involves the implementation of special irradiation devices (ID), which can be located either in radial or in axial blankets. In this case, the effect on core physics will be much less as compared with homogeneous recycling. But the efficiency of transmutation in these devices will be essentially lower.

The efficiency of these devices can be improved by introducing a rather large quantity of moderator. This is related to the fact that practically all FPs have maximum cross-sections in the thermal region and MA cross-sections increase as well when shifting the neutron spectrum to the thermal region. Possibilities for MA and FP efficient transmutation in ID containing moderator are considered in this report for a fast power reactor of BN-800 type as an example.

## 2. MA burning in special devices located in the radial blanket

Americium oxide located in a magnesium oxide inert matrix is considered as a fuel composition for MA burning. A ratio between volume fraction of americium oxide and magnesium oxide can be varied in such a way that it keeps the total americium loading in the ID.

We noted that fast reactors have an important advantage over thermal reactors for burning minor actinides because their neutron flux is two orders as higher. However, in a fast spectrum, actinides have lower cross-sections compared to a thermal spectrum.

On the basis of these two factors an idea appears to use moderated sub-assemblies (SAs) in fast reactors. In this case we conserve a rather high neutron flux and essentially increase the actinide cross-sections [1].

As an ID, we can consider a core SA in which part of the fuel pins are replaced by pins containing americium oxide in an inert matrix and others are replaced by pins containing a moderator. Varying the number of fuel pins with americium and moderator, one can change a moderator volume fraction in the SA. For a more essential moderator fraction increase, one can use a promising fuel pin design in which the central target material rod (of small diameter, with cladding or without it) is surrounded by a rather thick moderator layer. In this design, it is very easy to vary the ratio of fuel and moderator

volume fractions in a wide range conserving the fuel pin external diameter. This type SA design was used in further studies.

One of the tasks of optimisation studies was a search for moderator material allowing a more efficient actinides transmutation, which all other things being the same. Figure 1 presents the dependency of the capture and fission cross-sections of some actinides on material type and Figure 2 – the dependency of the same cross-sections on moderator volume fraction in the fuel pin for zirconium hydride as the most efficient moderator.

Figure 1. **Dependency of actinide cross-sections on a moderator type**

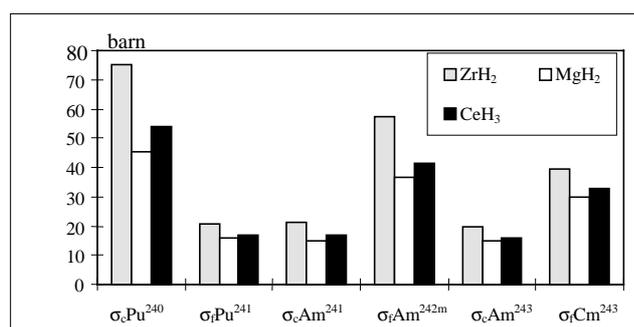
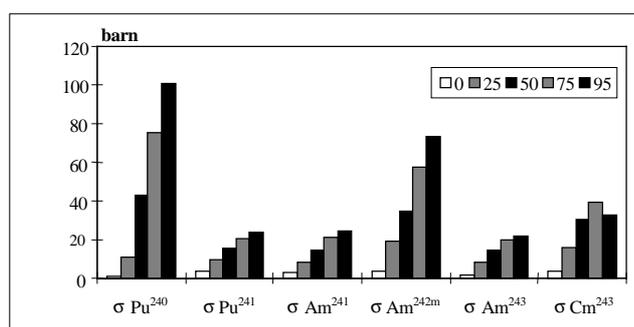
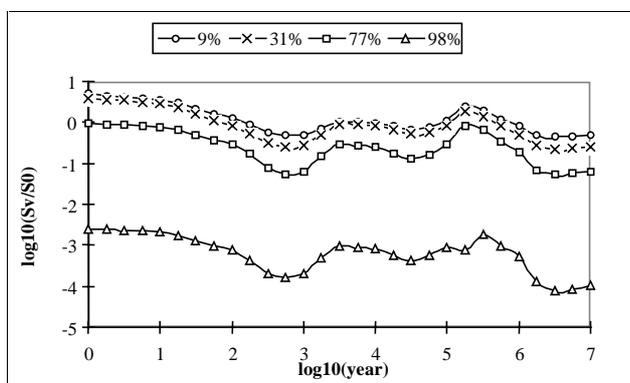


Figure 2. **Dependency of actinide cross-sections on a moderator volume fraction**



We noted that the transmutation of americium in such ID would be worthwhile only if the radiotoxicity of wastes remaining after irradiation is much less than the radiotoxicity of non-irradiated americium. Figure 3 presents the change in waste radiotoxicity for different ID burn-ups in reference to storage of non-irradiated americium

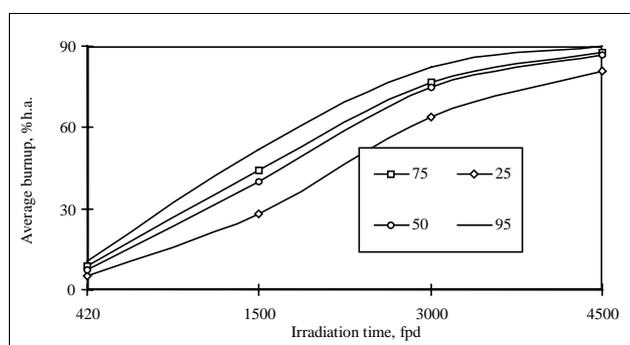
Figure 3. A change in radiotoxicity of ID wastes for different burn-up relating to storage of non-irradiated americium



The dependency presented shows that for a decrease in the waste radiotoxicity of at least two orders, it is necessary to reach 93-95% h.a. americium burn-up. Thus, when designing an ID for americium burning, we should start from the necessity to reach just this high burn-up with maximum possible americium loading, and at the same time not to fall outside the existing limitations for basic structure materials of fast reactor cores.

Figure 4 presents the dependency of americium burning value on irradiation time interval for different moderator volume fraction (zirconium hydride).

Figure 4. Average americium burn-up as a function of irradiation time



It is easy to see that handling the problem of reaching ~90% h.a. burning requires a long irradiation time (~10-15 years) and a rather high volume moderator fraction in fuel pins (>90%). We noted that long irradiation of SAs with americium will require the development of special reloading regimes. For example, in order to eliminate a high burn-up irregularity over a SA, it is necessary to turn it 180° during irradiation cycle. In this case the maximum damage dose will be ≈200 dpa, which will require a high performance for the structure materials used in these ID. Large changes in ID power with americium burn-up will require the development of a special regime for their cooling. Besides, the introduction of the ID with moderator in the first row of radial blanket will lead to increase in power of adjacent core SAs. The changing in power of these SAs can reach 20-30%. However, in our opinion, this power increase is not critical and will not require special measures.

Thus, in order that americium transmutation in ID is appropriate from the standpoint of essential decrease in actinide radiotoxicity, average americium burn-up should be not less than 90% h.a. This

requires irradiation times beyond 10 years, which, in turn, will require the structure material performing at high damage doses.

The second aspect of long-time irradiation is the dependence of actinide cross-sections and burning efficiency on cycle number. Taking into account that the irradiation time is more than 10 years and core lifetime is approximately one and half a year it is necessary to have ~10 cycles of moderated subassembly irradiations. Dependency of actinide cross-sections on cycle number for two type of sub-assemblies are shown on Figures 5a and 5b.

Figure 5a. Dependency of cross-sections on cycle number (without moderator)

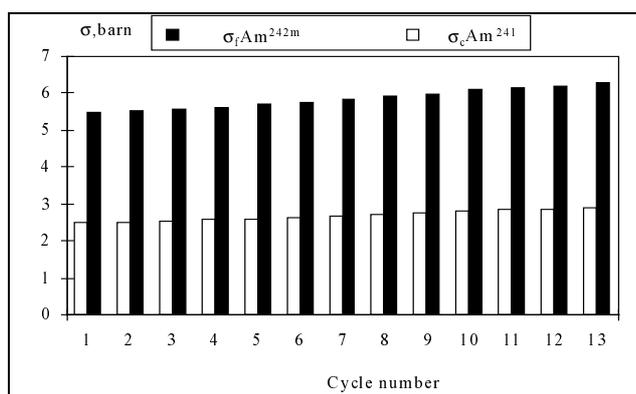
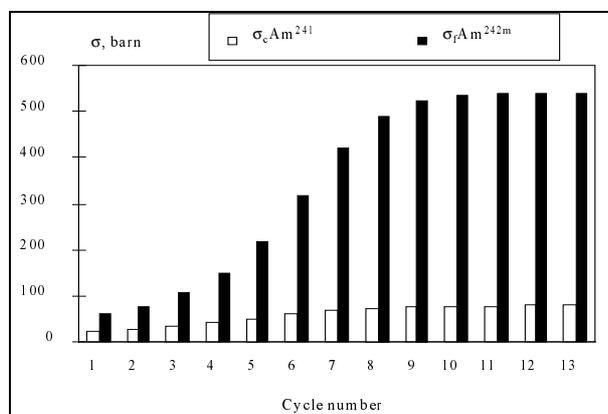


Figure 5b. Dependency of cross-sections on cycle number (with moderator)



This dependency shows the cross-sections in moderated sub-assembly reach the maximum value that after approximately 8 cycles. For the non-moderated sub-assembly the cross-sections do not practically depend on cycle number.

### 3. Influence of moderated sub-assembly on core parameters

To check the influence of moderated sub-assembly with americium on core parameters we investigate the dependency of multiplication factor, target burn-up and sub-assembly power on cycle number.

These dependencies are shown on Figures 6 to 8.

Figure 6. Dependence of multiplication factor on cycle number

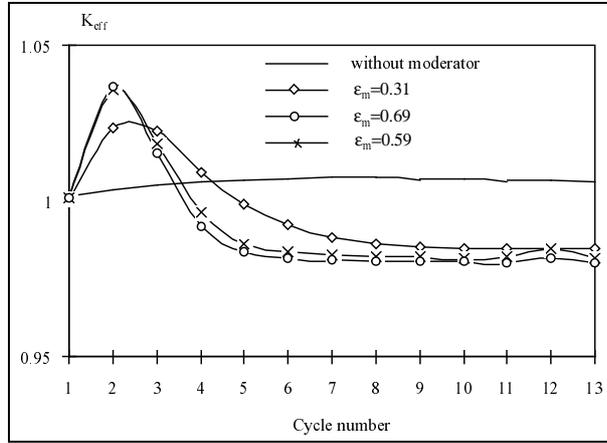


Figure 7. Dependence of target burn-up on cycle number

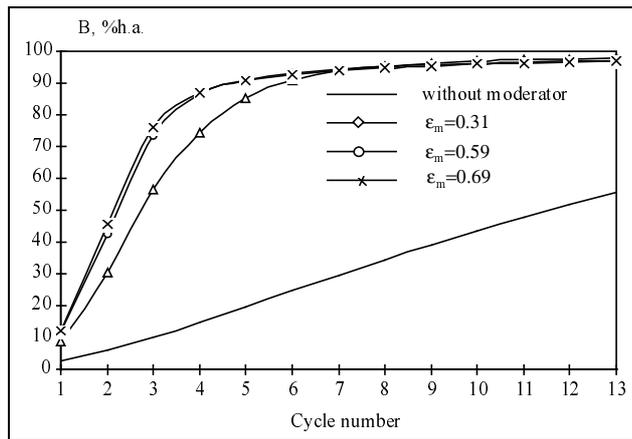
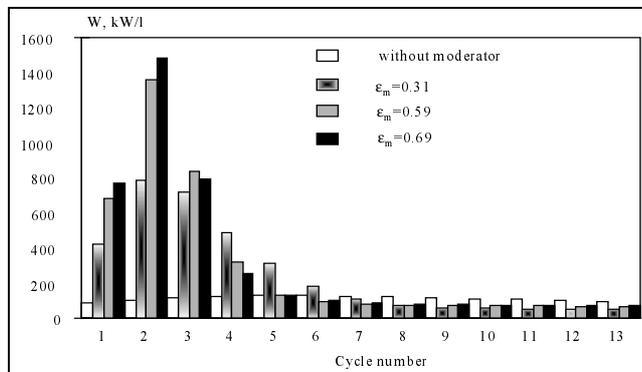


Figure 8. Dependence of sub-assembly power on cycle number



The dependencies presented allow making the following conclusions:

- The moderated sub-assemblies have a great influence on critical state of reactor and it is necessary to take special measures to avoid under and sub criticality.
- The power of a moderated sub-assembly is drastically increased during the first cycles of irradiation due to the formation of isotopes with large fission cross-sections (For example, Am242m) and than drops to the nominal level.
- The burn-up of 90% h.a. is reached after 5-6 cycle and the following 5% (up to 95%) requires approximately the same time.

So, the use of moderated sub-assembly for the americium transmutation faces large difficulties, the main of them being the drastically increase of sub-assembly power during irradiation. This can result in decreasing the temperature of target, moderator, steel cladding and possible melting of these components.

#### 4. Fission products transmutation in irradiation devices

Among different FPs, greatly contributing to a long-lived activity, there is the practice of separating a group of several isotopes:  $^{99}\text{Tc}$ ,  $^{107}\text{Pd}$ ,  $^{93}\text{Zr}$ ,  $^{135}\text{Cs}$ ,  $^{129}\text{I}$ ,  $^{126}\text{Sn}$  and  $^{79}\text{Se}$ . The problems of transmuting these isotopes both in thermal and fast reactors have been considered in detail in many publications [2-4]. There is no longer any doubt that a more efficient transmutation of these nuclides is reached in a thermal or close to thermal neutron spectra, since the basic resonance of these FPs are just in this energy range.

Two aspects are considered in this report, connected with FP transmutation in fast reactors with special ID implementation, containing a large moderator quantity:

- The effect of different moderators on the FP transmutation efficiency.
- The effect of irradiation devices on major neutronic core characteristics.

We considered two possible ways to locate the IDs:

- In the first row of the radial blanket.
- Without the lower blanket.

##### 4.1 Technetium-99 transmutation

Transmutation efficiency.  $^{99}\text{Tc}$  half-life period is  $2.13 \times 10^5$  years, and its production in spent fuel of modern power reactors comprises 3.0 kg/TWh for fast reactors and 3.2 kg/TWh for thermal reactors.

Table 1 presents a comparison of  $^{99}\text{Tc}$  transmutation when using different moderators.

It should be noted that the introduction of hydrated moderators makes it possible to obtain the most efficient transmutation. The effect of volume moderator fraction on the transmutation rate and absolute value of  $^{99}\text{Tc}$  transmutation is presented in Table 2.

When increasing the moderator fraction up to 95%, it is possible to transmute up to 80% of initial loading of the ID during a once-through irradiation, thus providing ~8% per year. Higher transmutation rates (up to 25%/year) can be provided by an ID located in the axial blanket.

Table 1. Comparison of  $^{99}\text{Tc}$  transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TWh	%/cycle	kg/TWh
$\text{CaH}_2$	77.3	2.29	35.7	5.33
$\text{MgH}_2$	77.3	2.29	35.7	5.33
$\text{TiH}_2$	71.5	2.12	31.2	4.66
$\text{CeH}_3$	74.6	2.21	33.5	5.01
$\text{ZrH}_2$	72.1	2.14	31.7	4.73
Be	64.0	1.90	26.2	3.92
C	60.1	1.78	24.0	3.58
$\text{Be}_2\text{C}$	65.3	1.94	27.1	4.04
$\text{NbBe}_{17}$	63.5	1.88	25.9	3.87

Table 2. Effect of moderator volume fraction on  $^{99}\text{Tc}$  transmutation efficiency (radial blanket/axial blanket)

	Moderator volume fraction ( $\text{CaH}_2$ ), %			
	0	22	80	95
%/cycle	11.2/3.4	15.4/4.8	44.0/15.6	77.3/35.7
kg/TW*h	6.0/9.6	7.3/11.1	5.9/10.2	2.3/5.3

However, a small irradiation time (~1.5 year) allows to burn in one cycle only 35% of the loaded technetium.

Thus, the introduction of a moderator in ID allows an important increase in the transmutation rate. However, a decrease in this case of the total FP results in a decrease of absolute value of transmuted technetium.

It should be noted that even an essential decrease of the quantity of long-lived FPs as a result of the irradiation does still not solve the problem of the activity decrease. It is due to the build-up of other nuclides from which the radioactivity could exceed the one of the target nuclide. In the case implying  $^{99}\text{Tc}$ , this problem does not exist, since during the irradiation a short-lived  $^{100}\text{Tc}$  isotope and two stable ruthenium isotopes are produced. The calculation results are presented in Table 3.

Table 3. Different isotope contribution into activity after irradiation of  $^{99}\text{Tc}$ , Cu/kg

Isotope	Loading	Discharge	After cooling, year		
			3	100	1000
$\text{Tc}^{99}$	17.05	3.5	3.5	3.5	3.5
$\text{Tc}^{100}$	–	5.26 + 5	–	–	–
Total	17.05	5.26 + 5	3.5	3.5	3.5

Thus, the ID activity will be defined by  $^{99}\text{Tc}$  both before and after irradiation.

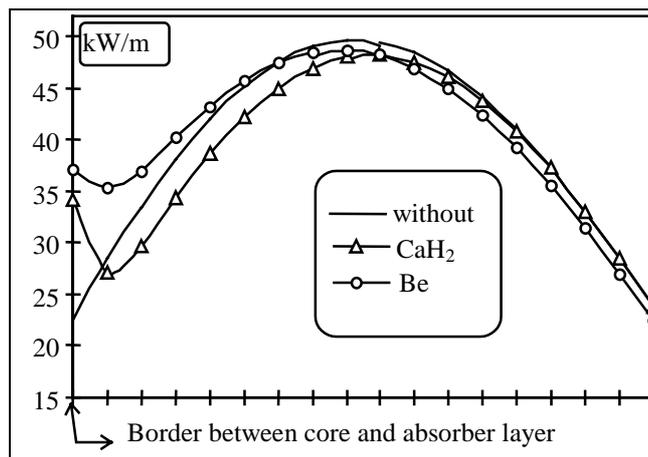
The presented results indicate a rather high efficiency of technetium transmutation in ID with a moderator. However the required transmutation efficiency should be determined from an economical point of view. Small technetium quantity transmuted in an irradiation cycle, even for a rather high transmutation rate, will require much more cycles of irradiated target processing, which increase irretrievable losses. Larger transmutation volumes, with low transmutation rate, require either a larger number of IDs in the reactor or an increase in reactor number, which should be loaded by IDs.

#### 4.2 The effect moderator in the blanket on core neutronic parameters

We will consider the effect of a moderator in the axial blanket on the core parameters for  $^{99}\text{Tc}$  transmutation.

First of all, we noted that the moderator location in the immediate vicinity of the core leads to the production of thermal neutrons in the moderator which re-enter the core, increasing sharply the fission rate and, therefore, the power in the nearest core layers. It is obvious in the Figure 9, which shows an axial power field in most fuel pins of the core.

Figure 9. Axial power distribution for various moderators in axial blanket

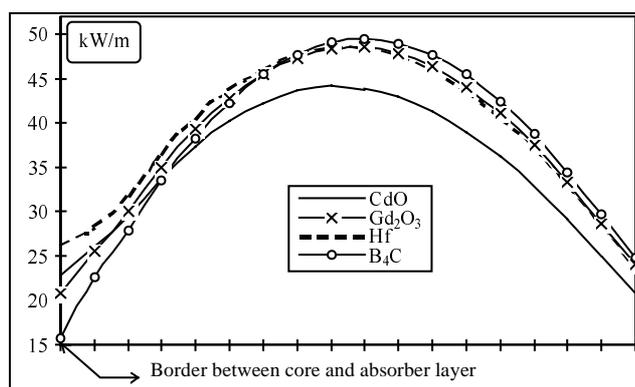


It is necessary in this case to pay attention to the fact that the use of hydrated moderators leads to a very sharp power increase in the region adjacent to the blanket. The power at the boundary between the core and the blanket increases to 80%.

Even greater power increases are observed when using a beryllium moderator, although in this case the power increase takes place smoothly along the height of the lower core half. The question if one of these cases is more dangerous from the standpoint of reactor safety requires the performance of detailed thermal-hydraulic calculations.

However, the effect of moderator on the power distribution can be essentially decreased by the introduction of an absorber layer between the core and the blanket with moderator. The Figures 9 and 10 present the axial power distributions when introducing a layer of different type absorber.

Figure 10. Axial power distribution for various absorber in layer between core and axial blanket (moderator CaH<sub>2</sub>)



These distributions show that best results are achieved when using an absorbing layer with cadmium oxide. In this case, not only the power at the boundary decreases but also the maximum power value decreases. We noted that when beryllium is used as a moderator, the effect of absorbers on the power fields is only observed in the immediate vicinity of the blanket.

The effect of the absorber introduction on transmutation efficiency and SVRE value are considered further on. The calculation results are presented in Table 4.

Table 4. The effect of absorber on transmutation efficiency and SVRE value

		Moderator CaH <sub>2</sub>				
Absorber	–	CdO	Gd <sub>2</sub> O <sub>3</sub>	Hf	B <sub>4</sub> C	
kg/TWh	5.29	4.59	3.36	3.79	1.90	
SVRE, %Δk/k	-0.169	+0.145	-0.100	-0.102	-0.311	
		Moderator Be				
Absorber	–	CdO	Gd <sub>2</sub> O <sub>3</sub>	Hf	B <sub>4</sub> C	
kg/TWh	3.76	3.56	2.13	2.38	1.79	
SVRE, %Δk/k	+0.339	+0.650	+0.171	+0.188	+0.109	

The results presented show that the introduction of CaH<sub>2</sub> as a moderator decreases SVRE value to ~-0.5%Δk/k comparing to the use of beryllium. The implementation of different absorbers changes the SVRE value in the limits of 0.5 %Δk/k.

Thus, the introduction of an absorber solves the power distribution problem and at the same time leads to a decrease in the FP transmutation efficiency. But if CdO is used as an absorber, the decrease of the transmutation efficiency does not exceed 15%.

### 4.3 Iodine-129 transmutation

$^{129}\text{I}$  has the largest among all long-lived wastes half-life period, equal to  $1.57 \times 10^7$  years, and its production amounts are about 0.7 kg/TWh for fast reactors and 0.66 kg/TWh for thermal reactors.

To consider  $^{129}\text{I}$  transmutation, it is necessary to choose the best chemical form for a material containing large quantities of  $^{129}\text{I}$ . Among different chemical compositions, three compositions are considered as target materials:  $\text{BeI}_2$ ,  $\text{NaI}$ ,  $\text{CeI}_3$ , the latter having the largest number of  $^{129}\text{I}$  nuclei [5]. Table 5 presents a comparison of  $^{129}\text{I}$  transmutation efficiency for different moderators and three chemical composition stabilising iodine

Table 5. A comparison of  $^{129}\text{I}$  transmutation for different chemical compositions

	<b>BeI<sub>2</sub></b>		<b>NaI</b>		<b>CeI<sub>3</sub></b>	
	kg/TWh	%/cycle	kg/TWh	%/cycle	kg/TWh	%/cycle
MgH <sub>2</sub>	0.71	29.3	1.17	28.6	1.25	28.8
ZrH <sub>2</sub>	0.71	29.2	1.17	28.6	1.25	28.8

It is necessary to pay attention to the fact that the introduction of  $\text{BeI}_2$  allows the assurance of a maximum transmutation rate, whereas introducing  $\text{CeI}_3$  provides the largest absolute transmutation efficiency. The results regarding the  $\text{BeI}_2$  composition are presented.

The comparison of  $^{129}\text{I}$  transmutation efficiency for implementation of different moderators is given in Table 6.

Table 6.  $\text{I}^{129}$  transmutation efficiency when using different type moderators

<b>Moderator</b>	<b>Radial blanket</b>		<b>Axial blanket</b>	
	%/cycle	kg/TWh	%/cycle	kg/TWh
CaH <sub>2</sub>	65.2	0.32	27.0	0.66
MgH <sub>2</sub>	68.7	0.33	29.3	0.71
TiH <sub>2</sub>	52.2	0.25	19.8	0.48
CeH <sub>3</sub>	67.8	0.33	28.7	0.70
ZrH <sub>2</sub>	68.6	0.33	29.2	0.71
Be	50.3	0.24	18.7	0.46
C	41.6	0.20	14.8	0.36
Be <sub>2</sub> C	51.3	0.25	19.3	0.47
NbBe <sub>17</sub>	47.7	0.23	17.6	0.43

Because the  $^{129}\text{I}$  capture cross-section has a clearly defined maximum in the thermal range, the best results are obtained when using hydrated moderators (MgH<sub>2</sub>, CeH<sub>3</sub>, ZrH<sub>2</sub>).

The effect of a moderator volume fraction on the transmutation rate and absolute quantity of iodine transmuted is presented in Table 7.

**Table 7. The effect of moderator volume fraction on  $^{129}\text{I}$  transmutation efficiency (radial blanket/axial blanket)**

	Moderator volume fraction ( $\text{ZrH}_2$ ) %			
	0	25	70	95
%/cycle	18.2/5.7	35.2/12.0	62.1/25.1	68.6/29.2
kg/TWh	1.8/2.3	2.7/4.6	1.3/2.6	0.33/0.71

An analysis of activity changing for  $^{129}\text{I}$  and the products of its irradiation is presented in Table 8.

**Table 8. Different isotope contribution to activity after  $^{129}\text{I}$  irradiation, Cu/kg**

Isotope	Loading	Discharge	After cooling, year		
			3	100	1000
$^{129}\text{I}$	0.18	0.05	0.05	0.05	0.05
$^{130\text{m}}\text{I}$	–	62 272.73	–	–	–
$^{130}\text{I}$	–	1.15+5	–	–	–
$^{131}\text{I}$	–	68.18	–	–	–
$^{131\text{m}}\text{Xe}$	–	595.45	–	–	–
$^{133\text{m}}\text{Xe}$	–	1.68	–	–	–
$^{133}\text{Xe}$	–	23,55	–	–	–
$^{134\text{m}}\text{Cs}$	–	0.8	0.64	–	–
$^{134}\text{Cs}$	–	1.75	–	–	–
Total	0.18	1.77+5	0.69	0.05	0.05

Contrary to  $^{99}\text{Tc}$ , an irradiation of  $^{129}\text{I}$  leads to the creation of e.g.  $^{134\text{m}}\text{Cs}$  while the activity of these will define the activity of irradiated targets activities during some time (~30 years). Nevertheless, the irradiation of iodine is also a rather efficient method to decrease the FP radioactivity. However, it is necessary to pay attention to the fact that the irradiation products are also gaseous xenon isotopes ( $^{130}\text{Xe}$ ,  $^{131}\text{Xe}$  and  $^{132}\text{Xe}$ ) creating a rather high pressure in the used targets.

#### 4.4 Palladium-107 transmutation

$^{107}\text{Pd}$  has a half-life period of  $6.5 \times 10^6$  years, and its production in the power reactor fuel amounts: 1.54 kg/TWh for fast reactors and 0.78 kg/TWh for thermal reactors.

The effect of different moderators on  $^{107}\text{Pd}$  transmutation efficiency is presented in Table 9.

Table 9.  $^{107}\text{Pd}$  transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TWh	%/cycle	kg/TWh
$\text{CaH}_2$	63.4	1.89	25.9	3.90
$\text{MgH}_2$	64.0	1.91	26.2	3.95
$\text{TiH}_2$	52.9	1.58	20.1	3.02
$\text{CeH}_3$	58.1	1.73	22.8	3.43
$\text{ZrH}_2$	57.1	1.71	22.3	3.36
Be	77.0	2.30	35.4	5.33
C	67.8	2.03	28.7	4.31
$\text{Be}_2\text{C}$	77.9	2.32	36.2	5.45
$\text{NbBe}_{17}$	74.6	2.23	33.5	5.05

Contrary to the technetium and iodine, the highest efficiency of palladium transmutation is provided by using beryllium containing moderators.

The effect of moderator volume fraction on the transmutation rate and absolute quantity of transmuted palladium is presented in Table 10.

Table 10. The effect of moderator volume fraction on  $^{107}\text{Pd}$  transmutation efficiency

	Moderator volume fraction( $\text{Be}_2\text{C}$ ) %			
	0	25	70	95
%/cycle	9.6/2.9	22.7/7.2	49.7/18.2	77.9/36.2
kg/TWh	5.8/8.7	10.6/16.9	6.5/12.0	2.32/5.45

The analysis results for palladium irradiation product activity are presented in Table 11.

Table 11. Different isotopes contribution to activity after  $^{107}\text{Pd}$  irradiation, Cu/kg

Isotope	Loading	Discharge	After cooling, year		
			3	100	1 000
$^{107}\text{Pd}$	0.51	0.11	0.11	0.11	0.11
$^{108}\text{Ag}$	–	0.33	–	–	–
$^{108\text{m}}\text{Pd}$	–	5641.60	–	–	–
$^{108}\text{Pd}$	–	77589.41	–	–	–
$^{109\text{m}}\text{Ag}$	–	3.53+5	–	–	–
$^{110\text{m}}\text{Ag}$	–	1.31+5	632.00	–	–
$^{110}\text{Ag}$	–	3.30+5	8.85	–	–
$^{111\text{m}}\text{Pd}$	–	5.0	–	–	–
$^{111}\text{Pd}$	–	31.76	–	–	–
$^{111}\text{Ag}$	–	32.4	–	–	–
$^{111\text{m}}\text{Cd}$	–	102.8	–	–	–
$^{113\text{m}}\text{Cd}$	–	0.73	0.63	–	–
$^{115}\text{Cd}$	–	2.87	–	0.01	–
Total	0.51	1.06+6	642.1	0.12	0.11

The results presented show that the irradiated palladium activity increases several orders due to the creation of short-lived nuclides ( $^{110\text{m}}\text{Ag}$ ,  $^{110}\text{Ag}$ ), which then falls quickly, and after approximately 30 years the targets activity will be again defined by palladium only.

#### 4.5 Caesium-135 transmutation

$^{135}\text{Cs}$  has a half-life time of  $2.3 \times 10^6$  years, and its production in the power reactor fuel amounts: 3.7 kg/TWh for fast reactors and 1.4 kg/TWh for thermal reactors.

When analysing the calculation results for possibility to transmute  $^{135}\text{Cs}$  in the targets with a moderator, as presented in Table 12, a conclusion can be made that in case of  $^{135}\text{Cs}$  irradiation the choice of moderator does not play an important role, since all moderators give approximately the same results.

It should be noted that the caesium transmutation rate is somewhat lower compared to the nuclides considered above, which is explained by a lower capture cross-section.

Table 12. <sup>135</sup>Cs transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TWh	%/cycle	kg/TWh
CaH <sub>2</sub>	44.7	0.40	16.2	0.72
MgH <sub>2</sub>	44.8	0.40	16.2	0.72
TiH <sub>2</sub>	33.6	0.30	11.5	0.51
CeH <sub>3</sub>	39.0	0.35	13.7	0.61
ZrH <sub>2</sub>	40.2	0.36	14.2	0.64
Be	40.3	0.36	14.2	0.64
C	41.3	0.37	14.7	0.65
Be <sub>2</sub> C	47.3	0.42	17.4	0.78
NbBe <sub>17</sub>	42.7	0.39	15.3	0.68

Besides, other caesium isotopes (<sup>133</sup>Cs, <sup>137</sup>Cs etc.) are accumulated in power reactor spent fuel and the Cs<sup>135</sup> fraction is ~10% only. When transmuting caesium without chemical isotope separation, the transmutation efficiency decreases one order due to creation secondary <sup>135</sup>Cs.

Thus, the results presented show that the issue on advising to transmute <sup>135</sup>Cs in reactor conditions remains open.

The transmutation of <sup>93</sup>Zr ( $T_{1/2} = 1.53 \times 10^6$  years, production 1.74 kg/TWh for fast reactors 2.8 kg/TWh for thermal reactors) was not considered in detail in this report due to large uncertainties in its nuclear data.

The transmutation of such elements <sup>79</sup>Se ( $T_{1/2} = 65\ 000$  years) and <sup>126</sup>Sn ( $T_{1/2} = 10^5$  years) is not considered because of their low transmutation rate.

The analysis results allow forming the final Table 13.

Table 13. A comparison of transmutation rate for different FP and their production in power reactors.

Isotope	T <sub>1/2</sub>	Production in reactors, kg/TWh	Transmutation efficiency kg/TWh	
			without moderator	with moderator
<sup>99</sup> Tc	$2.13 \times 10^5$	3.0/3.2	10.6	5.3
<sup>107</sup> Pd	$6.5 \times 10^6$	1.54/0.78	5.5	3.0
<sup>135</sup> Cs	$2.3 \times 10^6$	3.70/1.4	5.1	0.8
<sup>129</sup> I	$1.57 \times 10^7$	0.70/0.66	5.3	0.7
<sup>79</sup> Se	$6.5 \times 10^4$	0.03/0.02	0.09	0.01
<sup>126</sup> Sn	$10^5$	0.15/0.08	0.2	0.04
<sup>93</sup> Zr	$1.53 \times 10^6$	1.74/2.8	2.3	1.4

Thus the transmutation of such FPs  $^{99}\text{Tc}$ ,  $^{107}\text{Pd}$  and  $^{129}\text{I}$  with the use of moderators in IDs is rather proved, since their transmutation efficiency exceeds their production. The transmutation of  $^{135}\text{Cs}$ ,  $^{93}\text{Zr}$  in such blanket can turn out to be not useful, and a further optimisation of the moderator quantity is necessary. Moreover,  $^{135}\text{Cs}$  will require its separation from other Cs isotopes produced in spent fuel.

## 5. Conclusion

The considered method for radioactive wastes (actinides and fission products) transmutation in special irradiation devices containing large moderator quantity has essential advantages over the homogeneous method for these wastes to be recycled.

The results presented have shown that to decrease the americium radiotoxicity significantly, a very high actinide burn-up (up to 95% h.a.) should be achieved in these irradiation devices. In this case up to 60 kg of americium per year will be destroyed. Such quantity is accumulated presently in all VVER-100 reactors in Russia.

But this method faces large difficulties in realising such core with moderated sub-assemblies. It is necessary to take special measures to avoid the large power increase in moderated sub-assembly during irradiation.

The major long-lived fission products can be also transmuted in such irradiation devices. However, isotope separation will be needed to increase the transmutation efficiency of  $^{135}\text{Cs}$  and  $^{93}\text{Zr}$  isotopes, for example. The major advantage of the use of this fission product utilisation concept consists in the decrease of total losses in each step of waste reprocessing. Nevertheless, a serious contradiction should be pointed out between the transmutation rate and absolute quantity of utilised fission products.

The influence of irradiation devices with a moderator on some core neutronic parameters has been considered. And it has been shown that the implementation of absorbing blankets, made from cadmium oxide, will allow an essential decrease in the effect of such devices on the core parameters, decreasing slightly the transmutation efficiency.

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## **ASSESSMENT OF NUCLEAR POWER SCENARIOS ALLOWING FOR MATRIX BEHAVIOUR IN RADIOLOGICAL IMPACT MODELLING OF DISPOSAL SCENARIOS**

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### **Abstract**

The innovative scientific contribution of this study is to consider a third type of radiotoxic inventory: the potential radiotoxic inventory after conditioning, i.e. taking into account the containment capacity of the radionuclide conditioning matrices. The matrix fraction subjected to alteration over time determines the potential for radionuclide release, hence the notion of the potential radiotoxic inventory after conditioning. An initial comparison of possible scenarios is proposed by considering orders of magnitude for the radionuclide containment capacity of the disposal matrices and for their mobilisation potential. All the scenarios investigated are normalised to the same annual electric power production so that a legitimate comparison can be established for the ultimate waste forms produced per year of operation.

This approach reveals significant differences among the scenarios considered that do not appear when only the raw potential radiotoxic inventory is taken into account. The matrix containment performance has a decisive effect on the final impact of a given scenario or type of scenario. Pu recycling scenarios thus reduce the potential radiotoxicity by roughly a factor of 50 compared with an open cycle; the gain rises to a factor of about 300 for scenarios in which Pu and the minor actinides are recycled. Interestingly, the results obtained by the use of a dedicated containment matrix for the minor actinides in a scenario limited to Pu recycling were comparable to those provided by transmutation of the minor actinides.

## 1. Introduction

Under the provisions of the “separation-conditioning” option of the strategy and program defined under research topic 1 of the 1991 French radioactive waste management law, various fuel cycle scenarios will be assessed and compared [1] in terms of feasibility, flexibility, cost, and ultimate waste radiotoxic inventory. The latter criterion may be further broken down into “potential radiotoxic inventory” (the radiotoxic inventory of all the radionuclides produced) and “residual radiotoxic inventory” (the radionuclide fraction reaching the biosphere after migration from the repository).

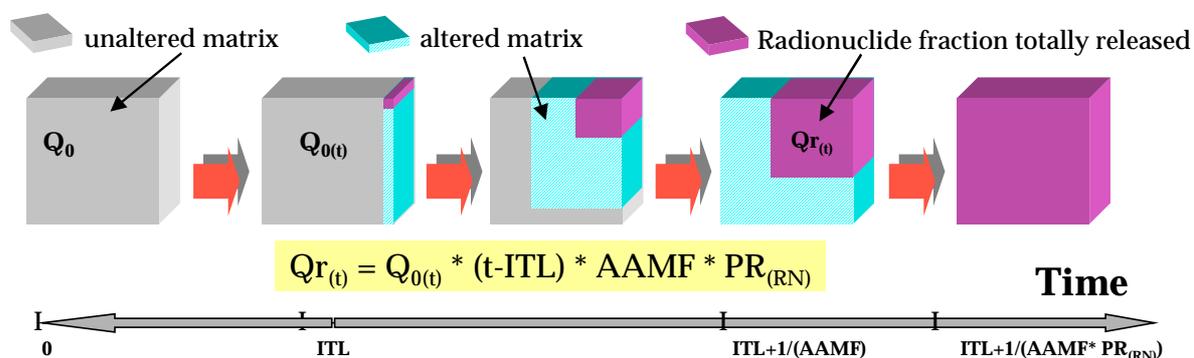
The innovative scientific contribution of this study is to consider a third type of radiotoxic inventory: the potential radiotoxic inventory after conditioning, i.e. taking into account the containment capacity of the radionuclide conditioning matrices. The source term therefore includes only the effects of the radionuclides released from the altered matrix. The matrix fraction subjected to alteration over time determines the potential for radionuclide release, hence the notion of their potential radiotoxic inventory after conditioning. The impact of the radionuclides on the human population is considered through ingestion alone, and not by inhalation.

## 2. Allowance for matrix containment capacity

The ultimate containment matrices in the various scenarios considered included *uranium oxide* in spent fuel and uranium ore, *glass* in waste packages containing fission products (FP) and minor actinides (MA), *new containment matrices* (NCM) or new glass compositions, and fuel assembly *structural materials* or compacted hulls and end-fittings containing structural activation products (SAP).

The potential release is taken into account by means of three parameters, as shown in Figure 1. The first parameter is the *integrity time limit* (ITL) after which matrix alteration begins; the ITL could correspond to the lifetime of the container (e.g. zircaloy cladding or steel package). The second is the matrix alteration rate, described by the *annual altered matrix fraction* (AAMF), which is constant over time; the reciprocal of the AAMF thus corresponds to the matrix lifetime. The third parameter involves the inherent behaviour of each radionuclide, as expressed by the *probability* that a particular radionuclide will be *released* from the altered portion of the matrix:  $PR_{(RN)}$ .

Figure 1. Schematic representation of radionuclide release over time



The equation postulates that the radiotoxic inventory of the radionuclides released from the matrix  $Q_{r(t)}$  is a fraction of the inventory over time in a closed system  $Q_{0(t)}$ : the fraction is the integral over time since the ITL of the product of the annual altered matrix fractions by the radionuclide

release probabilities. The time distribution of the radionuclide inventory in a matrix is thus a uniform and constant function of ITL up to  $ITL + 1/(AAMF \times PR_{(RN)})$ . The parameter values are indicated in Table 1.

The integrity time limit is first assumed constant for all the packages considered. The AAMF values for spent fuel and for glass reflect the minimum performance corresponding to their maximum leach rates; the figure for NCM represents a target value corresponding to 100 times better containment than currently estimated for glass. Uranium mine tailings were also added as a reference to assess the possible evolution of the equivalent of uranium ore under the same disposal conditions as the other matrices. The quantity considered was the natural uranium requirement necessary for one year of operation of a reactor population at equilibrium; the uranium was assumed to be at equilibrium with its decay products, and was taken into account here in oxide form.

With regard to their radionuclide release probability factors, the actinides characterised by their low mobility were assigned a value of  $10^{-2}$  over duration equal to  $1/AAMF$ . The release of the highly mobile fission products (iodine, caesium and technetium) was considered congruent with the matrix alteration, hence their  $PR_{(RN)}$  value of 1. The other less characteristic chemical elements were assigned intermediate AAMF values.

Table 1. **Parameter values for the radionuclide release equation:**  
(**boldface figures are the product of  $AAMF \times PR_{(RN)}$** )

Matrix	ITL (years)	AAMF (year <sup>-1</sup> )	$PR_{(RN)}$							
			Pu (10 <sup>-2</sup> )	Am (10 <sup>-2</sup> )	U (10 <sup>-2</sup> )	I (1)	Tc (1)	Cs (1)	Other FP (10 <sup>-1</sup> )	SAP (10 <sup>-1</sup> )
Uranium oxide	300	10 <sup>-4</sup>	10 <sup>-6</sup>	<b>10<sup>-6</sup></b>	<b>10<sup>-6</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-5</sup></b>	
Glass	300	10 <sup>-5</sup>	<b>10<sup>-7</sup></b>	<b>10<sup>-7</sup></b>	<b>10<sup>-7</sup></b>	<b>10<sup>-5</sup></b>	<b>10<sup>-5</sup></b>	<b>10<sup>-5</sup></b>	<b>10<sup>-6</sup></b>	
New containment matrices	300	10 <sup>-7</sup>	<b>10<sup>-9</sup></b>	<b>10<sup>-9</sup></b>	<b>10<sup>-9</sup></b>	<b>10<sup>-7</sup></b>	<b>10<sup>-7</sup></b>	<b>10<sup>-7</sup></b>	<b>10<sup>-8</sup></b>	
Structural materials	300	10 <sup>-4</sup>	<b>10<sup>-6</sup></b>	<b>10<sup>-6</sup></b>	<b>10<sup>-6</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-4</sup></b>	<b>10<sup>-5</sup></b>	<b>10<sup>-5</sup></b>

### 3. Application to fuel cycle scenarios

#### 3.1 Scenarios

All the scenarios considered were normalised with respect to an electric power production of 400 TWh/year. The scenarios also assumed quasi steady-state operation to allow valid comparisons of the ultimate waste production over one year of operation in each case. The twelve scenarios taken into account are briefly described below, and can be considered as belonging to four major types:

- *Scenarios resulting in large quantities of plutonium and minor actinides in the waste materials.* Open-cycle scenario, and once-through-Pu scenario in which plutonium is recycled once as MOX fuel without further reprocessing.
- *Scenarios eliminating the plutonium from the waste materials.* FNR-Pu, PWR/FNR-Pu, and PWR-Pu scenarios, in which plutonium is recycled repeatedly either as MOX fuel in pressurised water reactors (PWR) or fast neutron reactors (FNR), or as

PWR MIX (MOX with enriched uranium) fuel; a variant with isotopic separation of  $^{242}\text{Pu}$  was also considered: IS-Pu242.

- *Scenarios eliminating the plutonium from the waste materials, with separation and specific conditioning of the minor actinides.*

These are variants of the preceding scenarios with implementation of enhanced separation and conditioning (SC) techniques. The fuel is reprocessed in an enhanced reprocessing plant using the new DIAMEX and SANEX processes to separate americium (Am) and/or curium (Cm) for incorporation in a new containment matrix (NCM) with very high radionuclide retention performance (Table 1). The vitrified waste therefore contains only fission products (except for process losses). These scenarios are designated: PWR-Pu/MA-SC, FNR-Pu/MA-SC, and PWR/FNR-Pu/MA-SC.

- *Scenarios eliminating the plutonium and some or all of the minor actinides from the waste materials for transmutation in PWRs or FNRs.*

These are the PWR-Pu/MA, PWR-Pu/NpAm (Cm is sent to vitrification), FNR-Pu/MA, and PWR/FNR-Pu/MA scenarios in which the minor actinides are transmuted in homogeneous mode, and the PWR/FNR-AmCm-target and PWR/FNR-Am-target scenarios in which the actinides are transmuted in heterogeneous mode as once-through targets; after irradiation, 90% of the minor actinides are transmuted into fission products.

### 3.2 Fuels

The burn-up is assumed equal to  $60 \text{ GWd}\cdot\text{t}^{-1}$  for all the PWR fuels, and approximately  $140 \text{ GWd}\cdot\text{t}^{-1}$  for the fast neutron reactor fuels. The U and Pu reprocessing losses are assumed equal to 0.1%. All the fuel compositions and their annual flows are determined by neutronic feasibility analysis, calculated by the Reactor and Fuel Cycle Physics Department of the CEA's Nuclear Reactor Division (DRN/SPRC) [2]. The comparisons were performed under steady-state conditions based on the total annual production.

The potential radiotoxic inventory of the ultimate wasteforms over time were calculated by multiplying the activities of each radionuclide (determined by decay using the JEF2.2 data [3]) by the dose-per-unit-intake factors ( $\text{Sv}\cdot\text{Bq}^{-1}$ ) from ICRP72 [4].

## 4. Results

### 4.1 Mass balance and "raw" potential radiotoxic inventory

The mass balance was established for the fission products, minor actinides and plutonium released from the waste (Table 2). The structural activation products have a lower radioactive and radiotoxic impact. The reprocessed uranium is not considered as an ultimate waste form, unlike the uranium reprocessing losses (estimated at 0.1%).

Table 2. Annual heavy nuclide contribution (kg/year) to ultimate waste form for each scenario

Recycling policy	No recycling	Partial Pu	Pu recycling			
Scenario	Open cycle	Once-through Pu	Isotopic separation <sup>242</sup> Pu	MIX Pu	FNR Pu	PWR/FNR Pu
<b>U</b>	754 736	79 105	743	747	238	440
<b>Pu</b>	10 332	3 175	2 021	17	56	35
<b>Np</b>	746	683	632	674	172	499
<b>Am</b>	645	1 229	1 852	1 853	1 425	1 406
<b>Cm</b>	113	286	360	951	112	196
<b>Total (excl. U)</b>	11 835	8 373	4 865	3 495	1 776	2 136

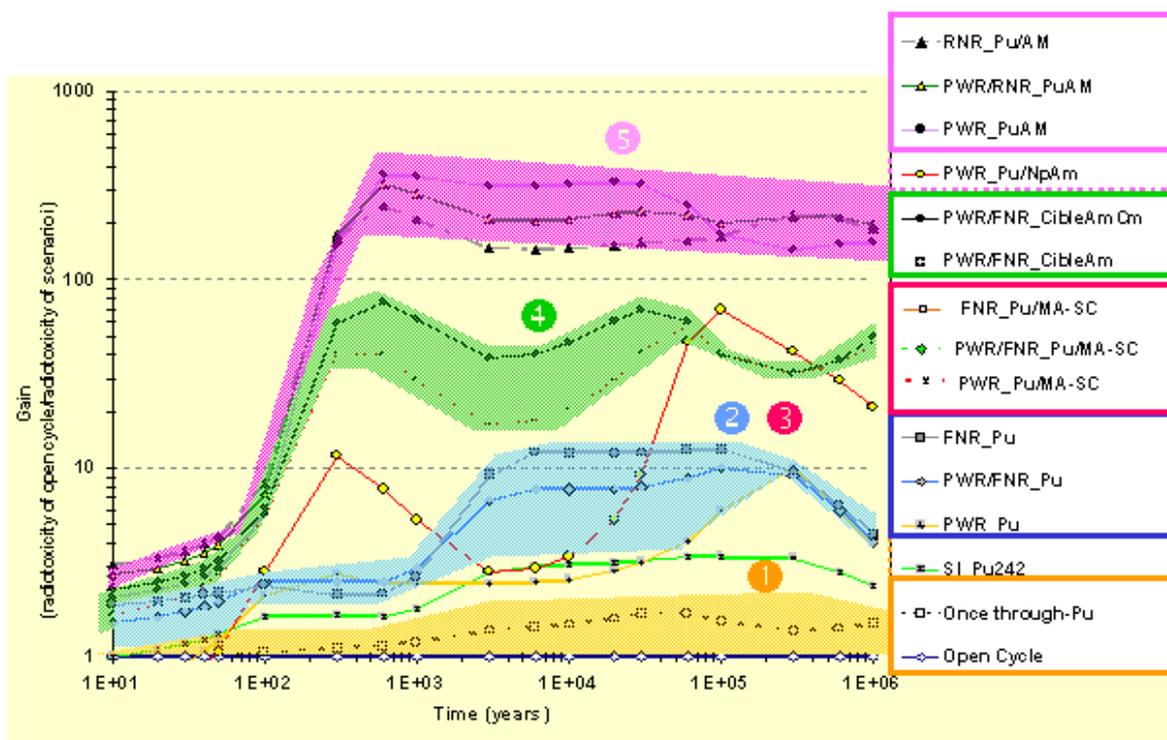
Recycling policy	Pu + MA recycling					
Scenario	MIX Pu + MA	MIX Pu + AmNp	PWR/FNR Am/Cm targets	PWR/FNR Am targets	PWR/FNR Pu + MA	FNR Pu + MA
<b>U</b>	735	720	437	438	378	236
<b>Pu</b>	25	22	96	95	39	58
<b>Np</b>	1	1	1	1	1	1
<b>Am</b>	3	3	16	15	2	2
<b>Cm</b>	3	1 668	94	252	1	1
<b>Total (excl. U)</b>	32	1 694	207	363	43	62

When the raw radiotoxic inventory results are plotted relative to the “open-cycle” reference scenario (Figure 2), three main categories of scenarios can be distinguished.

- The first includes the scenarios with significant quantities of residual plutonium (open and once-through cycles).
- The second, with a potential radiotoxic inventory 4 to 10 times lower, comprises the multiple plutonium recycling scenarios (MIX-Pu, FNR-Pu, and PWR/FNR-Pu).
- The third includes the scenarios in which both plutonium and the minor actinides are recycled (MIX-Pu/MA, FNR-Pu/MA, and PWR/FNR-Pu/MA), resulting in a potential radiotoxic inventory some 100 times lower than in the reference scenario.

*Note: From the standpoint of the potential radiotoxic inventory, there are no differences between the basic scenarios and the variants involving a separation and conditioning strategy.*

Figure 2. Reduction of potential raw radiotoxic inventory for each scenario compared with open cycle



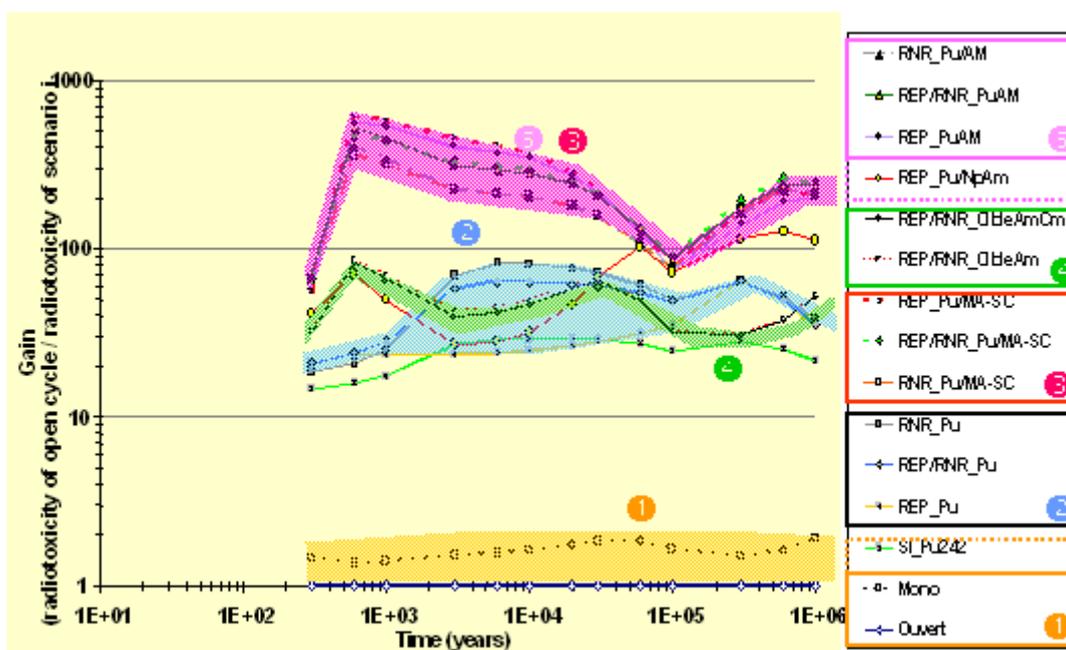
#### 4.2 Potential radiotoxic inventory after conditioning

Allowing for the containment capacity of the conditioning matrix, the logical outcome of the processes implemented in spent fuel reprocessing, provides a new basis for comparing the possible scenarios.

The matrix containment performance has a decisive influence on the final impact of a given scenario or group of scenarios. The Pu recycling scenarios provide a gain by a factor of about 50 over the open cycle in terms of the potential radiotoxic inventory after conditioning; the scenarios in which both plutonium and minor actinides are recycled result in a gain by a factor of about 300.

It is interesting to note that specific conditioning of the minor actinides in high-performance containment matrices in a scenario in which Pu alone is recycled would be as effective as transmuting the minor actinides. Moreover, the effectiveness of the scenarios in which the minor actinides are recycled as once-through targets would be no better, under the hypothetical conditions of this study, than recycling plutonium alone, as the glass matrix provides better containment than the unprocessed targets.

Figure 3. Reduction of potential radiotoxic inventory after conditioning for of each scenario compared with open cycle



## 5. Conclusion

The notion of the radiotoxic inventory after conditioning, by taking into account the respective containment properties of each ultimate wasteform, provides a means for distinguishing three categories of fuel cycle management routes according to the potential release of the radiotoxic inventory.

Fuel cycle management strategies in which plutonium is recycled partially or not at all yield the poorest performance; multiple plutonium recycling strategies are about 50 times more effective in this respect, and multiple recycling of plutonium and the minor actinides is even more effective (some 300 times more than the open cycle).

Enhanced reprocessing together with the use of dedicated matrices having a containment capacity 100 times better than glass taken into account in this study would result in performance factors equivalent to those of an enhanced reprocessing/transmutation cycle without requiring the use of burner reactors.

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**DISPOSAL OF PARTITIONING-TRANSMUTATION WASTES  
WITH SEPARATE MANAGEMENT OF HIGH-HEAT RADIONUCLIDES**

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**Abstract**

An alternative approach is proposed for disposing of partitioning-transmutation (P&T) wastes to (1) reduce repository costs and (2) improve repository performance. Radioactive decay heat controls the size and cost of the repository. It is proposed that P&T wastes be separated into a high-heat radionuclide (HHR) fraction and a very-low-heat-radionuclide (VLHR) fraction to bypass this repository design constraint. There are five repository HHRs in spent nuclear fuel: caesium, strontium, plutonium, americium, and curium. P&T, by destroying the long-lived HHRs (plutonium, americium, and curium), is an enabling technology for separate low-cost disposal of the remaining HHRs ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ), which have limited half-lives ( $T_{1/2} = \sim 30$  year), small volumes, and high heat-generation rates. These characteristics allow the use of lower-cost disposal methods for these HHR wastes. Eight HHR disposal options are identified and described. With the removal of the HHRs, there are lower-cost, higher performance methods for disposal of the remaining VLHRs.

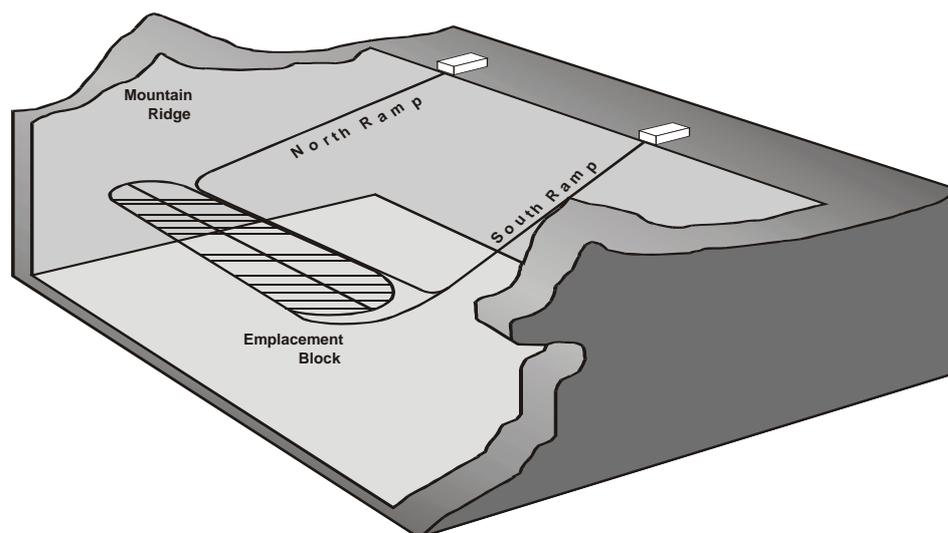
## 1. Introduction

Repository design and performance are primarily controlled by radioactive decay heat. Consider the proposed Yucca Mountain (YM) repository [1] in the United States. It is designed for ~70,000 t of spent nuclear fuel (SNF) and high-level waste (HLW). If there were no radioactive decay heat, the entire volume could be placed in a cube, which would be ~30 m on each side. The cost of such a repository would be very low. However, radioactive waste generates heat. To ensure repository performance, the repository temperatures are limited. The temperature is limited by packaging the wastes in ~11,000 long-lived, expensive waste packages (WPs) and dispersing the WPs over 100 km of tunnels. The repository program will cost several tens of billions of dollars.

From a distance, a schematic of the proposed YM repository (Figure 1) appears as a large planar structure—like a horizontal underground car radiator. This typical characteristic of geological repositories is a consequence of the need to limit repository temperatures and dissipate decay heat. If waste partitioning and transmutation (P&T) is to have a major impact on the repository cost, it must be by changing how decay heat is managed in a repository.

Figure 1. Schematic of the proposed YM repository

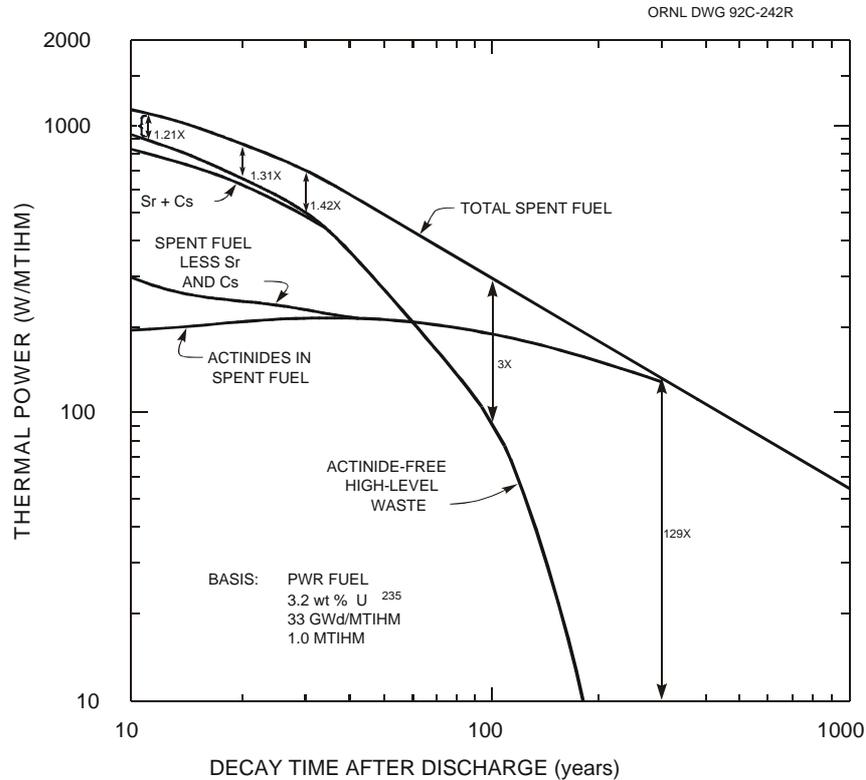
ORNL DWG 2000-272



## 2. Radioactive decay heat: sources and impacts

There are several temperature limits [2] on the repository: (1) waste-form limit, (2) package limit, (3) near-field rock limit, and (4) various far-field limits. Each limit is imposed to prevent damage to one or more barriers to radionuclide migration from the waste form to the accessible environment. Almost all repository decay heat from SNF (Figure 2) is produced from five elements: caesium ( $^{137}\text{Cs}$ ), strontium ( $^{90}\text{Sr}$ ), plutonium (Pu: multiple isotopes), americium (Am: multiple isotopes), and curium (Cm: multiple isotopes). While there are other heat-generating radionuclides, these decay away quickly. These high-heat radionuclides (HHRs) can be divided into two categories: shorter-lived HHRs ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ) and long-lived HHRs (Pu, Am, and Cm).

Figure 2. Decay heat from SNF



The temperature limits in and near the WP are controlled by decay heat from the shorter-lived HHRs –  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . The long-term temperature limits far from the WP are usually controlled by the longer-lived actinides. It takes a significant amount of decay heat over a long time to heat large quantities of rock to unacceptable temperatures. The removal of either the shorter-lived or longer-lived HHRs radionuclides from the waste provides some benefits to the repository, but the benefits are limited because both sets of radionuclides impose temperature limits on the repository—one set in the near term and the second set in the longer term.

If the HHRs are removed from the waste, alternative repository design options [3] exist that significantly reduce the size and cost of a repository. *A large repository is replaced with a mini-repository, and the size of the mini-repository is controlled by the fraction of the HHRs that are not removed.* For this to occur, alternative methods for management of the HHRs are required.

- *Long-lived HHRs.* This conference is examining P&T of actinides, including Pu, Am, and Cm. If the P&T technology is successful and economically viable, this approach can be used to destroy these troublesome HHRs.
- *Shorter-lived HHRs.* The  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  must be separately managed. Because the characteristics of shorter-lived HHR wastes are different than those of HLW and SNF, low-cost disposal methods may be available. These HHRs differ from SNF and HLW in four ways (characteristics):
  - *Half-life.* The short half-life ( $T_{1/2} \approx 30$  years) allows the use of options that are safe for disposal of these materials but that would be difficult to demonstrate as safe over geological times if disposing of SNF or HLW with their large inventories of long-lived radionuclides.

- *Waste volume.* The quantities of HHRs (caesium and strontium) are small. One tonne of 40 000-MWd light-water reactor (LWR) SNF contains 4.1 kg of caesium and strontium.
- *Heat-generation rate.* High heat-generation rates create options that require decay heat to function.
- *Fissile content.* These wastes include no fissile materials, and thus there are no safeguards or criticality concerns.

There is experience [4] in separating and packaging shorter-lived HHRs from HLW. Over  $10^8$  Ci of these HHRs were separated from defence HLW at Hanford, Washington, to minimise the cost of storing HLW in tanks. The decay heat, not the tank volume, limited tank capacity. The HHRs were packaged in 6.67-cm-diam capsules.

### **3. Management of shorter-lived HHRs**

There are many methods to manage  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . The method selected by any nation will depend upon institutional factors and the geology available to each nation to manage such wastes. *Near term and more speculative options* are described herein to emphasise that when the characteristics of the waste change, the disposal options change. This is an area of waste management where very few investigations have been conducted; thus, many of these options are not well understood. In parenthesis are the characteristics of the short-lived HHRs that are important for the disposal option.

#### **3.1 Long-term storage (half-life)**

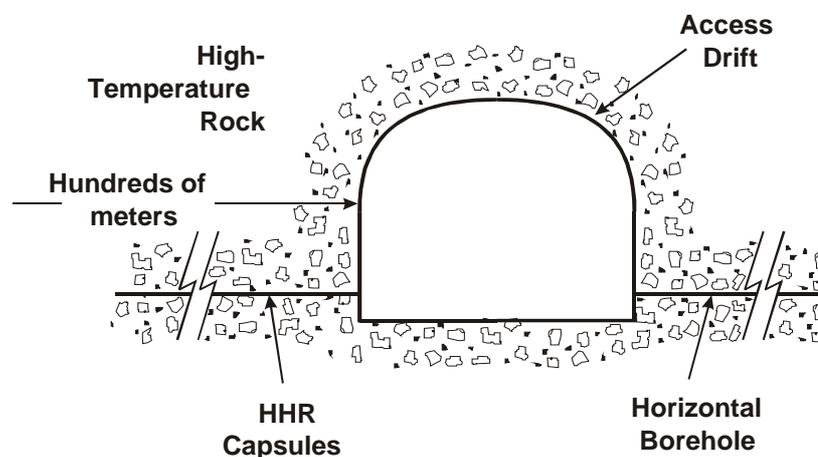
The HHR wastes can be placed in long-term, dry-storage facilities, which are similar to those used for HLW and SNF. After the decay of most of the HHRs, the wastes can be disposed of in the repository.

#### **3.2 Extended dry repository (waste volume, heat-generation rate, half-life)**

The HHR capsules could be disposed of in a separate section of a dry repository above the water table [5]. The proposed YM repository in the United States is of this type, and thus this is a potential option for the United States. Long boreholes would be drilled into the rock from a central tunnel and then filled with small-diameter HHR capsules. The heat load would be controlled by placing low-volume HHR capsules in small-diameter horizontal boreholes (<15 cm in diameter) rather than placing large, HLW or SNF WPs in 5.5-m-diam disposal tunnels. The holes could be drilled in a horizontal plane (Figure 3) or in a vertical array. Boreholes are less expensive than tunnels.

Figure 3. Shorter-lived HHR repository with boreholes (rather than tunnels) used to distribute decay heat load radionuclide

ORNL DWG 99C-391R



The HHR section of the repository would be designed as an “extended-dry” repository in unsaturated rock. By placing the boreholes closely together to obtain higher local heat loads and higher local temperatures (but sufficiently apart to avoid HHR capsule damage), the local rock temperature would be above the boiling point of water for thousands of years. If the rock temperature is above the boiling point of water, there can be no groundwater flow near the capsules and no migration of radionuclides in groundwater. The shorter-lived HHRs decay before the high-heat section of the repository cools below the boiling point of water. The large heat capacity of the rock maintains higher temperatures for extended periods of time. The need for high temperatures requires closer spacing of the HHRs than is used for SNF and HLW; and thus a correspondingly smaller repository section for these wastes.

The YM repository project investigated SNF extended-dry repository concepts [6] because of economic advantages. Such concepts have not been adopted for SNF or HLW because of the uncertainties in predicting long-term, extended-dry repository behaviour after the repository cools. These uncertainties do not exist for HHRs that decay before the high-heat section of the repository cools down.

### 3.3 Conventional repository (half-life, waste volume)

The HHR wastes could be disposed of in a conventional repository. However, the repository size and cost may be significantly reduced. Boreholes, not tunnels, are needed for HHR placement. Elimination of long-term heat-decay loads and most long-term performance requirements (>1 000 years) allows the use of simpler WPs and other simplifications.

### 3.4 Saltdiver (heat-generation rate, half-life)

Natural salt domes contain relatively pure salt in the shape of a mushroom with diameters measured in kilometers. The vertical dimension may be as large as 10 000 m. The saltdiver repository [3] uses the high-heat generation rates of HHR capsules to allow disposal at depths up to 10 000 m

underground in salt domes. The HHRs are packaged into moderately large containers (saltdivers) that are placed in a salt dome. The high-density, high-temperature WPs sink by heating the salt under the WP until the salt becomes plastic or melts (at 800°C).

### **3.5 *Rock melt (heat-generation rate, half-life)***

In the melt-rock repository [7], a large, spherical, underground cavity would be constructed several hundred to several thousand meters underground. Large quantities of HHRs would be placed in the cavity. During loading operations, active cooling systems control temperatures. After the cavity is loaded, the cavity would be sealed, and the cooling systems would be shut off. The HHRs would melt and then melt the surrounding rock. The radionuclides would then be incorporated into the molten rock. It is large-scale vitrification of waste. Ultimately, as the decay-heat levels decrease, the molten rock would solidify into solid rock.

During periods of high-temperature operations, the high temperatures cause plastic deformation of the rock beyond the melt zone, which seals all cracks. Several uncertainties [8] have been identified with this disposal option. However, the identified uncertainties apply only to HLW and SNF that contain long-lived radionuclides, not disposal of shorter-lived HHRs. Further analysis would be required to determine if there are unidentified failure modes when disposing of HHRs.

### **3.6 *Borehole (waste volume)***

The use of deep vertical boreholes (>5 km) has been considered for the disposal of various radioactive wastes. However, a major drawback is that a borehole has very limited volume. Drilling deep, wide boreholes is expensive. For short-lived HHRs, the volumes are very small; thus, this may be a viable low-cost option for these specific wastes.

### **3.7 *Seabed (waste volume, half-life)***

International programs [9] have investigated seabed disposal of SNF and HLW. Seabed disposal involves placing WPs into the clay layer, which covers most of the ocean's seabed. The clay layer has potentially excellent waste-isolation properties, and the ocean provides an independent backup mechanism (ocean dilution) if there were failures. There are major institutional problems and some technical problems associated with this option.

Demonstration of disposal viability of short-lived HHRs would be simpler than for other wastes because the shorter-lived HHRs remain hazardous for a much shorter period of time. Furthermore, there are no fissile materials associated with HHRs. Recent analysis has raised questions about the viability of disposal of wastes with fissile materials using this technology. New off-shore oil recovery technologies are making it increasingly easier to recover objects from the ocean seabed; thus, there is a concern about the recovery of any fissile materials by unknown parties if the disposal site is the ocean seabed.

### **3.8 *Shallow-land disposal, half-life***

The limited half-life may allow shallow-land disposal of the shorter-lived HHRs under some circumstances [3].

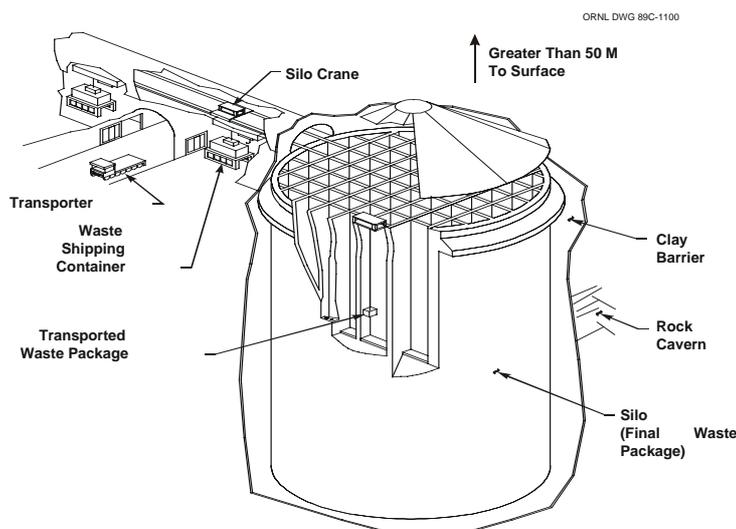
#### 4. Management of low-heat, long-lived radionuclides

With most of the HHRs removed, the repository for the remaining wastes becomes a small facility [3,5]. The required repository would contain two sections: a section for wastes with significant decay heat and a section for the very-low-heat radionuclides (VLHR). Existing vitrified HLW and some P&T target wastes (deep-burn, once-through targets; certain target-processing wastes) would be disposed of in a repository section similar to existing repository designs. Because of the small quantities of these wastes, this repository section would be relatively small.

The wastes from processing light-water reactor SNF, after removal of the shorter-lived ( $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ) and long-lived (Pu, Am, and Cm) HHRs would be a VLHR waste. These wastes may be disposed of in a few lower-cost, high-performance silos without exceeding temperature limits. Depending upon the geology and efficiency of removal of HHRs, such a silo might accept the wastes from up to 10 000 tonnes of SNF.

There is experience with waste silos [10]. Sweden (Figure 4) and Finland have constructed and are operating underground silos for the disposal of intermediate-activity wastes. The heat-generating characteristics of these wastes are somewhat similar to VLHR wastes. The Swedish waste silos are about 50 m high and 25 m in diameter. The costs per unit volume are a fraction of the cost of traditional WPs.

Figure 4. Swedish SFR silo for intermediate wastes



VLHR silos would be located in the middle of the repository at full repository depth to take advantage of the waste-isolation capabilities of the repository. The repository provides a major barrier against human intrusion, and the geology provides several barriers against radionuclide releases to the accessible environment. Silos are an alternative WP, not a replacement for the repository.

The replacement of WPs with large silos may result in significant improvements in the performance of the engineered barriers to radionuclide releases. The release of radionuclides from a failed WP is proportional to (1) the groundwater flow through the WP and the (2) solubility limits of the radionuclides in groundwater. By concentrating the VLHR wastes from up to 10 000 t of SNF in 1 silo rather than spreading it over ~1 000 WPs, the groundwater flow through the wastes per unit volume is reduced by a factor of 100 to 1 000. With the reduction of groundwater flow per unit

quantity of waste, radionuclide releases are proportionally reduced. The large waste silo has a smaller surface-to-volume ratio than does each WPs.

## 5. Other considerations

### 5.1 Scaling factors

No detailed economic analysis of these repository benefits has been conducted. However, some comparisons [3] between conventional repositories and these alternative designs can be made. Consider the case where (1) P&T destroys the long-lived HHRs, (2) the shorter-lived HHRs are disposed of in an extended-dry repository such as YM, and (3) the long-lived, low-heat wastes are disposed of in silos.

In the conventional YM repository design, the SNF is disposed of in large WPs in 5.5-m-diam tunnels. For every 100 m of tunnel required for disposal of SNF, about 71 m of boreholes would be required to dispose of the shorter-lived HHR wastes from the SNF. For every 100 SNF WPs, 71 HHR capsules of similar length and a small fraction of a silo would be required for disposal of the HHR-VLHR wastes from that SNF. In effect, there are three major changes: (1) substitution of 5.5-m disposal tunnels with 15-cm boreholes for the HHRs, (2) substitution of thousands of high-performance, expensive WPs with a few silos, and (3) reduced heat load from destruction of the longer-lived HHRs.

The impact of these changes would be to significantly reduce the operational costs for the repository. Operational costs include the mining of disposal drifts for the WPs. It may not impact siting or licensing costs – an important fraction of the total costs. The economic incentives are dependent upon the size of the repository. As the repository capacity increases and the cost per unit of waste decreases, operational costs become a larger fraction of disposal costs. Siting and licensing costs are essentially fixed costs.

The economic cost for the repository gains in an actinide P&T fuel cycle is the necessity to separate the caesium and strontium from the other waste streams. This cost is dependent upon the specific separation processes.

### 5.2 Caesium-135

The short-lived HHRs contain one long-lived radionuclide,  $^{135}\text{Cs}$ . It has a half-life of  $3 \times 10^6$  years. Performance assessments of proposed repositories [11] indicate that this long-lived radionuclide is not usually a significant risk to man nor a significant factor in terms of repository performance. There are several reasons for this:

- *Geochemistry.* Radionuclides, such as  $^{129}\text{I}$ ,  $^{237}\text{Np}$ , and  $^{99}\text{Tc}$ , which dominate the long-term risks from a repository are those most easily transported by groundwater with little retention by the geology. There is significant retention of caesium in most types of rock and ion-exchange of radioactive caesium isotopes with non-radioactive caesium in the rock.
- *Biological effects.* Differences in the accumulation rate of different radionuclides in specific human organs determines their relative hazards. The hazard from  $^{135}\text{Cs}$  is low compared to many other radionuclides because of its low rate of bioaccumulation.

For any HHR disposal option, a performance assessment of the risks from this radionuclide will be required. There are major engineering questions about the feasibility of isotopically separating this

isotope from other caesium isotopes; thus, disposal with the other caesium isotopes is likely to be the most practical route. Such an assessment would be significantly simpler to make than for HLW or SNF because there is only a single radionuclide.

## 6. Conclusions

Repository designs and costs are controlled by radioactive decay heat. Any P&T option that destroys long-lived HHRs (Pu, Am, and Cm) is an enabling technology that may allow for lower-cost, higher performance repositories by separate management of (1) the shorter-lived HHRs and (2) the VLHR wastes. These repository benefits may exceed the other waste management benefits of actinide P&T fuel cycles such as reductions in radiotoxicity. The cost for these benefits is the requirement to separate caesium and strontium from the other P&T wastes.

An understanding of the costs and benefits of separate management of shorter-lived HHRs should be a high priority within any investigation of actinide P&T fuel cycles. This is an appropriate area for international co-operation. Most of the issues (selection of radionuclides to be destroyed by P&T, transmutation efficiencies, solidification of short-lived HHRs, repository design for low-heat, long-lived radionuclides, etc.) are common issues for all. It is an area of waste management where only very limited studies have been undertaken.

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## THE AMSTER CONCEPT

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### Abstract

AMSTER is a concept for a graphite-moderated molten salt reactor, in which the salt treatment installation has been redesigned in order to reduce waste production. Using this concept, one can define a large number of configurations according to the products loaded and recycled. This document presents a configuration which self-consumes transuranium elements and generates fissile material with a mixed thorium and uranium support. This gives a reactor, which is highly economical in uranium and thorium consumption, leaving only a few grams of transuranium elements per billion KWhe in the ultimate wastes to be disposed of.

## 1. Introduction

On 30 December 1991 the French parliament adopted a law concerning research into the management of radioactive waste [1]. This law stipulated that three types of work should be conducted simultaneously:

- A search for solutions allowing the separation and transmutation of long-life radioactive elements present in this waste.
- A study into the possibility of reversible or irreversible storage in deep geological formations, in particular by creating underground laboratories.
- A study into long-term packaging and storage conditions for this waste on the surface.

The studies presented in this article aim to provide a foundation for work in the first area of research defined by this law.

Analysis of the problem of reducing long-life radioactive products leads us to propose a new concept: AMSTER (Actinides Molten Salt TransmutER). We will present this concept and the results of a preliminary study into the reactor physics associated with this concept.

## 2. Description of the AMSTER concept

AMSTER is a continuously reloaded, graphite-moderated molten salt critical reactor, using a Uranium 238 or Thorium 232 support, slightly enriched with  $^{235}\text{U}$  if necessary.

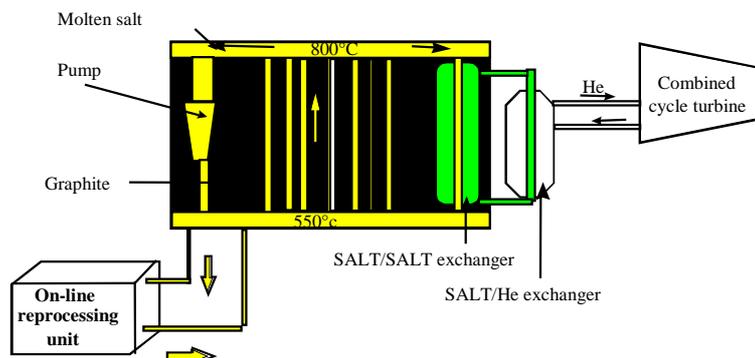
### 2.1 General presentation

Critical molten salt reactors were extensively studied in the 60s and 70s. Research was carried out in the Oak Ridge National Laboratory, where an 8 Mwth prototype was operated, the Molten Salt Reactor Experiment (MSRE). This experiment was followed by a 1 Gwe project, the Molten Salt Breeder Reactor (MSBR) [2], on which certain aspects of AMSTER are based. It should also be noted that the MSBR project was extensively examined in France (at the CEA and at EdF) in the 1970s.

The aim at the time was breeder reactors. Today, this type of reactor is again of interest to the specialists, owing to its incinerating capacity.

Figure 1 gives the basic layout of this type of reactor.

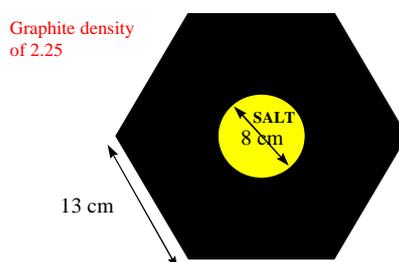
Figure 1. Layout diagram of the molten salt reactor



## 2.2 Reactor core

The core of a molten salt reactor consists of an array of graphite hexagons identical to those of the Saint-Laurent B1 UNGG reactor. Each hexagon contains a hole through which the salt circulates. We used the results of a study conducted by the EdF reactor physics department in 1976, which defines a salt hole diameter of 8 cm for a hexagon 13 cm on a side. The diameter of this hole was optimised to favour reactor conversion (transmuting as much  $^{238}\text{U}$  as possible into  $^{239}\text{Pu}$ ).

Figure 2. AMSTER reactor cell



We used salt of the same type as that of the MSBR project [2]. The composition adopted,  $61\text{LiF} - 21\text{BeF}_2 - 18\text{NLF}_4$ , enables a moderate quantity of uranium and transuranium nuclei to be introduced into the core (NL here stand for heavy nuclei).

## 2.3 Operating principle

When the salt enters the array, it becomes critical and heats up. It enters at a temperature of about 550°-600°C and leaves at a temperature of 800°C.

The core inlet temperature is determined by the salt melting temperature, which itself depends on the composition of the salt (500 to 600°C).

The outlet temperature is determined by the strength of the materials other than graphite (hastelloy).

Once heated, the salt is entrained by pumps and passes through salt/salt exchangers which enable the thermal energy produced to be recovered. **On leaving the core, an on-line reprocessing unit which we have entirely redesigned, takes a small fraction of the fuel for reprocessing, in other words it extracts the fission products from it. This reprocessing is accompanied by injection into the salt of new nuclei,  $^{235}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$  or transuranium elements, to replace the heavy nuclei already fissioned.**

The secondary salt heats up either steam or helium, which feeds either a combined cycle turbine plus alternator, or thermal applications such as the production of hydrogen if the salt temperature is high enough.

## 2.4 Configurations examined

A large number of configurations can be envisaged with this type of reactor, depending on:

- The products placed in the reactor: isotopes of uranium, transuranium elements, isotopes of Thorium, long-life fission products (LLFP).
- Substances multi-recycled in the reactor.
- The substances leaving the reactor:
  - Vitrified losses from reprocessing of transuranium elements, uranium, thorium, LLFP and all the short-life fission products (SLFP) considered being waste.
  - Depleted uranium when the  $^{235}\text{U}$  currently stored is used for enrichment.

The various configurations are characterised by:

- The type of the support: uranium, thorium, mixture of uranium and thorium. The thorium support has the advantage of producing significantly fewer transuranium elements than the uranium support and of having a thermal spectrum regenerating more fissile material than the uranium support.
- The presence of fertile blankets allowing regeneration of the fissile nuclei.
- The possible input of transuranium elements coming from other reactors, determining the incinerating capacity of the reactor. If no transuranium elements are input from the outside, the reactor consumes its own transuranium elements. The reactor loaded with outside transuranic become an incinerator.

We thus examined 7 configurations. Table 1 describes the various configurations examined. There are 4 configurations without fertile blanket and 3 configurations with fertile blanket.

Table 1. Definition of configurations examined

Without fertile blanket

Support	Uranium	Thorium	Uranium + thorium
Self consuming	<u>X</u>	<u>X</u>	
Incinerating	<u>X</u>	<u>X</u>	

With fertile blanket

Support	Uranium	Thorium	Uranium + thorium
Self consuming	<u>X</u>	<u>X</u>	<u>X</u>
Incinerating			

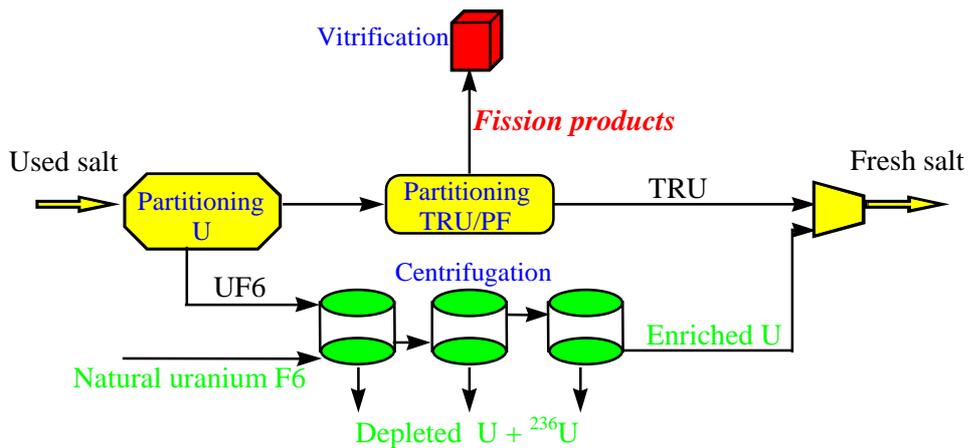
Below, we will present the transuranium elements consumption configuration, with fertile blanket and a mixed uranium-thorium support.

Processing unit: for each configuration examined, more or less complex salt reprocessing is required.

### 2.5 General recycling principle

Below, we present the recycling principle for the uranium support reactor. For the thorium support, the principle is the same, with the thorium separated after the transuranium elements and before the FP.

Figure 3. Layout diagram of the AMSTER salt processing unit



More specifically, the transuranium elements are confined in the core – reprocessing unit assembly by separate extraction of the transuranium elements and rare earths by a Bismuth counter-flow.

The salt processing unit includes the cycle front-end (salt enrichment) and back-end (FP extraction).

- Cycle front-end

Uranium is first of all extracted from the salt (Figure 3). It is in  $UF_6$  form, which is mixed with an adjusted mass of natural uranium (also in  $UF_6$  form). This mixture is enriched with  $^{235}U$  if necessary, to the required new salt  $^{235}U$  enrichment value, for example using an ultra-centrifuge.

The residual depleted uranium is evacuated, taking with it a large proportion of the  $^{236}U$  in the spent salt (about 35 %) [3]. This prevents the core being poisoned with this isotope.

This solution would require a small number of centrifuges, owing to the small quantity of  $^{235}U$  to be added.

- Cycle back-end

The transuranium elements are separated in a salt – liquid metal exchanger. Given the good separation factor in this operation (about 10), and by using 6 consecutive stages, the salt would only contain a residue of about  $10^{-5}$  times the initial mass of transuranium elements. Then the thorium and the fission products, except for the LLFP to be incinerated, are extracted from the salt with no need for a high separation capacity. The residual transuranium elements in the salt are extracted with the FP, which can be vitrified and stored in the same way as fission products today.

### 3. Numerical simulation principle

Numerical simulation of this type of reactor requires an iterative working method, the principle of which is described below.

We begin with an APOLLO 1 type calculation of a cell evolving in an infinite medium. We chose APOLLO 1, as this code requires little calculation time and has already been used to simulate and manage Gas Graphite Natural Uranium Reactors (UNGG). We checked the accuracy of the calculations against a reference calculation using the Monte Carlo TRIPOLI 4 code (discrepancy of 400 pcm for a  $k_{\infty}$  of 1.2).

At the end of this calculation step, we extract the FP from a fraction of the core. They are replaced by a mixture of TRU,  $^{235}U$  and  $^{238}U$  so as to guarantee the reactivity at the end of the step.

To limit the calculation time, the time between two reprocessing operations must be sufficiently long. We achieved initial equilibrium with a pitch of 10 days. Then, in the light of the first results (slight evolution of  $k_{\infty}$ ) we raised this pitch to 100 days.

We adopted a calculation pitch of 100 days and a cell  $k_{\infty}$  at the end of evolution of 1.05 (to take account of leaks), an electrical power produced by AMSTER of 1 GWe or 2 250 MWth, a salt volume of 48 m<sup>3</sup> (30 in the active core and 18 in the auxiliaries) for a reactor without blanket.

We defined a reference case in which we extract one third of the core every 100 days (burn-up of 300 efpd). This initial simulation showed that it was necessary to purge all or part of the  $^{236}U$  formed by  $^{235}U$  capture.

We therefore adopted a partial purge (30%) of the  $^{236}\text{U}$  for the rest of the study.

The transuranium elements, enriched uranium, thorium and possibly other transuranium elements, would be re-injected into the salt and then into the reactor.

#### **4. Feasibility of an AMSTER self-generating uranium concept**

##### **4.1 The concept**

In the case of the thorium support, the consumption of fissile uranium is low enough for it to be produced in the form of  $^{233}\text{U}$ , in an extra core zone. This fertile zone located on the core periphery, would be under-moderated by increasing the diameter of the salt hole.

In this concept, the size of the fertile zone would be adapted to make the reactor only just a  $^{233}\text{U}$  self-generator (the production of the fertile zone would exactly compensate the consumption of the fissile zone).

Salt processing would simply be by removal of the FP and replacement by the same mass of  $^{232}\text{Th}$  or (and)  $^{238}\text{U}$  support.

##### **4.2 Calculation method**

Only an equilibrium situation using a complete core calculation would be able to validate this concept completely. However, its feasibility can be evaluated by cells calculations.

In the fertile zone, the radius of the cylinder in which the salt circulates is set at 8 cm. This value will then be optimised when the core calculation verification is made.

We also supposed that the power density in the fertile zone was half that in the fissile zone. An exact determination of this ratio is only possible with a core calculation.

We calculate the volume of fissile and fertile salt to obtain the power level sought.

In these conditions, the mean power density of the fertile salt is equal to one-eighth the mean power density of the fissile salt. So that heating of the fertile salt is the same as that of the fissile salt, we slow down the salt flowrate in the blanket by a factor of 8 and thus multiply the time the salt spends in the blanket by 8. The production of heavy nuclei in the blanket is thus equivalent to the production from a volume of salt during the time the salt passes through the fissile core.

Given these hypotheses, the reactor was thus balanced for a given ratio of fertile and fissile salt volumes. At each time step  $\Delta t$ :

- The fuel was placed in each zone for  $\Delta t$ , and the 2 zones were mixed pro rata the core volumes.
- Part of the fission products is removed and is replaced by the same mass of a mixture of  $^{233}\text{U}$  and  $^{232}\text{Th}$ .
- The enrichment of this mixture is calculated to keep the fissile core  $k_{\infty}$  at 1.05.

The volume of salt in the fertile area is that for which the mass of  $^{233}\text{U}$  to be added at equilibrium is zero.

Equilibrium was achieved for several ratios of salt volume in fertile zone to salt volume in fissile zone. For reasons of precision, we took a time pitch of 10 efpd

### **4.3 The thorium support**

In the fertile zone the  $^{233}\text{U}$  balance is positive: More  $^{233}\text{U}$  comes out of the blanket than goes in. If we increase the blanket volume, we increase the quantity of  $^{233}\text{U}$  produced and at any given moment, the  $^{233}\text{U}$  contribution of the blankets will compensate the  $^{233}\text{U}$  consumption by the fissile core. The core will generate its own uranium. To determine the volume of the blankets needed, we varied the ratio between the fertile salt volume and the fissile salt volume between 1.5 and 2.5.

For each of these ratios, we give (Table 2) the consumption of  $^{233}\text{U}$ , the masses in the reactor at equilibrium, the increase in core size (volume and radius) necessary, and the proportion of power given off in the fissile zone.

These calculations show the feasibility of the concept. This configuration is particularly interesting because:

- It does away with the uranium enrichment phase.
- It requires no extraction of  $^{236}\text{U}$ .
- It offers a very small transuranium elements inventory.
- It reduces consumption of heavy nuclei to 100 kg of  $^{232}\text{Th}$  per TWhe, thus reducing mining waste accordingly.
- It considerably reduces the masses of depleted and reprocessed uranium.

### **4.4 The thorium-uranium support**

It is also possible to add uranium (i.e. depleted U in stock) to the thorium support. We thus increase the quantity of transuranium elements at equilibrium, while remaining within a reasonable range. But we then burn  $^{238}\text{U}$  and make  $^{232}\text{Th}$  savings. We significantly reduce the fissile nuclei enrichment of the uranium in the core, thus making the reactor non-proliferating.

As a counterpart, we will have to increase the production of  $^{233}\text{U}$  and thus the volume of the blankets.

We thus varied the proportion of  $^{238}\text{U}$  added to the load from 0 to 100% for 2 volumes of fertile salts: 2 times the fissile volume and 2.5 times the fissile volume.

Figure 4 shows the mass of  $^{233}\text{U}$  consumed (or produced) versus the percentage of uranium 238 in the fertile material added;

The more uranium is placed in the support, the more  $^{233}\text{U}$  must be added and the more the volume of the blanket must be increased. Between 0 and 50 %  $^{238}\text{U}$  the  $^{233}\text{U}$  input is small enough to be

conceivable. Thus by increasing the relative volume of fertile salt to 2.5, self-generation is achieved for 50%  $^{238}\text{U}$ .

Table 2. Characteristics of the two-zone AMSTER, depending on the ratio of salt volume in fertile core to the salt volume in fissile core with a 100% thorium support

Fertile salt volume to fissile salt volume ratio	1.5	2	2.5
Core inventories (kg/GWe)			
$^{232}\text{Th}$	138 800	153 580	167 360
Uranium	3 354	3 650	3 900
Transuranium elements	53	54	54
$^{233}\text{U}$ consumption (kg/Twhe)	2.11	-0.12	+1.91
Core radius (m)	4.95	5.04	5.11
Total volume of salt in the reactor ( $\text{m}^3$ )	82	91	99
Power produced in the fissile area (MWe)	1 895	1 800	1 714

The presence of  $^{238}\text{U}$  increases the mass of transuranium elements at equilibrium. This rises almost linearly with the percentage of  $^{238}\text{U}$  and is equal to 1 500 kg for a percentage of 50%.

The presence of  $^{238}\text{U}$  also modifies the isotopic composition of the transuranium elements. Figure 5 shows the different masses in the reactor for 0%, 50% and 100 %  $^{238}\text{U}$  in the fertile material inputs.

Figure 4. Mass of  $^{233}\text{U}$  consumed (or produced) versus the percentage of  $^{238}\text{U}$  in the fertile material input

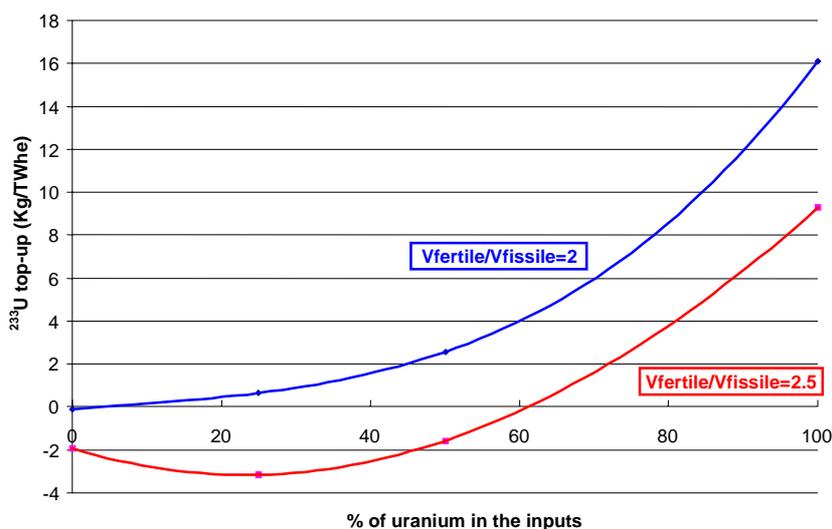
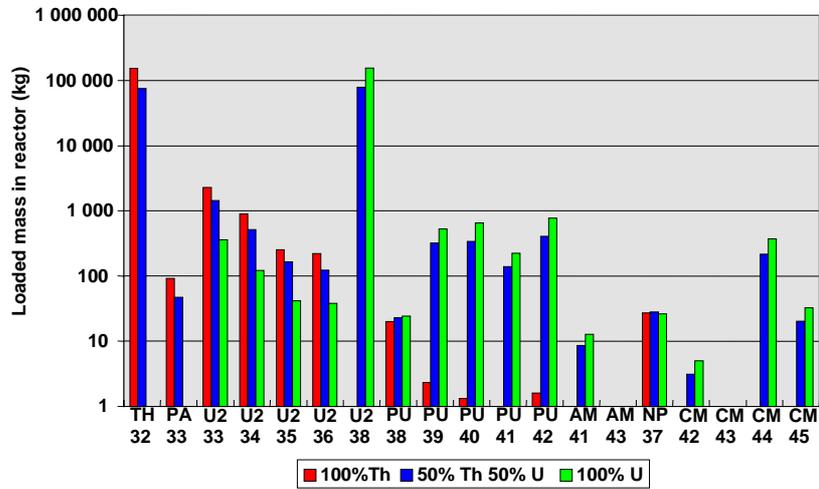


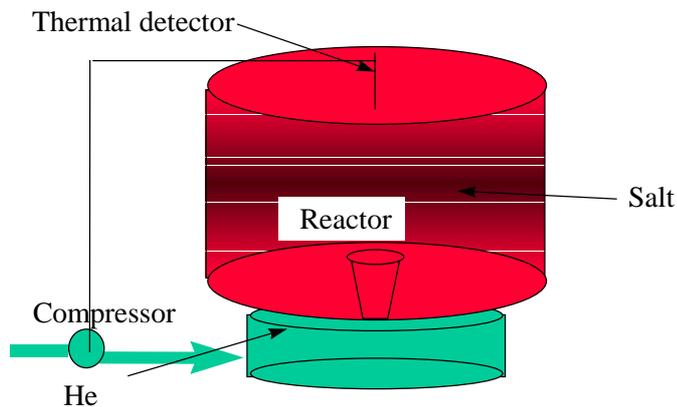
Figure 5. Core mass balance for 0.50% and 100%  $^{238}\text{U}$  in the fertile material input for  $V_{\text{fertile}}/V_{\text{fissile}} = 2$



### 5. A major safety asset: core drainage

The salt is at its maximum reactivity in the graphite. A safe fallback position can thus be obtained by draining the core. As the fuel is liquid, it can be extracted from the core at any moment. For this, we adopted a concept proposed by EdF and the CEA, which consists in placing a drain tank under the core, which is permanently connected to it. Salt is confined within the core by a helium back-pressure (Figure 6). One therefore need simply interrupt the electric power supply to the He compressor for gravity drainage of the core. This feature, allied with the considerable thermal inertia of the reactor and the difficulty of rapidly inserting reactivity, should make the reactor particularly safe.

Figure 6. Core drainage principle



## 6. R&D needed to validate the AMSTER concept

To make the transition from concept to technology, much research and experimentation is required.

Among the subjects covered will be:

- Salt chemistry, structural corrosion by salt.
- Processing chemistry, containment of transuranium elements in the salt.
- Reactor dynamics.
- Safety.

It should be recalled that major R&D work was already carried out in the 60s/70s and a prototype functioned remarkably well (MSRE). A detailed preliminary project for a breeder reactor (MSBR) was conducted. The AMSTER concept is similar to these two reactors and the experience acquired would be directly applicable to it.

## 7. Fertile and fissile material utilisation

The AMSTER concept should allow the production of energy with very reduced quantities of transuranium element waste (a few g per TWhe), and with no transportation of highly radiotoxic substances.

AMSTER with a mixed thorium-uranium support with a peripheral fertile zone, breeding its own uranium, should further improve this performance. This reactor should in fact offer incinerating performance identical to that of the thorium AMSTER supplied with  $^{235}\text{U}$ , while eliminating the need for uranium enrichment and making the reactor non-proliferating. Furthermore, this self-breeding reactor would consume only 50 kg of thorium and 50 kg of  $^{238}\text{U}$  per TWhe, which in the light of estimated resources (1 to 4 million tonnes of thorium and 1 to 3 million tonnes of uranium), would allow the production of 20 to 70 million TWhe (the world's annual electricity consumption from all sources together is about 15 000 TWhe); The depleted uranium stored in France (200 000 t), together with 200 000 t of thorium, could produce 4 million TWhe (annual production is 400 TWhe).

Figure 7 compares the mass balance entering and leaving the reactor for a standard open-cycle PWR and a self-generating AMSTER with a support of 50% U and 50% Th.

The inputs are natural or depleted uranium and thorium.

The outputs comprise:

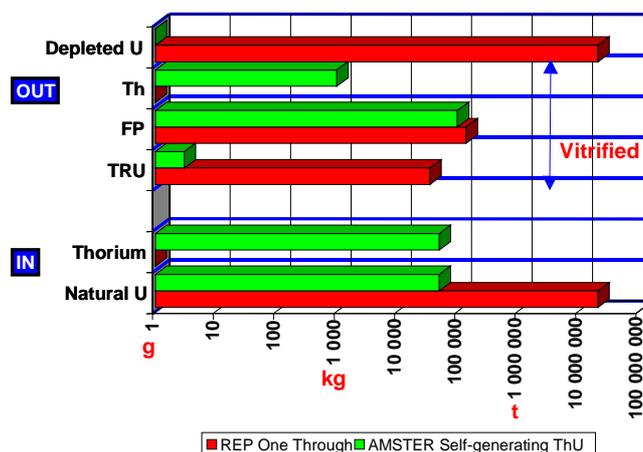
- Transuranium element losses, U and Th vitrified with the FP.
- Depleted uranium produced at enrichment of the support.

This theoretical study shows that:

- AMSTER would be far “cleaner” than a PWR (4 decades reduction of transuranium element waste).

- As we saw earlier, it offers “virtually inexhaustible” resources.
- It should be safer, owing to the fact that the fuel can be rapidly extracted from the core if necessary.

Figure 7. **Mass balance entering and leaving the reactor for a standard open-cycle PWR and for a self-generating AMSTER with a support of 50% U and 50% thorium**



## 8. Initial conclusions

Although new, molten salt reactor technology was already experimented at Oak Ridge in the 1960s. The prototype built at the time operated remarkably well. Furthermore, EdF and the CEA studied molten salt reactors until 1983. This preliminary study should be followed by a minimal R&D and engineering program in order to evaluate both the pyrochemical reprocessing process (loss rates to be confirmed) and the technological feasibility and safety of AMSTER.

The reactor specifications would then have to be optimised: unit power, size of graphite array, salt burn-up, quantity of transuranium elements in the reactor, etc., and its economics evaluated.

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