

# THORIUM FUEL CYCLES: A GRAPHITE-MODERATED MOLTEN SALT REACTOR VERSUS A FAST SPECTRUM SOLID FUEL SYSTEM

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## ABSTRACT

The aim of this paper is to compare two main options dedicated to long-term energy production with Thorium: solid fuel with fast spectrum and molten salt with moderated neutrons. In both cases, we have set up a specific system and studied its behaviour until it reaches the  $^{232}\text{Th}/^{233}\text{U}$  equilibrium from two different starting fuels:  $^{232}\text{Th}/^{233}\text{U}$  and  $^{232}\text{Th}/\text{Pu}$ , with Pu coming from PWR spent fuel.

All this work was done with the same computing methods based on the coupling between a validated neutron transport code and a numerical resolution of the evolution equations which takes into account the time dependence of the mean cross sections. Our results consist in precise time evolutions of physical parameters, inventories and waste production, allowing comparison of breeding performance and induced radiotoxicities. From this comparison, the molten salt solution appears to be more interesting than the solid fuel reactor for a quick transition to a significant Thorium fuel cycle.

## I. INTRODUCTION

Thorium is an attractive way to produce long-term nuclear energy with low radiotoxicity waste. Moreover, the transition to Thorium could be done through the incineration of industrial Plutonium. Contrary to the  $^{238}\text{U}/^{239}\text{Pu}$  cycle in which breeding can be obtained only with fast spectra, the  $^{232}\text{Th}/^{233}\text{U}$  cycle can operate with either fast or epithermal spectra. As a consequence, Thorium fuel cycle reactors can use, with suitable reprocessing, either solid fuel with fast neutrons or molten salt with moderated neutrons.

A lead-cooled fast Accelerator-Driven System (ADS) has already been studied in our group in the frame of a comparative analysis of the Thorium and Uranium fuel cycles [1]. The results of this study concerning Thorium led us to compare them to the case of a critical Molten Salt Reactor (MSR) coupled to an online reprocessing unit, in order to complete our study of the possible Thorium fuel cycles. For our MSR, we chose a classical design, close to the Molten Salt Breeder Reactor (MSBR) concept which was studied in the early seventies at the Oak Ridge National Laboratory [2].

Identical simulation protocols were used for both studies. Evolution programs which ensure the coupling between the resolution of Bateman equations and the Monte-Carlo code MCNP [3] aim at fully integrating the constraints and flexibility of each system. This is especially important in the case of the MSR whose simulated feeding and extracting processes can be adjusted during the calculation. All the data are thus known precisely from start-up to equilibrium.

In this paper we describe first the two specific systems. Next comes a general presentation of the methods used in our simulations. Once systems and methods are defined, we detail our studies with the  $^{232}\text{Th}/^{233}\text{U}$  starting fuel and compare their main results in terms of breeding performance and induced radiotoxicities. For both options, we discuss then the possibility of using Plutonium extracted from PWR spent fuel as an initial fissile inventory in order to reach the  $^{232}\text{Th}/^{233}\text{U}$  equilibrium. Finally, we compare the two systems starting with Plutonium or  $^{233}\text{U}$  if available within a French scenario of transition to Thorium.

## II. SYSTEMS

### A. Main Characteristics of the Fast ADS

The system is subcritical ( $k_s \simeq 0.98$ ), driven by a 1 GeV proton beam incident on a lead target where neutrons are produced by spallation (spectrum calculated by the code FLUKA [4]). MCNP transports neutrons into the core, where a hexagonal array of steel needles contains the fuel. The reflector and the coolant are lead. The detailed geometry is described in Ref. [1].

Depending on its fuel, the reactor has different thermal powers. In the following, if not otherwise specified, all results are normalized to the same thermal power as the MSR: 2500 MWth.

### B. The Graphite-Moderated MSR Design

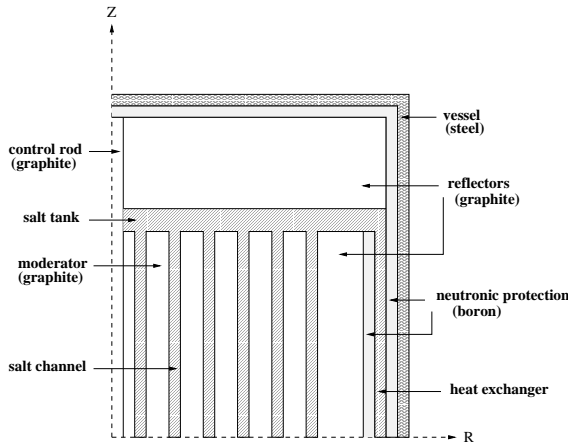


FIG. 1 – Schematic view of the simulated MSR.

The MSR design has been chosen as classical as possible (Fig. 1). Its MCNP description is nevertheless very precise. Preliminary evaluations, in particular of heat transfers and neutron losses, provided the main dimensions of the device.

The core is a cylindrical assembly (2.3 m radius, 4.6 m high) of high density graphite ( $2.3 \text{ g.cm}^{-3}$ ) hexagons (15 cm side), each pierced by a channel (15 cm diameter) for molten salt circulation. The graphite radial reflector is 50 cm thick. The heat exchangers are simulated by a 10 cm thick crown of salt sandwiched between boron loaded graphite ( $\text{B}_4\text{C}$ ). Above and under the core are located salt tanks (30 cm high each) and graphite axial reflectors (1.3 m high each). The total salt volume (molten salt circuit) is  $46.0 \text{ m}^3 = 20.5$  (core channels) + 10.2 (tanks) + 15.3 (heat exchanger).

## III. CALCULATION METHODS

### A. Basic Principles

The general method is described and justified in Ref [1]. MCNP is a static code, which means that it cannot take into account a possible important time evolution of the neutron spectrum and consequently of the mean cross sections. We have thus to regularly compute new mean cross sections, especially with moderated neutrons.

In all our studies, we have coupled MCNP to a program which solves the evolution differential equations by a 4<sup>th</sup> order Runge-Kutta method. A MCNP calculation is done each time the user-defined precision on the calculated abundances of nuclides requires it.

### B. Improvements for MSR studies

#### 1. Exploitation of Nuclear Data Bases

In order to achieve precise calculations, we had to use explicit rather than pseudo Fission Product cross sections. This need led us to integrate the nuclear data processing code NJOY [5] in our programs to build our own MCNP data files from the existing bases (ENDF-B/VI, JENDL3.2 and JEF2.2).

Moreover, NJOY allows us to compute effects like Doppler and thermal scattering in graphite. We chose 900 K as the temperature for the salt and all other materials in the core, 600 K for the axial reflectors and 300 K for the steel vessel.

#### 2. Modelisation of the MSR Reprocessing

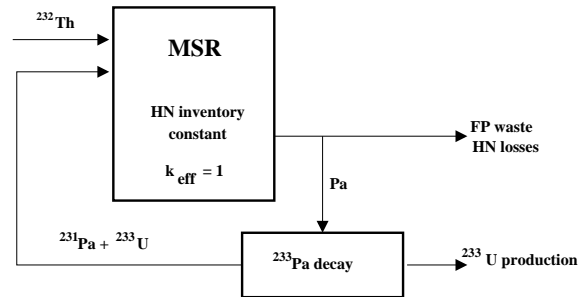


FIG. 2 – Flow diagram of the simulated reprocessing.

Contrary to the ADS for which the fuel reprocessing takes place every 5 years, we simulate in the MSR case a MSBR-like online fuel reprocessing unit. Fig. 2 shows the reprocessing material flows that we take into account. HN stands for Heavy Nuclei (nuclei from Thorium to Californium), and FP for Fission Products. There are two types of such flows: feeding and extraction.

In all studies, the total Heavy Nuclei inventory is kept constant by means of a variable  $^{232}\text{Th}$  fertile feeding. Concerning the fissile feeding ( $^{233}\text{U}$  for instance), its flow is regularly adjusted in function of the evolution of  $k_{eff}$  (partial derivatives of  $k_{eff}$  with time and  $^{233}\text{U}$  flow), in order to maintain criticality. Finally, Li, Be and F additions aim at taking care of chemical constraints.

The extraction of the isotope  $i$  of a chemical element  $e$  is simulated by a pseudo-decay whose constant is

$$\lambda_i = \frac{\varepsilon_e}{\tau_r},$$

where  $\varepsilon_e$  is the extraction efficiency for  $e$  and  $\tau_r$  the time it takes for the whole salt volume to be reprocessed (as in the MSBR project, we take  $\tau_r = 10$  days). We assume that all extractions are based on the liquid-liquid extraction process, which amounts to exchanging Thorium and Lithium dissolved in molten Bismuth for the constituents to be removed from the salt (FPs and Pa).

Some FPs are chemically so close to Thorium that their extraction efficiencies are lowered by this technique [2]. FPs are thus subdivided in 3 categories whose respective efficiencies are 20% (halogens and rare earths, like for example, in decreasing order of concentration in a  $^{232}\text{Th}/^{233}\text{U}$  salt at equilibrium: Ce, Nd, Y, La, Pr, ...), 5% (Zr and semi-noble metals: Sn, Sb, ...) and 1% (alkaline elements: Sr, Cs, Ba, Rb, ...) [6]. Rare gases and noble metal dust escape from the salt circuit in less than one minute with flowing Helium [7].

In the following, Protactinium is extracted with 100% efficiency within the 10-day reprocessing time. This is the main way to breed  $^{233}\text{U}$ , as we will see in the next section. After sufficient  $^{233}\text{Pa}$  decay, Protactinium is reinjected with the  $^{233}\text{U}$  feeding, in order to limit the radiotoxic  $^{231}\text{Pa}$  inventory.

Likewise, Heavy Nuclei reprocessing losses are simulated by a pseudo-decay with an optimistic but conceivable efficiency  $\varepsilon_{HN}$  of  $10^{-5}$  [8].

## IV. $^{232}\text{Th}/^{233}\text{U}$ REACTORS

### A. Breeding Conditions

#### 1. Solid Fuel Case: reactivity variations impose reprocessing rates

Thanks to a suitable initial fissile proportion, the multiplication factor  $k_s$  remains constant during about 6 years but then starts to drop because of the decreasing breeding performance and the increasing FP poisoning. Recycling for the ADS consists thus in removing every 5 years all Fission Products from the fuel and replacing the used mass of Thorium.

In the breeding configuration, a Thorium blanket is added around the core and replaced by a new one every two months in order to prevent any excessive reactivity increase [9]. The  $^{233}\text{U}$  produced is extracted and stored.

This way, about 10% of the initial  $^{233}\text{U}$  inventory are produced within 5 years in the blanket. The fuel used in the core during this 5-year period is extracted, replaced by a fresh one, let cool down during 5 years and then topped with  $^{232}\text{Th}$  and a little  $^{233}\text{U}$  and reinserted in the core for a new 5-year burn-up. As a consequence, it takes 10 years to breed 10% of the whole fissile inventory, which means the doubling time is about 100 years.

### 2. Molten Salt Case: suitable salt composition and reprocessing allow good breeding

The carrier salt that we chose is LiF-BeF<sub>2</sub>, with LiF at 80 mol% (“4LiF-BeF<sub>2</sub>”). Lithium is  $^7\text{Li}$  at 99.995 mol%. In this salt, the Heavy Nuclei (only  $^{232}\text{Th}$  and  $^{233}\text{U}$  at start-up) are at 12.5 mol%: LiF-BeF<sub>2</sub>-(HN)F<sub>4</sub> at 70-17.5-12.5 mol% respectively.

This composition is close to the MSBR one, whose main physico-chemical properties like density (3.3 g.cm<sup>-3</sup> at 900 K) are well-known.

Heavy Nuclei proportion in 4LiF-BeF <sub>2</sub> (mol%)	10	<b>12.5</b>	15
initial $^{232}\text{Th}$ inventory (tons)	56	<b>67</b>	77
initial $^{233}\text{U}$ inventory (tons)	1.0	<b>1.1</b>	1.3
$^{233}\text{U}$ production after 50 years (tons)	1.4	<b>2.1</b>	2.6
average doubling time after 50 years (y)	36	<b>26</b>	25

TABLE 1 – *Initial inventories and breeding performance for different salt compositions.*

Table 1 shows that the chosen Heavy Nuclei proportion in 4LiF-BeF<sub>2</sub> ensures minimal doubling time and initial fissile inventory to within about a percent.  $^{233}\text{U}$  production after 50 years is simply the difference between the total mass of  $^{233}\text{U}$  produced by Pa decay and the total mass of  $^{233}\text{U}$  used in the fissile feeding for reactivity control (Fig. 2). The average doubling time after 50 years is obtained directly from the number of  $^{233}\text{U}$  initial inventories contained in this production.

The conversion ratio C at equilibrium is obtained by dividing the  $^{233}\text{U}$  production rate (2.14 kg/day from Pa decay) by the  $^{233}\text{U}$  feeding flow (2.04 kg/day), which gives  $C = 1.05$  ( $\pm 0.01$ , due to uncertainties in nuclear data).

The  $^{233}\text{Pa}$  inventory at equilibrium is only 20 kg with the reprocessing chosen. We find that without Pa extraction, the  $^{233}\text{Pa}$  inventory stabilizes to about 100 kg, which is similar to that of the solid fuel ADS. In this case,  $C = 0.99$  at equilibrium, which means that the MSR is no longer breeding but just converting.

Criticality imposes 98.4%  $^{232}\text{Th}$  - 1.6%  $^{233}\text{U}$  as the starting Heavy Nuclei molar composition with the salt chosen. The reactor with solid fuel has a similar Heavy Nuclei inventory in mass; about 70 tons of Thorium and Uranium oxides. However, the  $^{233}\text{U}$  proportion is much larger. Indeed the initial molar composition in the ADS fuel is 90.5%  $^{232}\text{ThO}_2$  - 9.5%  $^{233}\text{UO}_2$ , with about 6 tons of  $^{233}\text{U}$ .

## B. Transitions to Equilibrium

### 1. Fission Products

In the solid fuel case, the FP inventory increases linearly from 0 to about 5 tons in 5 full power years. The inventory averaged over 5 years is  $2.05 \cdot 10^4$  moles, the mean capture cross section 0.15 barn and the mean flux in the fuel  $1.70 \cdot 10^{15} \text{ n.cm}^{-2}.\text{s}^{-1}$ . The mean FP capture rate is thus 0.45 mol/day.

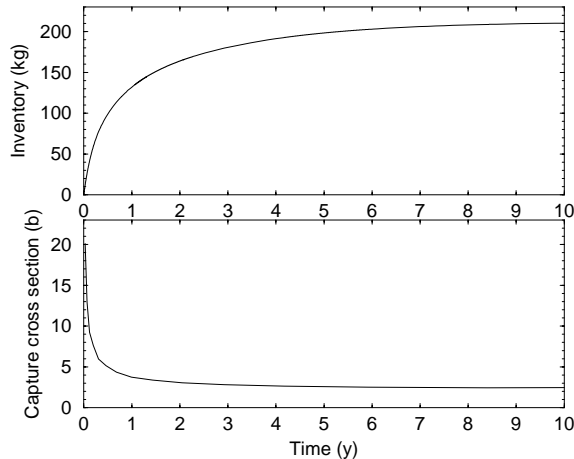


FIG. 3 – FP transitions in the MSR.

With molten salt, there is only one FP transient episode. Fig. 3 shows that the FP inventory stabilizes to 210 kg (1910 moles) within about 5 years. The mean capture cross section at equilibrium is 2.4 barn and the flux averaged over the whole molten salt circuit is  $3.2 \cdot 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$ , which gives a mean FP capture rate of 0.13 mol/day.

Contrary to the solid fuel FP poisoning, that of the molten salt can be limited more easily by means of an almost continuous fuel reprocessing and is actually 3 times lower here.

## 2. Uranium and Transuranic Elements

The Uranium inventories at equilibrium are much lower in the MSR case (Fig. 4) than with the fast spectrum solid fuel system (5800 kg of  $^{233}\text{U}$ , 1900 kg of  $^{234}\text{U}$ , 460 kg of  $^{235}\text{U}$  and 380 kg of  $^{236}\text{U}$ ).

In the MSR, we find that, through the continuous fissile feeding, the  $^{233}\text{U}$  inventory increases slowly from 1.1 to about 1.2 tons, essentially because of the transient FP poisoning.

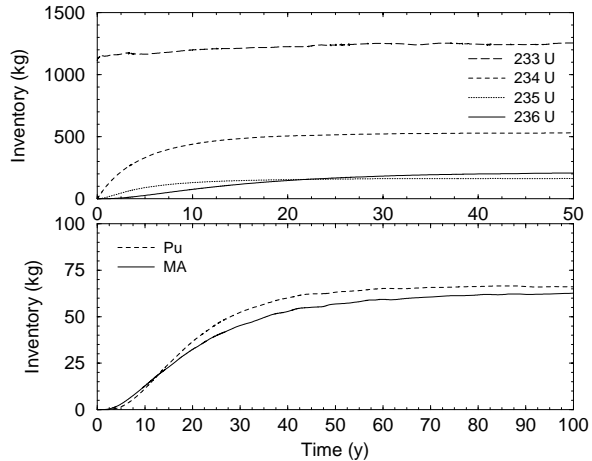


FIG. 4 – U, Pu and MA inventories in the MSR.

The main difference concerning transuranic elements is the time it takes for their inventories to reach equilibrium. Fig. 4 shows that in the molten salt case, Minor Actinides (MA = Np, Am and Cm) stabilize after about 50 years of energy production. In the solid fuel reactor, these elements do not stabilize before 100 years.

The two main transuranic nuclei in the solid fuel fast reactor at equilibrium are  $^{237}\text{Np}$  (83 kg) and  $^{238}\text{Pu}$  (58 kg). The spectrum is fast enough to prevent the inventories of Am and Cm isotopes from being significant (less than 1 kg after 100 years of energy production).

In the epithermal spectrum case of the MSR, the Np inventory is lower (44 kg of  $^{237}\text{Np}$ ) and the Pu inventory is similar to that of the fast ADS with the same main component (49 kg of  $^{238}\text{Pu}$  in 66 kg of Pu). But the Am and Cm inventories are much higher (about 4 kg of  $^{243}\text{Am}$ , 8 kg of  $^{244}\text{Cm}$  and 5 kg of  $^{246}\text{Cm}$  at equilibrium).

## C. Neutronics at Equilibrium

### 1. Neutron balance

Table 2 shows the neutron balance in the MSR fuel circuit at equilibrium. We find that  $^{235}\text{U}$  contributes to almost 10% of fissions. As for captures,

$^{233}\text{U}$  (7.8%) and  $^{234}\text{U}$  (8.1%) are the most capturing nuclei after  $^{232}\text{Th}$ . FPs and Pa capture together less than the light components of the salt (“salt” = Li, Be and F), whose total neutron consumption is itself very low compared to that of Heavy Nuclei.

	Production	Disappearance	
Fission	2.50	1.00	$^{233}\text{U}$ 87.8%
			$^{235}\text{U}$ 9.2%
			$^{239}\text{Pu}$ 1.2%
			$^{241}\text{Pu}$ 0.5%
			others 1.3%
$(n, 2n)$	0.02	0.01	$^9\text{Be}$ 91.8%
			$^{232}\text{Th}$ 8.2%
$(n, \gamma)$	-	1.41	salt 2.2%
			FP 1.1%
			$^{232}\text{Th}$ 73.0%
			$^{233}\text{Pa}$ 0.4%
			U 18.9%
			Pu 2.6%
			MA 1.8%
Losses	-	0.10	-

TAB. 2 – Detailed neutron balance (neutrons per fission) in the molten salt circuit at equilibrium. The last column gives the probabilities of absorption by different nuclei for each reaction.

In the fast spectrum case, the  $^{232}\text{Th}/^{233}\text{U}$  fuel has the same value for  $\nu$  (2.50) as in the molten salt case. But the ADS subcriticality amounts, in terms of neutron balance, to dividing  $\nu$  by the multiplication factor  $k_s$  [10]. This gives to the ADS about 0.05 more neutrons per fission available for breeding than the MSR.

However, neutron losses from the fuel are higher in the ADS without Thorium blanket (0.16), which cancels the gain obtained from subcriticality. These neutron losses from the fuel can be lowered in both cases by adding a Thorium blanket, which is mandatory for breeding in the solid fuel case. Finally, compared to critical solid fuel reactors, MSRs need a much smaller reactivity reserve at start-up thanks to the regular fissile feeding.

## 2. Neutron spectrum

Fig. 5 shows the neutron spectra at equilibrium averaged over the solid fuel of the ADS and the whole molten salt circuit of the MSR. On the MSR spectrum, we notice resonance self-shielding effects due to capture resonances of  $^{232}\text{Th}$  (at 20 eV) and  $^{19}\text{F}$  (above a few tens of keV). This spectrum is said to be epithermal, relatively flat (from 0.1 eV up to 1 MeV) compared to the ADS classical fast spectrum, whose maximum is around 200 keV.

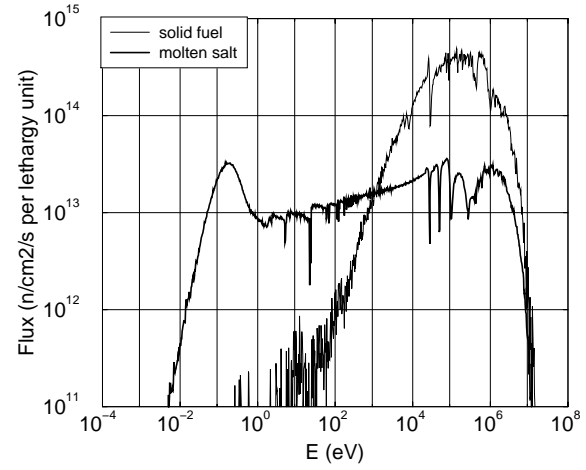


FIG. 5 – Average neutron spectra in the solid and molten salt fuels at equilibrium.

Table 3 shows the mean cross sections of important reactions of the  $^{232}\text{Th}/^{233}\text{U}$  cycle. Some remarkable differences in these data illustrate the respective constraints and advantages of both systems towards  $^{233}\text{U}$  breeding.

	solid fuel		molten salt	
	fission	capture	fission	capture
$^{232}\text{Th}$	0.008	0.32	0.008	1.36
$^{233}\text{Pa}$	0.055	1.05	0.055	20.1
$^{233}\text{U}$	2.73	0.27	49.6	6.2
Flux	1.70 $10^{15}$		3.2 $10^{14}$	

TAB. 3 – Mean cross sections (fission and  $(n, \gamma)$  capture, barn) and fluxes ( $n \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ ) in the whole fuel of each system at equilibrium.

The  $^{232}\text{Th}$  inventories at equilibrium are about  $2.4 \cdot 10^5$  moles in the ADS core and  $2.9 \cdot 10^5$  moles in the MSR molten salt circuit. Looking at the  $^{232}\text{Th}$  capture cross sections and at the mean fluxes, we notice that both  $^{232}\text{Th}$  capture rates are equal to about  $1.3 \cdot 10^{-4}$  mol/s, which is close to the total fission rate of 11.2 mol/day imposed by the constant power of 2500 MWth.

Concerning Protactinium, it is obvious that its reprocessing is a necessity for the MSR, because of a much higher capture cross section.

Finally, the higher  $^{233}\text{U}$  fission cross section in the molten salt fuel explains that the  $^{233}\text{U}$  inventory is about 6 times lower in the MSR, which is the main reason for its good breeding performance. The better neutron balance of  $^{233}\text{U}$  fission in the ADS (1.10 neutrons captured for one fission against 1.13 in the MSR) has no significant influence.

## D. Radiotoxicities

The radiotoxicity by ingestion of a mixture of radionuclides  $R$  (Sv) is calculated as

$$R = \sum_i F_i A_i,$$

where  $A_i$  is the activity (Bq) of the  $i^{th}$  ingested nucleus and  $F_i$  its dose coefficient (Sv/Bq). We used  $F_i$  values for workers appearing in the Publication 68 [11] of the International Commission on Radiological Protection (ICRP). By means of the code DECAY [12], we obtain for each isotope present in the initial mixture the time evolution of the radiotoxicity due to its decay and to the decay of its daughters.

### 1. Waste Radiotoxicities at Equilibrium

The assumed fractions of each element going to waste are comparable for the two types of fuel. In the solid fuel case, 0.1 % of Uranium and Plutonium and 1.0 % of Protactinium and Minor Actinides are lost every 5 years [1]. Within the same period, the MSR reprocessing detailed in III.B.2. amounts to losses of 0.2 % for all Heavy Nuclei.

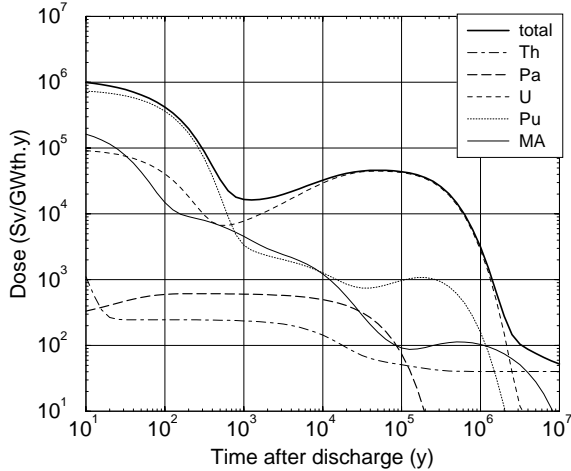


FIG. 6 – Radiotoxicity due to the Heavy Nuclei losses of the MSR at equilibrium.

Fig. 6 shows the radiotoxicity due to the Heavy Nuclei losses at equilibrium normalized to an energy production of 1 GWth.year. The initial level is, as with the solid fuel ADS, about 30 times lower than in the  $^{238}\text{U}/^{239}\text{Pu}$  cycle [1].

In both cases, at equilibrium, the principal contributions to waste radiotoxicity come mainly from  $^{238}\text{Pu}$  (during the first 1000 years after discharge) and then from  $^{233}\text{U}$ . At  $t=0$  and  $10^5$  y, the maximal levels respectively due to  $^{238}\text{Pu}$  and  $^{233}\text{U}$  are approximately the same for the two systems.

At  $t=10^3$  y, the total radiotoxicity is minimal. In the fast ADS case,  $^{231}\text{Pa}$  dominates waste radiotoxicity at this time. In the MSR softer spectrum,  $^{231}\text{Pa}$  production (by (n,2n) on  $^{232}\text{Th}$ ) is slower and  $^{231}\text{Pa}$  disappearance by capture faster, which explains that the  $^{231}\text{Pa}$  inventory is about 20 times smaller than in the ADS. We find that the  $^{231}\text{Pa}$  contribution to waste radiotoxicity 1000 years after discharge is about 100 times lower for the MSR, which is consistent with the respective Pa reprocessing losses (5 times lower in the MSR case).

### 2. Total Radiotoxicities for 200 Years of Energy Production

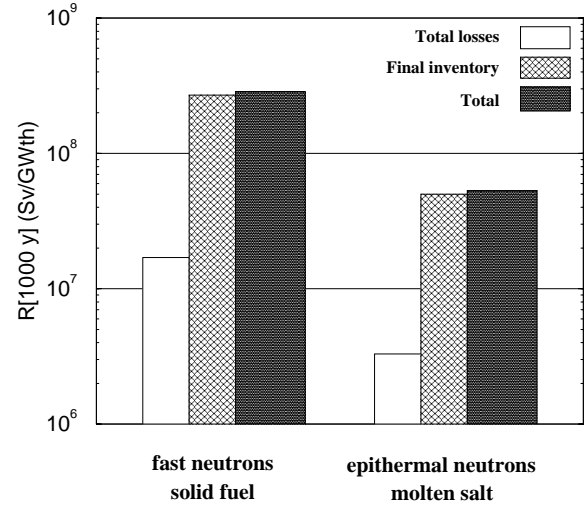


FIG. 7 – Radiotoxicities, 1000 years after discharge, due to the total Heavy Nuclei losses and the final inventory after 200 years of energy production.

Fig. 7 shows the radiotoxicities generated 1000 years after discharge by 200 years of continuous energy production, per unit of installed thermal capacity (Sv/GWth). The contributions of the accumulated HN losses and the final inventory are given for each system. All values are much lower than the  $R[1000\text{ y}]$  total values of PWR UOX and  $^{238}\text{U}/^{239}\text{Pu}$  spent fuels, respectively estimated to be  $3.4 \cdot 10^{10}$  and  $1.2 \cdot 10^{10}$  Sv/GWth.

Thanks to a lower HN inventory (in particular for  $^{231}\text{Pa}$ ), the  $R[1000\text{ y}]$  value of the MSR total losses is about 5 times lower than that obtained with the solid fuel system. We find the same factor in favour of the molten salt option for the final inventory contribution and the total value.

## V. STARTING WITH Pu FROM PWR

In order to start a Thorium fuel cycle, Plutonium from PWR spent fuel can be used in both

systems as the initial fissile inventory with Thorium in “ $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$ ” reactors. While Plutonium is burnt,  $^{233}\text{U}$  is produced and accumulates in the fuel until the  $^{232}\text{Th}/^{233}\text{U}$  equilibrium is reached.

Concerning the MSR, keeping the HN inventory constant amounts to replacing the burnt Plutonium ( $\text{PuF}_3$ ) by addition of Thorium ( $\text{ThF}_4$ ) in the salt. We chose thus the same HN proportion in the salt as with the  $^{232}\text{Th}/^{233}\text{U}$  fuel, so that the same optimal equilibrium towards breeding is reached.

$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$	$^{241}\text{Pu}$	$^{242}\text{Pu}$
3.1	52.5	24.5	12.2	7.7

TAB. 4 – Isotopic composition (mol%) of Pu extracted from 5-year-old PWR UOX spent fuel.

In the following, the Plutonium that we use in our solid fuel and molten salt  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  reactors is let cool down during 5 years within the PWR UOX spent fuel before its extraction and use. Table 4 gives the isotopic molar composition of this Plutonium (“Pu” from now).

We need about 12 tons of this Pu to start the  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  ADS, whereas the MSR needs only 4 tons of the same Pu in order to be critical at start-up. Pu concentration in the molten salt stays below 1 mol%, which prevents precipitation problems.

### A. From Pu burning to $^{233}\text{U}$ breeding

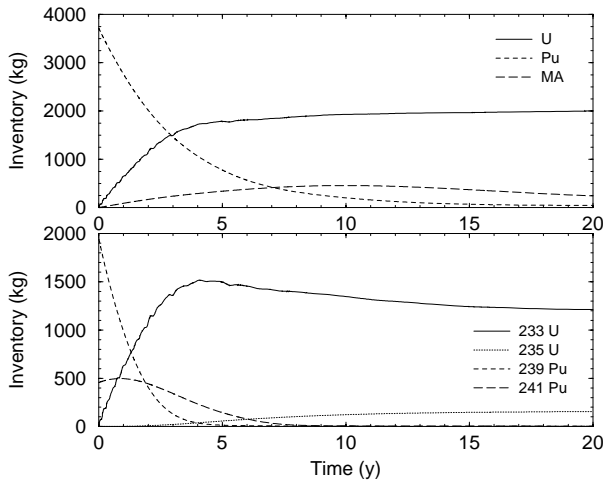


FIG. 8 – Principal inventories within the first 20 years of the  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  MSR.

Fig. 8 illustrates the Pu burning episode in the MSR, first through the evolution of the total U, Pu and MA inventories. From  $t=15$  y, the Pu inventory is below its  $^{232}\text{Th}/^{233}\text{U}$  equilibrium value. In the following, we consider thus that from this time the

initial Pu is forgotten and that the system operates roughly as with the  $^{232}\text{Th}/^{233}\text{U}$  fuel. This is only an approximation, in so far as the MA inventory is longer to reach its  $^{232}\text{Th}/^{233}\text{U}$  equilibrium value of about 60 kg. It is maximal at 10 years (450 kg) and then starts to drop (240 kg at 20 years, 120 kg at 30 years, ...).

The lower part of Fig. 8 shows the inventories of the principal fissile nuclei. During the first 5 years,  $^{233}\text{U}$  accumulates up to 1.5 tons through its feeding, in order to maintain criticality in spite of the increasing MA poisoning. At 15 years,  $^{233}\text{U}$  and  $^{235}\text{U}$  have definitively replaced the fissile isotopes of the initial Pu and reached their  $^{232}\text{Th}/^{233}\text{U}$  equilibrium inventories.

Thorium replaces progressively the burnt Plutonium in order to keep the total HN inventory constant. In consequence, the Thorium capture rate (8 mol/day at start-up) increases until it reaches its  $^{232}\text{Th}/^{233}\text{U}$  equilibrium value of about 11 mol/day at 10 years.

### B. French Transition to Thorium at Constant Power

Here, we want to compare the two systems by putting them in a context of transition to Thorium applied to the present French situation. The important point is that our simplifying hypotheses are the same for both scenarios. The main constraint on each scenario is that the total installed power is constant and equal to 60 GWe.

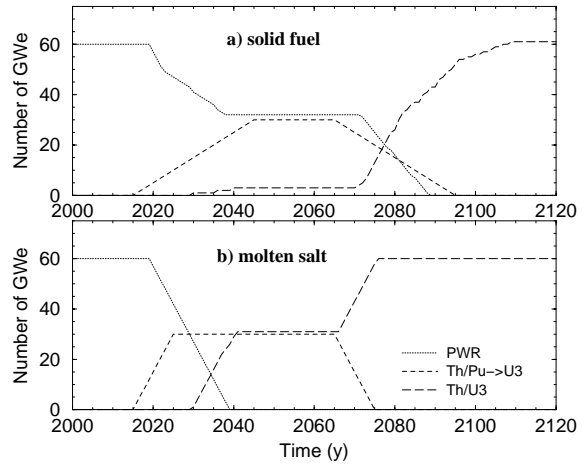


FIG. 9 – Evolution of the number of installed GWe in French transition to Thorium scenarios based on solid fuel (a) or molten salt (b) reactors.

We assume that the Pu reprocessing into MOX fuel stops in 2010. From 2015, Pu extracted from

5-year-old PWR UOX spent fuel is thus available at the rate of 12 tons per year for  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  reactors (only this “Pu” is used for the transition). As soon as  $^{233}\text{U}$  is produced, it may be used in order to start directly the  $^{232}\text{Th}/^{233}\text{U}$  reactors. If the total power is still below 60 GWe, new PWRs have to be installed. Every reactor has a 1 GWe power and a 50-year lifetime.

Fig. 9 shows that, with these constraints, more than 30 new PWRs are necessary in the scenario based on the solid fuel reactors. Indeed, the initial Pu inventory of the  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  solid fuel reactor is 12 tons and only one can be started-up every year. In this scenario, the last PWR should be installed in 2040, and therefore the 100% Thorium-based fuel cycle should be reached in 2090. The long doubling time (about 100 years against 25 years for the MSR) is not really a problem, since we just want to keep the total power constant.

Thanks to a 3 times lower Pu initial inventory (4 tons), the molten salt scenario needs no additional PWR. In 2040, when the last PWR should stop, the youngest  $^{232}\text{Th}/\text{Pu}\rightarrow^{233}\text{U}$  reactor should be 15 years old. At this time, the whole fuel cycle should be close to the  $^{232}\text{Th}/^{233}\text{U}$  equilibrium.

## VI. CONCLUSION

The results presented in this paper emphasize the main advantages of using molten salt rather than solid fuel reactors for transition to Thorium. The better breeding and radiotoxicity performance of the MSR are mainly due to much lower inventories and a greater freedom in fuel reprocessing.

In the case of a transition to Thorium scenario at constant nuclear power for an already developed country, the molten salt option is faster but solid fuel remains a solution. This is no longer true in world-wide scenarios, when we take into account the general evolution of energy needs and ecological problems. From this point of view, an intensive contribution of nuclear power seems to be a realistic way to reduce the  $\text{CO}_2$  emissions while satisfying the increasing demand for energy in emerging countries [13]. In this context, the  $^{232}\text{Th}/^{233}\text{U}$  fast reactor is no longer competitive, because of too slow breeding. The  $^{238}\text{U}/^{239}\text{Pu}$  fast reactor is then the only competitor of the  $^{232}\text{Th}/^{233}\text{U}$  MSR, but its induced radiotoxicities are considerably higher.

Our detailed studies remind us of the well-known advantages of the MSR with Thorium. It is all the more interesting to rediscover the molten salt option since it can play a significant role in future nuclear energy production. Therefore, the already considerable knowledge of molten salt systems should be

completed by new simulations and experiments, particularly on safety and chemistry aspects.

## ACKNOWLEDGEMENTS

One of the authors (A.N.) acknowledges partial support from EDF, Clamart, France. Thanks to M. Asghar and E. Huffer for their valuable remarks.

## REFERENCES

1. S. David et al., Fast subcritical hybrid reactors for energy production: evolution of physical parameters and induced radiotoxicities, Nucl. Instr. Meth. A 443 (2000) 510-530.
2. M. W. Rosenthal et al., Molten-Salt Reactors-History, Status, and Potential, Nucl. Appl. Tech. vol. 8 (1970) 107-117.
3. J. F. Briesmeister, MCNP4B - A General Monte Carlo N Particle Transport Code, Los Alamos Laboratory report LA-12625-M (1997).
4. A. Fassò et al, Nucl. Instr. Meth. A 332 (1993) 459.
5. R. E. MacFarlane, NJOY97, Code System for Producing Pointwise and Multigroup Neutron and Photon Cross Sections from ENDF/B Data, LANL PSR-368 (1997).
6. EDF/DER, “Analyse critique du projet MSBR”, HT-12/24/77.
7. P. N. Haubenreich et al., Experience With the Molten-Salt Reactor Experiment, Nucl. Appl. Tech. vol. 8 (1970) 118-136.
8. P. Faugeras, “Revue des domaines clefs d’étude sur les réacteurs à sels fondus”, note technique CEA N.T. SPRC/LEDC 98/405 (1998).
9. S. David, H. Flocard, F. Naulin, J-P. Schapira, U-Pu cycle waste incineration within a transition towards a Th-U cycle, GLOBAL 2001 (Paris).
10. H. Nifenecker et al., Hybrid Nuclear Reactors, Prog. Part. Nucl. Phys. 43 (1999) 753.
11. ICRP Publication 68, Pergamon Press (1995).
12. S. Ménard, J-P. Schapira, in “Impact Radiologique à Long-Terme de l’Extraction du Thorium”, IPNO DRE 95-07, IPN Orsay, France.
13. H. Nifenecker et al., Scenarios with an intensive contribution of nuclear energy to the world energy supply, GLOBAL 2001 (Paris).