MASTERY OF THE PLUTONIUM IN HIGH TEMPERATURE REACTOR

F. DAMIAN [1], *X. RAEPSAET* [1], *M. LECOMTE* [2]

[1] CEA-Saclay DEN/DM2S/SERMA 91191 Gif-sur-Yvette Cedex (France) (33) 1 69 08 46 64
[2] Framatome Tour Framatome 92084 Paris La Defense (France) (33) 1 47 96 56 73

ABSTRACT

Whatever the future of the civil nuclear programme in France may be, the plutonium reprocessing and recycling option has been chosen 14 years ago and the control of the plutonium inventory appears today as a major R&D issue. Many studies in progress at CEA attempt to improve plutonium recycling in PWR by increasing the amount of plutonium fed in the core, using inert matrix, new design, ...[1,2,3,4]. Moreover, in spite of their good performances and safe behaviour, innovative reactor concepts considered at the present time must also demonstrate their capacity to use at best the plutonium matter that represents at the same time a great energetic potential and strong radiotoxic source in spent fuel. In this context and with regard to the renewed interest in the High Temperature Gas-cooled Reactor (HTGR) concept, the problem of the mastery of the plutonium stock with the help of the HTGR has been undertaken at CEA in collaboration with Framatome.

Keywords: HTGR, prismatic core type, plutonium cycle, multi-recycling, radiotoxicity

INTRODUCTION

Preliminary studies concerning mono-recycling of the plutonium in HTGR have been performed in the past [5]. It has been shown that due to its highly versatile and flexible core that can fulfil a wide range of diverse fuel cycles by accommodating different neutronic parameters, such as fissile/fertile fuel particle fraction, particle volume fraction in the graphite matrix, type of fuel (enrichment, plutonium quality and content...), burnable poison, ... the HTGR appears as an attractive way to exploit the merits of the plutonium.

In the first part of this paper, we will try to highlight the main trends and the physical characteristics of HTGR fully loaded with plutonium. The potential of several fuel cycles with a large variety of plutonium grades are compared. Then, on the basis that improvements in the future fuel performances and technology should be accomplished and assuming that the fuel particles reprocessing does not represent a killing issue [6], it is interesting to investigate the possibility of multi-recycling the plutonium in HTGR without, as a first step, limiting themselves by the fuel technological constraints.

Two scenarii based on pure plutonium fully loaded core configurations have been considered here. The first one was focused on the possibility of achieving an equilibrium cycle considering that all the plutonium discharged from an HTGR core would be re-used. It could be mixed with a certain amount of fresh plutonium (coming from PWR). This amount is determined to maintain a sufficient reactivity level. An iterative calculation process has been adopted to predict this equilibrium cycle with its characteristics (time to reach the equilibrium, cycle length, fuel burnup, ...). The second scenario is established on a more pragmatic approach. Assuming a typical annual LWR production of plutonium, the problem concerns the evaluation of the number of HTGR able to burn this production considering a similar cycle length (280 EFPD). As in the previous scenario all the plutonium discharged from the HTGR is mixed with fresh plutonium and thus waste coming out from this recycling does not contain plutonium.

1 - PLUTONIUM FUEL CYCLE CHARACTERISTICS

The aim of this first part is the analysis of the GT-MHR core behaviour with regards to the fuel cycle length and the plutonium and minor actinides balance whatever the mass of plutonium loaded into the core or the isotopic composition of the plutonium are.

1.1 - Categories of fuel investigated

Several plutonium compositions coming from the spent Light Water Reactor fuel (LWR) have been envisaged. Partially burnt in France in the same LWR as Mixed-Oxide (MOX) fuel, it generates a second generation of plutonium that has also been taken into account in the present study. Plutonium containing typically 36 % to

54 % of ^{239}Pu (50 to 66 % in fissile Pu) allows to cover a wide scale of isotopic vectors that could be found today or in a near future.

Cycle name	*Plutonium origin*	*Pu_f/Pu_t*
Pu1	PWR# 3,7 % (U5/U) 42 GWj/t - 1/4	66,2 %
Pu2	EPR# 3,53 % (U5/U) 45 GWj/t - 1/5	64,2 %
Pu3	PWR 4,5 % (U5/U) 55 GWj/t - 1/6	62,7 %
Pu4	1st generation MOX-PWR 45 GWj/t - 1/4	58,5 %
Mox1	Second generation. MOX-PWR 33 GWj/t - 1/3	56,2 %
Mox2	Second generation MOX-EPR 60 GWj/t	50,15 %

#PWR, EPR : Pressurized Water Reactor and European Pressurized Reactor

Table 1 : Plutonium isotopic composition

1.2 - Computer Codes and Methodology

For the following calculations, the French reactor physics code system SAPHYR is used. SAPHYR gathers several CEA codes like APOLLO2 (transport) based on a database produced with THEMIS/NJOY, CRONOS2 (diffusion-transport), FLICA4 (3D- thermal hydraulics), ..., which are interconnected. The calculations have been performed on the Gas Turbine Modular Helium-cooled Reactor (GT-MHR) 600 MWth concept [7]. The core neutronics analysis is essentially based on a specific calculation process employing the APOLLO2 transport code [8]. The standard 172-groups cross section library issued mainly from JEF 2.2 is used in the present study. The core depletion calculations are essentially based on a representation in infinite medium of the fuel element treated with the P_{ij} transport method. The geometry representative of the standard fuel element is a hexahedral multicell geometry (Fig. 1).

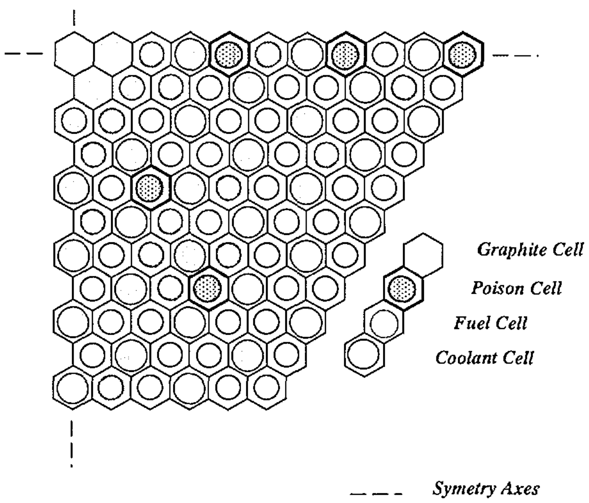

Figure 1 : Multicell geometry – ¼ element

In order to estimate the multiplication coefficient (keff) of the reactor core during evolution, a similar method than the one described in [5] has been used. This permits to take into account the reflector efficiency function of the neutron spectrum variation during the fuel depletion. The discharged burnup was determined in order to achieve a reactivity margin of 2000 pcm at the end of cycle (k_{eff} = 1.02) embracing the possible uncertainties.

1.3 – Plutonium Fuel Cycle Results

Impact of the plutonium loading and the isotopic content on the reactivity margin

Some of the characteristics of the fundamental mode are presented hereafter. Figure 2 shows the evolution of infinite multiplication coefficient for three types of plutonium loading and two reactor grade plutonium with the highest and the lowest fissile isotopic content (Pu1 and Mox2). On the one hand, the increase of the plutonium loaded in the fuel element leads to a spectrum hardening that induces a decrease of the reactivity margin at the beginning of cycle (decrease of the anti-trap factor). On the other hand, the decrease of the fissile isotopic content leads to a similar effect, i.e. a strong decrease of the reactivity margin that restraint the use of highly degraded fuel.

Fuel cycle length

As far as the fuel cycle length is concerned, the consequences of an increase of the total mass fed into the core have been analysed for various types of fuel. All the results are gathered in the Figure 3. Whatever the plutonium isotopic content is, the fuel cycle length is proportional to the total mass loaded into the core. The higher the plutonium loaded into the core is, the longer the fuel cycle length. Nevertheless, an increase of the plutonium loaded into the core is limited by technological and physical criteria. For example, the particles volume fraction in the compact represents a technological limit to the plutonium loading capacity. Besides, the reactivity margin at the beginning of cycle appears as a physical limit to the use of highly degraded plutonium or important fuel loading. Indeed, higher plutonium loading imply an increase of great absorbers like ^{240}Pu in a similar core geometry and reduce the reactivity margin although the fissile isotopes content increases. By increasing the loaded fuel mass, the neutron spectrum becomes harder and favours the neutron absorption in the fertile isotopes.

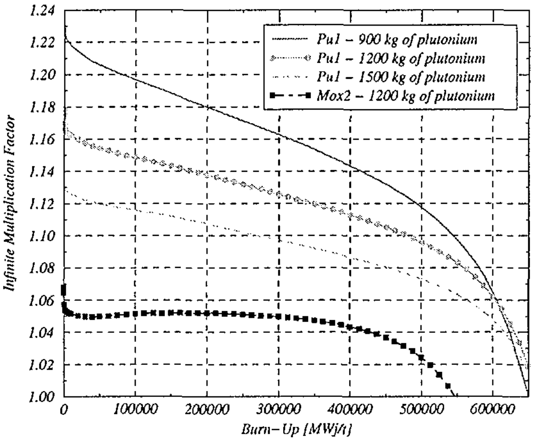

Figure 2 : Infinite multiplication factor in evolution

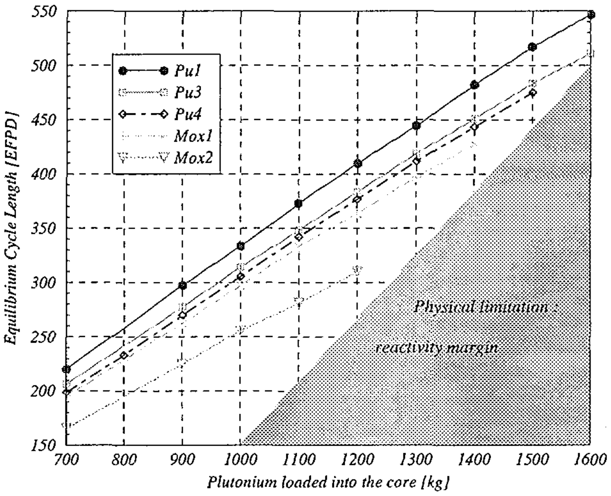

Figure 3 : Impact of the plutonium loaded into the core on the fuel cycle length for different isotopic vectors

Plutonium and minor actinides balance

The isotopic balances have been calculated for various type of fuel and are gathered in Table 2 and Table 3. All the isotopic balances are given for a cooling time of 5 years after the fuel discharge. A general trend emphasized in these tables is a plutonium consumption of about 100 kg/TWhe whatever the plutonium isotopic contents and the fuel loading are. This plutonium balance corresponds to a plutonium consumption of about 210 kg/year. Indeed, the high conversion process existing inside this type of core implies that a great part of the initial plutonium loaded into the core is burnt (between 55 to 72 %), even for highly degraded isotopic vectors of plutonium. Furthermore, it should be noticed that the fissile plutonium contents at the end of life are constant whatever the type of the loaded plutonium is. The stabilization of the fissile plutonium content (around 30 % for the discharged plutonium) seems to be very attractive in the way of the plutonium multi-recycling in high temperature reactor.

Fuel cycle	**Pu1**	**Pu3**	**Pu4**	**Mox1**	**Mox2**
Core loading [kg]	1200	1200	1200	1200	1200
Plutonium balance					
[%]	– 74,4	– 71,7	– 71,5	– 70,4	– 63,9
[kg/TWhe]	– 100,2	– 101,9	– 102,5	– 103,6	– 106,6
Pu_f / Pu_{total} at EOL [%]	30,0	29,4	28,4	27,5	28,4
Minor actinides balance					
[kg/TWhe]	+ 10,2	+ 11,9	+ 12,7	+ 13,8	+ 16,9
In % of metal burnt	10,2	11,7	12,4	13,4	15,8

Table 2 : Plutonium and minor actinides balances as a function of the plutonium composition

Fuel cycle	**Pu1**				
Core loading [kg]	701	900	1200	1500	1800
Plutonium balance					
[%]	– 67,4	– 71,3	– 74,4	– 75,4	– 75,1
[kg/TWhe]	– 98,7	– 99,3	– 100,2	– 101,0	– 102,0
Pu_f / Pu_{total} at EOL [%]	28,3	28,6	30,0	32,7	36,7
Minor actinides balance					
[kg/TWhe]	+ 8,25	+ 9,1	+ 10,2	+ 11,2	+ 12,2
In % of metal burnt	8,3	9,2	10,2	11,1	12,0

Table 3 : Plutonium and minor actinides balances as a function of the initial plutonium loading

However, in spite of a linear increase of the fuel cycle length with the plutonium loaded into the core, the plutonium balance for the discharge core reaches an optimum depending on the reached burn up (e.g. the optimum for the plutonium Pu1 with respect to the plutonium balance is about 1400 kg of loaded fuel). As it is shown in Figure 4, there is a strong correlation between the plutonium balance optimum and the fuel discharge burnup behaviour with regards to the mass loaded into the core. With respect to the total fuel loading, this optimum decreases with the plutonium fissile isotopic content (Fig. 4). As far as the minor actinides balance is concerned, the increase of the loaded plutonium induces an increase of the minor actinides production higher than the increase of the plutonium consumption (due to spectrum hardening). Thus, the minor actinides balances increase linearly whatever the plutonium contents or the mass loaded into the core are.

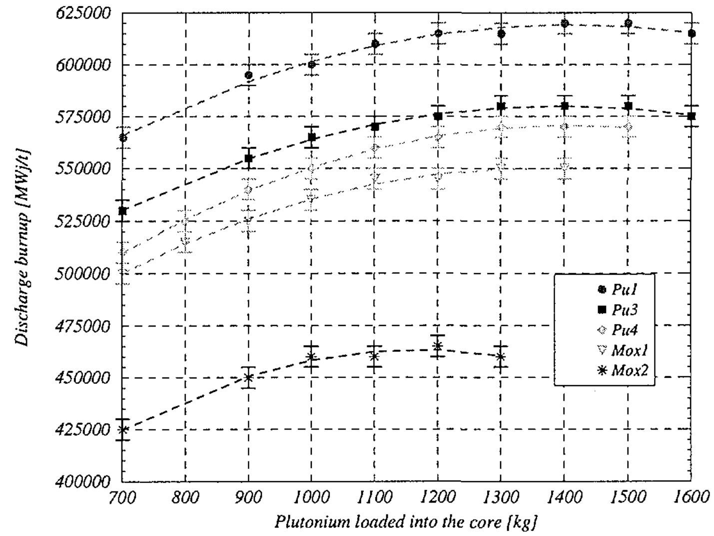

Figure 4 : Impact of plutonium loaded into the core on discharge burn up

2 - PLUTONIUM MULTI-RECYCLING IN HIGH TEMPERATURE REACTOR

Scenarii examined

The plutonium multi-recycling analysis has been done considering two different scenarii. However, the schematic diagram is still the same whatever the multi-recycling scenario is. For both cases, after the first fuel irradiation, the plutonium balance is evaluated in order to determine the plutonium fraction that has not been used. This plutonium fraction is mixed with a certain amount of fresh fuel, i.e. first generation plutonium coming from LWR, before being loaded in core. This process is run up to ten times (Fig. 5). In all the study, the time spent between two successive core irradiations takes into account a manufacturing delay of about two years and a cooling time after irradiation of five years.

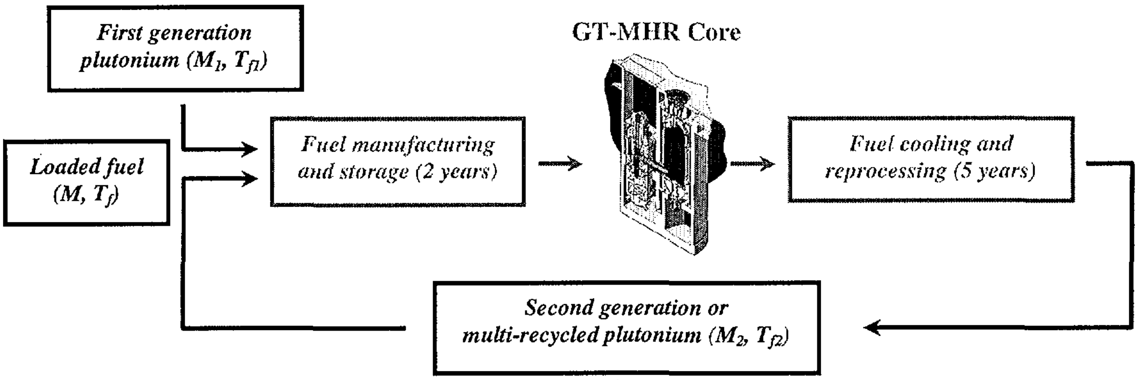

Figure 5 : Schematic diagram of plutonium multi-recycling

Scenario 1

As far as the first scenario is concerned, the possibility of achieving an equilibrium cycle has been evaluated assuming that all the plutonium discharged from the core is mixed with fresh plutonium (Pu1), the initial idea being to keep constant the fissile isotope content in the fuel loading. It was achieved by an adjustment of the amount of fresh fuel i.e. an adjustment of the total mass of the plutonium loaded into the core. However, no equilibrium cycles were found because more and more plutonium was required to respect this criterion. This led, as it is shown in Figure 3, to a too small reactivity margin at the beginning of cycle.

Finally, the calculations were performed with a constant mass of fuel fed into the core. The amount of fresh plutonium has then been determined to keep constant the core loading at 1200 kg. It is noteworthy that this plutonium loading has been chosen close to the optimum shown on Figure 4. All the results are gathered in the Table 4.

Fuel cycle number	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
Core loading [kg]	1200	1200	1200	1200	1200	1200	1200	1200	1200	**1200**
First generation plutonium fraction [%]	100	71,3	60,3	53,5	48,9	46,3	44,1	41,9	40,8	**39,6**
Multi-recycled plutonium fraction [%]	*	28,7	39,7	46,5	51,1	53,7	55,9	58,1	59,2	**60,4**
Fissile isotopic content for fuel loaded into the core [%]	66,2	56,7	52,3	49,7	48,0	46,8	45,9	45,2	44,7	**44,2**
Fissile isotopic content for fuel discharged from the core [%]	35,1	32,9	32,3	32,1	31,5	31,3	31,5	31,3	31,2	**31,0**
Discharge burnup [GWj/t]	615	485	415	370	345	325	305	295	285	**280**
Equilibrium cycle length [EFPD]	410	323	276	246	230	216	203	196	190	**186**

Table 4 : Equilibrium fuel cycle characteristics – Scenario 1

The tenth fuel cycle presented in Table 4 can be considered as an equilibrium cycle. Indeed, the amount of fresh plutonium is stabilized to 40 % of the total amount of plutonium loaded into the core (480 kg). The fissile isotopic content of the multi-recycled plutonium (60 % of the total amount) decreases up to 31 % while the average fissile isotopic content of the total plutonium loaded into the core is about 44,5 %. However, the isotopic degradation of the plutonium fed into the core induces a decrease of the equilibrium cycle length (~ 180 EFPD). As far as the minor actinides balance is concerned, the equilibrium of the discharged plutonium composition leads to an equilibrium production balance of the americium and the curium respectively of 24 and 6 kg/TWhe as it is shown on Figure 6.

The conclusion of this scenario is that an HTGR core can re-used and recycled all its own spent plutonium with a burning capacity of about 248 kg/year and a cycle length of ~180 EFPD. Considering that this burning capacity is not so far from the one of an annual plutonium production of a LWR, it is interesting to investigate if it is possible to determine the equilibrium cycle corresponding to a GT-MHR with an higher cycle length (then with a greater plutonium loading) and with a similar burning capacity. This has been tackled in the second scenario presented below.

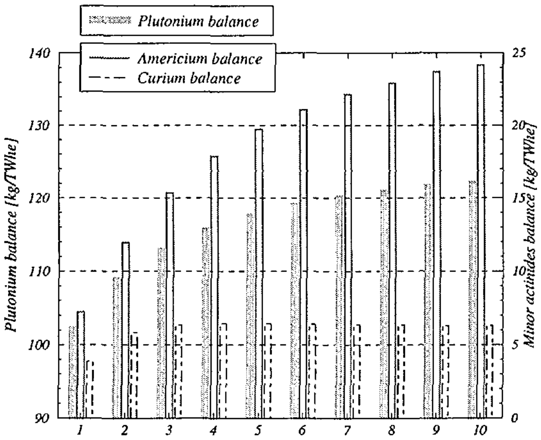

Figure 6 : Plutonium and minor actinides balances as a function of the fuel cycle number – Scenario 1

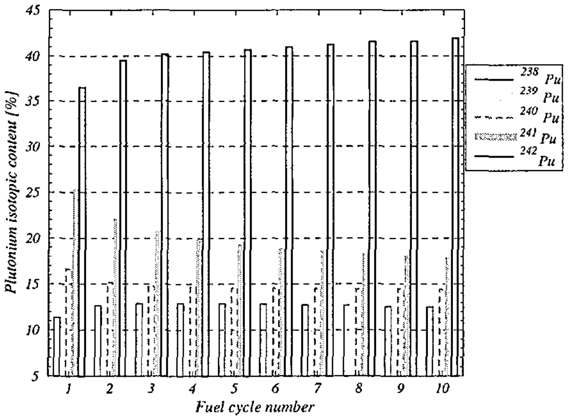

Figure 7 : Plutonium isotopic content for discharged fuel

Scenario 2

The second scenario is then established on a more pragmatic approach. Assuming a typical annual LWR reactor production of plutonium (1450 MWe-EPR), the problem concerns the evaluation of the number of GT-MHR able to burn annually this production. The major constraint for this scenario is the fuel cycle length that is imposed by LWR fuel management (280 EFPD). Assuming a fuel management by 1/5 for LWR and knowing the amount of fresh plutonium that is available each year, it is necessary to evaluate the total amount of plutonium to be loaded into the core in order to reach a constant fuel cycle length of 280 EFPD. In this study, the amount of fresh plutonium (Pu2 in Table 1) is fixed to 245 kg/year. Of course, as in the first scenario, all the irradiated plutonium coming out from the HTGR is entirely re-used at the end of each cycle. All the results concerning this scenario are gathered in Table 5.

Fuel cycle number	**1**	**2**	**3**	**4**	**5**
Core loading [kg]	900	1008,1	1162,6	1319,8	1433,2
Fissile isotopic content for fuel loaded into the core [%]	63,2	55,0	51,3	49,6	49,1
Fissile isotopic content for fuel discharged from the core [%]	32,5	31,0	32,0	34,3	36,3
First generation plutonium fraction [%]	100,0	74,9	64,8	58,7	53,9
Multi-recycled plutonium fraction [%]	*	25,1	35,2	41,3	46,1
Equilibrium cycle length [EFPD]	287	272	274	275	271
Discharged burnup [GWj/t]	575	485	425	375	340

Table 5 : Equilibrium fuel cycle characteristics – Scenario 2

With a fixed cycle length of 280 EFPD, it has been necessary to increase and adjust the plutonium loading at the beginning of each fuel cycle in order to maintain the cycle length. However, if the fissile plutonium content of the loaded fuel might be considered as stabilized at the end of the fifth cycle, the decrease of the core reactivity margin restraints the use of the discharged plutonium for the sixth cycle. Moreover, the increase of the total plutonium loading between the third and the fifth cycles is high (+ 157 kg and + 113 kg) whereas the plutonium fissile content decreasing in the fuel loading is rather low (about 1 %). These results indicate that the optimum with regards to the plutonium loading and its quality has been exceeded (see Fig. 4). The number of HTGR required to burn all the plutonium discharged from the LWR has also been evaluated. This calculated fraction is constant around 0.9 and corresponds roughly to one HTGR per LWR.

Discussion

Despite the difficulties to reach an equilibrium cycle in the second multi-recycling scenario, this analysis provided many characteristics concerning the behaviour of high temperature reactors with regards to the plutonium multi-recycling. Indeed, the fuel cycle length criteria imposed in the second scenario seems to be too much constraining. Further calculations have been done in order to evaluate the main tendencies in the case of a lower fuel cycle length criteria. The results are gathered in Figure 8.

Whereas a fuel cycle length of 280 EFPD (case a) induces a constant increase of the plutonium loading with the number of fuel cycles, it should be stressed that a released fuel cycle criteria allows reaching a higher number of cycles (case c & d). Furthermore, the increase of the loaded plutonium at each fuel cycle becomes lower. The last case d (180 EFPD) let appear an equilibrium cycle. It corresponds to the possibility of having an HTGR absorbing 245 kg/year of plutonium and having a cycle length on the order of 180 EFPD. It is noticeable that this case is very similar to the equilibrium cycle defined in the first scenario and leads to an equivalent fraction (0.9) of HTGR per LWR needed to burn all the plutonium coming from these LWRs.

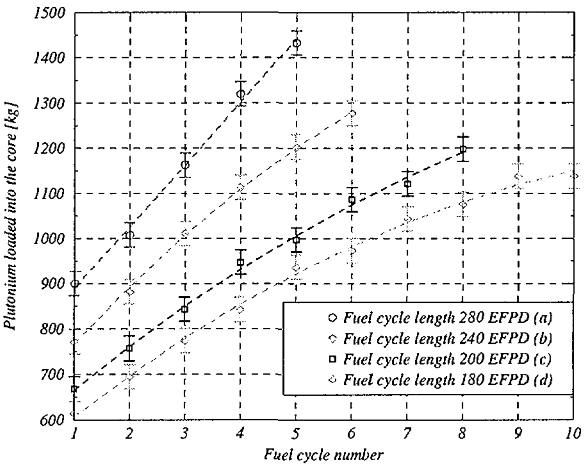

Figure 8 : Fuel cycle length impact on plutonium loaded into the core

Another solution to the fuel cycle length problem could be the change of the fuel management. In the present study, a fuel management by 1/3 has been chosen. This fuel management is required when highly enriched plutonium (Pu1 for example) is loaded into the core (high reactivity margin). When the plutonium fissile content decreases or the plutonium loading increases, both reactivity margin and reactivity depletion during irradiation reduce. Thus, a fuel management by 1/2 could be envisaged provided that all the problems imposed by this change could be solved (fuel loading scheme, core spectrum transient, power peaking...).

CONCLUSION

This study allowed highlighting the major trends of the plutonium multi-recycling in an HTGR. A calculation method dedicated to a parametric analysis has been set up to evaluate the core reactivity evolution during the fuel depletion. The core modeling takes into account particular features of the HTGR such as the double geometric heterogeneity and neutron leakage evolution in the annular core.

The first results obtained with a wide spectrum of plutonium isotopic compositions indicate good HTGR potentials to use highly degraded plutonium. It also proves that the fissile content of discharged plutonium is constant whatever the plutonium isotopic composition is. Moreover, high fuel cycle length can be reached even with highly degraded fuel. All these results indicate that the HTGR is well adapted to the plutonium multi-recycling.

The plutonium multi-recycling analysis carried out in this study emphasized the difficulties to conciliate an imposed plutonium burning rate with a relatively long fuel cycle length (second scenario). Indeed, the recycling of all the plutonium discharged from an HTGR induces invariably a decrease of the fuel cycle length if the plutonium loading is kept constant. An increase of this total amount of plutonium loaded into the core allows reaching the fuel cycle length criteria but the consequences are a strong decrease of the reactivity margin at the beginning of cycle.

As a conclusion, both scenarii have demonstrated the possibility of having an HTGR loaded with a constant fuel mass of 1200 kg that would burn about 245 kg/year with a cycle length ranging from 180 to 200 EFPD. Nevertheless, it should be noticed that the time spent to reach the equilibrium cycle is relatively high (between 5 and 10 fuel cycles) and that the amount of plutonium loaded into the core is rather low with regards to the plutonium stockpiles available today.

As far as the heavy nuclei are concerned, the only wastes generated by this fuel cycle are the minor actinides (Am, Cm and Np). The estimation of the wastes radiotoxicity has been done in each case. In the code system SAPHYR, the DARWIN/PEPIN2 code [9] solves, by an analytical method, the radioactive decay equations for 762 fission products and 88 heavy nuclei. Figure 9 and Table 6 show that the total radiotoxicity generated by the multi-recycling of plutonium does not decrease significantly in the long term (case d). Moreover, the important plutonium destruction capacity of the HTGR shows that the mono-recycling in HTGR (case c) does not justify the fuel particles reprocessing. All these analyses are made on a 400 TWhe park of mixed LWR-HTR power plants and are compared to the scenario of the LWR in open-cycle and of the present plutonium LWR mono-recycling (case a and b).

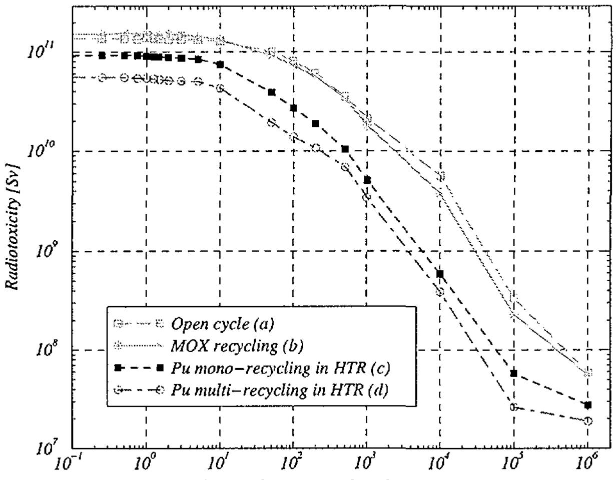

Figure 9 : Evolution of radiotoxicity (Sv) for different strategies in comparison with direct storage

Time (year)	$\frac{\textit{Direct storage}}{\textit{Pu mono} - \textit{recycling}}$	$\frac{\textit{Direct storage}}{\textit{Pu multi} - \textit{recycling}}$
1	1.5	2.7
10	1.7	3.4
100	3.0	14.1
1000	4.2	16.1
10000	9.6	17.4
100000	5.9	20.6

Table 6 : Gain for different strategies in comparison with direct storage

Due to the strong radiotoxicity of the generated minor actinides by plutonium multi-recycling, only one order of magnitude is achieved in better cases. The problem of minor actinides multi-recycled with plutonium remains then to be addressed in order to reduce more significantly the total waste radiotoxicity.

ACKNOLEDGMENTS

This work is being performed at Systems and Structures Modelling Department of the Commissariat à l'Energie Atomique, within the frame of a joint FRAMATOME ANP-CEA study program.

REFERENCES

[1] G. YOUINOU, J. L. GUILLET and A. PUILL, "Plutonium Management and Multi-recycling in LWRs using a ^{235}U support", Proc. Int. Conf., Global'99, Jackson Hole, USA (1999).

[2] A. PUILL, J. BERGERON and M. ROHART, "Mastery of the plutonium inventory in PWRs: the APA concept", International Conference on Future Nuclear System - Global'99 (Vol.1) (1999).

[3] J.P. GROUILLE et al., "Different possible scenarios for plutonium recycling in PWRs", International Conference on Future Nuclear System - Global'01, Paris, France (2001).

[4] H. GOLFIER, J. BERGERON, A. PUILL and M. ROHART, "Multi-recycling of plutonium and incineration of Americium, Curium, and Technetium in PWRs", International Conference – Safewaste'2000, Montpelier, France (2000).

[5] X. RAEPSAET, F. DAMIAN and M. LECOMTE, "Fuel cycle performances in High Temperature Reactor", International Conference on Future Nuclear System - Global'99 (Vol.1)

[6] N. D. HOLDER, W. S. LESSIG, "Advances in HTGR spent fuel treatment technology", GENERAL ATOMICS, Report A17625, August 1984.

[7] A.I. KIRYUSHIN, N.G. KODOCHIGOV, N.G. KOUZAVKOV, N.N. PONOMAREV-STEPNOI E.S. GLOUSHKOV, V.N. GREBENNIK, "Project of the GT-MHR high-temperature helium reactor with gas turbine", Nuclear Engineering and Design 173, 119-129 (1997)

[8] R. SANCHEZ, A. HEBERT, Z. STANKOVSKI, M. COSTE, S. LOUBIERE, C VAN DER GUCHT and I. ZMIJAREVIC, "APOLLO2 Twelve Years Later", Mathematics and Computation, Reactor Physics and Environmental Analysis in Nuclear Applications - Madrid - (September 99)

[9] A. TSILANIZARA, B. NIMAL, C. GARZENNE, P. MARIMBEAU, V. ALLAIS, C. MONIER, "The Darwin fuel cycle package - Procedures for material balance calculation and qualification" ENC'98 - Nice, France (1998)