

UTILISATION/DISPOSITION OF REACTOR-GRADE PLUTONIUM IN HTR'S

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ABSTRACT

High Temperature Gas Cooled reactors (HTRs) are able to accommodate a wide variety of mixtures of fissile and fertile materials without any significant modification of the core design. This flexibility is due to an uncoupling between the parameters of cooling geometry, and the parameters which characterize neutronic optimisation (moderation ratio or heavy nuclide concentration and distribution).

Among other advantageous features, an HTR core has a better neutron economy than a LWR because there is much less parasitic capture in the moderator (capture cross section of graphite is 100 times less than the one of water), in internal structures, and in fission products (because of a harder spectrum).

Moreover, thanks to the high strength of the coated particles, HTR fuels are able to reach very high burn-ups, far beyond the possibilities offered by other fuels (except the special case of molten salt reactors).

These features make HTRs especially interesting for closing the nuclear fuel cycle and *stabilizing the plutonium inventory*.

A large number of fuel cycle studies are already available today, on 3 main categories of fuel cycles involving HTRs : i) High enriched uranium cycle, based on thorium utilization as a fertile material and HEU as a fissile material; ii) Low enriched uranium cycle, where only LEU is used (from 5% to 15 %); iii) Plutonium cycle based on the utilization of plutonium only as a fissile material, with (or without) fertile materials.

Plutonium consumption at high burnups in HTRs has already been tested with encouraging results under the DRAGON project and at Peach Bottom. To maximize plutonium consumption, recent core studies have also been performed on plutonium HTR cores, with special emphasis on weapon-grade plutonium consumption. In the following, we complete the picture by a core study for a HTR burning reactor-grade plutonium. Limits in burnup due to core neutronics are investigated for this type of core.

With these limits in mind, we study in some detail the Pu cycle in the special case of a reactor fleet made of a mixture of LWRs and HTRs. It is reasonable to assume that if HTRs are to be deployed on an industrial scale, they will co-exist during a long period of time with already existing LWRs. The present paper investigates the symbiotic behaviour of LWRs producing plutonium, and of HTRs burning it.

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1. Introduction

High Temperature, Gas-Cooled reactors (HTRs) have several fundamental features which distinguish them from other types of reactors, and provide significant operational advantages.

HTRs are able to accommodate a wide variety of mixtures of fuels without any significant modification of the core design. This flexibility is due to an uncoupling between the parameters of cooling geometry, and the parameters which characterize neutronic optimization (that is moderation ratio or heavy nuclide concentration and distribution). In fact, it is possible to modify the packing fraction of coated particles in the fuel within the graphite matrix without changing the dimensions of the fuel elements.

Furthermore, different kinds of particles with various kernel diameters, containing either fertile or fissile isotopes may be accommodated in the same fuel. Other physical reasons favour the much better adaptability of HTRs with regard to the fuel cycle in comparison with reactors using moderators in the liquid form, such as LWRs. An illustration of that is the void coefficient which limits the plutonium content of PWR MOX fuels and which is not a constraint for HTRs. It is to be noted also that an HTR core has a better neutron economy than a LWR because there is much less parasitic capture in the moderator (capture cross section of graphite is 100 times less than the one of water), in internal structures, and in fission products (because of a harder spectrum).

Finally it must be noted that HTR fuels are able to reach very high burn-ups, which are far beyond the possibilities offered by other thermal reactors (except the particular case of molten salt reactors). This capability allows for essentially complete plutonium fission without recycle and minimizes the proliferation risk in the use of this fuel form.

2. - A classification of fuel cycles in HTRs

Numerous studies have been carried out in the past to assess and compare merits and drawbacks of all solutions which may be considered in HTRs, and which depend on:

- the type of fissile material (U233, U235, Plutonium), fertile material (Th232, U238), and enrichments,
- utilization conditions in the reactor: moderation ratio, frequency of reloads, fraction of the core replaced at each cycle, distribution of fertile and fissile materials among various categories of particles, etc.
- fissile material recycling strategy

A large number of results are available today [1], and these can be summarized by considering 3 main categories of fuel cycles :

- 1) High Enriched Uranium (HEU) cycle based on thorium utilization as a fertile material, and high enriched uranium (typically 93 %) as a fissile material
- 2) Low Enriched Uranium (LEU) cycle where only enriched uranium is used (from 5 % to 15 %)
- 3) Plutonium cycle based on the utilization of plutonium only as a fissile material, with (or without) fertile materials.

For completeness, it is to be noted that a kind of "hybrid" cycle called MEU (involving a mixture of medium enriched uranium (20%) with thorium) has also been considered. In the following, we shall discuss in more detail each of the three categories mentioned above.

2.1 – The HEU + Th cycle

Thorium, being the fertile material utilized in this cycle, generates U233, which is by far the best fissile isotope for thermal spectrum reactors. Furthermore, there are probably more thorium than uranium resources, and its utilization as a fertile isotope in reactors has been extensively studied, particularly for HTRs.

For these reasons, the HEU + Th cycle was considered as the reference cycle at the very beginning of HTR development in both USA and Germany. As a result, 4 HTR prototype power reactors having operated in the past (AVR [2] and THTR in Germany, Peach Bottom and Fort Saint Vrain in USA) were first loaded with fuel containing thorium in various forms such as carbides, oxides, and single thorium particles or mixtures with uranium.

The main advantage of this cycle is to significantly reduce uranium consumptions (typically from 10 % for an open cycle to a factor 2 or more when high conversion ratios are used in a closed cycle). Another advantage, which was not underlined in the past but which may become an important argument, is the significant reduction of minor actinide production if U233 is recycled in reactors, as would occur in reactors operated with Th-U233 cycles.

On the other hand, the competitiveness of an HEU cycle is questionable today, all the more so because there is a large uncertainty on thorium costs, since the market for this material is very limited. The main technical hurdle for the development of the HEU cycle comes from U233 recycling. The difficulty arises from the significant energetic γ emission of some daughter products of U232 (70 year half life), which is produced along with U233. This harmful emission essentially requires a remote fabrication process of U233 fuels. Technically this operation is certainly feasible with modern technologies, but significant R&D effort would be necessary to industrialize it and to ensure its profitability.

2.2 – The LEU cycle

The LEU cycle uses uranium with a minimum enrichment of 5 to 6 %, which is more than the highest enrichments usually utilized in other current thermal reactors such as LWRs (for HTR's, the uranium enrichment for LEU cycles usually ranges from 5 % to 15 %). This is due to a rather diluted and homogeneous uranium distribution in HTR fuels which favours U238 resonance captures (self shielding effect is reduced). This greater neutron absorption must be compensated for by a higher enrichment.

The LEU cycle was studied during the 60's and 70's in USA and Germany as well as in England and in France. Some LEU fuels were loaded in the European experimental reactor DRAGON. Furthermore, Germany decided in the 80's to select this fuel for their future projects, and this kind of fuel is also the one used for the South-African PBMR project [3,4]. Japan has also selected this fuel as well for their HTTR experimental reactor, which is in operation today [5].

2.3 Plutonium fuel cycles with or without thorium

From a physics standpoint, the interest of plutonium use in HTRs comes from flexible features already discussed above. It is known that plutonium isotopes have very large capture resonances near the thermal range of the neutron spectrum. This is the reason why plutonium reactivity and evolution as function of time heavily depends upon its initial concentration and distribution in the fuel (because of self-shielding effect mentioned above). In that respect, a HTR provides a large margin to the designer for optimising fuel cycle characteristics.

The idea to use plutonium as the only fissile material but still with thorium as a fertile material was considered very early in the 60's within the framework of the DRAGON project [6]. The General Atomics company took up the studies in 1968 in a program with Edison Electric Institute. This program was carried out up to the manufacturing of a test fuel element and its irradiation in the Peach Bottom HTR.

To maximize plutonium consumption, more recent studies have been performed on plutonium HTR cores with no fertile material at all. This solution has been particularly considered in the framework of weapon-grade plutonium disposition [7,8]. For the reasons already discussed above, only HTRs offer such a possibility (a part from fast neutron reactors for which it seems theoretically feasible to design core containing only plutonium).

As far as net plutonium consumption is concerned, performances claimed for this type of fuel cycle are remarkable. For example, if we refer to joint studies between GA and MINATOM on what they call the "PC-MHR" (Plutonium Consumption – Modular Helium Reactor), plutonium consumption reaches 90 kg/TW(e)hr¹. As a guide, an EPR (European Pressurized Reactor) loaded with 100% MOX fuel could theoretically consume 65 kg/TW(e)hr. One can also compare with a Fast Neutron Reactor operating in "under-breeding" mode which could achieve a theoretical consumption of 80 kg/TW(e)hr, provided that advanced fuel containing 45% of plutonium are developed (see French "CAPRA" program).

From this short overview, we can draw the following main conclusions:

In spite of its potential advantages, the HEU + Th cycle appears to be handicapped because of proliferation concerns. However it should be worthwhile to carry out further studies in order to better assess its potential benefits with regard to other questions, such as minor actinide generation, which has taken much importance in recent years.

The LEU cycle benefits from the largest experience but needs an uranium enrichment beyond 5% which is the upper limit for most existing fuel cycle facilities in many countries. However, the centrifuge enrichment technology, which is presently emerging as the reference technology for uranium enrichment, is especially well suited to fulfill the HTR needs. Thanks to its intrinsic flexibility, it enables the production of uranium enriched in a wide range of grades with only minor modifications in the cascade arrangement. Therefore, production of enriched uranium for LEU HTRs on an industrial scale is already possible in the existing centrifuge plants, and should be even easier in the future ones. The

¹ It is easy to verify this figure by using a 50 % efficiency of the plant, and elementary fission energy release of 210 MeV. In fact, the net plutonium consumption reaches about 100 kg/TWhe due to the the formation of americium and curium from the plutonium by neutron capture.

development of new fuel fabrication plants, needed to manufacture HTR fuel particles and fuel elements, is probably a more delicate issue.

Thorium use is not specific to HTRs even though they are better adapted than other reactors to take advantage of thorium properties, because of a much higher resonance absorption rate of thorium in an HTR fuel. Recycling of U233 however would require more industrial experience.

For plutonium consumption, HTRs are, without doubt one of the best types of reactors. Only fast neutron reactors may compete in this domain, but at the cost of as much, or more, development as HTRs. Taking into account the acquired background on HTR fuels, it seems that there is no technological hurdle to achieve expected performances of plutonium consumption. Moreover, 20 years ago, the context was not the same and in particular, questions such as plutonium consumption or long-term waste management were not so acute. Therefore, it is worthwhile to reconsider this option in light of these new challenges.

3. Burning reactor-grade plutonium in HTRs. A fuel cycle scenario.

3.1 Assumptions

In this paper, we studied the fuel cycle for a plausible scenario describing the progressive introduction of Pu-burning HTRs in a fleet initially composed of LWRs.

In this preliminary step of the study, we did not try to treat the subject exhaustively. Indeed, we restricted ourselves to the assumption that the HTRs are exclusive Pu burners, and that the Pu comes from the LWRs.

Fuel cycle studies in steady state regime are of limited interest, because time constants for the introduction of new nuclear reactors are always very long. For the long-term future, fast neutron reactors might possibly become available before a steady state regime for any existing reactor fleet can be established. For the sake of plausibility, the scenario in this paper has thus been studied in transient regime, and covers only the mid-term future.

We are interested mainly in the plutonium balance, because Pu is an important energetic resource, and because it is the major contributor of the radiotoxic fuel inventory.

Hypotheses underlying the scenario under study are as follows :

- The studied scenario is deemed to be realistic for the next 50 years. However, in order to gain some insight into the asymptotic behavior of the reactor fleet, the studied time window has been extended to 100 years, that is between 2000 and 2100.
- The total electric power produced by the reactor fleet is kept constant during this period.
- We start with a realistic initial total plutonium inventory in 2025, supposed to be representative of the French situation at that time, namely 400 tons.

-We assume a gradual replacement of existing LWRs by Pu-burning HTRs. The rate of replacement is 2.1 % per year, between years 2025 and 2035. At that time, the reactor fleet is stabilized, and 21.25 % of the total installed electric power is produced by HTRs (Fig. 1) (This figure has been chosen because, as will be seen later, it ensures a quasi-stabilization of the total Pu inventory).

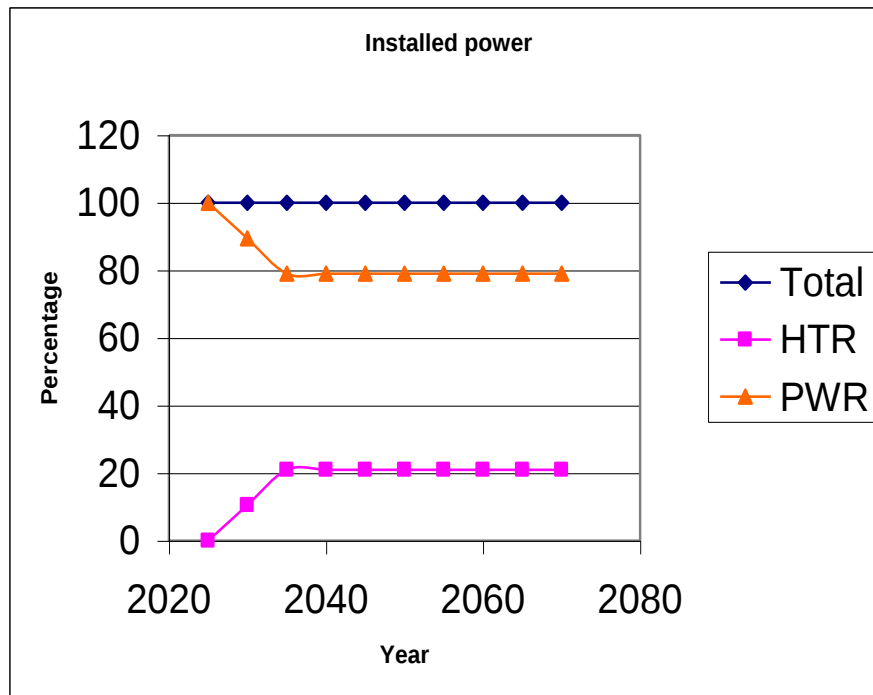


Fig. 1: Composition of the reactor fleet, vs time

-We assume that MOX fuel is used in 42% of the LWR fleet during the early years of the period under study (until 2025). After that date, we assume that the LWRs become exclusive UOX burners (noMOXing in LWRs after 2025). However, all the UOX fuel burnt in the LWRs is still processed to produce the Pu fuel needed for HTRs (in the following, this first-generation plutonium will be referred to as "Pu1"). All the MOX fuel is reprocessed as well, in order to feed HTRs with its second-generation plutonium (Pu2).

-The burnup of the UOX fuel in LWRs is assumed to be 55 GW.d/t, with a fuel renewed by one fifth every year. This corresponds to a net production of plutonium of 25 kg/TWhe.

-Since the high burnup achievable in HTRs does not encourage multiple recycling, we assume a once-through cycle of Pu in HTRs (discharged plutonium then becomes a waste).

-We assume a net consumption of plutonium in HTRs of 100 kg/TWhe. The Pu burnup achievable in a thermal HTR has been assumed to be 500 GW.d/t. As will be justified in section 4, this is probably a rather conservative value.

3.2 Results of the fuel cycle study in this scenario

Stabilization of Pu inventory is achieved around year 2030. This Pu inventory has two components : in and out of the reactors. The plutonium inventory out of the reactors is plotted vs time in fig. 2. The mass of Pu discharged from the HTRs grows linearly with time, while the Pu mass still available for HTR fuel fabrication decreases linearly after year 2040.

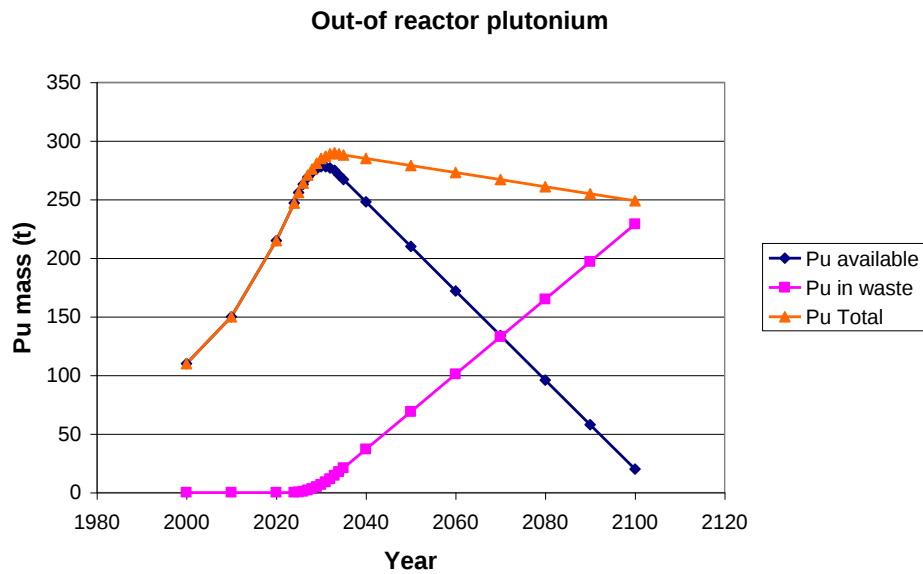


Fig 2: Evolution of the out-of-reactor Pu inventory for a fleet producing 400 TWhe/year

It can be seen that the plutonium mass available for HTR fuel fabrication almost vanishes around year 2100. We reach here the limits of a realistic scenario, since other types of reactors might possibly enter into the fleet long before this date.

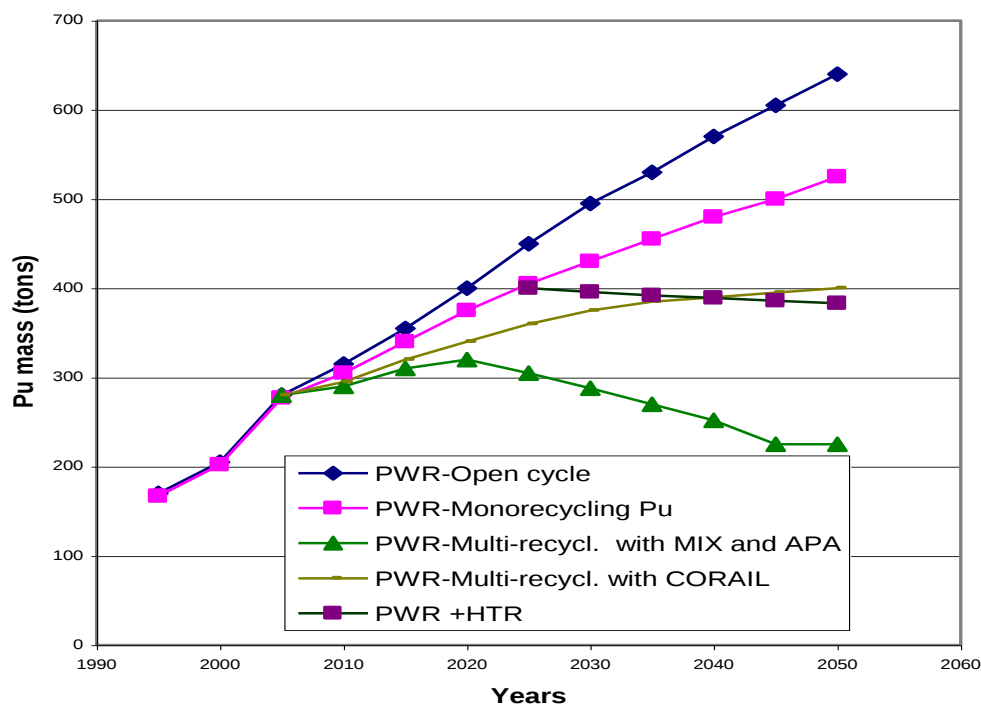


Fig. 3 Total plutonium inventory for a reactor fleet producing 400 TWhe/year

During the period under study in the present scenario, other fuel cycle options could be envisaged for light water reactors. For example, advanced fuels like CORAIL and APA, whose goal is precisely to enhance the Pu consumption in LWRs [9], could be introduced. It is thus interesting to compare

these options, which assume a pure LWR reactor fleet, with our symbiotic scenario LWR+HTR. The total Pu inventory versus time is plotted in Fig. 3 for all these options.

It can be seen here that for Pu burning, HTRs compare favorably to LWRs. The stabilization of Pu inventory is possible with LWRs only if advanced fuels are used and with multirecycling. A mixed fleet of LWRs and HTRs easily achieves the same result with standard fuel (for LWRs) and without multirecycling. However, the comparison between the various options is not straightforward, and needs a few comments. One should note that the isotopic composition of the Pu left as "waste" by the LWR + HTR symbiotic fleet is so degraded that the proliferation risk becomes negligible (Table 1).

Isotopic composition of the discharged plutonium	PWR/UOX 42 GWd/t	HTR/ Pu 575 GWd/t
Pu 238	2.7	10
Pu 239	54.5	8
Pu 240	22.9	23
Pu 241	11.7	24
Pu 242	7	35
% Pu fissile	66.2	32

Table 1. The plutonium isotopic composition of a typical spent UOX fuel (Pu1) and of a typical spent HTR fuel.

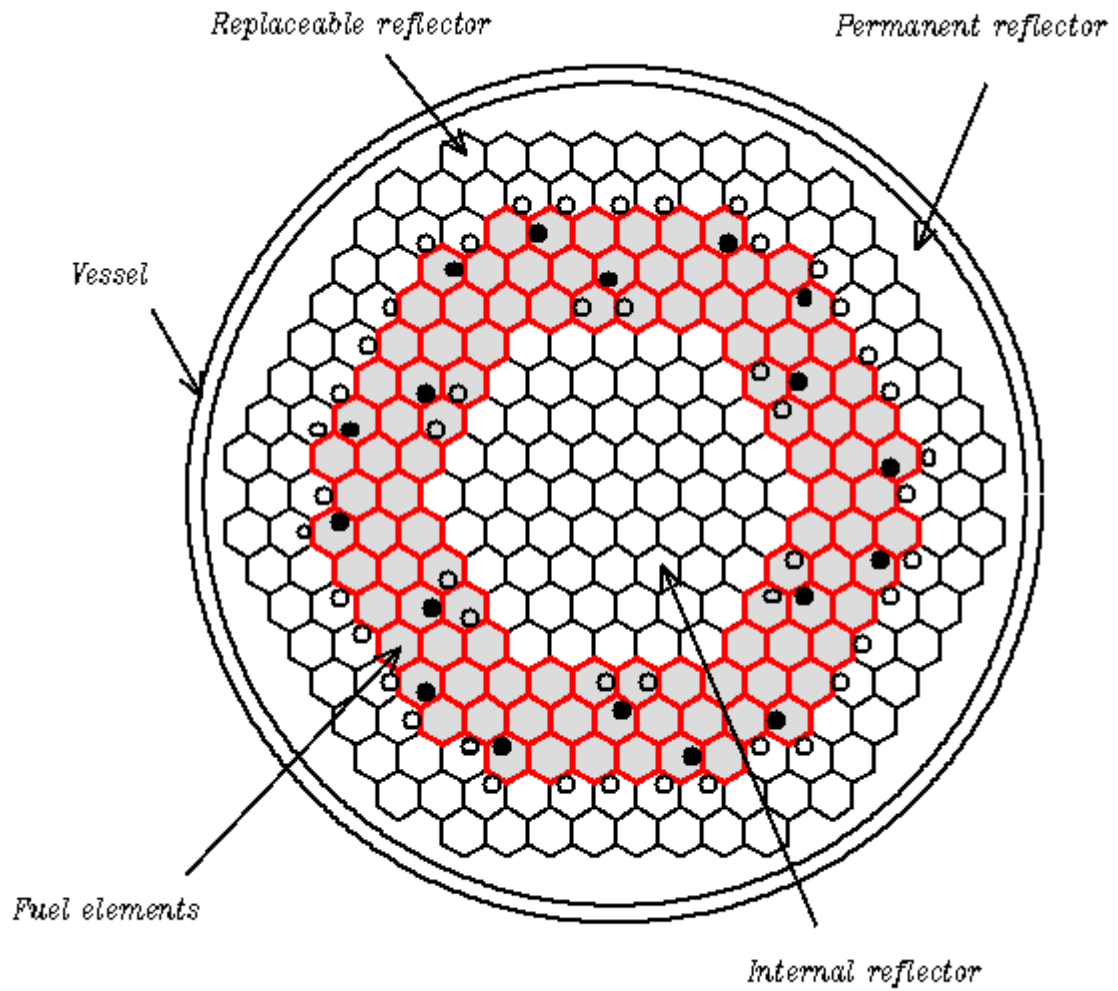
Moreover, whereas the advanced fuel discharged from LWRs may need some further conditioning, the Pu discharged from HTRs is under the form of coated microspheres, which can be considered already as a suitable waste form for interim storage or for final disposal. However, it must be noted that the deep burning of plutonium produces large quantities of minor actinides. Typically, the production of 1 TWhe by fission of 1st –generation plutonium results in the production of 12 kg of americium and curium. Calculations of the decay heat and radiotoxicity of spent fuel are under way.

4. Core studies for a Pu-burning HTR. Maximum Pu burnup achievable in a HTR

A key assumption in the above fuel cycle studies is the Pu burnup achievable in HTRs. The burnup limitation can come from two main causes : the stability of the fuel particles under irradiation, and the neutronic behaviour of the core. The fuel particle themselves do not seem to be a severe limitation, since burnups as high as 750 GW.d/t have been achieved without degradation of the fuel particles in previous experiments [6]. This encouraging –but isolated- result must be confirmed by new experiments.

In order to assess the feasibility of HTRs with an "allplutonium" core, preliminary studies, mainly based on a fuel element calculation, [10, 11, 12] were carried out at CEA in the special case of the Gas Turbine Modular Helium-cooled Reactor (GT-MHR) 600 MWth General Atomics concept. The annular core geometry of the GT-MHR (fig. 4) was selected to maximize the power density and still permit passive core heat removal while maintaining reasonable fuel temperatures during accident conditions.

Fig. 4 GT-MHR 600 MWth core



Cycle name	Plutonium origin [#]	Pu_f / Pu_t
Pu1	PWR 3,7 % (U5/U) 42 GWd/t - 1/4	66.2 %
Pu2	Second generation MOX-EPR 60 GWd/t	50.15 %
WPu	Weapons grade	94.6 %

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

Table 2 : Plutonium isotopic compositions

4.1 Categories of Pu fuels investigated

The Pu composition considered in this study is of course the first-generation Pu coming from the spent LWR UOX fuel (Pu1), but we thought it would be worthwhile to perform core calculations for other Pu compositions as well (Table 2). MOX burning in LWRs generates a second generation of plutonium (Pu2) that has also been considered in the present core study. Plutonium coming from the dismantling of nuclear warheads has also been studied (WPu).

4.2 Computer codes and methodology for core calculation

For the following calculations, the French reactor physics code system SAPHYR developed at CEA has been used [13, 14]. It is composed of several codes: APOLLO2 (transport), CRONOS2 (diffusion-transport), FLICA4 (3D thermal hydraulics), which are interconnected.

4.3 Plutonium fuel cycle characteristics

Fuel element analyses: reactivity margin and burnable poison impact

Figure 5 shows the evolution of infinite multiplication coefficient for Pu1, for two different masses of Pu loaded in the core, and with or without burnable poison (Erbium). On the one hand, it should be stressed that compared to a similar fuel depletion without erbium (Pu1–701 kg), the fuel with poison presents an initial reactivity reduction of around 9000 pcm which almost disappears towards 600 GWd/t, corresponding to the loss of 90 % of ^{167}Er . The cycle lengths are however comparable. This important

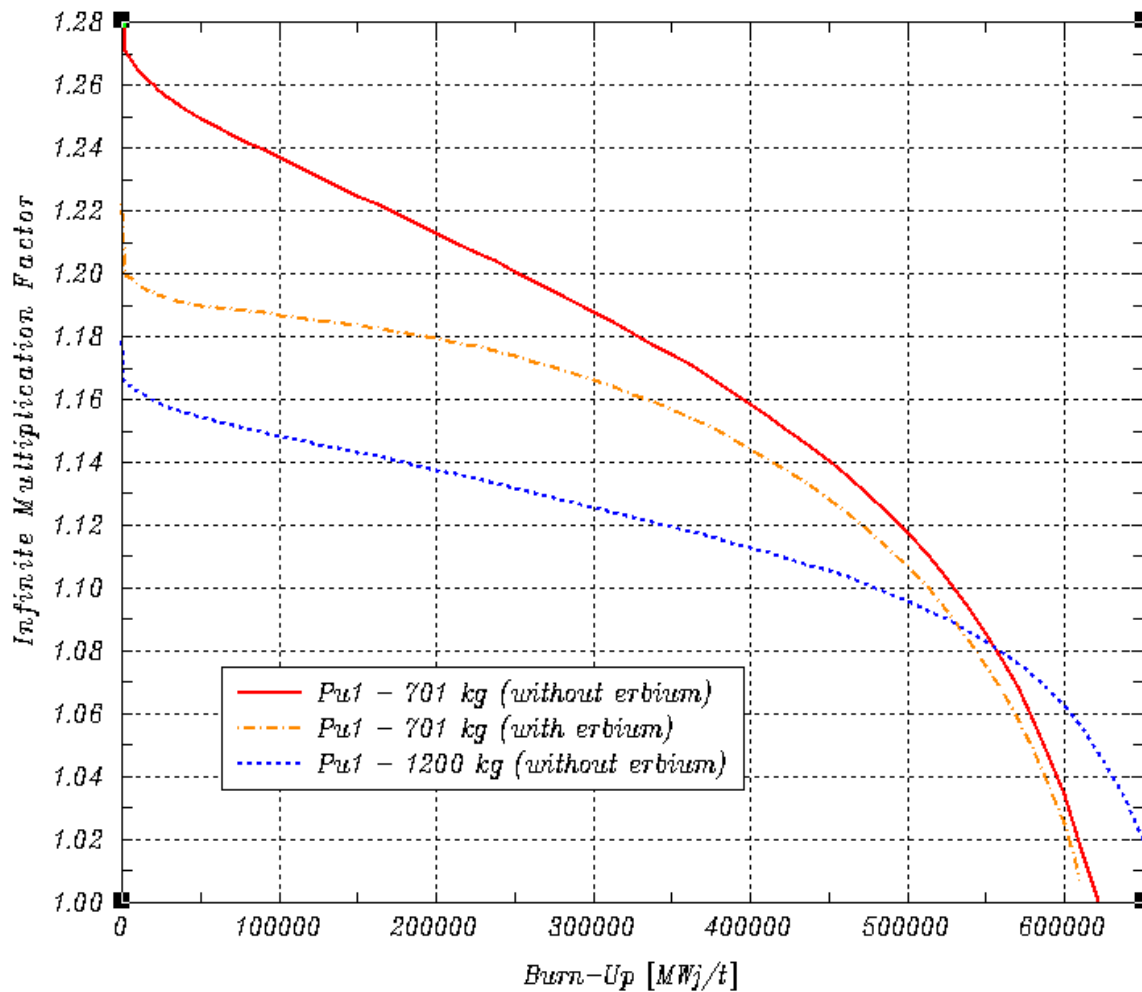


Fig. 5 k_{inf} in evolution

feature of the burnable poison equivalent to that observed with ^{10}B allows adjusting the initial reactivity

of the different fuel cycle without changing the cycle lengths. On the other 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 (increase of the resonance capture). Then, it is possible to adjust the initial reactivity by adjusting the plutonium loaded into the core. Nevertheless, we should not forget that the presence of the Erbium in fuel elements may play an important part in the core behaviour with regard to the negative reactivity feedback especially with highly enriched plutonium.

Fuel element temperature coefficient (infinite medium)

The results presented hereafter (Table 3) gather the average temperature coefficient (Doppler and moderator coefficient) between a cold state ($T_{\text{fuel}} = T_{\text{moderator}} = 20\text{ }^{\circ}\text{C}$) and an arbitrary nominal state ($T_{\text{fuel}} = 900\text{ }^{\circ}\text{C} / T_{\text{moderator}} = 500\text{ }^{\circ}\text{C}$). The calculations have been performed for various types of fuel in order to evaluate both isotopic and core loading effects on this reactivity balance.

Fuel	WPu - 701 kg with Er	Pu1 - 701 kg without Er	Pu1 - 1200 kg without Er	Pu1 - 1800 kg without Er	Pu2 - 1200 kg without Er
Doppler coefficient (pcm/K)					
BOC	- 1.33	- 2.76	- 3.49	- 3.70	- 3.99
EOC	- 1.09 (700 GWd/t)	- 0.98 (625 GWd/t)	- 0.92 (650 GWd/t)	- 1.14 (650 GWd/t)	- 1.61 (550 GWd/t)
Moderator temperature coefficient (pcm/K)					
BOC	- 5.33	- 2.29	- 1.91	- 1.46	- 1.83
EOC	+ 8.60 (700 GWd/t)	+ 8.15 (625 GWd/t)	+ 4.47 (650 GWd/t)	+ 1.74 (650 GWd/t)	+ 4.02 (550 GWd/t)

Table 3 : Fuel element temperature coefficient at the beginning of cycle (BOC) and at the end (EOC)

Finally, these calculations show that regardless of the plutonium quality, the Doppler coefficient remains negative throughout the fuel cycle. This is probably the most important factor for the pilotability and safety of the reactor. On the other hand, it should be noted that the moderator temperature coefficient becomes less negative with irradiation, and even becomes positive at the end of the fuel cycle. This is due to the modification of the isotopic composition of the core under irradiation, which induces in turn a thermalization of the neutron spectrum; The influence of the temperature on the reactivity is more important for a thermalized spectrum. This effect also occurs albeit to a lesser extent for LEU cores, and is probably of minor concern, given the large thermal inertia of the moderator, but a more detailed safety study would be needed to ascertain the viability of the "all plutonium cores", for the three types of Pu composition and for the burnups mentioned above.

Residual decay heat

For various types of fuel, Figure 6 shows detailed evolution of the stored energy during a Loss of Coolant accident. This energy comes from the core decay heat of the GT-MHR-600 MWth calculated after a reactor shutdown occurring at the end of cycle. Due to the difference in fission yield between the Pu isotopes, the calculation shows some differences in decay heat depending on the plutonium composition and loading. More important, for high Pu loading, the calculated values are significantly higher than for a typical "all uranium" core. The result will be an increase of the maximum temperature reached by the Pu core in the course of the accident. No detailed thermal calculation has been undertaken so far but our first guess is that this temperature increase is still acceptable for a Pu fuel element.

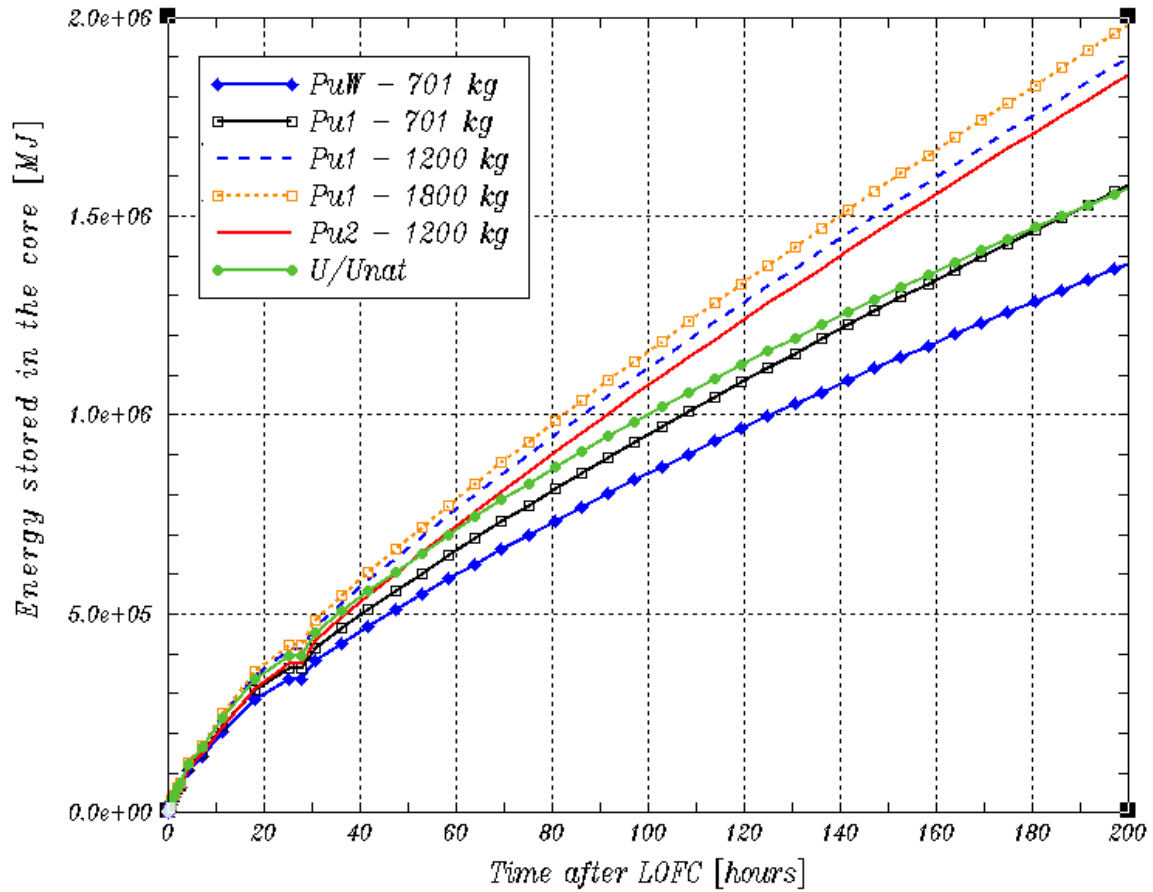


Fig. 6 Cumulated core decay heat after a reactor shutdown

5. Conclusion

Core studies with a wide range of plutonium isotopic compositions suggest that High-Temperature Reactors might be able to use the plutonium as fuel with no major hurdle from the safety standpoint. For each plutonium composition, limits on the possible applications of long fuel cycle lengths have been assessed. Long cycles and high burnups are possible if fluences of about 12 n/kb (a factor 2 more than the common requirements) are technologically feasible. Nevertheless, more detailed core neutronic analysis and thermal calculations are necessary to assess the reactivity control aspects, to define the appropriate fuel management and to answer the issues related to power distribution, which are especially important in the case of the plutonium use.

The present study confirms that a symbiotic reactor fleet involving LWRs and Pu-burning HTRs might be a very effective way to stabilize the plutonium inventory while providing energy in safe and (hopefully) economical conditions.

However, it must be stressed that even though a rather large experience has already been gained in HTR fuels, significant R&D effort would still be necessary to perfect and qualify a high performance plutonium fuel on an industrial scale for this type of reactor.

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