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Plutonium potential use in bloc type reactors

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INTRODUCTION

For a few years, the High Temperature Gas cooled Reactor (HTGR) technology has gained worldwide new interest due to its specific characteristics. It is a promising reactor concept for the next generation of nuclear power applications. In addition to the studies performed on the industrial concept (uranium core type), there is a strong interest in the investigations carried out on both

- a plutonium version of HTGRs emphasis on the use of civil plutonium from spent light water reactor (first generation of plutonium) and from spent LWR Mixed OXide fuel (second generation of plutonium)
- a «Deep Burner» version of HTGRs, dedicated to minor actinides destruction.

Indeed, it has been shown that HTGR have attractive characteristics concerning the use of plutonium. It has especially a flexible core that can fulfil a wide range of diverse fuel cycles. Until now several analyses of fuel cycles have been carried out without really taking into account common fuel particle performance limits (burnup, fast fluence, temperature). The use of a wide spectrum of plutonium isotopic compositions prove HTGR potentials to use at best the plutonium as fuel without generating large amounts of minor actinides.

However, long cycles and associated high level of Pu-destruction are possible if burnups as high as 700 GWd/t and fluences in the order of 12 n/kb (a factor 2 with the common requirements) sustained by the fuel particles are technologically feasible. The use of high-burnup plutonium particles cannot be regarded as proven technology today and important fuel characterisation program including irradiation will be required to demonstrate that a burnup equal about 80 % "fissions per initial metal atom" (FIMA) can be achieved for the Pu-particles without an inadmissible failure rate of the fuel coating.

Assuming such high fuel performances would be achieved not so far in a near future, the bloc type reactors with **batch-wise reloading scheme** have been studied in the frame of the **WP3 of HTRN**. **Different fuel cycles** based on pure plutonium or plutonium mixed with Minor Actinides (MA) have been investigated. In conjunction with these investigations, the **optimisation of burnable poison particle designs** (mainly required for batch-loaded HTRs) has been conducted. Indeed, it is well known that the use of the burnable poison, diminishes the role of the active reactivity control mechanisms, allow flattening the typical reactivity to time behaviour of the bloc type cores.

The present report provides only an overview of the work that has been done through the WP3 of HTR-N concerning the <u>bloc type reactors</u>.

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1. CALCULATIONAL METHODS

For the GT-MHR-based reference reactor, CEA investigated the Pu (and minor actinide) incineration capability. It is noteworthy that to give information such as cycle length, mass balance, peak power, core flux distribution, etc. for a specific block-type HTR loaded with plutonium fuels suppose an important *optimisation stage of the core*: use burnable poison or not, flattening the flux distribution in the annular zone (different filling fraction in the compact close to the reflector, different enrichment, burnable poison in the reflector, etc.), number of the control rods, etc. Moreover, this optimisation stage must be based on an equilibrium fuel cycle assuming a specific fuel-reshuffling scheme.

Besides, it should be stressed that the european calculational methods used in PBR and bloc type HTGR are not at the same level of maturity. If the <u>3D core burn-up calculations</u> are available for the PBR and largely and commonly used to evaluate the core performances assuming different pebble shuffling modes, those concerning the <u>HTGR block-type</u> appears a little bit immature. They are not so widely used and <u>remain to be validated and qualified</u>. Through the past few years, an important effort has been devoted in France to these validation steps especially as far as the **uranium fuel** is concerned. These validations are essentially based on Monte Carlo comparisons at each level of the calculation scheme (particles, compact, fuel element, 2D and 3D core calculations). A qualification step of this calculation scheme has also been done through the WP1 activities of the present HTR-N contract and this is also true for the others HTR-N partners. Even though <u>core burn-up calculations</u> are being validated on the base of *reference 2D core transport calculations*, this first validation stage for **uranium fuel** has not been carried out for all other sort of fuels containing **plutonium and minor actinides**.

The huge effort associated with these validation and qualification steps could not be planed in the present HTR-N contract. Just as the allocated effort to this WP3 could not allow using such new 3D core calculation scheme for block-type reactors. A simplest methods than 3D core burn-up calculations have been employed for the present study. 2D transport detailed calculations allowed to compute the fuel depletion. Nevertheless, in order to get the fuel element discharged burn-up, the core reactivity was calculated during fuel depletion using a simplified 2D annular core configuration on which also transport calculations have been done. It is important to note that all these calculations have been performed without taking into account temperature feedback. The same 2-D annular core configuration was used for the temperature coefficient estimations. The plutonium and minor actinides balances were calculated considering a thermal efficiency of 48 % and a loading factor of 0.85.

A complete description of the codes, methods and modelling hypotheses as well as the detailed analyses and interpretations of the obtained results are available in the <u>Task</u> <u>Reports</u> HTR-N-04/07-D-3.3.1 and HTR-N-04/07-D-3.3.2.

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2. PLUTONIUM INCENERATION CAPABILITY

All the fuel cycle investigations have been conducted on the basis of the bloc type reactor depicted on Figure 1, 2 and 3. The core consists of 102 columns of fuel comprising 72 standard element columns and 30 control element columns. The reflector and fuel columns consist of stacks of prismatic blocks with a height of 80 cm and 36.0 cm across opposite sides. The core also includes a reflector at the top and the bottom with a height of 130 cm.



Figure 3: Geometry of a control fuel element

In this study, two types of plutonium have been analysed. The first one (Pu1) corresponds to a first generation plutonium coming from LWR UOx Fuel [1]. As far as the second generation plutonium is concerned, the composition has been previously defined in [2].

2.1. PLUTONIUM INCENERATION

Preliminary investigations showed that:

- The fuel cycle length increases linearly with the mass of plutonium loaded into the core
- There is an optimum for the fuel fed into the core with respect to the discharge burnup, which allows using at best the plutonium

Indeed, an increase of the total mass fed into the core has been analyzed for both types of plutonium fuel. All the results are gathered in the Table 1. 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, the longer the fuel cycle length. Nevertheless, an increase of the plutonium loaded into the core will be limited by technological and physical criteria. For example,

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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.

 TABLE 1 Plutonium and minor actinides balance for 1st and 2nd generation plutonium fuel.

Type of fuel	1st gene	ration pluto	nium (66,2 9	%)	
Mass of fuel loaded into the core [kg]	701	900	1200	1500	1800
Plutonium balance					
[%]	- 67,4	– 71,3	- 74,4	- 75,4	- 75,1
Pu _f / Pu _{total} at EOL [%]	28,3	28,6	30,0	32,7	36,7
Minor actinides balance					
In % of metal burnt	8,3	9,2	10,2	11,1	12,0

Type of fuel	2nd gener	ration pluton	ium (42,2 %)
Mass of fuel loaded into the core [kg]	700	900	1100
Equilibrium cycle length	180	234	275
Average discharged BU	460,7	468,0	450,7
Plutonium balance			
[%]	- 56,1	- 58,2	- 57,6
[kg/TWhe]	- 107,4	- 110,3	- 113,5
Pu _f / Pu _{total} at EOL [%]	19,35	22,4	27,0
Minor actinides balance			
Americium [kg/TWhe]	+ 13,57	+ 14,88	+ 16,93
Curium [kg/TWhe]	+ 3,90	+ 5,29	+ 6,43
Total [kg/TWhe]	+ 17,47	+ 20,17	+ 23,36
In % of metal burnt	16,3	18,3	20,5

In fact, higher plutonium loading imply an increase of great absorbers like ²⁴⁰Pu 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 favors the neutron absorption in the fertile isotopes. It should be noted that if the plutonium balance reaches an optimum with respect to the plutonium loaded into the core, it is not the case with the minor actinide balance, which increases linearly with the mass of plutonium. One could have thought that **maximize the burn-up should minimize both discharged masses of Pu and minor actinides**. In fact, as shown in Table 1, the production of minor actinides raises continuously with the Puloading. Consequently, the optimum burn-up obtained from the critical calculations, which leads to an optimum of the plutonium consumption with respect to the fuel loading, can be explained as follow:

- Despite a smaller initial reactivity, the increasing of the Pu-loading leads to a neutron spectrum hardening that will enhance the Pu conversion and thus increase the cycle length (then the burn-up);
- At a certain level of Pu-loadings a too hard neutron spectrum (deteriorating the fission rate) and the important amount of minor actinides in the fuel will limit again the cycle length and thus the burn-up.

Therefore, for each isotopic Pu-composition, an optimum Pu-loading exists that maximizes the burn-up and then minimizes the Pu-discharge despite a constant MA-discharge mass increasing.

Finally, as far as the 1st generation plutonium, the temperature effect (Doppler and moderator) has also been evaluated on the fuel element geometry between 20 and 900 °C. As far as the moderator temperature coefficient is concerned, the calculated value is an average between 20 and 500 °C. Despite the strong decrease of the moderator temperature coefficient during fuel irradiation, the

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results have shown that the global core temperature effect is negative and therefore selfstabilizing, with a fuel management by 1/3rd where the average core burnup ranges roughly from 200 and 400 GWd/t between the beginning and the end of cycle. [3]

2.2. MINOR ACTINIDE TRANSMUTATION

Further studies have also been conducted concerning the incineration of minor actinides in

prismatic block HTRs [4]. Final conclusions on this application cannot be drawn yet at the present day. The purpose of this study was essentially to evaluate some physical aspects of the Deep-Burn concept initially presented by the US on the basis of a bloc type reactor loaded heterogeneously with plutonium and minor actinides in the fuel elements. The calculations aimed at evaluating, in a simplified configuration, the impact of some design options such as plutonium and minor actinides located in different coated fuel particles and fuel compact. By using simplified calculations, we have tried to find the optimum configuration for which the minor actinides consumption plutonium and is enhanced.

As to conclude in the <u>once-through cycle</u> case, the use of two types of coated fuel particles in the Deep-Burn

concept allows to reach a **consumption rate of roughly 60 % in the DRIVER fuel** (the DRIVER fuel is loaded with first generation plutonium) and a **consumption rate of 25 to 30 % in the TRANSMUTER fuel** (loaded with DRIVER fuel that has not been used in a previous irradiation and minor actinides coming from LWR irradiated fuel).

As far as the <u>multi-recycling option</u> is concerned, **different scenarios** have been analysed with regard to the targeted cycle length, the mass of recycled minor actinides and the amount of minor actinides coming from LWR. From the preliminary results obtained with these different assessed scenarios, **no major positive trends** have been identified for the plutonium and minor actinides multirecycling. After four fuel cycles, the curium mass balance is still positive.





3. OPTIMISATION OF BURNABLE POISON DESIGN

3.1. CONTEXT

During the operation of a nuclear reactor, the reactivity effect of fuel burnup must be compensated by some means of long-term reactivity control, especially when the reactor operates with batchwise fuel loads. An elegant way for such a control is the use of burnable poison in the fuel elements to balance the reactivity loss caused by fuel burnup and fission product poisoning by the reactivity gain due to the disappearance of the burnable poison.

Burnup calculations have been performed [5] on a standard **HTR fuel pebble** (fuel zone with radius of 2.5 cm surrounded with a 0.5 cm thick graphite layer) and burnable poison particles (BPPs) containing B_4C made of pure ¹⁰B or containing Gd_2O_3 made of natural Gd. Two types of fuel were considered: UO₂ fuel made of 8% enriched uranium and PuO₂ fuel made of plutonium from LWR spent fuel (1st generation Pu). The radius of the BPP and the number of particles per fuel pebble were varied to find the flattest reactivity-to-time curve.

3.2. RESULTS AND TRENDS

With Burnable Poison Particles (BPPs) mixed in the fuel of an HTR, it is possible to control the excess reactivity present at beginning of life. For 8% enriched UO_2 fuel, mixing 1070 spherical BPPs containing B_4C with radius of 75 μ m through the fuel zone of a standard HTR fuel pebble with outer radius of 3 cm, the reactivity swing is 2% at a k_{∞} of 1.1. This means the burnable poison occupies a volume 60,000 less than that of the fuel pebble (FVR=60,000).

Using Gd_2O_3 as a burnable poison gives an optimum radius of about 840 µm and an FVR of only 5,000. This latter number corresponds to 9 spherical BPPs per fuel pebble. The low number for the FVR reflects the fact that the natural Gd in the particle absorbs fewer neutrons despite the fact that the thermal cross sections of the ¹⁵⁵Gd and ¹⁵⁷Gd isotopes are much larger than that of the ¹⁰B. This is due to the relatively large microscopic absorption cross section of ¹⁰B in the epithermal range and the high atomic number density of the boron in B₄C. For the Gd₂O₃ particles, the resulting reactivity swing is 3%, which is very similar to that obtained with the B₄C particles. The bigger size of the Gd₂O₃ particles could be advantageous for the manufacturing process of the BPPs.

The B₄C particles used in UO₂ fuel (radius between 70 and 90 μ m) can also be used to reduce the reactivity swing in PuO₂ particles. The reactivity swing at a target k_∞ of 1.1 is about 4% for spherical BPPs with radius of 85 μ m and an FVR of 27,500 (corresponding to 1600 BPPs per fuel pebble). The uniform temperature coefficient is comparable to that of the UO₂ fuel (-7 to -8 pcm/K). From theory, the burnup behavior of cylindrical BPPs differs from that of spherical ones, if the particles are large enough to be considered as 'black'. The reactivity swing calculated for these burnable 'needles' is, however, larger than that of the spherical particles, although it is not clear this will be the case for all values of the target k_∞.

The application of 'hollow' or coated BPPs, which consist of a non-poisonous graphite kernel covered with a B_4C burnable poison layer, gives results similar to the spherical particles. If not for other reasons like the manufacturing process, these particles have no large benefits compared to the solid spherical burnable particles.

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A complete description of the codes, methods and modelling hypotheses as well as the detailed analyses and interpretations of the obtained results are available in the <u>Task</u> <u>Report HTR-N-03/05-D-3.3.3</u>.

3.3. REMARKS ON PURE PLUTONIUM CYCLE IN HTR

It should be stressed that compared to a similar fuel depletion without erbium (1st generation plutonium Pu1 in Figure 4), the fuel with poison presents an initial negative reactivity of around 9000 pcm which strongly decreases towards 600 GWd/t, corresponding to the loss of 90 % of ¹⁶⁷Er. The cycle lengths are however comparable. This important feature of the burnable poison equivalent to that observed with ¹⁰B allows adjusting the initial reactivity of the different fuel cycle without changing the cycle lengths.

Likewise, one should note that the presence of Pu²⁴⁰ in all the fuel analysed play an important role in the reactivity margin control as it is shown in Figure 5. It acts as a burnable poison. By increasing the loaded fuel mass, the neutron spectrum becomes harder and favours the neutron absorption in the fertile isotopes.



Figure 4: Impact of burnable poison on fuel element k_{infinity}

Consequently, different approaches are possible with regard to the complex definition of what would be a reference **pure plutonium cycle**:

- Adjust the Pu mass to fit the maximum initial reactivity. In that case the optimum of plutonium incineration in term of mass balances won't be reached.
- Adjust the Pu mass as a function of the optimum burn-up that might be achieved (or maximum tolerable MA discharged mass) and then tailor the initial excess of reactivity by the use of BP.





Figure 5: Impact of fuel loading on fuel element depletion

Only additional studies would allow to precise the actual performances of such *pure* Pu cycles. It is clear that the optimum load of fixed burnable poison has not been estimated in the present study. It will result, for a bloc type reactor, from complex compromises between fuel performances, targeted mass balances (Pu and MA) and cycle length and the peak power mastery in the core and in the fuel element.... The 3d core calculations taking into account different fuel reloading management schemes will allow in a near future to provide additional information and to confirm the promising HTR performances with respect to the plutonium fuel.



4. CONCLUSION

It should be stressed that precaution must be taken with regard to the preliminary results given in this report. Indeed, the indicated mass balances have not been estimated from 3D full core calculations and remain to be confirmed. Nevertheless, such a 3D core calculation is inferred that a core optimisation approach close to conceptual design studies is needed for a block-type reactor fully loaded with plutonium fuel. This has not been carried out in the present study.

Likewise, it is noticeable that further detailed core physic analyses will be required in the future in order to assess the dynamic features of such a reactor. Additional studies concerning also, the reactivity control aspects, the temperature coefficients, the decay heat associated with plutonium fuel, the appropriate fuel management and the associated power distributions related issues (especially important in the case of the plutonium use) should allow to precise that pure plutonium cycles will respect the current high level of safety of the HTGR.

Finally, fuel back end cycle studies in PWR are usually tackled on the basis of a fuel assembly model. These calculations performed in infinite medium and based on the fundamental mode hypothesis, allow reaching rapidly the fuel mass balance avoiding core calculations. In the present WP3 studies, information is provided with regard to the precautions that must be taken in transuranic cycle studies carried out on annular core configurations as the one encountered in the GT-MHR. It turns out that the transuranic mass balances in a GT-MHR cannot be estimated easily from fuel element calculations but rather need the use of a core modelling approach taking into account the presence of the graphite reflectors.

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