DESIGN OF B\textsubscript{4}C BURNABLE PARTICLES MIXED IN LEU FUEL FOR HTRS

V. Berthou, J.L. Kloosterman, H. Van Dam, T.H.J.J. Van der Hagen.
Delft University of Technology
Interfaculty Reactor Institute
Email: v.berthou@iri.tudelft.nl

Abstract

The purpose of this study is to design burnable particles in the fuel elements of High Temperature Reactors (HTR) in order to control reactivity as a function of burn up. We focus on heterogeneous poisoning in which burnable particles (particles containing only burnable poison) are mixed with fuel particles in a graphite matrix. There are many degrees of freedom in the design of the burnable particle and the emphasis is put here on the optimization of its geometry (spherical or cylindrical), its size (different radius), and its composition (B\textsubscript{4}C with either natural boron or 100\% enriched in \textsuperscript{10}B). As a result, we have designed burnable particles that reduce considerably the reactivity swing during irradiation (up to 2.5\%).

Introduction

During the operation of a nuclear reactor, the reactivity effect of fuel burnup must be compensated by some means of long-term reactivity control. One way for such a control is the use of burnable poison in the fuel elements. In this way, it is possible to balance the reactivity loss due to fuel burnup and fission product poisoning by the reactivity gain due to the disappearance of the burnable poison.

With homogeneous poisoning it seems impossible to obtain a flat reactivity curve as a function of burnup [1]. More promising is heterogeneous poisoning, in which burnable particles (particles containing only burnable poison) are being mixed with the fuel particles in the graphite matrix. Because of the many degrees of freedom, correct dimensioning of the burnable particles and the ratio of burnable particles and fuel particles is not straightforward. In this study, the emphasis is put on optimization of the geometry, size and composition of the burnable particle as well as on the optimal ratio of burnable particles and fuel particles, in order to minimize the reactivity swing as a function of irradiation time.

The first part of this paper presents the description of the fuel and the burnable particles as well as the computational model. The second part shows the calculations for two different geometries: spherical and cylindrical.

Computational Model and Reactor Type

Computational Model
The reactor physics codes used are from the SCALE system [2]. The BONAMI code, applying the Bondarenko method, is used for the resonance treatment in the unresolved energy region, and the NITAWL code, applying the Nordheim integral method, is used for the resonance treatment in the resolved energy region. XSDRNPM, a 1-D discrete-ordinates transport code, is used for the cell-weighting calculations, while the actual burnup calculations are performed by the ORIGEN-S fuel depletion code. All data used are based on the JEF-2.2 nuclear data library [3].

In all calculations, a macro cell is modeled containing one burnable particle surrounded with several (typical two or three) fuel layers. The total thickness of all the fuel layers determines the effective number of fuel particles per burnable particle. In the macro-cell calculation, the burnable particle is subdivided in burnable layers with each layer having its own characteristic nuclide composition.

Because the SCALE system cannot treat the double heterogeneity of the fuel explicitly, the cell-weighting procedure is split up into two parts. First, the homogenized (resonance-shielded and cell-weighted) cross sections of each fuel layer are calculated in a micro-cell calculation. The micro unit cell consists of a three-zone coated fuel particle with a nuclide composition (particle and graphite) characteristic for the actual burnup time step. Secondly, if necessary, the resonance shielding calculations for each layer of the burnable particle are performed. Finally, a macro-cell calculation is done to calculate the neutron flux density and the neutron spectrum in each burnable layer, and the fission power density in each fuel layer. In these calculations, the average core power density is 3 MW.m$^{-3}$, and the temperature is 900°C.

Besides the resonance-shielded cell-weighted cross sections to be used in the macro-cell calculation, also the resonance-shielded zone-averaged nuclide cross sections are calculated for each fuel layer, and passed to the burnup data library. Subsequently, burnup calculations are performed for each fuel layer and for each layer of the burnable particle using the nuclide cross sections updated at each burnup time step. For the burnable layers, the depletion calculation is done using the constant flux approximation, while for the fuel layers the constant power approximation has been applied. As mentioned before, both the neutron flux density and spectrum, and the fission power density are calculated in the macro-cell calculation.

The whole sequence of micro-cell calculations, macro-cell calculation, and burnup calculations for both the fuel layers and the burnable layers are repeated many times (typically 20) to calculate the composition of each layer and the k-infinite of the system as function of burnup. Furthermore, at each burnup time step, the temperature coefficient of reactivity is being calculated with the VAREX code [4].

**Reactor Type**

For the fuel considered, the ESKOM PMBR [5,6] has been chosen with an average core power density of 3 MW.m$^{-3}$. The fuel particles contain 8% enriched UO$_2$ with a uranium mass of 9 gram per pebble. One pebble contains more than 13,000 fuel particles, each with a radius of 0.046 cm. For the poison in the burnable particle, we have chosen B$_4$C with either natural boron or 100% enriched $^{10}$B.

The chosen poison has a high thermal absorption cross-section, and, in addition, the absorption-to-scattering ratio is more than 1000. This implies that the burnable particle can be assumed to be purely absorbing. If the radius of the particles is large in units of thermal neutron absorption mean free path, then we can consider the particles as “black” particles, which means that every neutron hitting its surface will be absorbed. The effective absorption cross section for a small “black” particle is related to its geometrical cross section, i.e. for a sphere or a cylinder (with a radius R), it is calculated as [1]:

$$
\sigma_{\text{eff}} = 4\pi R^2 \left(1 - \frac{2}{3} \frac{\sqrt{3}}{\pi} \frac{R}{\lambda} + \frac{1}{12} \frac{\sqrt{3}}{\pi} \frac{R^3}{\lambda^2} \right) 
$$
\[ A_{\text{sphere}} = \pi R^2 \quad A_{\text{cylinder}} = \frac{\pi R}{2} \text{ (per unit of length)} \]

The target is to reach a \( k_{\infty} \) of 1.05 constant during the whole irradiation time.

**Spherical Burnable Particles**

*Evolution of Reactivity as a function of the Burn Up*

The burnable particles considered here are similar to the fuel particles: spherical geometry with a radius of 0.046 cm. The burnable poison is B\(_4\)C with either natural boron or enriched boron in 100% of \(^{10}\)B.

**B\(_4\)C with Natural Boron**

The evolution of the reactivity during the irradiation of the fuel has been studied (figure 1) for different volume ratios between fuel and burnable particles. The case without any burnable particle in the fuel is taken as a reference.

We can notice that after 1300 EFPD (Equivalent Full Power Days), the different curves do not converge: burnable particles are not fully burnt yet. The drop of the reactivity at the beginning of the irradiation is due to the short lived fission products (Xe and Sm effect).

The case with a ratio of 10 300 between the fuel volume and the poison volume starts with a correct reactivity level of 1.05, but the reactivity swing is still too large during the irradiation: 7% up to 1300 EFPD.

The different studied cases correspond to an insertion of only 1 or 2 burnable particles in the pebble: few burnable particles have already an significant impact on the reactivity.

![Figure 1: Reactivity as a function of burn up for different values of the volume ratio between fuel and burnable particles, for spherical burnable particles (B\(_4\)C with natural boron)](image)

**B\(_4\)C with Enriched Boron (100% of \(^{10}\)B)**

The same calculations have been done with burnable particles made of B\(_4\)C with enriched boron (100% of \(^{10}\)B).

Because the neutron mean free path for this poison is smaller (0.0064 cm), the burnable particle has been divided into more zones for the calculations (14) in order to have two layers per mean free path).

Figure 2 shows the reactivity as a function of the burn up for a volume ratio of 22 600 between the fuel and the poison in order to compare the burnable particles made of natural and enriched boron.
The two curves do not start at the same reactivity level, as it would be for a completely black particle. At first sight, this contradicts a previous study [1] in which gadolinium was used as a poison. In that study, the burnable particle (Gd) had a neutron mean free path of $8 \times 10^{-3} \text{ cm}$, which means that the ratio between the burnable particle diameter and the neutron mean free path is about $\frac{D}{\lambda} = 120$. Consequently the particle is black.

In our study, the neutron mean free path in natural boron is $3.24 \times 10^{-2} \text{ cm}$, and in enriched boron, $6.4 \times 10^{-3} \text{ cm}$. That leads to a ratio $\frac{D}{\lambda} = 3$ and 14 respectively.

It is clear that a burnable particle with natural boron is not black, that is why we notice a gap between the two curves at the beginning of life (BOL).

This feature can also be observed on the spectrum of the burnable particle inner zone (figure 3). For a $\text{B}_4\text{C}$ poison with enriched boron, there is no thermal neutron in the inner zone of the burnable particle, but for a $\text{B}_4\text{C}$ poison with natural boron, there are still some thermal neutrons that can reach the inner zone of the burnable particle. This means that this particle cannot be considered as a black particle.

![Figure 2: Reactivity for spherical burnable particles as a function of burn up for a volume ratio between the fuel and the burnable poison of 22 600 (B$_4$C with natural and enriched boron)](image)

![Figure 3: Spectrum of the burnable particle inner zone at the Beginning Of Life (BOL) for B$_4$C with natural boron and for enriched boron)](image)

**Evolution of $^{10}\text{B}$ concentration as a function of the Burnable Particle Radius**
Figures 4 and 5 show the concentration of $^{10}$B as a function of the distance from the center of the burnable particle for each burn up step (given in EFPD), for respectively the case with natural boron and enriched boron. The volume ratio between the fuel and the poison is 10300.

In the case of B$_4$C with natural boron, we notice that the burning of the particle is quite homogeneous whatever the distance from the center of the burnable particle is, which confirms the fact that the burnable particle is not black for thermal neutrons.

In the case of B$_4$C with enriched boron, the external zone burns much faster than the internal zone, the thermal neutrons are all absorbed when they hit the surface of the burnable particle. We can also notice that the burnable particle is not fully burnt at the end of life (EOL) as previously pointed out.

![Figure 4: $^{10}$B concentration as a function of distance from the center of the burnable particle, with the burn up as a parameter, for a B$_4$C spherical burnable particle made of natural boron](image)

![Figure 5: $^{10}$B concentration as a function of distance from the center of the burnable particle, with the burn up as a parameter, for a B$_4$C spherical burnable particle made of enriched boron](image)

**Variation of the Burnable Particle Diameter**
We have seen that the effective macroscopic absorption cross section of a black particle is related to its geometrical cross section, which means that particles with different radii would have different behavior.

In spherical geometry, different radii of the burnable particle are considered with a constant volume ratio of the fuel and the burnable particles. This means that the number of burnable particles per pebble increases when the radius of each particle decreases. The smaller the burnable particle, the more homogeneous the poison is distributed, and the faster the particles will burn.

In figure 6, the reactivity is shown as a function of time for the reference case without poison and for two radii of the burnable particles, 0.046 cm and 0.03 cm. The initial reactivity is lower in the case of smaller particles because of more burnable particles present in the pebble that are distributed more homogeneously. In addition, the effective absorption cross sections are respectively \( A = \pi R^2 = 0.0066 \text{ cm}^2 \) for a radius of 0.046 cm and \( A = \pi R^2 = 0.0028 \text{ cm}^2 \) for a radius of 0.03 cm.

Consequently the reactivity swing is smaller for the burnable particle with a radius of 0.03 cm (2.5% compared to 6% with a radius of 0.046 cm).

\[ \text{Figure 6: } K_{\text{inf}} \text{ as a function of irradiation time for the reference case without poison and for spherical burnable particles with different radii} \]

Different volume ratios between the fuel and the poison have been studied, as well as other burnable particle radii (0.046, 0.03, 0.02 cm). The case of burnable particles with a small radius (0.02 cm) and a volume ratio of 10 300 has a reactivity swing of 3.6% up to 1600 EFPD. The burnable particles are almost fully burnt at 1600 EFPD, but the level of reactivity is too low (between 1 and 1.03).

**Uniform Temperature Coefficient (UTC)**

Since the kernel of each fuel particle has a high surface-to-volume ratio, an effective heat transfer from the fuel to the moderator is realized. This means that there is only a small temperature difference between the kernel and the surrounding graphite, namely 1K [7]. Since the temperature of the kernel and the graphite is almost equal, this allows us, in view of reactivity feedback effects, to consider a uniform temperature coefficient.

Figure 7 shows the UTC as a function of the irradiation time for the reference case without burnable particles and for two different values of volume ratio between fuel and poison (case of spherical burnable particles with a 0.046 cm radius, and containing B\(_4\)C with enriched boron).
The UTC is relatively constant during the irradiation, around –10 pcm/K. In presence of burnable particles, the UTC is more constant, and a little bit more negative due to a slight shift of the spectrum towards higher energies. At the end of the irradiation the spectra are the same, and then the UTC are equal.

**Figure 7: Uniform-Temperature Coefficient (UTC) for the reference case and for two different volume ratio between the fuel and the spherical BP (Containing B₄C with enriched boron)**

**Cylindrical Burnable Particles**

*Evolution of Reactivity as a function of the Burn Up*

The burnable particles considered here have a cylindrical geometry with a diameter of 0.092 cm. The burnable poison is B₄C with either natural boron or boron 100% enriched in ¹⁰B.

**B₄C with Natural Boron**

The evolution of the reactivity during the irradiation of the fuel has been studied (figure 8) for different values of the volume ratio between fuel and burnable particles. The case without any poison in the fuel is taken as a reference.

We can notice that, after already 1000 EFPD, the different curves converge: the burnable particles are fully burnt.

The case with a volume ratio of 22 600 between the fuel and the poison has a lower reactivity swing: 6% up to 1000 EFPD.
Figure 8: Reactivity as a function of the burn up for different values of the volume ratio between the fuel and the burnable poison and for the reference case, for cylindrical burnable particles (B$_4$C with natural boron)

If we consider now two different shapes of burnable particles (spherical and cylindrical) but with the same effective absorption cross section (that means a smaller radius for the cylindrical particle) and the same ratio of fuel volume and poison volume, the reactivity swing is smallest for the cylindrical particle (much more cylindrical burnable particles homogeneously distributed in a pebble).

$B_4C$ with Enriched Boron (100% of $^{10}$B)

The burnable particle containing B$_4$C with natural boron is fully burnt after 1000 EFPD. With enriched boron, a flatter curve can be obtained, with a reactivity swing of 2.2% (for a volume ratio between fuel and poison of 10 300) up to 1600 EFPD. Then the burnable particle is fully burnt at 1600 EFPD (figure 9). The only drawback is the level of reactivity, around 1.03, which is a little bit too low.

For the same burnable particle radius, and for a black particle, the effective absorption cross section is larger for a cylindrical shape than for a spherical one ($A_{\text{sphere}} = \pi R^2 = 0.0066$ cm$^2$ and $A_{\text{cylinder}} = \frac{\pi}{2} R = 0.072$ cm$^2$). As a result, the cylindrical particle is fully burnt at the end of the irradiation while the spherical particle still contains significant amounts of $^{10}$B after 1600 EFPD.
Figure 9: Reactivity as a function of burn up for different values of the volume ratio between the fuel and the burnable poison and for the reference case, for cylindrical burnable particles (B$_4$C with enriched boron)

**Evolution of $^{10}$B concentration as a function of the Burnable Particle Radius**

Figure 10 shows the concentration of $^{10}$B as a function of the distance from the center of the burnable particle for each burn up step (given in EFPD), for the case of enriched boron. The volume ratio between the fuel and the poison is 10 300. Also here we can notice that the cylindrical burnable particle is fully burnt faster than the spherical one: after 1000 EFPD the $^{10}$B concentration has decreased both in the outer and inner zone of the burnable particle.

![Figure 10: $^{10}$B concentration as a function of the distance from the center of the burnable particle, with the burn up as a parameter, for a B$_4$C cylindrical burnable particle with enriched boron](image)

**Conclusion**

The purpose of this study is the use of burnable particles in the fuel elements of HTR in order to control the reactivity effect of the fuel burn up. Heterogeneous poisoning in which burnable particles (particles containing only burnable poison) are mixed with the fuel particles in
a graphite matrix is considered. In order to obtain the flattest reactivity-to-time curve numerous parametrical studies have been carried out to tailor the burnable particle.

The general trends are the following:
- The use of B\textsubscript{4}C with enriched boron (100\% of \textsuperscript{10}B) is required in order to have a “black” particle.
- The smaller the radius for the burnable particle, the more homogeneous the poison is distributed, and the faster the particle will be burnt.
- For a spherical shape, a small reactivity swing is obtained (2.5\%) with a burnable particle radius of 0.03 cm.
- The cylindrical shape leads to the lowest reactivity swing as a function of irradiation time: 2.2\% up to 1600 EFPD for burnable particles containing B\textsubscript{4}C 100\% enriched in \textsuperscript{10}B with a radius of 0.046 cm, and for a volume ratio between fuel and poison of 10 300).

Heterogeneous poisoning of HTR fuel seems quite promising. We have designed burnable particles containing B\textsubscript{4}C that reduce considerably the reactivity swing. Further studies will focus on burnable particles that contain graphite or fuel surrounded with a thin layer of poison, on different poison materials (e.g. Er or Gd), and on different fuel compositions. The ultimate aim is to abandon active long-term reactivity control in HTRs.

The authors like to acknowledge the European Commission for co-funding this research that is performed in the project “High Temperature Reactor Physics and Fuel Cycle Studies (HTR-N)” [8] together with other European Organizations.

References