

**APPLICATION OF BORON AND GADOLINIUM BURNABLE POISON
PARTICLES IN UO₂ AND PUO₂ FUELS IN HTRs**

J.L. KLOOSTERMAN

Delft University of Technology

Interfaculty Reactor Institute

Mekelweg 15, NL 2629 JB Delft

The Netherlands

E-mail: J.L.Kloosterman@iri.tudelft.nl

Tel: ++31-15-27-81191

Fax: ++31-15-27-89534

ABSTRACT

Burnup calculations have been performed 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 ^{10}B or containing Gd_2O_3 made of natural Gd. Two types of fuel were considered: UO_2 fuel made of 8% enriched uranium and PuO_2 fuel made of plutonium from LWR spent fuel. The radius of the BPP and the number of particles per fuel pebble were varied to find the flattest reactivity-to-time curve.

For the UO_2 fuel, the reactivity swing is lowest (around 2%) for BPPs made of B_4C with radius of 75 μm . In this case around 1070 BPPs per fuel pebble are needed. For the PuO_2 fuel to get a reactivity swing below 4%, the optimal radius of the BPP is the same, but the number of particles per fuel pebble should be around 1600. The optimal radius of the Gd_2O_3 particles in the UO_2 fuel is about ten times that of the B_4C particles. The reactivity swing is around 3% when each fuel pebble contains only 9 BPPs with radius of 840 μm . The results of the Gd particles illustrate nicely the usage of black burnable poison particles introduced by Van Dam (2000).

1. INTRODUCTION

During the operation of a nuclear reactor, especially when it operates with batch-wise fuel loads, the reactivity loss due to fuel burnup and fission product poisoning must be compensated by some means of long-term reactivity control. An elegant way to accomplish this is to use burnable poison particles (BPP) originally introduced by Van Dam (2000). This is especially of interest to the gas-cooled High Temperature Reactor (HTR), because this reactor has some attractive properties not offered by other reactors. The application of TRISO coated fuel particles makes it possible to reach very high burnup values and to ensure the containment of fission products up to a fuel temperature of 1600 °C (Kugeler, 1989). Together with the use of BPPs to reduce or eliminate the role of the active reactivity control mechanisms, this paves the way for long-term unattended reactor operation for HTRs with batch-wise fuel loads, like a common block-type HTR or a cartridge-fueled pebble-bed HTR. This could make HTR technology applicable to areas like ship propulsion and (co) generation of heat and electricity in remote areas.

By lumping the burnable poison into spherical particles, one has two design parameters to achieve a flat reactivity curve (size and concentration of the BPP) instead of only one for the case of homogeneous poisoning (concentration of the burnable poison). For a small BPP that absorbs all impinging neutrons, a so-called black particle, the effective absorption cross section is equal to its geometrical cross section independent of the composition of the particle. This means that the neutron absorption rate depends on the size of the particle, but not on its actual composition. Then, the effective absorption cross section for a spherical particle is a quadratic function of time (Van Dam, 2000):

$$\Sigma_a(t) = N_p \pi R_0^2 \left(1 - \frac{\Phi_0 t}{4NR_0} \right)^2. \quad (1)$$

Here, N_p is the number density of the burnable poison particles, R_0 the initial radius of the particle, and N the atomic density of the absorbing nuclide in the particle. For more details, see the paper of Van Dam (2000) or Kloosterman et al. (2003). The BPP themselves contain either B_4C made of pure ^{10}B or Gd_2O_3 made of natural Gd (see Table I). From a neutronics point of view, the main difference between these two materials is the mean free path for thermal neutrons, which is about four times larger for the B_4C particle. As a fuel we used either UO_2 or PuO_2 particles embedded in a graphite pebble with a diameter of 5 cm surrounded by a graphite layer with thickness of 0.5 cm. More details about the fuel composition are given in Table II.

The aim of this paper is to find the radius and the concentration of the BPPs that minimize the reactivity swing at a specific value for k_∞ . To this end, we have modeled one BPP surrounded by a smeared fuel layer for which we calculated the k_∞ as a function of time with the BPP radius and the outer radius of the smeared fuel layer as parameters. This paper is an extension to a previous one (Kloosterman et al., 2003) that considered spherical, cylindrical and coated burnable poison particles made of B_4C in UO_2 fuel.

2. CALCULATION SCHEME

From the SCALE code system (SCALE-4.2, 1994), we used the BONAMI-S code for the resonance treatment in the unresolved energy region, the NITAWL-II code for the resonance treatment in the resolved energy region, and the XSDRNPM-S code for the cell-weighting calculations. The actual burnup calculations were done using the ORIGEN-S fuel depletion code. All codes were coupled to each other via PERL scripts and via own-made FORTRAN programs.

All nuclear data used, both for the burnup calculations as well as for the neutron spectrum calculations, are based on the JEF-2.2 nuclear data library (Hoogenboom and Kloosterman, 1997, and JEF-2.2, 2000).

In all calculations, a macro cell was modeled containing one BPP (made of either B_4C or Gd_2O_3) surrounded by smeared fuel. The latter contains all nuclides present in a standard fuel pebble, i.e. the nuclides from the TRISO coated fuel particles (containing 25 fission products and actinide nuclides), the carbon from the graphite matrix in between the fuel particles, and the carbon from the 0.5 cm thick outer layer.

Two sets of calculations were done: global parameter studies using a macro cell containing one burnable particle zone surrounded by one smeared fuel layer. The results of these studies are the k_∞ as a function of time. A more accurate set of calculations was done with the BPP divided into ten concentric zones (see Fig. 1). The k_∞ values of these two sets of calculations are almost the same, but because of the larger number of zones, the latter set of calculations gives a better insight into the spatial distribution of the burnable poison as a function of time. Furthermore, in that case the fuel temperature coefficient of reactivity and other parameters were calculated at various time steps during the irradiation.

Because the SCALE code system cannot handle explicitly the double heterogeneity of the fuel, the cell-weighting procedure had to be split into two parts (see Fig. 1). First, the homogenized resonance-shielded and cell-weighted cross sections of the fuel layer were calculated in a micro-cell calculation with the unit cell made up of a TRISO coated fuel particle with a nuclide composition characteristic for the actual burnup time step surrounded by an equivalent amount of graphite. The Dancoff factor used in the resonance shielding was calculated by an analytical procedure (Bende, 1999) and takes into account the double heterogeneity of the fuel. Secondly, a

macro-cell calculation was done to calculate the neutron flux density in the BPP, and to calculate the fission power density in the smeared fuel layer.

Besides the resonance-shielded cell-weighted cross sections to be used in the macro-cell calculation, also the resonance-shielded *zone*-averaged cross sections were calculated, and passed to the burnup data library. Burnup calculations were done for each fuel layer and for each layer of the BPP using the nuclide cross sections updated at each time step. The whole sequence of micro-cell calculations, macro-cell calculations, and burnup calculations for the fuel layer and the burnable particle layers were repeated ten times to calculate the k_{∞} and the composition of each layer as a function of burnup. Furthermore, at each burnup time step, the temperature reactivity coefficient was calculated.

3. RESULTS

3.1 UO₂ fuel with B₄C

This section presents the results for the BPP made of B₄C. For one choice of the BPP and the fuel, there remain only two parameters to be varied: the radius of the BPP and the number of BPPs per fuel pebble. The latter number is presented in a more abstract way by the fuel volume ratio (FVR), i.e. the ratio of the volume of one fuel pebble with radius of 3 cm and the volume of all BPPs in that pebble.

There are two output parameters of major interest: the *initial* k_{∞} and the *reactivity swing* $\Delta\rho$. Because the definition of these parameters is not unambiguous, we will clarify their definition. The *initial* k_{∞} is the value of the k_{∞} curve directly after saturation of all short-lived fission products like Xe and Sm. In practice, the value of the k_{∞} after 10 days of burnup is used for this.

The *reactivity swing* $\Delta\rho$ is defined as the maximum difference of the k_∞ between 10 days of burnup and the burnup value for which the k_∞ of the reference curve (with no BPP) crosses the *initial* k_∞ value. This makes sense as one would like to achieve a reactivity curve as flat as possible. Figure 2 illustrates the parameters defined above.

Figure 3 shows the initial k_∞ and the reactivity swing $\Delta\rho$ of the fuel for radii of the BPP varying from 10 to 200 μm and for FVR varying from 10,000 to 200,000. The initial k_∞ increases when either the FVR or the BPP radius increases. The latter can be understood from the theory: for a fixed volume of the poison (constant FVR), the larger the radius of the BPP, the more heterogeneously distributed the poison, and the smaller the effective absorption cross-section. On the other hand, the reactivity swing shows a 'valley' ranging from the upper-left corner to the lower-right corner. The optimization procedure would be to first find the combinations of BPP radius and FVR that fulfill the requirement on the initial k_∞ (this could be, for example, that in order to have a critical reactor, the initial k_∞ must be equal to 1.1). Secondly, from these results found, one should extract those values for the BPP radius and the FVR that give the lowest reactivity swing. The third plot in Fig. 3 shows the absolute deviation of the initial k_∞ and its target value (arbitrarily chosen to be 1.1), while the fourth plot shows this parameter summed to the reactivity swing. Two regions are most favorable: a BPP radius of 75 μm with a FVR of 60,000 and a BPP radius of 90 μm with a FVR of 50,000. The reactivity swing in these two regions is below 2% while the initial k_∞ deviates from 1.1 no more than 1%. However, most probably a broad range of BPP radii (say between 70 and 90 μm) can be applied, which relaxes the requirements in the manufacturing process of the particles. Actually, the k_∞ for a BPP radius 70 μm and FVR=50,000 is shown in Fig. 2 as a function of time. More results for this case

among which the fuel temperature coefficient as a function of burnup are given in Kloosterman et al. (2003).

3.2 UO₂ fuel with Gd₂O₃

Figure 4 shows the results for the BPP containing Gd₂O₃. Surprisingly, for a specific radius of the BPP, the FVR is smaller for the Gd₂O₃ case, which means that this kind of particles is less absorbing than the B₄C ones. For example, for a BPP with radius of 200 μm to obtain $k_{\infty}=1.1$ we must chose FVR=30,000 for the B₄C particle, but only FVR=17,200 for the Gd₂O₃ particle. Figure 5 shows the reason for this unexpected behavior. The macroscopic absorption cross section of Gd is larger than that of ¹⁰B only for energies below 0.2 eV. Above this energy the reverse is true, which is also due to the fact that the atomic density of boron in B₄C is about 15 times (!) larger than the atomic densities of the two absorbing Gd isotopes in Gd₂O₃. Because the configurations chosen give the same k_{∞} , the fraction of neutrons absorbed in the BPP is also the same (about 0.16). However, for the Gd₂O₃ particle these are only thermal neutrons with energy below 0.2 eV, while for the B₄C particle about one third of the absorption rate is due to neutrons with energy above 0.2 eV. In conclusion: although the Gd₂O₃ particle is more absorbing (less grey) for thermal neutrons, the total spectrum-averaged neutron absorption rate is larger in the B₄C particle.

From Fig. 4 it seems that with the use of a Gd₂O₃ particle, it is not possible to reach a reactivity swing less than 5% at a target k_{∞} of 1.1. Fortunately, more accurate calculations with more zones in the BPP and more neutron spectrum calculations during the lifetime of the fuel show slightly better results. Figure 6 shows the k_{∞} for a BPP with a radius of 840 μm and a FVR of 5,000. The reactivity swing is about 3%, which is comparable to the results obtained with the B₄C particles

(see Section 3.1). The most notable difference with the B_4C particles is the fact that the optimum radius of the Gd_2O_3 particles is about 10 times as large, which could be an advantage for the manufacturing process of the BPPs.

3.3 PuO_2 fuel with B_4C

Finally, the B_4C burnable poison was also applied in combination with PuO_2 fuel with composition given in Table II. The burnup behaviour of the PuO_2 fuel differs considerably from that of the UO_2 fuel. The k_∞ curve of the latter fuel decreases almost linearly in time to a value of about unity (see Figs. 2 and 6), while that of the PuO_2 fuel decreases gradually in time till $k_\infty \approx 1.25$, after which it suddenly drops (see Fig. 7). This effect is due to the consumption of all the actinides in the particle.

The results of these studies can be seen in Fig. 8, which shows the k_∞ , the reactivity swing $\Delta\rho$, the deviation of the k_∞ from its target value (1.1) and the sum of this parameter and the reactivity swing ($\Delta k + \Delta\rho$). The optimum size of the BPP is around 90 μm , which is very similar to the results obtained for the UO_2 fuel (see Section 3.1 and Fig. 3), although the FVR is lower than that of the UO_2 fuel (25,000 versus 60,000 for UO_2). This last difference is not that easy to understand. Before saturation of the short-lived fission products, the k_∞ values for the UO_2 and PuO_2 fuels with no BPP are 1.44 and 1.38, respectively (after saturation these numbers are 1.39 and 1.34). This means that for the UO_2 fuel to reach $k_\infty = 1.1$, a larger fraction of neutrons needs to be absorbed in the BPP, from which at first instance one would expect that the FVR for the UO_2 fuel would be smaller. But the reverse is true! Figure 9 shows the neutron spectra in the fuel zones for both fuel types, together with the cumulative neutron absorption rate in the BPP. Two interesting points can be observed. First, because of the harder neutron spectrum in the

PuO₂ fuel, neutron absorption in the BPP takes place at higher energies where the absorption cross section of the ¹⁰B is lower. Thus more ¹⁰B or a lower FVR is needed to achieve a sufficiently large reduction of the k_∞. Secondly, for both cases the neutron fraction absorbed in the BPP is the same (about 0.16) while the k_∞ with no BPP is about 5% larger for the UO₂ fuel. Apparently, the BPP in the PuO₂ fuel absorbs many neutrons that would otherwise be captured at lower energies by the plutonium isotopes. For this effect, the strong capture resonance of ²⁴¹Pu at 1 eV could be responsible, which gives a large value for the capture-to-fission ratio of reactor grade plutonium around 1 eV (see Fig. 10).

Figure 7 shows the k_∞ as a function of irradiation time for two values of the BPP radius and the FVR in the region of the minimum in Fig. 8. Clearly the reactivity swing is considerably reduced to values of about 4%. Because of the different burnup behavior of the plutonium in the particle, the reactivity swing in the PuO₂ fuel is slightly larger than for the UO₂ fuel.

Up to 700 EFPD, the uniform temperature coefficient of the standard PuO₂ fuel is between -7 and -8 pcm/K, while this is between -5 and -6 pcm/K for the case of a BPP with radius of 90 μm and a FVR of 25,000. Despite the fact that the fuel contains no uranium, these values are negative and sufficiently large for the safe operation of an HTR. For the UO₂ fuel considered in Section 3.1, the uniform temperature coefficient has a similar value of about -8 pcm/K (Kloosterman et al., 2003).

4. CONCLUSIONS

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 BPPs containing B_4C with radius of $75\text{ }\mu\text{m}$ 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 ($\text{FVR}=60,000$).

Using Gd_2O_3 as a burnable poison gives an optimum radius of about $840\text{ }\mu\text{m}$ and an FVR of only 5,000. This latter number corresponds to 9 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 ^{155}Gd and ^{157}Gd isotopes are much larger than that of the ^{10}B . This is due to the relatively large microscopic absorption cross section of ^{10}B in the epithermal range and the high atomic number density of the boron in B_4C . For the Gd_2O_3 particles, the resulting reactivity swing is 3%, which is very similar to that obtained with the B_4C particles. The bigger size of the Gd_2O_3 particles could be advantageous for the manufacturing process of the BPPs.

The B_4C particles used in UO_2 fuel (radius between 70 and $90\text{ }\mu\text{m}$) can also be used to reduce the reactivity swing in PuO_2 particles. The reactivity swing at a target k_∞ of 1.1 is about 4% for BPPs with radius of $85\text{ }\mu\text{m}$ and an FVR of 27,500 (corresponding to 1600 BPPs per fuel pebble). The uniform temperature coefficient is comparable to that of the UO_2 fuel (-7 to -8 pcm/K).

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REFERENCES

Bende E.E., Hogenbirk A.H., Kloosterman J.L., Van Dam H., 1999, Analytical Calculation of the Average Dancoff Factor for a Fuel Kernel in a Pebble-Type High-Temperature Reactor, *Nuclear Science and Engineering*, **133**, 1-16.

Hoogenboom J.E., Kloosterman J.L., 1997, Generation and Validation of ORIGEN-S Libraries for Depletion and Transmutation Calculations Based on JEF2.2 and EAF3 Basic Data, *Nuclear Engineering&Design*, **170**,107-118.

JEF-2.2, 2000, The JEF-2.2 nuclear data library, report JEFF17, Nuclear Energy Agency, Paris.

Kloosterman J.L., Van Dam H., Hagen T.H.J.J. van der, 2003, Applying Burnable Poison Particles to Reduce the Reactivity Swing in High Temperature Reactors, *Nuclear Engineering&Design*, accepted for publication.

Kugeler K., Schulten R., 1989, *Hochtemperaturreaktortechnik*, Springer-Verlag.

Ruetten H.J., Kuijper J.C., 2003, Plutonium (1st and 2nd generation) cell burnup benchmark specification, report HTR-N1-02/06-S-3.1.1, benchmark defined within the framework of the HTR-N1 project (see Acknowledgements).

SCALE-4.2, 1994, Modular code system for performing standardized computer analyses for licensing evaluations, Oak Ridge National Laboratory, Tennessee.

Van Dam H., 2000, Long-term Control of Excess Reactivity by Burnable Particles, *Annals of Nuclear Energy*, **27**, 733-743.

Table I:
Definition of the burnable poison particles used.

Parameter	B ₄ C	Gd ₂ O ₃
Density (g.cm ⁻³)	2.52	7.4
at dens ¹⁰ B (cm ⁻¹ barn ⁻¹)	0.11	0
at dens ¹⁵⁵ Gd (cm ⁻¹ barn ⁻¹)	0	3.64·10 ⁻³
at dens ¹⁵⁷ Gd (cm ⁻¹ barn ⁻¹)	0	3.85·10 ⁻³
Thermal mean free path (μm)	23.6	8.3

Table II:

Definition of the fuel used. The UO₂ fuel is very similar to that foreseen in the PBMR, the PuO₂ fuel has been defined within the framework of the HTR-N1 project (Ruetten and Kuijper, 2003), and can be assumed to be extracted from LWR spent UO₂ fuel.

Parameter	UO ₂	PuO ₂
²³⁵ U (w%)	8	0
²³⁸ U (w%)	92	0
²³⁸ Pu (w%)	0	2.59
²³⁹ Pu (w%)	0	53.85
²⁴⁰ Pu (w%)	0	23.66
²⁴¹ Pu (w%)	0	13.13
²⁴² Pu (w%)	0	6.78
Fuel kernel diameter (μm)	500	240
Actinide mass (g/pebble)	9	1
Power rating (W/pebble)	340	1000
Temperature (K)	900	1073

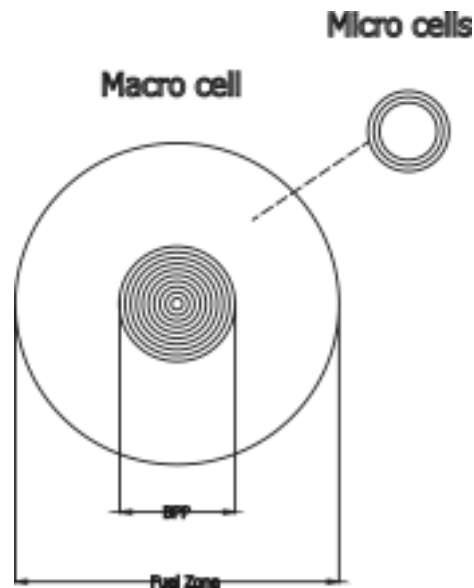


Figure 1: Schematic representation of the calculation model for the accurate set of calculations with the BPP divided into ten concentric zones surrounded with a smeared fuel layer. For the fuel layer, micro cell calculations on a TRISO coated particle with graphite are done to determine the zone-weighted and cell-weighted cross sections for each fuel layer. The cell-weighted cross sections are subsequently used in the macro cell calculations to determine the power and flux spatial distribution, while the zone-weighted cross sections of the fuel are used to update the burnup data library. This process is repeated each time step with updated atomic nuclide densities.

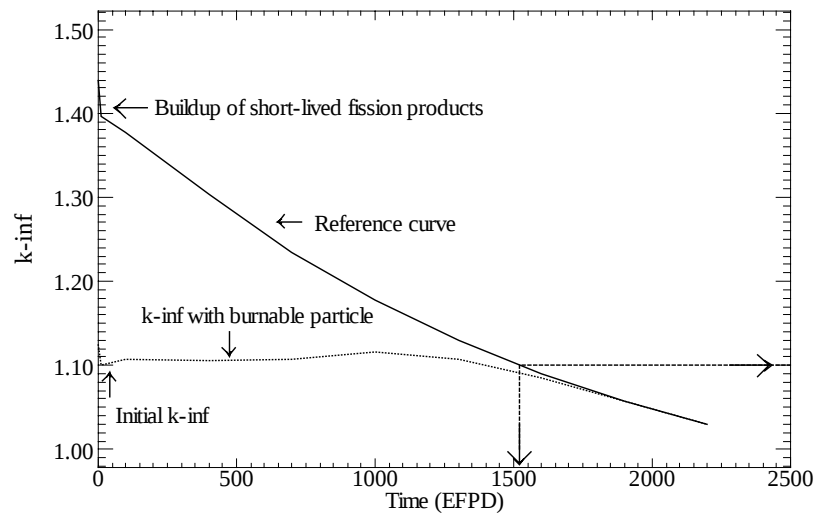


Figure 2. Illustration of the parameters used to characterize the fuel with BPPs. The initial k_{∞} is the value after saturation of short-lived fission products (in this paper after 10 days of burnup). The maximum irradiation time is achieved when the reference k_{∞} curve (with no BPP) reaches the value of the initial k_{∞} . In this plot this occurs after 1520 days of burnup. The reactivity swing is the spread of the k_{∞} between 10 and 1520 Effective Full Power Days (EFPD). Here, 1520 EFPD correspond to a burnup of 57,422 MWd/tU.

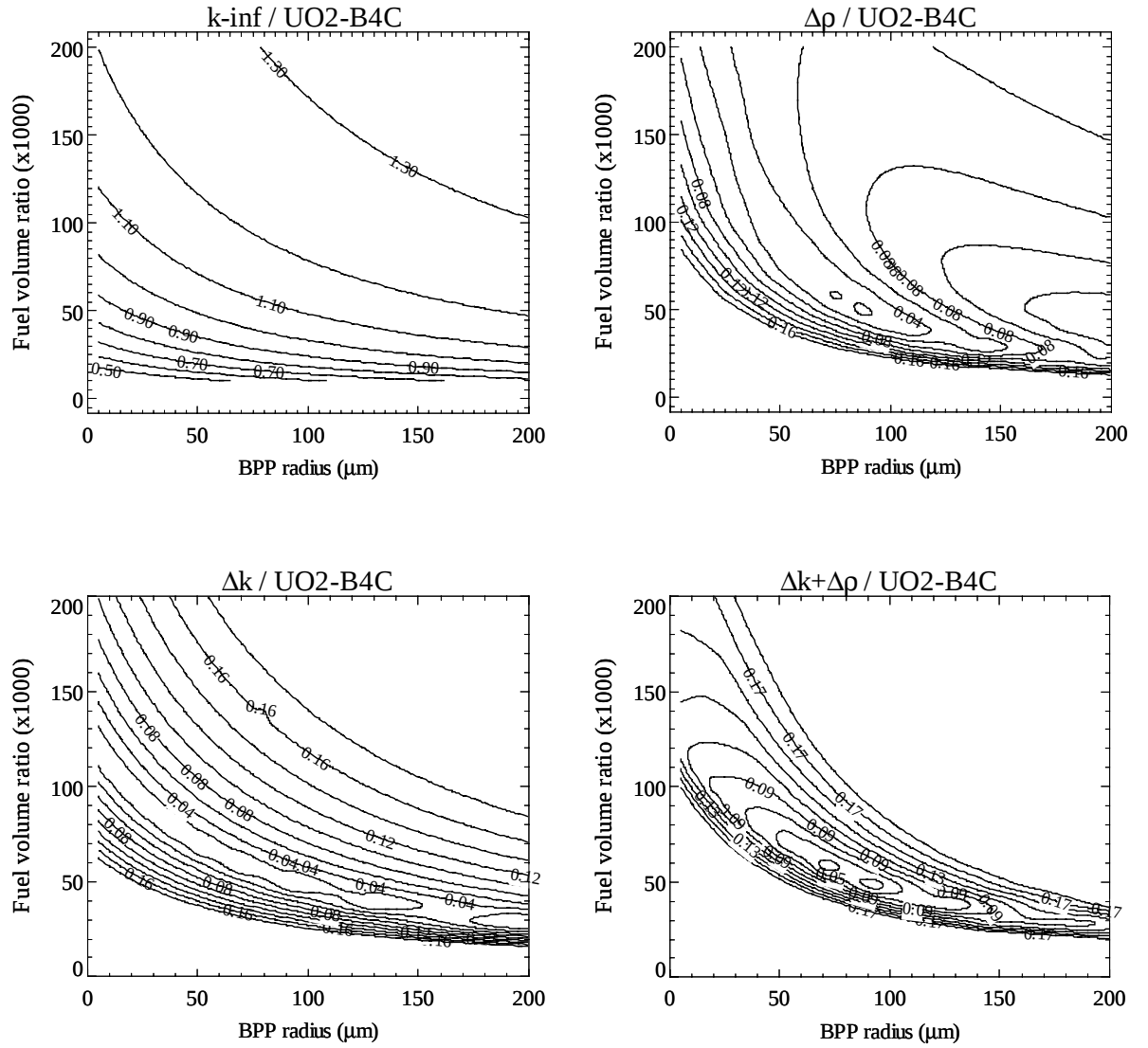


Figure 3: The k_{∞} (top left) and the reactivity swing $\Delta\rho$ (top right) as a function of the BPP radius and the fuel volume fraction. The bottom left plot shows the deviation of the initial k_{∞} from the target value (1.1), while the bottom right plot shows the sum of this and the reactivity swing (bottom right). The minimum in the latter plot is observed at a BPP radius of about 75 μm and a fuel volume ratio of 60,000 at which the reactivity swing is about 2%.

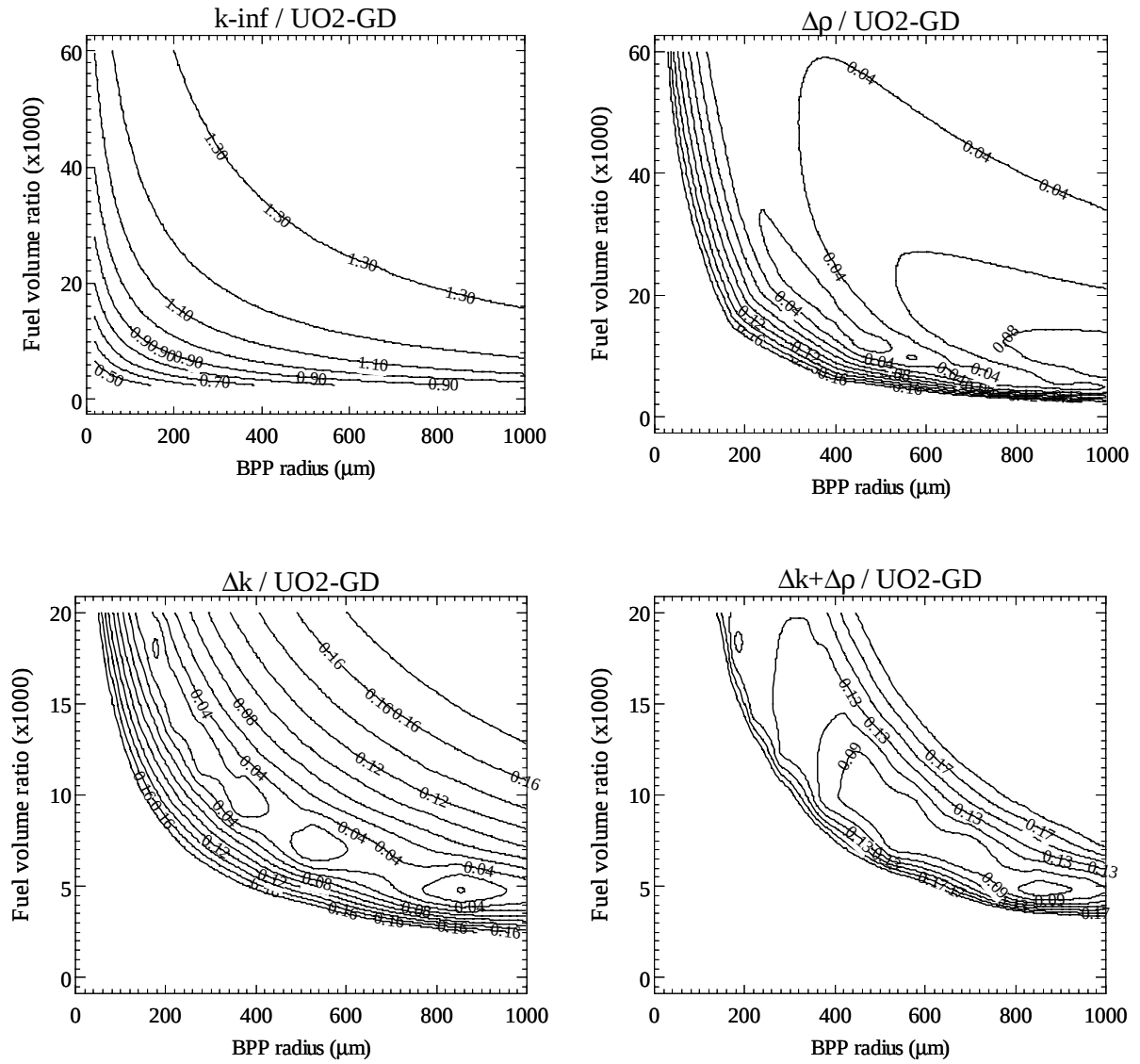


Figure 4: The k_{∞} (top left) and the reactivity swing $\Delta\rho$ (top right) as a function of the BPP radius and the fuel volume fraction. The bottom left plot shows the deviation of the initial k_{∞} from the target value 1.1, while the bottom right plot shows the sum of this and the reactivity swing (bottom right). The minimum in the latter plot is observed at a BPP radius between 840 and 880 μm and a fuel volume ratio of 5,000 at which the reactivity swing is almost 6%. Note the different scales along the y axes.

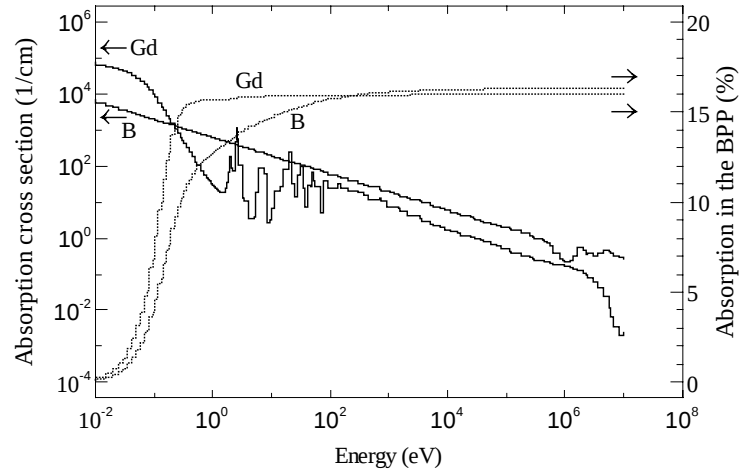


Figure 5: Macroscopic absorption cross section for the two burnable poison materials. Below 0.2 eV the absorption cross section of the Gd is largest, which also shows up in the cumulative absorption rate per fission neutron (right axis). The latter two curves are for a BPP with radius of 200 μm and a FVR of 17,200 for the Gd_2O_3 particle and 30,000 for the B_4C . The k_∞ in both cases is the same, which can be derived from the fact that the cumulative absorption in the BPP is the same (0.16 per fission neutron).

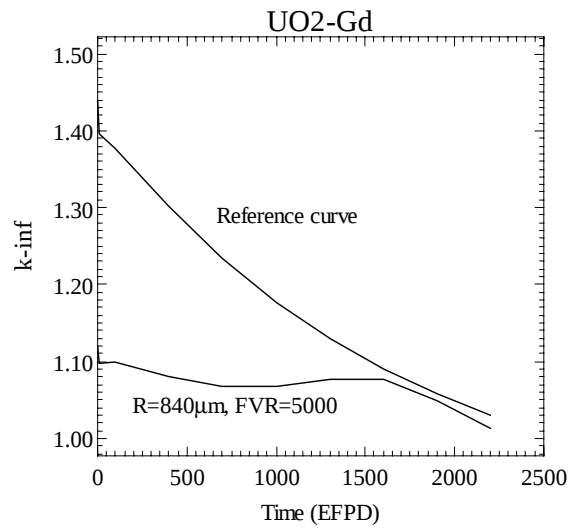


Figure 6: The k_{∞} as a function of irradiation time for the UO_2 fuel with a BPP made of Gd_2O_3 . For a BPP with radius of $840\text{ }\mu\text{m}$ and an FVR of 5000 the reactivity swing is about 3% between 10 and 1520 EFPD. Here, 1520 EFPD correspond to a burnup of 57,422 MWD/tU.

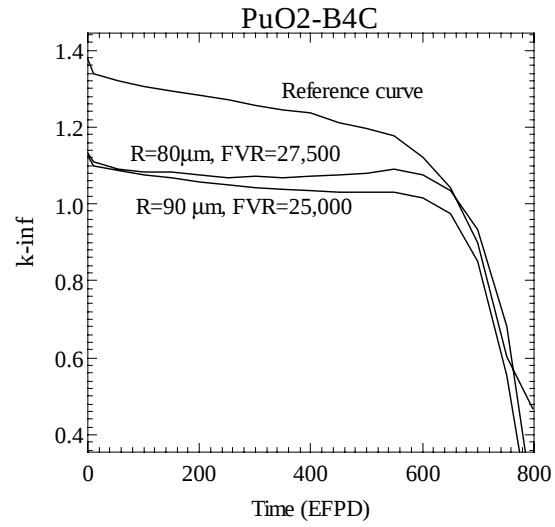


Figure 7: The k_{∞} as a function of irradiation time for the PuO₂ fuel with a BPP made of B₄C. For a BPP with radius of 80 μm and an FVR of 27,5000 the reactivity swing is just below 4% between 10 and 620 EFPD. Here, 620 EFPD correspond to a burnup of 620 MWD/tPu.

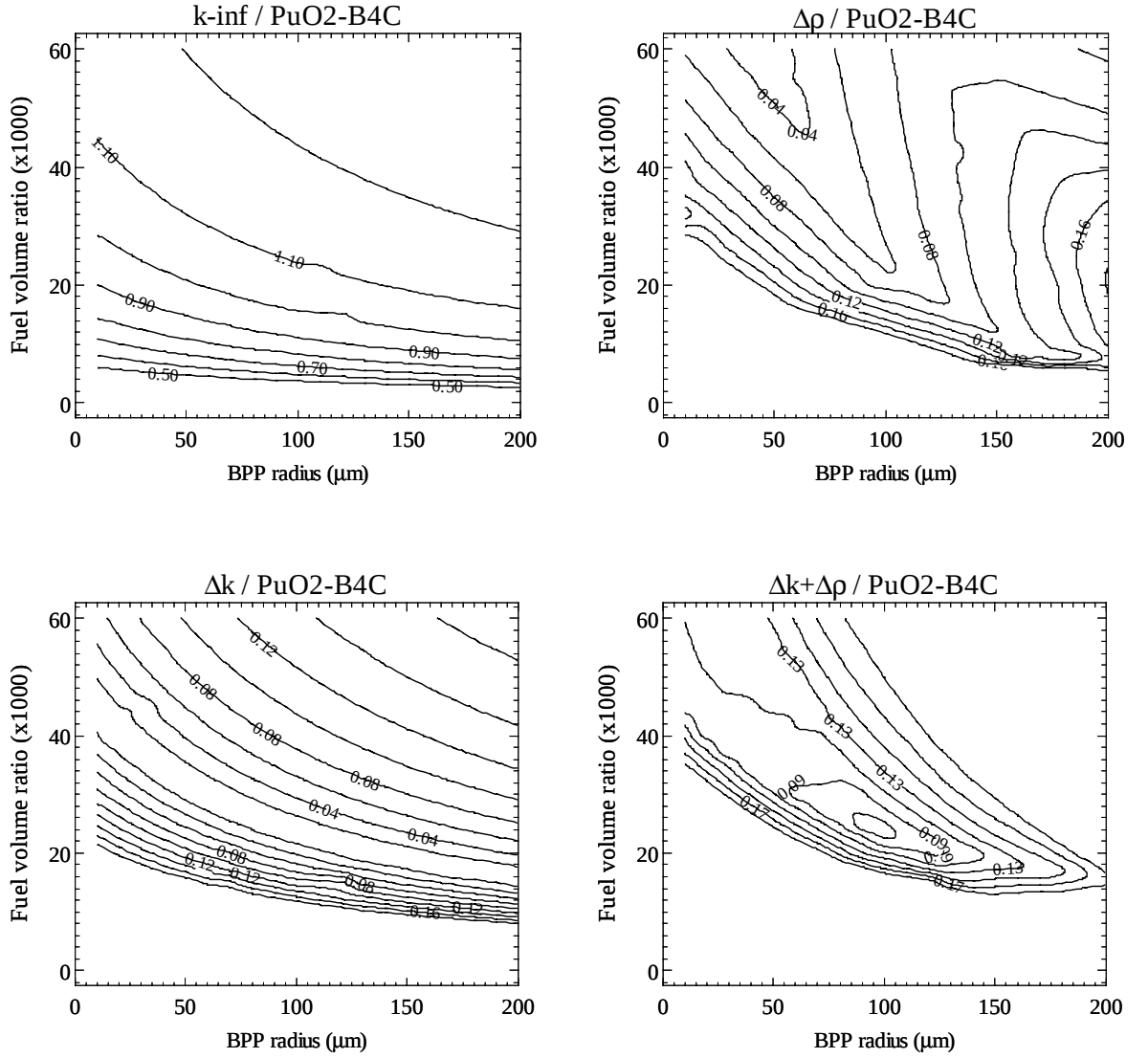


Figure 8: The k_{∞} (top left) and the reactivity swing $\Delta\rho$ (top right) as a function of the BPP radius and the fuel volume fraction. The bottom left plot shows the deviation of the initial k_{∞} from the target value 1.1, while the bottom right plot shows the sum of this and the reactivity swing (bottom right). The minimum in the latter plot is observed at a BPP radius of about 90 μm and a fuel volume ratio of 25,000 at which the reactivity swing is less than 6%.

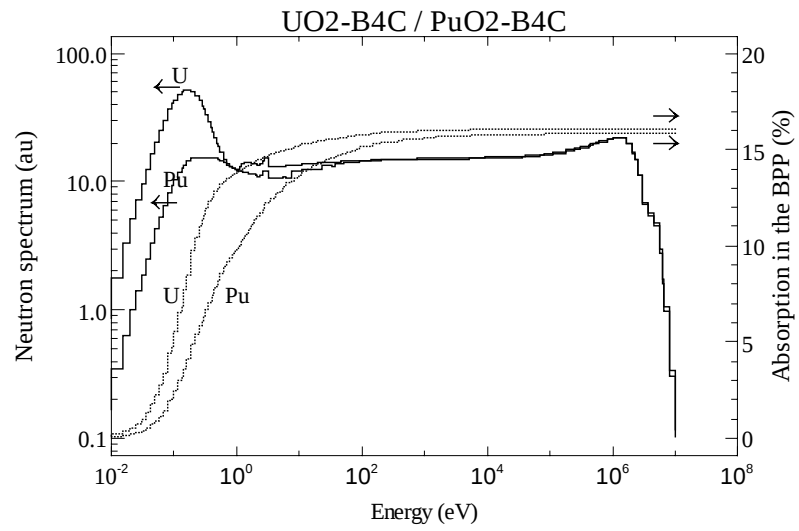


Figure 9: The neutron spectrum in the fuel zone for the both fuels with a B₄C particle in the center. Also shown is the fraction of neutrons absorbed in the BPP (right axis).

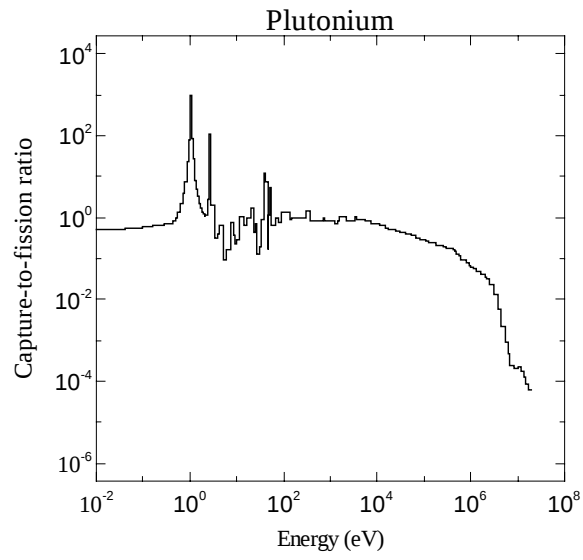


Figure 10: The capture-to-fission ratio (α) of reactor grade plutonium (see Table II) as a function of energy.