



## Section 3. Reactor physics studies

**Reactor physics aspects of plutonium burning in inert matrix fuels**J.L. Kloosterman<sup>a,\*</sup>, P.M.G. Damen<sup>b,1</sup><sup>a</sup> *Technical University of Delft, Interfaculty Reactor Institute (IRI), Mekelweg 15, NL-2629 JB Delft, Netherlands*<sup>b</sup> *NRG, P.O. Box 25, NL-1755 ZG Petten, Netherlands***Abstract**

Burnup calculations have been performed on fuels containing either reactor grade or weapons grade plutonium mixed in an inert matrix or mixed in a thorium oxide matrix. At each branching during burnup, the fuel temperature coefficient, the moderator void coefficient and the boron reactivity worth have been calculated. From the reactor physics point of view, use of thorium oxide as a matrix compares best with irradiation of plutonium in ‘ordinary’ (U, Pu) mixed oxide fuel. Because the thermal properties and the irradiation resistance of thorium oxide are generally better than of uranium oxide, the irradiation of plutonium (either reactor or weapons grade) in a thorium oxide matrix seems without problem. The use of an inert matrix to irradiate plutonium reduces the fuel temperature coefficient by a factor of two to three, which is beneficial from the viewpoint of the power reactivity defect, but disadvantageous from the viewpoint of reactivity induced accidents. Furthermore, the absence of <sup>238</sup>U or <sup>232</sup>Th in the matrix deteriorates the moderator void coefficient. Whether inert matrix fuels are feasible or not depends to a large extent on the possibilities to increase the magnitude of the moderator void coefficient and on the thermal properties of the matrix, like heat conductivity and melting point. © 1999 Elsevier Science B.V. All rights reserved.

**1. Introduction**

Burning of plutonium is most effectively done in a matrix with no uranium present, which can be accomplished in so-called inert matrices or in thorium oxide. Several matrices have been proposed and the characterisation of these is subject of worldwide research [1–3]. Besides the materials behaviour of inert matrices, the reactor physics behaviour of uranium-free fuels is also investigated, as this may be quite different from ‘conventional’ uranium oxide fuels [4–6]. For example, the absence of uranium in the fuel significantly influences the burnup reactivity swing, the reactivity coefficients and the kinetic parameters of the fuel.

In this paper the fuel temperature coefficient (FTC), the moderator void coefficient (MVC) and the boron reactivity worth (BRW) have been calculated as a

function of burnup for plutonium oxide mixed in an inert matrix and for plutonium oxide mixed in thorium oxide. Two different plutonium compositions have been used: reactor grade (RG) plutonium and weapons grade (WG). The fuel compositions are those described in the international benchmark on non-fertile fuels organised by PSI [5].

**2. Benchmark description**

The geometry consists of a PWR fuel pin with a moderator to fuel ratio of 1.9, similar to that of present day reactors. Four different matrix compositions have been used, as shown in Table 1.

As mentioned, two different plutonium compositions have been used: RG and WG. The isotopic compositions are given in Table 2.

The plutonium density in the fuel is the same for either RG or WG plutonium, but differs for each matrix to compensate for the extra neutron absorption by the matrix. Table 1 gives the plutonium density for each of the six different matrix/fuel compositions calculated.

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Table 1

Matrix/fuel compositions used in the benchmark. In total six burnup calculations have been performed (two for matrix I, two for matrix II, one for matrix III and one for matrix IV). RG means reactor grade plutonium and WG weapons grade (see Table 2)

Number	Composition	Short name	Pu composition	Pu density (g cm <sup>-3</sup> )	Linear power (W cm <sup>-1</sup> )
I	PuO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> -ZrO <sub>2</sub> -MgO	Inert matrix	RG/WG	0.5	150
II	PuO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> -ThO <sub>2</sub> -MgO	Thorium matrix	RG/WG	0.6	150
III	PuO <sub>2</sub> -ZrO <sub>2</sub> -Er <sub>2</sub> O <sub>3</sub>	Matrix with erbium	RG	0.7	190
IV	PuO <sub>2</sub> -ZrO <sub>2</sub> - <sup>10</sup> B	Matrix with boron	RG	0.7	190

Table 2

Isotopic compositions of the RG and WG plutonium (wt%)

Isotope	RG	WG
<sup>239</sup> Pu	58	93
<sup>240</sup> Pu	24	6
<sup>241</sup> Pu	13	0.8
<sup>242</sup> Pu	5	0.2

### 3. Calculations

All burnup calculations have been performed with the OCTOPUS burnup and criticality code system [7] applying the BONAMI-NITAWL-XSDRNPM codes for the resonance shielding and 1-D spectrum calculations, respectively, and the ORIGEN-S code for burnup calculations. In the spectrum calculations,  $k_{\infty}$  of the fuel, one-group resonance-shielded cross sections of the fuel nuclides, and the fine-group neutron spectrum is calculated. The latter is used to collapse fine-group cross sections of nuclides at infinite dilution to one group. These cross sections together with the one-group cross sections of the fuel nuclides explicitly modelled in the spectrum calculations are passed to the burnup module. In the spectrum calculations, an artificial pseudo nuclide is used that accounts for the neutron absorption and production rates in the fuel due to all nuclides not explicitly modelled. This way, an accurate prediction of  $k_{\infty}$  can be obtained without taking into account all actinide and fission product nuclides. In the burnup module, the densities of the fuel nuclides are updated and subsequently passed to the spectrum module. This sequence of spectrum calculations and burnup calculations is repeated many times to calculate the nuclide densities in the fuel zone at the end of the irradiation time. In the calculations, all data used were based on the JEF2.2 evaluated nuclear data file. More details about the calculations can be found in Ref. [8].

For each matrix/fuel composition shown in Table 1, reactivity coefficients have been calculated as a function of irradiation time up to 1200 days. The fuel temperature coefficient (FTC) has been calculated by increasing

the fuel temperature from 873 to 1173 K, the moderator void coefficient (MVC) by increasing the moderator void fraction up to 99.9%, and the boron reactivity worth (BRW) by increasing the boron density from zero to 500 ppm. All coefficients were calculated as  $\delta k/(k\delta P)$ , where  $k$  is the  $k$ -infinite of the fuel and  $\delta P$  the parameter change (change of fuel temperature, moderator density, or boron concentration). Furthermore, the contributions of the individual nuclides to the FTC have been calculated by means of first-order perturbation theory [9].

### 4. Results

#### 4.1. RG plutonium in the inert matrix (fuell\_rg)

When plutonium is mixed in an inert matrix without any burnable poison, the reactivity swing will be quite large due to the low conversion ratio. Some even plutonium isotopes will be converted to odd fissile ones, but the production of <sup>239</sup>Pu by neutron capture reactions in <sup>238</sup>U is absent. Fig. 1 shows the  $k_{\infty}$  as a function of irradiation time. Taking into account the  $k_{\infty}$  range of (U, Pu) mixed oxide (MOX) fuel (from about 1.2–1.0), the burnup reactivity swing of fuell\_rg shown in Fig. 1 is indeed quite large.

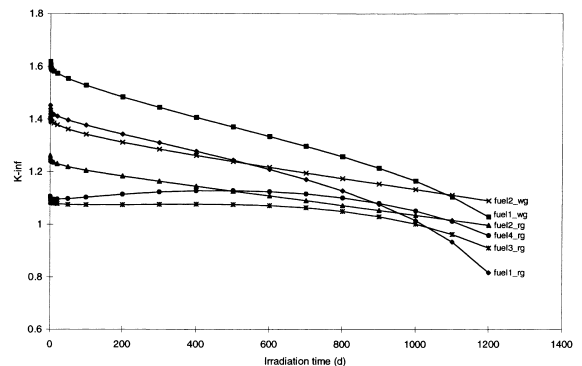


Fig. 1.  $k_{\infty}$  as a function of irradiation time for all six calculations.

#### 4.1.1. FTC

Table 3 gives the FTC as a function of irradiation time. The FTC is rather small compared to that of (U, Pu) MOX fuel ( $-2.6 \text{ pcm K}^{-1}$  [9]). Furthermore, it decreases as a function of irradiation time, while that of (U, Pu) MOX fuel increases up to  $-3.1 \text{ pcm K}^{-1}$  at a burnup of 50 MWd/kgHM.

The contributions of the individual nuclides to the FTC are shown in Fig. 2 as a function of irradiation time. The isotope  $^{240}\text{Pu}$  dominates the FTC followed by  $^{242}\text{Pu}$ . The magnitude of the latter contribution increases as a function of the irradiation time due to build-up of this isotope, which leads to more resonance shielding and a larger Doppler effect. The flux depression at 2.67 eV due to  $^{242}\text{Pu}$  increases considerably with irradiation time.

Surprisingly, the nuclide  $^{16}\text{O}$  gives a significant negative contribution to the FTC up to about 600 days, after which this contribution becomes positive. This effect is caused by upscattering due to the oxygen in the fuel, which may give a positive or negative reactivity contribution depending on the fuel composition. Fig. 3 shows the ratio of the fission and absorption cross sections of both  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  as a function of energy in the thermal range. At a moderator temperature of  $T_m = 573 \text{ K}$  and a fuel temperature of  $T_f = 873 \text{ K}$ , as was the case for these calculations, the neutrons have a mean energy of about 0.08 eV ( $3/2 k_B T$  with  $k_B$  Boltzman's constant and  $T$  the temperature). Increasing the fuel temperature shifts the Maxwell spectrum to higher energies. If the isotope  $^{239}\text{Pu}$  dominates the fission rate a shift of the Maxwell spectrum to higher energies will give a decrease of the fission-to-absorption ratio in the fuel. When  $^{241}\text{Pu}$  dominates the fission rate, which is generally the case at high burnup, the fission-to-absorption cross section ratio increases with higher temperature.

Also the isotope  $^{239}\text{Pu}$  gives a small negative contribution to the FTC, due to the fact that  $^{239}\text{Pu}$  dominates both the fission and neutron absorption rates. This can be seen from the expression for the one-group notation for  $k_\infty$  of a homogeneous system:

$$k_\infty = \frac{\nu \cdot \Sigma_f^9 + \nu \cdot \Sigma_f^0}{\Sigma_a^9 + \Sigma_a^0}, \quad (1)$$

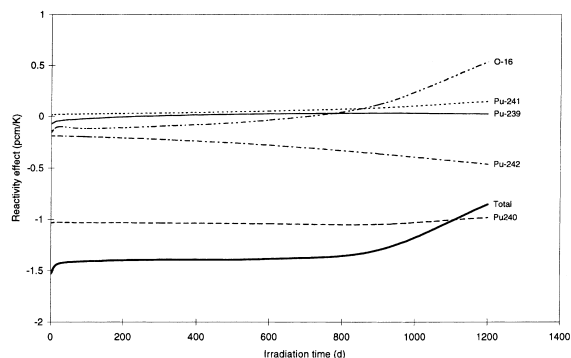


Fig. 2. Contributions of individual nuclides to the FTC as functions of irradiation time for fuel1\_rg.

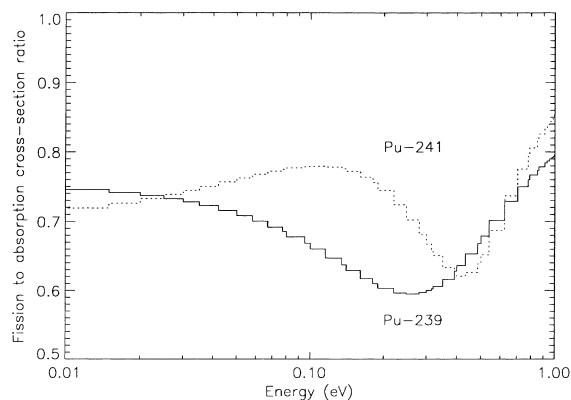


Fig. 3. Ratio of the fission and absorption cross sections of  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  as functions of energy in the thermal energy range.

where  $\nu$  is the number of neutrons released per fission,  $\Sigma_f^9$  is the macroscopic fission cross section of  $^{239}\text{Pu}$  and  $\Sigma_f^0$  the macroscopic fission cross section of all the other isotopes. The  $\Sigma_a^9$  and  $\Sigma_a^0$  are the corresponding macroscopic absorption cross sections. Upon a small disturbance of the  $^{239}\text{Pu}$  cross sections,  $k_\infty$  becomes

$$k'_\infty = \frac{\nu \cdot \Sigma_f^9 + \nu \cdot \Sigma_f^0 + \nu \cdot \delta \Sigma_f^9}{\Sigma_a^9 + \Sigma_a^0 + \delta \Sigma_a^9}. \quad (2)$$

Table 3

Fuel temperature coefficient (FTC) in units of ( $\text{pcm K}^{-1}$ ) between 873 and 1173 K as a function of irradiation time

Time (day)	Fuel1_rg	Fuel1_wg	Fuel2_rg	Fuel2_wg	Fuel3_rg	Fuel4_rg
0	-1.32	-1.21	-3.26	-3.18	-1.45	-0.74
20	-1.22	-1.13	-3.18	-3.11	-1.36	-0.71
100	-1.20	-1.12	-3.14	-3.08	-1.32	-0.76
300	-1.18	-1.09	-3.12	-3.04	-1.29	-0.91
600	-1.17	-1.04	-3.13	-2.99	-1.30	-1.11
900	-1.12	-0.97	-3.15	-2.96	-1.37	-1.26
1200	-0.77	-0.74	-3.19	-2.96	-1.34	-1.24

In case  $^{239}\text{Pu}$  dominates both the fission and absorption rates, the reactivity change  $\delta\rho$  due to a  $^{239}\text{Pu}$  cross section change becomes

$$\delta\rho \approx \frac{k'_\infty - k_\infty}{k_\infty} \approx \frac{1}{k_\infty} \cdot \frac{v \cdot \delta\Sigma_f^9}{\Sigma_a^9 + \Sigma_a^0} - \frac{\delta\Sigma_a^9}{\Sigma_a^9 + \Sigma_a^0} \approx \frac{\delta\Sigma_f^9}{\Sigma_f^9} - \frac{\delta\Sigma_a^9}{\Sigma_a^9} \quad (3)$$

If the cross sections increase, this reactivity change may be positive or negative, depending on the relative cross section changes. On the other hand, when  $^{239}\text{Pu}$  dominates the fission rate but not the absorption rate, the reactivity change reads

$$\delta\rho \approx \frac{1}{k_\infty} \cdot \frac{v \cdot \delta\Sigma_f^9}{\Sigma_a^9 + \Sigma_a^0} \approx \frac{\delta\Sigma_f^9}{\Sigma_f^9} \quad (4)$$

which is always positive upon increase of the  $^{239}\text{Pu}$  fission cross section. From this we expect that the contribution to the FTC due to  $^{239}\text{Pu}$  is stronger negative when WG plutonium is used, and that it becomes positive when other strong absorbers like burnable poisons are mixed in the fuel.

#### 4.1.2. MVC

The values for the MVC are given in Table 4. The MVC is positive up to a few hundred days of irradiation, but becomes negative afterwards. Because a whole core contains fuel batches with different burnup values, this probably poses no serious problems.

It is interesting to note the contribution due to zirconium. When nuclear data of natural zirconium at infinite dilution is used, the MVC is negative at BOL, while this coefficient is positive when nuclear data of the individual zirconium isotopes with resonance shielding treatment are used (the latter values are shown in Table 4). The fractional absorption rate by zirconium is shown in Table 5. When nuclear data of natural zirconium at infinite dilution are used, zirconium in the fuel and cladding regions absorbs 1.3% of the neutrons in the nominal (non-voided) case and 18.0% of all neutrons in the voided case. These numbers become 1.2% and 14.8%, respectively, when nuclear data of the individual zirconium isotopes with resonance shielding treatment are used. Because the absorption rate by all nuclides except the plutonium and zirconium isotopes is very

Table 5

Fractional absorption rate (%) by zirconium for fuel1\_rg in both the fuel and cladding regions. Natural zirconium has no resonance shielding treatment, while the individual zirconium isotopes have

Zr in fuel	Zr in clad	Nominal case	Voided case
Natural	Natural	1.3	18.0
Natural	Isotopes	1.2	15.8
Isotopes	Isotopes	1.2	14.8

small ( $\approx 2.5\%$ ), this means that in the latter case, the absorption rate by the plutonium isotopes is about 3.2% higher (18.0–14.8%) when nuclear data of individual zirconium isotopes with proper resonance shielding treatment are used.

#### 4.1.3. BRW

Due to burnup of plutonium and the corresponding softening of the neutron spectrum, the BRW increases significantly in magnitude. Table 6 shows the BRW as a function of irradiation time. In general, the burnup averaged BRW compares reasonably well with that of  $\text{UO}_2$  fuel (about  $-8 \text{ pcm ppm}^{-1}$ ).

#### 4.2. WG plutonium in the inert matrix (fuel1\_wg)

When WG plutonium is used instead of RG, the fissile contents in the fuel are much higher (94% vs. 71%, see Table 2) which gives higher values for  $k_\infty$  (see Fig. 1). Tables 3, 4 and 6 give the FTC, MVC and BRW, respectively, as a function of irradiation time. In general, these values do not differ much from those of the previous case (fuel1\_rg), which vitiates the generally believed opinion that WG plutonium in an inert matrix always has positive FTC values. Fig. 4 shows the contributions of individual nuclides to the FTC as a function of irradiation time. Despite the lower concentration of the  $^{240}\text{Pu}$ , its contribution to the FTC is about the same as for fuel1\_rg ( $-1 \text{ pcm K}^{-1}$ ). Again the isotopes  $^{239}\text{Pu}$  and  $^{16}\text{O}$  give a negative contribution at BOL and a (small) positive contribution at EOL for the same reason as before. Table 4 shows that the MVC is always negative in this case.

Table 4

Moderator void coefficient (MVC) in units of ( $\text{pcm \%}^{-1}$ ) between 0 and 99.9% void as a function of irradiation time

Time (day)	Fuel1_rg	Fuel1_wg	Fuel2_rg	Fuel2_wg	Fuel3_rg	Fuel4_rg
0	25	-28	-334	-347	102	114
20	46	-7.6	-323	-337	116	122
100	36	-8.4	-328	-337	99	102
300	-9.0	-33	-344	-349	35	37
600	-114	-104	-375	-375	-88	-78
900	-297	-222	-410	-407	-241	-228
1200	-532	-416	-446	-444	-439	-433

Table 6

Boron reactivity worth (BRW) in units of (pcm ppm<sup>-1</sup>) between 0 and 500 ppm as a function of irradiation time

Time (day)	Fuel1_rg	Fuel1_wg	Fuel2_rg	Fuel2_wg	Fuel3_rg	Fuel4_rg
0	-4.0	-3.8	-3.4	-3.2	-2.7	-2.4
20	-3.9	-3.7	-3.3	-3.1	-2.7	-2.4
100	-4.1	-3.8	-3.3	-3.2	-2.8	-2.5
300	-4.6	-4.1	-3.5	-3.3	-3.1	-2.9
600	-6.0	-5.0	-3.9	-3.5	-3.9	-3.8
900	-9.1	-6.9	-4.5	-4.0	-5.4	-5.5
1200	-18.6	-11.8	-5.4	-4.6	-8.9	-9.4

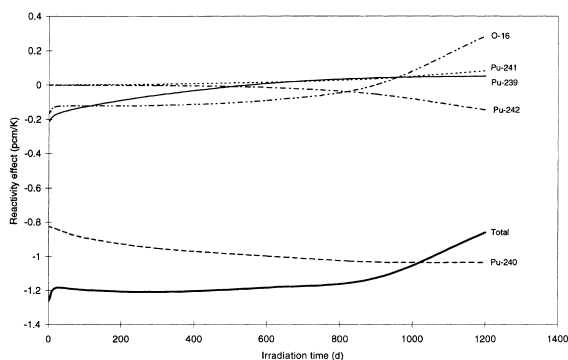


Fig. 4. Contributions of individual nuclides to the FTC as functions of irradiation time for fuel1\_wg.

Reducing the initial plutonium density in this fuel does not have a large impact on the calculated FTC and BRW values. After about 400 days of irradiation, the nuclide density of <sup>239</sup>Pu as well as the  $k_{\infty}$  of this fuel equals that of fuel1\_rg at beginning of life (BOL). Because the FTC and BRW do not change much with irradiation time during this period, reducing the initial <sup>239</sup>Pu density to a value comparable with that of fuel1\_rg at BOL ( $7.3 \times 10^{-4}$  barn<sup>-1</sup> cm<sup>-1</sup>) does not have a large impact on the FTC and BRW. However, calculations confirmed that the MVC does change. This reactivity coefficient becomes much stronger negative (factor of 4) when the <sup>239</sup>Pu nuclide density reduces to  $7.3 \times 10^{-4}$  barn<sup>-1</sup> cm<sup>-1</sup>.

#### 4.3. RG plutonium in the thorium matrix (fuel2\_rg)

When a fertile material like thorium oxide replaces the zirconium oxide in the matrix, the burnup reactivity swing will decrease due to conversion of fertile <sup>232</sup>Th to fissile <sup>233</sup>U. Furthermore, due to the larger resonance absorption rate the FTC generally increases in magnitude. The first effect is seen in Fig. 1, which shows  $k_{\infty}$  as a function of irradiation time. The burnup reactivity swing now compares much better with that of ordinary (U, Pu) MOX fuel.

The reactivity coefficients are given in Tables 3, 4 and 6. Due to the <sup>232</sup>Th in the fuel, the FTC (Table 3) and MVC (Table 4) increase to values comparable with those of ordinary (U, Pu) MOX fuel. Furthermore, these coefficients and the BRW (Table 6) do not vary much with irradiation time. Fig. 5 shows the contributions of individual nuclides to the FTC. Clearly, <sup>232</sup>Th dominates the FTC with a large negative contribution of  $-2.1$  pcm K<sup>-1</sup> during the whole irradiation time. This contribution compares with that of <sup>238</sup>U in (U, Pu) MOX fuel, which equals about  $-2.2$  pcm K<sup>-1</sup> [10].

Table 4 shows that <sup>232</sup>Th in the fuel has a mitigating effect on the MVC, like <sup>238</sup>U has in (U, Pu) MOX fuel. This is due to the relative large neutron capture cross section of <sup>232</sup>Th in the voided case comparable to that of the plutonium isotopes. When voiding occurs, <sup>232</sup>Th captures about 50% of all neutrons, while this is only 13% in the non-voided case.

#### 4.4. WG plutonium in the thorium matrix (fuel2\_wg)

Because <sup>232</sup>Th dominates the reactor physics behaviour of the fuel, the use of WG plutonium instead of RG has only minor impacts. Due to the higher fissile inventory, both the  $k_{\infty}$  values and the burnup reactivity swing are higher than in the previous case, but the FTC,

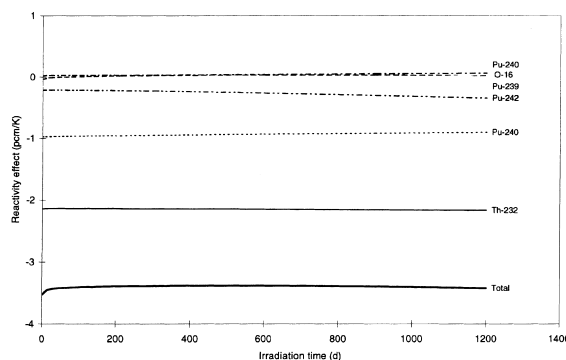


Fig. 5. Contributions of individual nuclides to the FTC as functions of irradiation time for fuel2\_rg.

MVC and the BRW are comparable with those of the RG case, as shown in Tables 3, 4 and 6, respectively.

4.5. RG plutonium in the inert matrix with erbium (fuel3\_rg)

Erbium contains several major isotopes of which only <sup>166</sup>Er and <sup>167</sup>Er have cross sections in the JEF2.2 evaluated nuclear data file. Due to its strong resonance capture integral of about 3000 barns, the latter isotope can serve as a burnable poison with a possible contribution to the FTC. Due to the presence of erbium in the matrix, the burnup reactivity loss is much less compared with the previous fuels. Fig. 1 shows that the erbium isotopes suppress considerably the initial reactivity of the fuel.

4.5.1. FTC

In Table 3, the FTC is given as a function of irradiation time. These values, which are only slightly larger than those of fuel 1, indicate that the contributions due to the Er isotopes are not very large. This is confirmed by Fig. 6, which shows the contributions of individual isotopes to the FTC. The isotope <sup>167</sup>Er contributes only  $-0.4 \text{ pcm K}^{-1}$  at BOL, and this contribution decreases rapidly with time. Already after 600 days it is negligible. The isotope <sup>166</sup>Er, which has a much smaller resonance integral of 140 barn, contributes only  $-0.1 \text{ pcm K}^{-1}$  during the whole fuel lifetime. After 1200 days of irradiation, the nuclide density of the isotope <sup>166</sup>Er has decreased with 13%, while that of <sup>167</sup>Er has decreased with a factor of 9 due to the much larger resonance integral and thermal cross section.

As shown in Fig. 6, <sup>239</sup>Pu gives a positive contribution to the FTC due to the strong burnable absorbers in the fuel, which make the relative change of the <sup>239</sup>Pu capture cross section very small compared to the relative change of the <sup>239</sup>Pu fission cross section. As can be deduced from Eq. (4), the reactivity effect due to Doppler broadening of the <sup>239</sup>Pu resonances is positive in that case.

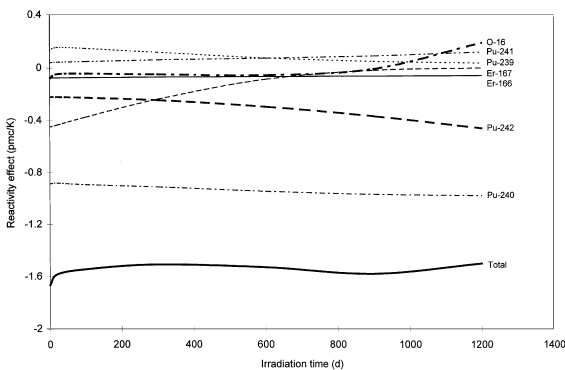


Fig. 6. Contributions of individual nuclides to the FTC as functions of irradiation time for fuel3\_rg.

The magnitude of the contributions of <sup>240</sup>Pu and <sup>242</sup>Pu increase with irradiation time. For <sup>240</sup>Pu this occurs despite the fact that the nuclide density of this isotope decreases with about a factor of two during irradiation (from  $4.2 \times 10^{-4}$  to  $2.0 \times 10^{-4} \text{ barn}^{-1} \text{ cm}^{-1}$ ). One should realise that the Doppler effect is mainly due to a change of the resonance escape probability, which depends on both the absorber nuclide density and the temperature derivative of the resonance integral [11]

$$\alpha_T = - \frac{N_F}{\xi \cdot \Sigma_p} \cdot \frac{dI}{dT_F}, \quad (5)$$

where  $N_F$  is the nuclide density of the resonance isotope,  $\xi$  the logarithmic energy decrement per collision with a moderator nuclide,  $\Sigma_p$  the potential scatter cross section of the moderator,  $I$  the effective resonance integral of the absorber, and  $T_F$  the temperature of the fuel lump. The nuclide density and the derivative of the resonance integral are given in Table 7 for both <sup>240</sup>Pu and <sup>242</sup>Pu. For the first isotope the product of these two factors decreases no more than 20%, despite the fact that the <sup>240</sup>Pu nuclide density decreases more than a factor of two. For <sup>242</sup>Pu, the temperature derivative of the resonance integral is hardly dependent on the irradiation time, but the nuclide density increases with a factor of two.

4.5.2. MVC/BRW

The presence of Er in the fuel has a deteriorating effect on the MVC (see Table 4). At BOL, this coefficient has a large positive value of about  $100 \text{ pcm } \%^{-1}$ . Due to the erbium in the fuel, the neutron spectrum is relatively hard, which gives quite low values for the BRW (see Table 6).

4.6. RG plutonium in the inert matrix with B-10 (fuel4\_rg)

The sixth fuel composition is characterised by the presence of <sup>10</sup>B as a burnable poison. Again the burn-

Table 7  
Nuclide density  $N_F$  and derivative of the resonance integral  $dI/dT_F$  for fuel3\_rg as functions of irradiation time

Irradiation time (day)	$N_F$ ( $\text{barn}^{-1} \text{ cm}^{-1}$ )	$dI/dT_F^a$ ( $\text{barn K}^{-1}$ )	Product ( $\text{K}^{-1} \text{ cm}^{-1}$ )
<b><sup>240</sup>Pu</b>			
0	$4.22 \times 10^{-4}$	$2.74 \times 10^{-2}$	$1.15 \times 10^{-5}$
600	$3.66 \times 10^{-4}$	$3.04 \times 10^{-2}$	$1.11 \times 10^{-5}$
1200	$2.00 \times 10^{-4}$	$4.96 \times 10^{-2}$	$9.92 \times 10^{-6}$
<b><sup>242</sup>Pu</b>			
0	$8.71 \times 10^{-5}$	$7.24 \times 10^{-2}$	$6.31 \times 10^{-6}$
600	$1.06 \times 10^{-4}$	$7.25 \times 10^{-2}$	$7.70 \times 10^{-6}$
1200	$1.51 \times 10^{-4}$	$6.89 \times 10^{-2}$	$1.04 \times 10^{-5}$

<sup>a</sup> Derived from the NITAWL-II output, valid only for the resolved energy range.

able poison depresses the initial fuel reactivity. Fig. 1 shows that  $k_{\infty}$  increases slightly up to 500 irradiation days. After about 800 days,  $k_{\infty}$  decreases rapidly with time, which indicates that the fissile plutonium isotopes are burned rapidly then. The FTC, MVC and BRW are shown in Tables 3, 4 and 6, respectively. The FTC is much smaller than in the previous case, and also much smaller than in the first fuel (RG plutonium mixed in an inert matrix). This is mainly due to the relative small contributions of the even non-fissile plutonium isotopes and the positive contributions of the odd fissile isotopes. As explained before, the latter effect is due to the large neutron absorption rate by the boron. Fig. 7 shows the contributions of individual nuclides to the FTC. Similarly to the previous fuel, the MVC has a large positive value at BOL due to the presence of the  $^{10}\text{B}$ .

## 5. Discussion and conclusions

1. Irradiation of plutonium in an inert matrix gives values for the fuel temperature coefficient (FTC) which are smaller by a factor of two to three compared with those of (U, Pu) MOX or  $\text{UO}_2$  fuel. It depends to a large extent on the thermal properties of the matrix, like the thermal conductivity and the melting point, whether these lower FTC values are acceptable or not. In general, lower FTC values lead to a lower power defect (reactivity gain when going from hot full power to hot zero power), but to larger heat deposition in the fuel in case of reactivity induced accidents.
2. Irradiation of plutonium in an inert matrix leads to very small but negative moderator void coefficients (MVC). The results presented here are based on pin-cell calculations without leakage. This implies that the uncertainty in this parameter may range up to  $50 \text{ pcm } \%^{-1}$  [12]. Therefore, it is recommended

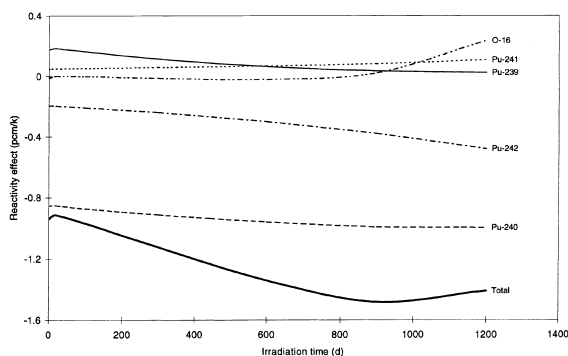


Fig. 7. Contributions of individual nuclides to the FTC as functions of irradiation time for fuel4\_rg.

to investigate the MVC with more advanced full core calculations.

3. Irradiation of plutonium in an inert matrix and in thorium oxide leads to values of the boron reactivity worth (BRW) that compare with those of (U, Pu) MOX and  $\text{UO}_2$  fuels.
4. The use of weapons grade plutonium instead of reactor grade has no large impact on the FTC, the MVC or the BRW. Although the  $^{240}\text{Pu}$  concentration in weapons grade plutonium is three times less than in reactor grade plutonium (compare fuel1\_rg with fuel1\_wg, or fuel2\_rg with fuel2\_wg), the contributions of this isotope to the FTC are about the same in both cases ( $-1.0 \text{ pcm } \text{K}^{-1}$ ).
5. The use of a thorium oxide matrix considerably improves the FTC and the MVC to values comparable with those of  $\text{UO}_2$  fuel. The BRW has values in between those of (U, Pu) MOX fuel and of  $\text{UO}_2$  fuel.
6. The use of erbium as a burnable poison considerably reduces the burnup reactivity swing and gives a negative contribution to the FTC (magnitude of the FTC increases) up to about 600 irradiation days. However, erbium deteriorates the MVC and reduces the BRW with several tens of percents compared to the case with no burnable poison. The same conclusion holds for the use of  $^{10}\text{B}$  as a burnable poison, except that this nuclide does not give a contribution to the FTC.
7. From the reactor physics standpoint, irradiation of plutonium in thorium oxide seems feasible, while this cannot be concluded from this paper for the irradiation of plutonium in an inert matrix. In the latter case, the thermal conductivity and melting point of the matrix will determine the feasibility of such a material in case of accidents, like the important classes of reactivity induced accidents and moderator voiding accidents. Detailed investigation of such accident scenarios is necessary to assess the safety of these innovative fuel types.

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