

Neutronics calculations on the impact of burnable poisons to safety and non-proliferation aspects of inert matrix fuel

C. Pistner ^{a,*}, W. Liebert ^a, F. Fujara ^b

^a *Interdisciplinary Research Group Science, Technology and Security (IANUS), Darmstadt University of Technology, Hochschulstrasse 4a, D-64289 Darmstadt, Germany*

^b *Institute of Condensed Matter Physics, Darmstadt University of Technology, D-64289 Darmstadt, Germany*

Abstract

Inert matrix fuels (IMF) with plutonium may play a significant role to dispose of stockpiles of separated plutonium from military or civilian origin. For reasons of reactivity control of such fuels, burnable poisons (BP) will have to be used. The impact of different possible BP candidates (B, Eu, Er and Gd) on the achievable burnup as well as on safety and non-proliferation aspects of IMF are analyzed. To this end, cell burnup calculations have been performed and burnup dependent reactivity coefficients (boron worth, fuel temperature and moderator void coefficient) were calculated. All BP candidates were analyzed for one initial BP concentration and a range of different initial plutonium-concentrations (0.4–1.0 g cm⁻³) for reactor-grade plutonium isotopic composition as well as for weapon-grade plutonium. For the two most promising BP candidates (Er and Gd), a range of different BP concentrations was investigated to study the impact of BP concentration on fuel burnup. A set of reference fuels was identified to compare the performance of uranium-fuels, MOX and IMF with respect to (1) the fraction of initial plutonium being burned, (2) the remaining absolute plutonium concentration in the spent fuel and (3) the shift in the isotopic composition of the remaining plutonium leading to differences in the heat and neutron rate produced. In the case of IMF, the remaining Pu in spent fuel is unattractive for a would be proliferator. This underlines the attractiveness of an IMF approach for disposal of Pu from a non-proliferation perspective. © 2006 Elsevier B.V. All rights reserved.

1. Introduction

The existing stockpiles of separated plutonium – military or civilian – pose a proliferation threat that has to be dealt with urgently. Today, about 250 tonnes of military plutonium exist worldwide [1], most

of it in Russia and in the US. The civilian stockpiles of separated Pu are of about the same size. However, only about 4–6 kg of plutonium are needed to build a nuclear weapon.

Uranium-free Pu fuels with inert matrix (IMF) are under research worldwide which could be used to achieve an efficient reduction of Pu stockpiles [2–4]. From a non-proliferation viewpoint, this fuel may be advantageous in comparison to conventional MOX fuels if it leads to (1) a higher fraction of initial Pu being burned, (2) a lower absolute Pu

* Corresponding author. Tel.: +49 6151 16 2873; fax: +49 6151 16 6039.

E-mail address: christoph.pistner@physik.tu-darmstadt.de (C. Pistner).

concentration in the spent fuel and (3) a considerable shift in the isotopic composition of the remaining Pu [5,6]. The isotopic composition determines the heat- and neutron-rate of Pu and thus influences the attractiveness for weapons diversion [7,8].

Apart from advantages with respect to non-proliferation aspects, IMF may show differences to currently used MOX fuels with regard to reactor physics aspects (especially concerning reactivity behavior and reactivity coefficients) that will give limitations for possible fuel concepts. To this end, burnup dependent reactivity coefficients are analyzed in this paper.

2. Computational system

To estimate the potential of IMF for Pu elimination in existing light-water reactors (LWR), we performed a set of burnup calculations for fuels with different initial compositions regarding the Pu concentration and burnable neutron poisons (BP).

The calculations are performed with the program system MCMATH [9], based on the neutron transport code MCNP [10] and burnup routines programmed in Mathematica [11]. This system takes into account about 50 actinide nuclides and 150 fission product isotopes. All calculations have been performed on the basis of cell burnup calculations with an infinite lattice, thus no leakage is taken into account. The geometry of the fuel moderator cell has been chosen according to [12], i.e., the radius of the fuel rod is 0.4095 cm, the outer cladding radius is 0.4750 cm and the pitch of the fuel cell is 1.3133 cm. The temperature of the fuel has been set to 900 K, moderator and cladding temperature are taken to be 600 K. A constant average boron concentration in the moderator of 500 ppm was used during the burnup calculations to estimate the nuclide composition of the fuels. For all other calculations presented here, no boron was assumed in the moderator.

For reference purposes, calculations for MOX fuels based on depleted uranium with an initial fissionable plutonium content of 3.2–6.0% and a heavy metal density of 9.0 g(HM) cm⁻³ were investigated. The calculations were performed at a constant power level of 38.3 W g⁻¹ up to a maximum burnup of 50 MW d kg⁻¹(HM).

As fuel matrix for IMF, yttria-stabilized zirconia with a density of 5.0 g cm⁻³ is chosen as discussed e.g., in [13]. The initial Pu density of the fuels investigated is varied from 0.4 to 1.0 g cm⁻³ in steps of

Table 1
RPu and WPu isotopic abundancies (%)

Isotopes (%)	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
RPu	1.80	59.00	23.00	12.20	4.00
WPu	0.05	93.60	6.00	0.30	0.05

0.05 g cm⁻³. Two isotopic composition are used, typical reactor-grade Pu (RPu) and weapons-grade Pu (WPu), see Table 1 [12]. For IMF, the calculations are performed at a power density of 360 W cm⁻³ up to a maximum burnup of 1250 full power days, corresponding to a fuel burnup of 450 kW d cm⁻³ (equivalent to 50 MW d kg⁻¹(HM) in MOX fuel with a density of 9 g cm⁻³).

The primary aim of our calculations is the comparison of MOX fuels with possible IMF from a non-proliferation perspective. For this purpose the fuel compositions for fresh and spent fuel have to be known. For a given initial inventory, the spent fuel composition is determined mainly by the achieved discharge burnup B_d of the fuel. The maximum B_d achievable is restricted by either reactivity constraints or the possible fuel lifetime. In the following discussion we assume a possible fuel lifetime of 450 kW d cm⁻³ for MOX and IMF as a target burnup.

Whether such a burnup is achievable for a given initial fuel composition is then depending on the reactivity change of the fuel during burnup. The average core reactivity of a reactor can be estimated assuming a linear reactivity model [14]. If the core consists of n batches of fuel, the burnup dependant core average reactivity during one cycle will be given by

$$\rho_{av}(t) = \frac{1}{n} \sum_{i=1}^n \rho_{\infty}(t + (i-1) \cdot t_{\text{cycle}}), \quad t \in [0, t_{\text{cycle}}], \quad (1)$$

with $\rho_{\infty}(t)$ being the calculated burnup dependant reactivity of the fuel and t_{cycle} being the cycle length. For a given fuel, the achievable cycle length is then determined after subtracting the reactivity loss due to leakage $\Delta\rho_L$, which has not been taken into account in the cell calculations and is assumed here to be 0.03. This value is subtracted from the calculated $\rho_{av}(\text{EOC})$ and the achievable cycle length is calculated such that the resulting value is zero at the end of the cycle, i.e., $\rho_{av}(\text{EOC}) - \Delta\rho_L = 0$ for the calculated cycle length. This equation is solved with the help of Mathematica for all fuels under consideration.

3. Burnup behavior of inert matrix fuels with different burnable poisons

To compensate the high initial reactivity of fresh IMF, BP will have to be used. To analyze the impact of BP on burnup, four BP candidates are analyzed for IMF with RPu as well as WPu, with one specific initial BP concentration and a range of different initial Pu-concentrations ($0.4\text{--}1.0\text{ g cm}^{-3}$). Thereby, a homogenous mixture of the BP with the fuel is assumed. Since Er and Gd will turn out to be the two most promising BP candidates, four different concentrations are investigated to further study the impact of their concentration on fuel burnup, see

Table 2. The concentrations have been chosen to encompass a broad range of initial reactivities for the fuels under consideration.

Some representative results for the RPu-fuels will be discussed in the following. Fig. 1 shows $\rho_{\infty}(t)$ for

Table 2

Reference concentrations for analyzed BP (g cm^{-3})

	B	Eu	Gd	Er
RPu	0.02	0.04	0.03, 0.06, 0.09, 0.12	0.15, 0.20, 0.28, 0.40
WPu	0.03	0.06	0.02, 0.04, 0.08, 0.12	0.25, 0.40, 0.55, 0.70

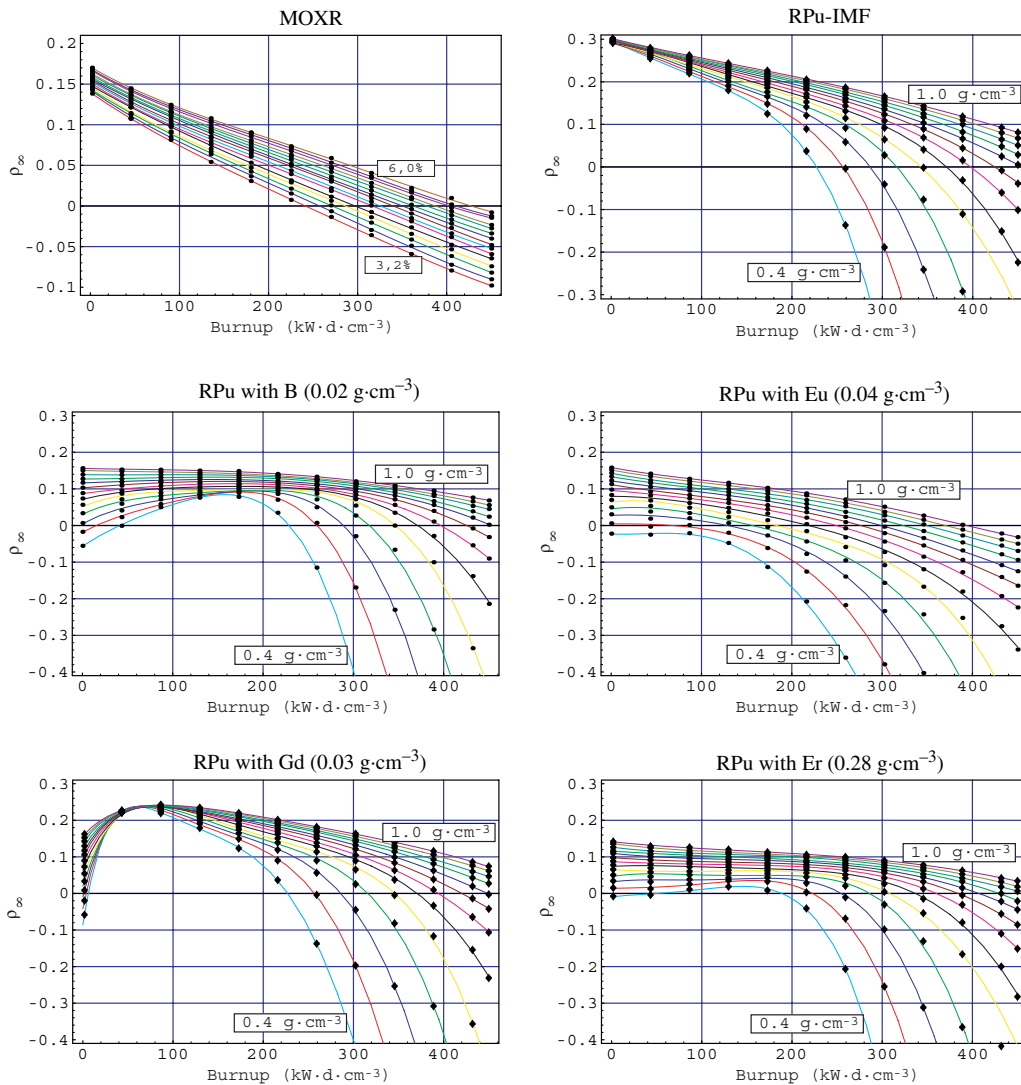


Fig. 1. ρ_{∞} for MOXR with initial fissile RPu concentrations from 3.2% to 6.0% and IMF with initial RPu concentrations from 0.4 to 1.0 g cm^{-3} (higher Pu concentrations correspond to higher reactivity) and different BP.

RPu–MOX (MOXR) and RPu–IMF. For MOXR, values of $\rho_{\infty}(\text{BOL})$ are between 0.14 and 0.17, depending on the initial Pu-concentrations. A basically linear reactivity loss can be seen for all MOXR up the maximum burnup. For all IMF, ρ_{∞} drops rather rapidly when the contained Pu is exhausted. While $\rho_{\infty}(\text{BOL})$ is about 0.3 for all RPu–IMF without BP, if BP are used it depends strongly on the combination of BP- and Pu-concentrations. Gd has the strongest impact on $\rho_{\infty}(\text{BOL})$ and burns fastest and nearly complete, as could be expected because of the very high thermal cross-sections of its most important isotopes. B also has a strong impact on ρ_{∞} up to a burnup of about 200 kW d cm^{-3} , thereafter most of the initial B has been transmuted. With Eu, ρ_{∞} is lowered throughout the whole burnup, showing that Eu is not a very good BP. In the case of Er, a nearly constant ρ_{∞} is resulting during the first half of the burnup.

In Fig. 2 the discharge burnup B_d for IMF fuels with different initial Pu- and Er-concentrations is shown. To this end, a value of $n = 4$ is chosen in Eq. (1), assuming that the high target burnup of 450 kW d cm^{-3} chosen in Section 2 will have to be achieved in a four cycle burnup. Off course, any other combination of target burnup and number of cycles the user is interested in can be chosen in the calculational system. As can be seen, B_d increases approx. linearly with increasing initial Pu. Increasing the initial Er concentration will reduce the achievable B_d . This is due to the fact, that Er is not fully burned after one cycle, thus reducing ρ_{∞} in Eq. (1). To achieve the reference burnup of 450 kW d cm^{-3} (denoted by the horizontal line in Fig. 2), one has to increase the initial Pu-concentration in the fuel (see vertical dashed lines in Fig. 2).

4. Analysis of reactivity coefficients

To further investigate the neutronics behavior of IMF, burnup dependent values for three reactivity coefficients have been calculated, i.e., the boron worth (BWC), fuel temperature (FTC) and moderator void coefficient (MVC).

4.1. Boron worth coefficient

Knowledge of the burnup dependant BWC is necessary to estimate, whether the $\rho_{\text{av}}(\text{BOC})$ for an IMF under consideration can be compensated in today's PWR by the use of soluble boron in the moderator. As an example, Table 3 summarizes results for RPu–IMF that might achieve 450 kW d cm^{-3} in a four cycle burnup (for a broader discussion, especially concerning results of WPu–IMF cf [9]).

As discussed in Section 3, for increasing initial Er concentration, correspondingly more initial Pu has to be used to achieve a reference burnup of 450 kW d cm^{-3} . Still, $\rho_{\text{av}}(\text{BOC})$ decreases with increasing Er-concentration as can be seen in Table 3. At the same time, due to the increasing Pu-concentration, the average boron worth $\text{BWC}_{\text{av}}(\text{BOC})$ becomes less negative. It is assumed here that to maintain criticality, a critical boron concentration $c_B = (\rho_{\text{av}} - \Delta\rho_L)/\text{BWC}_{\text{av}}$ is needed, again taking a $\Delta\rho_L$ of 0.03 for leakage into account. If a maximum tolerable value of 2000 ppm boron in the moderator is then assumed for today's PWR, a reference fuel with 0.7 g cm^{-3} initial RPu and 0.2 g cm^{-3} Er can be identified (case RPuEr7020 in Table 3).

In comparison to Er, for Gd the necessary Pu-concentration increases only slightly with increasing initial Gd-concentration. In contrast to other BP,

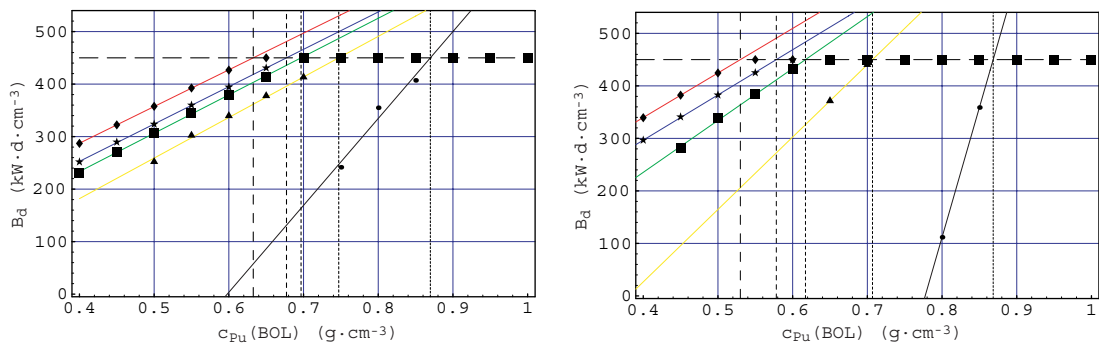


Fig. 2. Reactivity limited discharge burnup B_d for IMF in dependence of initial Pu-concentration $c_{\text{Pu}}(\text{BOL})$ for fuels without BP and with increasing concentrations of Er (from left to right, see Table 2). The horizontal line depicts the chosen reference burnup of 450 kW d cm^{-3} , the vertical dashed lines give the corresponding $c_{\text{Pu}}(\text{BOL})$ necessary to achieve the reference burnup for the different concentrations of Er.

Table 3

 ρ_{av} , BWC_{av} and corresponding c_B for RPu–IMF achieving 450 kW d cm^{-3} burnup within 4 cycles

Fuel	$c_{Pu}(BOL)/c_{Er}(BOL)$ ($g\ cm^{-3}$)	$\rho_{av}(BOC)/\rho_{av}(Max.)$	$BWC_{av}(BOC)/$ $BWC_{av}(Max.)$ ($10^{-5}\ ppm^{-1}$)	$c_B(BOC)/c_B(Max.)$ (ppm)
RPu064	0.64/–	0.181/0.181	–4.2/–4.2	3600/3600
RPuEr6815	0.68/0.15	0.116/0.116	–3.9/–3.9	2200/2200
RPuEr7020	0.70/0.20	0.099/0.099	–3.7/–3.7	1900/1900
RPuEr7528	0.75/0.28	0.075/0.075	–3.4/–3.4	1300/1300
RPuEr8740	0.87/0.40	0.050/0.050	–2.8/–2.8	700/700
RPuGd6403	0.64/0.03	0.125/0.130	–4.0/–4.5	2400/2200
RPuGd6406	0.64/0.06	0.093/0.107	–4.0/–4.7	1600/1600
RPuGd6509	0.65/0.09	0.069/0.091	–3.8/–4.8	1000/1300
RPuGd6612	0.66/0.12	0.049/0.074	–3.7/–4.7	500/900

Gd is burned rather rapidly, leading to an increase of ρ_{av} after BOC. Thus in Table 3 the values at maximum reactivity during a cycle are given in addition to those at BOC. It can be seen that ρ_{av} decreases considerably with increasing Gd, while BWC_{av} nearly remains constant. Again, assuming a maximum acceptable value of 2000 ppm for c_B , a fuel with 0.64 $g\ cm^{-3}$ initial RPu and 0.06 $g\ cm^{-3}$ Gd (case RPuGd6406) can be identified from Table 3 as a reference fuel.

4.2. Fuel temperature coefficient

Furthermore, the burnup dependant FTC for a change in fuel temperature from 900 to 300 K has been calculated for all fuels. The FTC is primarily determined by the isotopic composition of the Pu and/or U in the fuel or, in case of IMF with BP, may be influenced by the properties of the BP.

Our calculations show that, while for typical uranium fuel the FTC changes from initially about -2×10^{-5} to $-4 \times 10^{-5}\ K^{-1}$ at EOL, corresponding values for MOX range from -3×10^{-5} to about $-4 \times 10^{-5}\ K^{-1}$ in case of RPu–MOX and

are about $0.2 \times 10^{-5}\ K^{-1}$ less negative for WPU–MOX. For RPu–IMF without BP the FTC changes from about $-1 \times 10^{-5}\ K^{-1}$ at BOL to about $-2 \times 10^{-5}\ K^{-1}$ at EOL, again, WPU–IMF being about $0.2 \times 10^{-5}\ K^{-1}$ less negative. Thus the FTC for IMF is considerably less negative than in uranium fuels or MOX. Adding a BP can have an additional impact on the FTC at least during the first part of the burnup. Fig. 3 shows the influence of different BP concentrations of Gd and Er on FTC(BOL) for IMF with RPu or WPU respectively.

Already for very small concentrations ($0.01\ g\ cm^{-3}$) of Gd, the FTC changes to about $-0.6 \times 10^{-5}\ K^{-1}$ for WPU or $-0.7 \times 10^{-5}\ K^{-1}$ for RPu respectively and becomes less negative with further increasing Gd-concentration. For Er as BP, the opposite behavior can be seen, with the FTC nearly linearly decreasing with increasing Er-concentration. This is due to the fact, that Gd is primarily a thermal neutron absorber, leading to a much harder neutron spectrum, while Er is a strong resonance absorber, thus giving a considerable negative contribution to the FTC. For relevant Er concentrations of $0.2\text{--}0.3\ g\ cm^{-3}$ as calculated in

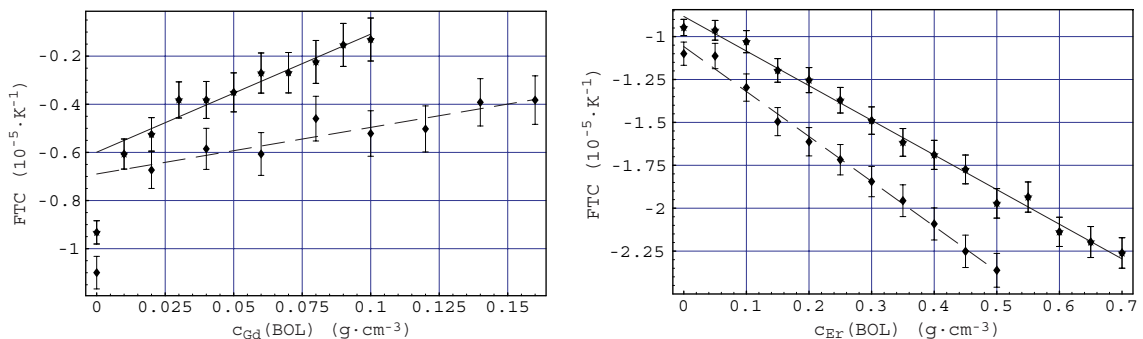


Fig. 3. Dependence of FTC(BOL) on concentration of Gd or Er. Lines correspond to IMF with $0.55\ g\ cm^{-3}$ initial WPU (solid line) and $0.65\ g\ cm^{-3}$ RPU (dashed line) for Gd, $0.60\ g\ cm^{-3}$ WPU and $0.65\ g\ cm^{-3}$ RPU for Er.

Section 4.1, the value of the FTC at BOL is still less negative than in uranium fuel, but the difference with respect to uranium fuel amounts to only about 25% while in the case of RPu–IMF without BP the FTC was 50% less negative.

4.3. Moderator void coefficient

The MVC is mainly determined by the moderator to fuel ratio, i.e., the concentration of Pu in the fuel. Thus it typically has its highest values at BOL becoming more negative with burnup when the Pu-concentration decreases. Fig. 4 shows values of MVC(BOL) for RPu–IMF and WPu–IMF without BP for different initial Pu-concentration and two void-fractions, 10% and 90%. Especially for 90% void the calculated MVC will yield to pessimistic values, since no leakage is taken into account. Leakage will increase for high voids, giving a negative contribution to the real MVC. Nevertheless, the calculated values of the MVC will allow to compare the different Pu containing fuels.

While the absolute values of the MVC(BOL) for small voids (10%) are small, they stay negative for all concentrations of Pu investigated. For a situation with large voids (90%), the values are still negative for low Pu concentrations. For more than about 0.62 g cm^{-3} RPu or 0.78 g cm^{-3} WPu however, the values of MVC(BOL) become positive. The addition of B, Eu or Gd as BP to the fuel results in increasingly positive values of the MVC, especially for high voids. Er gives a small negative contribution to the MVC in case of small voids and the least positive contribution to the MVC in the case of high voids. Thus, for the reference fuels discussed in Section 4.1, for RPuEr7020 the MVC in the case of a large void-fraction (90%) has a small

positive value during the first 100 kW d cm^{-3} burnup. For RPuGd6406 the MVC(BOL) is positive for 10% void as well as for 90% void, with an absolute value of about $200 \times 10^{-5} \%^{-1}$. Negative values are only reached after $50\text{--}100 \text{ kW d cm}^{-3}$ burnup.

Thus, considering reactivity coefficients, Er shows a far superior behavior than Gd as a BP. To assure strictly negative values of the MVC, a lower initial Pu concentration corresponding to a reduced achievable B_d may have to be used.

5. Comparison of fuels

Table 4 summarizes the specifics of the reference fuels with respect to the non-proliferation criteria introduced in Section 1 for the example of RPu. Comparable values are achieved for WPu fuels (cf. [9]).

For comparison, our calculations show that in conventional uranium fuel with a burnup of 450 kW d cm^{-3} , the spent fuel contains about 0.11 g cm^{-3} Pu. The heat- and neutron rate of that Pu would be about 30% higher than that of today's typical RPu as defined in Table 1. For MOXR fuel to achieve the same burnup, the initial Pu concentration would have to be 0.62 g cm^{-3} . About 30% less Pu would remain in spent fuel, also having about 30% higher heat- and 50% higher neutron-rate than standard RPu. An IMF without BP would need 0.64 g cm^{-3} initial Pu, but nearly 77% less Pu would remain after irradiation. Practically all ^{239}Pu would have been either fissioned or transmuted, such that the heat-rate of the remaining Pu would be more than a factor of 2, the neutron-rate nearly a factor of 3.5 higher than for RPu.

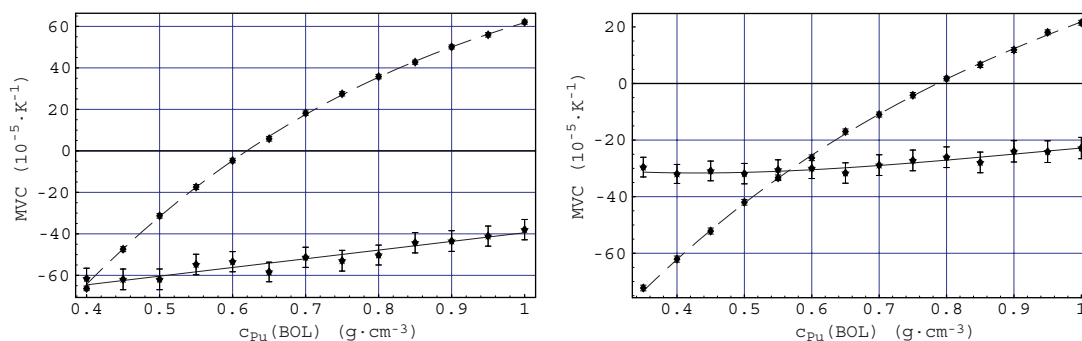


Fig. 4. MVC(BOL) for varying initial Pu concentrations $c_{\text{Pu}}(\text{BOL})$ for RPu- and WPu–IMF. Lines correspond to 10% (solid line) and 90% (dashed line) void.

Table 4
RPu–IMF in comparison with MOX and uranium-fuel

Fuel		c_{Pu} (g cm ⁻³)	$c_{^{239}\text{Pu}}$ (g cm ⁻³)	Heat-rate (W kg ⁻¹ (Pu))	Neutron-rate (10 ³ s ⁻¹ kg ⁻¹ (Pu))
UO _x	(EOL)	0.11	0.05	17.8	420
RPu–MOX	(BOL)	0.62	0.37	13.3	324
	(EOL)	0.43	0.17	17.3	497
		-30.1%	-53.5%	×1.30	×1.53
RPu064	(BOL)	0.64	0.38	13.3	324
	(EOL)	0.15	0.00	29.1	1110
		-76.9%	-98.7%	×2.19	×3.42
RPuGd6406	(BOL)	0.64	0.38	13.3	324
	(EOL)	0.15	0.01	30.0	1107
		-77.0%	-98.6%	×2.26	×3.42
RPuEr7020	(BOL)	0.70	0.41	13.3	324
	(EOL)	0.21	0.02	28.6	925
		-70.4%	-96.1%	×2.15	×2.85

Values for BOL and EOL as well as relations between BOL and EOL.

Because such a fuel will probably not be feasible in a current LWR, addition of BP is necessary as discussed in Section 4.1. When using Gd, no impact on initial and remaining Pu-concentrations can be seen for a fuel capable to achieve the same burnup, the isotopic composition of the remaining Pu also being nearly identical to the case of pure RPu–IMF. As discussed in Sections 4.2 and 4.3, this type of fuel may be uncomfortable due to drastically reduced FTC and MVC at BOL, making it less attractive from a safety point of view. Finally, when using Er as a BP, a higher initial Pu concentration of 0.7 g cm⁻³ has to be used to achieve the reference burnup. About 0.21 g cm⁻³ remain at EOL, which is nearly twice the amount in spent uranium fuel but only about one half of that in spent MOXR. With this fuel, a total Pu reduction of 70% seems feasible. Also, the remaining Pu has a more than a factor of two higher heat-rate and nearly a factor of three higher neutron-rate compared to the initial RPu. Thus this Pu would be very unattractive for a would be proliferator.

6. Conclusions

Burnup calculations for a variety of Pu containing fuels have been performed. By the use of the linear reactivity model, the necessary Pu concentration to achieve a given discharge burnup can be estimated. Based on calculations of the burnup dependent boron worth, the needed initial boron concentration has been calculated and feasible combinations of initial Pu and BP concentra-

tions are defined. For a set of reference fuels, the fuel and moderator temperature coefficient have been investigated, showing the superior behavior of Er as BP compared to Gd with respect to acceptable values of the reactivity coefficients. Finally, when comparing the non-proliferation aspects of the fuels, the clear advantage of IMF in comparison to MOXR can be shown. In a suitable IMF, only about 30% of the initial Pu will remain in spent fuel, the isotopic composition of this Pu is shifted to the heavier Pu-isotopes, making this Pu very unattractive for a would be proliferator.

Acknowledgement

Work on this project has been made possible by funding of the German Federal Ministry of Education and Research (BMBF) and the German Foundation for Peace Research (DSF).

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