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Dissolution, Reactor, and Environmental Behavior of ZrO2-MgO Inert Fuel Matrix: Neutronic Evaluation of ZrO2-MgO Inert Fuels

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Dissolution, Reactor, and Environmental Behavior of ZrO_2-MgO Inert Fuel Matrix

Neutronic Evaluation of MgO-ZrO_2 Inert Matrix Fuels

4th Progress Report

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I. Summary

This report presents the results of the Task 4, defined in working program as: evaluation of reactivity feedback coefficients. Three main parameters of the Fertile-Free Fuel (FFF) lattices were evaluated: Moderator Temperature Coefficient (MTC), Fuel Temperature Coefficient due to Doppler Effect (DC), and soluble Boron reactivity worth (BW).

One of the major design challenges associated with utilization of FFF is deterioration of the temperature coefficients and control materials reactivity worth caused by high thermal cross-section of Pu and consequent hardening of the neutron spectrum. The purpose of the investigation reported in this section is to estimate the potential of addition of different burnable poison (BP) materials to improve reactivity feedback coefficients without significant deterioration of control materials worth. Therefore, each parameter was evaluated for all BP design options. For each design option, i.e. BP material and geometrical arrangement, one design was selected, that with sufficient loading of BP material to ensure operationally acceptable maximum soluble boron concentration. These BP loading values were determined in Task 3 of the current project.

List of calculated cases is presented in the following section, the case identification and associated design parameters are detailed in the previous progress report.

II. List of calculated cases

This section presents a list of all BP designs considered for evaluation of reactivity feedback coefficients. Main design parameters are summarized in Table 1 for WABA (1.A), IFBA (1.b), and Homogeneous (1.C) design options. Additional details may be found in previous progress reports; designations of the design options shown as the case #, are kept consistent with the Task 3 progress report.

In addition to cases with practical BP loading for each design option and selected from the previous Task 3, we also calculated the same set of reactivity coefficients for cases with BP loading reduced roughly by 20%. This is in order estimate the sensitivity of the reactivity coefficients to the BP concentrations and by that to evaluate the potential of each BP material and design to improve these coefficients.

In Tables 1.A – 1.C, the cases with the reduced BP loading are designated as "x.1", where x denotes the case number consistent with the Task 3 designations.

Table 1.A: WABA cases

Case #	Case designation	Inner / outer radii of BP ring (cm)	BP material in BP region (vol./o)	Number of BP rods per assembly	Total weight of BP (kg/assembly)
1	No BP	-	-	0	0.00
13	WABA-Gd-9	0.33379 / 0.46895	100	24	21.08
13.1	WABA-Gd-9.1	0.33379 / 0.46895	80	24	16.86
16	WABA-Hf-3	0.33379 / 0.46895	100	24	27.24
16.1	WABA-Hf-3.1	0.33379 / 0.46895	80	24	21.79
19	WABA-Er-3	0.33379 / 0.46895	100	24	24.32
19.1	WABA-Er-3.1	0.33379 / 0.46895	80	24	19.46

Table 1.B: IFBA cases

Case #	Case designation	IFBA coating thickness (mm)	BP material in BP region (vol./o)	Number of BP rods per assembly	Total weight of BP (kg/assembly)
25	IFBA-Gd-4	0.0160	100	264	2.78
25.1	IFBA-Gd-4.1	0.0160	80	264	2.22
26	IFBA-Hf-1	0.0160	100	264	3.24
26.1	IFBA-Hf-1.1	0.0160	80	264	2.59
27	IFBA-Er-1	0.0160	100	264	3.63
27.1	IFBA-Er-1.1	0.0160	80	264	2.90

Table 1.C: Homogeneous cases

Case #	Case designation	IFBA coating thickness (mm)	BP material in BP region (vol./o)	Number of BP rods per assembly	Total weight of BP (kg/assembly)
30	HOMO-Gd-3	-	2.0	264	7.17
30.1	HOMO-Gd-3.1	-	1.6	264	5.74
37	HOMO-Hf-1	-	1.0	264	9.26
37.1	HOMO-Hf-1.1	-	0.8	264	7.41
46	HOMO-Er-1	-	2.0	264	4.13
46.1	HOMO-Er-1.1	-	1.6	264	3.30

III. Methodology

As in the previous analyses, BOXER computer code was used in this task. Also, all the neutronic calculations were performed for a single fuel assembly of a typical PWR (17x17 pins) geometry with reflective boundary conditions (infinite medium).

The MTC, DC, and BW were calculated at three time points: Beginning of Life (BOL - 1 EFPD), Middle of Life (MOL - 700 EFPD), and End of Life (EOL - 1400 EFPD). The soluble boron concentration was taken to be equal to 2000 ppm, 1000 ppm, and 0 ppm at BOL, MOL and EOL respectively in order to approximate conditions close to realistic core.

All reactivity coefficients were calculated at Hot-Full-Power (HFP), Xe-equilibrium, All-Rods-Out (ARO) operating conditions.

In addition, standard UO_2, 4.21% enriched, fuel reactivity coefficients were evaluated for the comparison purposes. It is also important to note that the values of reactivity coefficients obtained from the assembly level calculations cannot provide a reliable estimate of the real core with finite dimensions and multiple fuel types. Therefore, the assembly calculation results should be used only for the comparison of different burnable poison designs against the reference UO_2 fuel evaluated on the same basis.

Moderator temperature coefficient

The MTC relates a change in reactivity to a change in reactor coolant temperature. It is defined as the change in reactivity per degree change in moderator temperature and calculated as:

$$\frac{\Delta\rho}{1^{\circ}C} = \frac{k_{\inf}(T_2) - k_{\inf}(T_1)}{k_{\inf}(T_1) \times k_{\inf}(T_2) \times (T_2 - T_1)} \times 10^5 \left[\frac{pcm}{^{\circ}C}\right]$$

where T_1 and T_2 are two moderator temperature values, k_{inf} (T_1) and k_{inf} (T_2) are corresponding criticality values, while MTC values are attributed to the middle of the corresponding (T_1 - T_2) range and is measured in terms of pcm per 1°C. In these calculations, T_1=307.5 °C, and T_2=312.5 °C were used.

Fuel temperature coefficient due to Doppler Effect

The Doppler coefficient (DC) is defined as the change in reactivity per degree change in effective fuel temperature due to the Doppler resonance broadening and calculated as:

$$\frac{\Delta\rho}{1^{\circ}C} = \frac{k_{\inf}(T_2) - k_{\inf}(T_1)}{k_{\inf}(T_1) \times k_{\inf}(T_2) \times (T_2 - T_1)} \times 10^5 \left[\frac{pcm}{^{\circ}C}\right]$$

where T_1 and T_2 are two fuel temperatures, while k_{inf} (T_1) and k_{inf} (T_2) are corresponding criticality values. The Doppler coefficient is measured in terms of pcm per 1°C. Here, we used T_1=605.0 °C, and T_2=645.0 °C.

Boron reactivity worth coefficient

The boron worth (BW) coefficient is defined as the change in reactivity per one ppm change in the soluble boron concentration and calculated as:

$$\frac{\Delta\rho}{1ppm} = \frac{k_{\inf}(B_2) - k_{\inf}(B_1)}{k_{\inf}(B_1) \times k_{\inf}(B_2) \times (B_2 - B_1)} \times 10^5 \left[\frac{pcm}{ppm}\right]$$

where B_1 and B_2 are two boron concentrations, k_{inf} (B_1) and k_{inf} (B_2) are corresponding criticality values, and is measured in terms of pcm per 1ppm. In this case, we used the reference boron concentration ±50 ppm for the values of B_1 and B_2 in each time point.

IV. Results of calculations

The results of the reactivity coefficients calculations for all considered cases are presented in Tables 2 through 4.

Moderator temperature coefficient

Moderator temperature coefficients are reported in Table 2. For all calculated burnable poison designs the MTC remains negative throughout the fuel lifetime with exception of Gd in IFBA and Homogeneous designs. In these Gd BP cases, the MTC at BOL is very strongly positive (on the order of +100 pcm/°C). This is due to the fact that in contrast to Er and Hf burnable poisons, Gd is a strong thermal absorber rather than resonance neutrons absorber. Therefore, in case of spectrum hardening due to the lower water density, absorption in Gd decreases resulting in an increase in reactivity.
In the case of reference "All-U" fuel the MTC is also positive at BOL (although only slightly: +11 pcm/°C). However, this is due to the fact that the assumed soluble boron concentration at BOL is relatively high – 2000 ppm.

All-Uranium fuel exhibits somewhat stronger dependence of MTC on the concentration of soluble boron than investigated Pu-FF fuel with Gd, as illustrated by Figure 1. As can be observed, the All-U fuel MTC becomes negative at SB concentration in the coolant below 1500 ppm. Whereas, MTC at BOL for the Pu-FFF-Gd fuel remains positive for the whole range of the SB concentration.

Additional illustration of the fact that Gd is largely responsible for the positive MTC in FFF is presented in Figure 2. The Figure shows the dependence of MTC at BOL on Gd_2O_3 loading in Pu-FFF at 2000 ppm of soluble boron in the coolant. The MTC is slightly negative only for "No BP" case. So that even small addition of Gd results in a positive MTC. The effect of MTC variation with Gd loading calculated with BOXER code was verified by performing the Monte-Carlo simulations with MCNP-4C code [1]. The MTC values obtained with the two codes agree within the statistical error of Monte Carlo calculations.
In WABA cases, the MTC at BOL is negative for all BP materials including Gd. This is due to the fact that the fuel, in this case, does not compete directly with BP for

neutron absorption as a result of the spatial separation of the fuel and BP. In fact, Gd-WABA case exhibits the most negative MTC at BOL among all the calculated cases. At the MOL point, the MTC is negative for all considered BP materials and designs. In addition, all cases show the same general trend of becoming more negative with fuel burnup. This indicates that in a real core consisting of a mixture of fresh and partially burned fuel assemblies, the core average MTC may still be negative at BOL even for the Gd-IFBA and Gd-HOMO designs. Such an assumption can be reliably verified only by performing a full core 3-dimensional analysis. The Er and Hf cases in IFBA and Homogeneous geometries have MTC values very similar to those of the reference UO_2 fuel case. Although, Er has slightly higher potential of improving the MTC than Hf.

Table 2: MTC (pcm/°C)

Case No.	Case Designation	BOL	MOL	EOL
Ref.	UO_2, e=4.21%	11.2	-23.3	-59.9
1	No BP	0.61	-14.03	-27.0
13	WABA-Gd-9	-20.7	-33.76	-39.6
13.1	WABA-Gd-9.1	-16.5	-27.8	-38.0
16	WABA-Hf-3	-17.3	-35.6	-60.8
16.1	WABA-Hf-3.1	-15.2	-32.6	-54.9
19	WABA-Er-3	-12.6	-30.6	-53.2
19.1	WABA-Er-3.1	-11.0	-28.6	-47.4
25	IFBA-Gd-4	96.9	-16.3	-32.6
25.1	IFBA-Gd-4.1	86.7	-16.3	-32.2
26	IFBA-Hf-1	-9.7	-24.0	-40.0
26.1	IFBA-Hf-1.1	-8.2	-22.4	-38.2
27	IFBA-Er-1	-18.8	-32.6	-43.8
27.1	IFBA-Er-1.1	-16.2	-30.2	-40.9
30	HOMO-Gd-3	120.7	-16.7	-32.0
30.1	HOMO-Gd-3.1	111.8	-16.7	-31.8
37	HOMO-Hf-1	-7.0	-19.2	-34.9
37.1	HOMO-Hf-1.1	-5.8	-18.6	-34.3
46	HOMO-Er-1	-13.2	-24.1	-37.8
46.1	HOMO-Er-1.1	-11.3	-22.5	-36.3

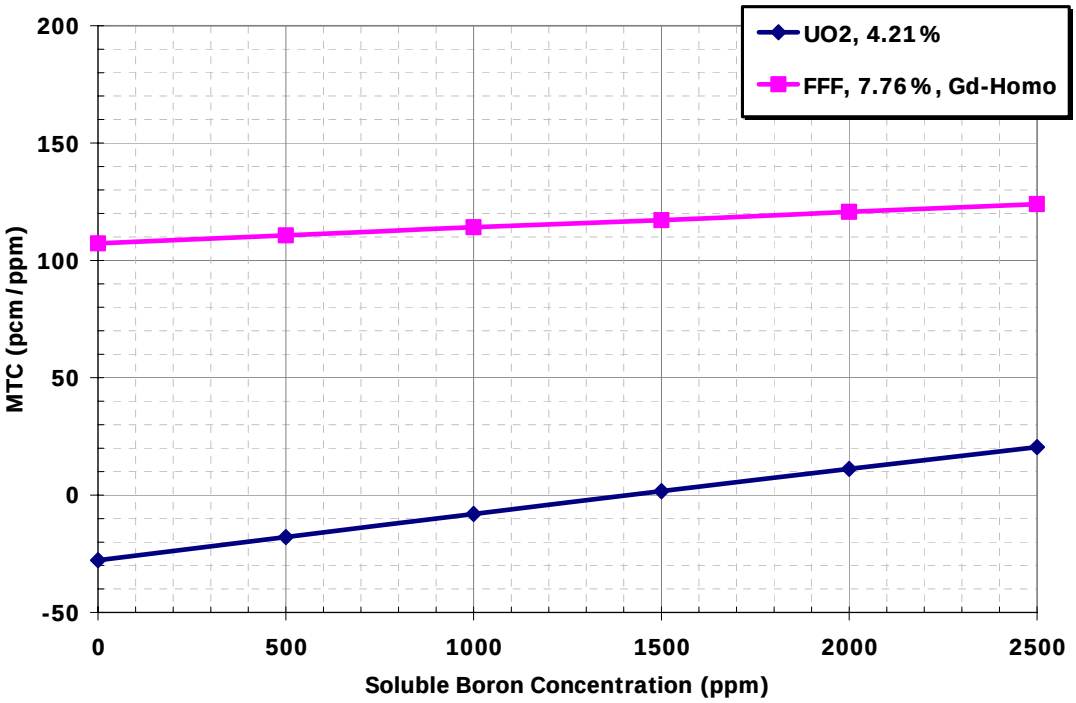

Figure 1. Effect of SB concentration on MTC

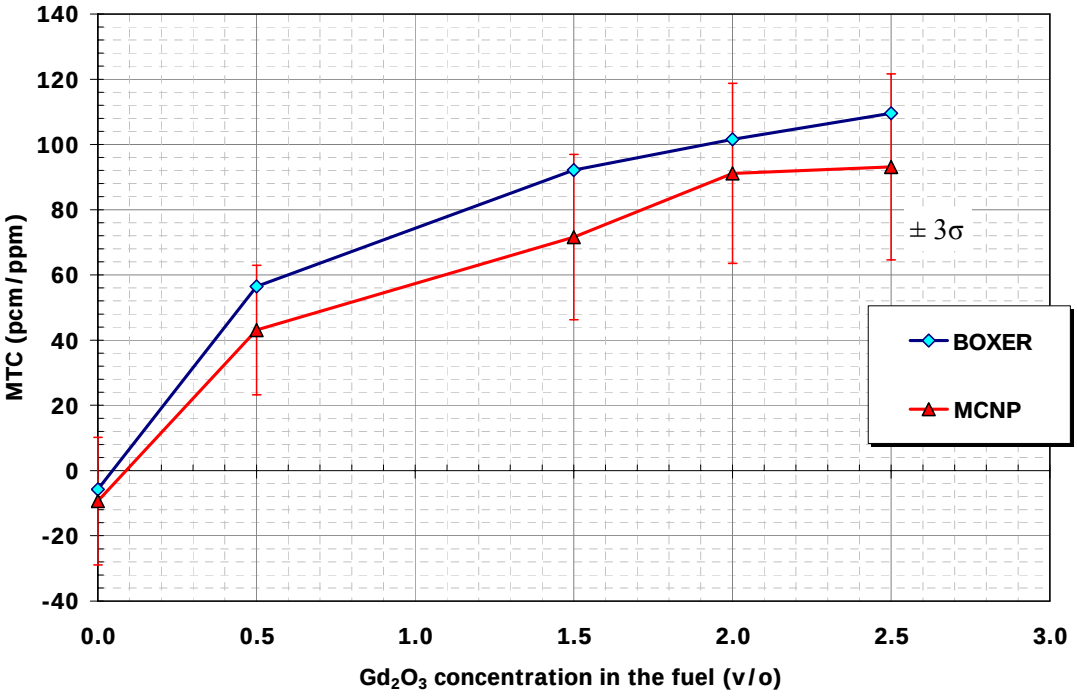

Figure 2. Effect of Gd loading on MTC: Boxer vs. MCNP comparison

Doppler Coefficient

Degradation of Doppler Coefficient in FFF in comparison with conventional UO_2 fuel is one of the major reactor safety related concerns. With no BP, the DC is less negative for FFF than for UO_2 by a factor of two (Table 3). At normal reactor operation, no specific requirements are imposed on the magnitude of DC except for the requirement that it should be negative at all times. However, the magnitude of DC has significant impact on the reactor safety in various accident scenarios and particularly important in rapid Reactivity Initiated Accidents (RIA). This is because DC is the only prompt reactivity feedback preventing the power runaway. For example, in PWR control rod ejection accident, the total energy deposition in the fuel can be roughly approximated by the adiabatic Fuchs-Nordheim Model [2] as:

$$E = \frac{2\,\rho_{fuel} C_p\,(\rho_0 - \beta)}{\alpha_{DC}} \qquad (1)$$

where $\rho_{fuel}\ C_p$ is the volumetric fuel heat capacity [J/cm^3-K], ρ_0 is the initial reactivity inserted (by ejected rod), β is the effective delayed neutron fraction, and α_{DC} is the Doppler coefficient.

The total energy deposition in the fuel provides a measure of the fuel performance during the accident. The NRC specifies the value of 280 cal/g at any axial location in any fuel pin for UO_2 fuel as a threshold value above which fuel damage and release of FP into the coolant is expected. [3]

The relation (1) clearly shows that the energy deposition is inversely proportional to the magnitude of the Doppler Coefficient. However, it should also be noted that the energy deposition depends on a combination of parameters which differ considerably for the conventional UO_2 and FFF cores. The smaller α_{DC} and β_{eff} for the FFF loaded core should make a negative contribution to the fuel performance in reactivity initiated accidents increasing the energy deposition in the fuel. However, lower, due to the harder spectrum, ejected control rod reactivity worth (ρ_0) of the FFF will compensate for the negative effects of the smaller α_{DC} and β_{eff} to some extent.

To summarize, the acceptable, with regards to safety requirements, value of DC for FFF core depends on a combination of the core neutronic characteristics and material properties. The assessment of such safety criteria is beyond the scope of the current research task but must be addressed in the future. In order to perform such an assessment the following will be required:

- Data on thermal properties of FFF matrix materials (e.g. thermal conductivity and specific heat as a function of temperature)
- Detailed reactor dynamic simulation of the RIA accidents with thermal feedbacks in order to establish the core locations where the maximum energy deposition would occur.
- Data on FFF performance under accidents conditions in order to establish a new correlation between the fuel failure and the total energy deposition. The US NRC 280 cal/g of heavy metal in UO_2 cannot obviously be used for FFF because neither FF matrix materials performance nor fuel failure mechanisms are not very well understood yet.

Although the DC values for FFF calculated in this task cannot be directly compared with UO_2 fuel values, it is clear from the above discussion that the large negative value of DC is beneficial for the reactor safety.

The DC values calculated for all BP designs and materials considered in this task are presented in Table 3. As can be observed from the Table, the most effective BP design is Er in IFBA geometry. Such a configuration, allows an increase in absolute value of FFF DC from -1pcm/°C for the No BP case to -1.6 pcm/°C for the IFBA-Er case. The effect can be explained by the fact that Er-167 is a strong resonance absorber (IR≈3000b [4]) with its first absorption resonance overlapping with fission resonance of Pu-239 (Figure 3), so that the DC is enhanced due to the mutual shielding of Pu and Er resonances. The IFBA geometry also improves the DC because most of the resonance absorption in the fuel occurs at the outer rim of the fuel pellet. Therefore, using Er as a fuel pellet coating increases the resonance neutron absorption in it relative to the resonance absorption in the fuel.

Other considered burnable poisons are much less effective and have almost no effect on the Doppler Coefficient.

It is also worth noting that similar to MTC, DC also becomes more negative with the fuel burnup, so that the lowest DC value is always observed at BOL. Therefore, the BOC core average DC values (Uniform and Distributed DC) should be more negative than the BOL values presented in Table 3 since the core contains a mixture of assemblies with different burnups.

Table 3: DC (pcm/°C)

Case No.	Case Designation	BOL	MOL	EOL
Ref.	UO_2, e=4.21%	-2.0	-2.9	-3.4
1	No BP	-1.0	-1.1	-1.5
13	WABA-Gd-9	-1.0	-1.4	-1.6
13.1	WABA-Gd-9.1	-1.1	-1.3	-1.7
16	WABA-Hf-3	-1.0	-1.2	-1.8
16.1	WABA-Hf-3.1	-1.0	-1.2	-1.8
19	WABA-Er-3	-1.1	-1.3	-1.8
19.1	WABA-Er-3.1	-1.1	-1.3	-1.8
25	IFBA-Gd-4	-0.9	-1.1	-1.5
25.1	IFBA-Gd-4.1	-0.9	-1.1	-1.5
26	IFBA-Hf-1	-1.2	-1.3	-1.6
26.1	IFBA-Hf-1.1	-1.2	-1.3	-1.6
27	IFBA-Er-1	-1.6	-1.6	-2.0
27.1	IFBA-Er-1.1	-1.5	-1.5	-1.9
30	HOMO-Gd-3	-1.0	-1.2	-1.5
30.1	HOMO-Gd-3.1	-1.0	-1.2	-1.5
37	HOMO-Hf-1	-1.1	-1.1	-1.5
37.1	HOMO-Hf-1.1	-1.1	-1.1	-1.5
46	HOMO-Er-1	-1.3	-1.2	-1.5
46.1	HOMO-Er-1.1	-1.2	-1.2	-1.5

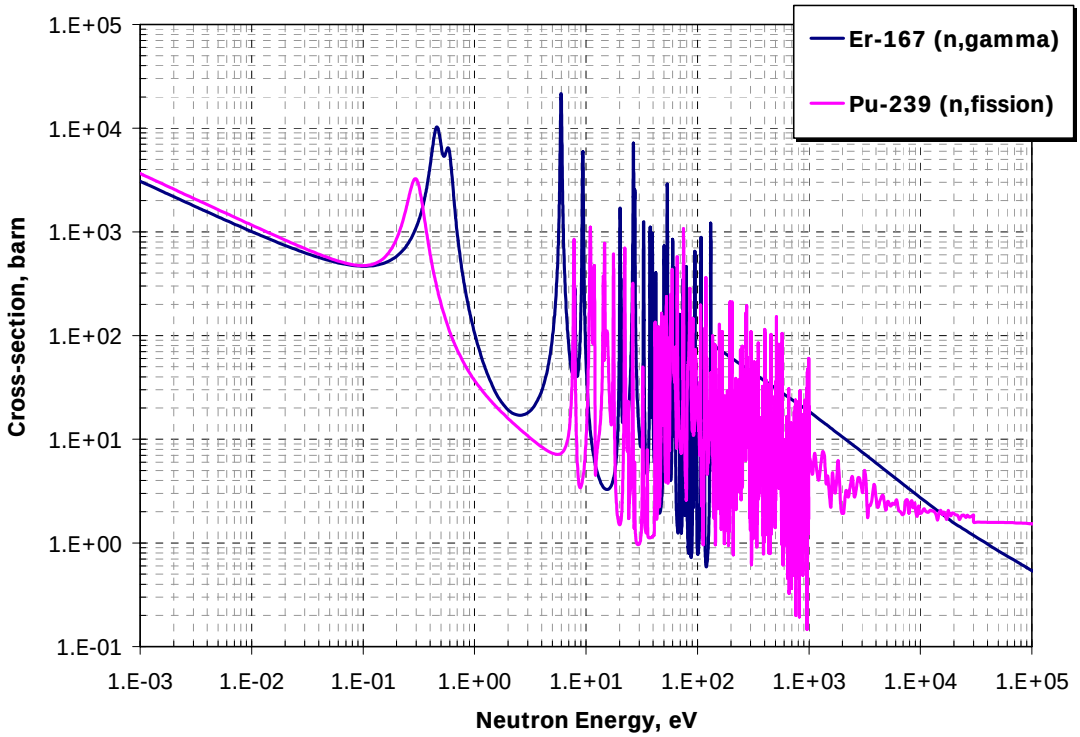

Figure 3. Er-167 (n,γ) and Pu-239 (n,f) Microscopic Cross-sections (JEF-2.2) [4].

Soluble Boron Reactivity Worth

The results of the soluble boron reactivity worth coefficients (BW) are summarized in Table 4.

As already noted, Pu containing fuels have factor of 2 to 3 lower BW than typically observed in the conventional All-U cores due to the fact that Pu is much stronger thermal neutrons absorber than Uranium. Therefore, the BW increases with the depletion of fissile material and corresponding "softening" of the spectrum. This effect is also much stronger in FFF-Pu than in All-U fuel. While the BW of UO_2 fuel increases almost linearly with burnup due the the buildup of Pu, the FFF exhibits a sharp increase in BW at EOL when most of the fissile Pu is depleted. This effect may cause a power peaking problem in the FFF-Pu core.

In general, BP material and geometrical arrangement have limited effect on the BW of FFF-Pu fuel. Hf and Er tend to improve the BW slightly (from -2.4pcm/ppm in NoBP case to about -2.5pcm/ppm in Er and Hf cases in IFBA and Homogeneous

geometries), while Gd, generally, reduces it. This is due to the competition between Gd and Boron for thermal neutron absorption as both of these materials are mostly thermal neutron absorbers.

Table 4: BW (pcm/ppm)

Case No.	Case Designation	BOC	MOC	EOC
Ref.	All U	-5.8	-7.0	-9.5
1	No BP	-2.4	-4.0	-15.3
13	WABA-Gd-9	-2.2	-3.5	-12.9
13.1	WABA-Gd-9.1	-2.2	-3.5	-13.3
16	WABA-Hf-3	-2.2	-3.8	-13.8
16.1	WABA-Hf-3.1	-2.2	-3.8	-13.9
19	WABA-Er-3	-2.2	-3.6	-13.3
19.1	WABA-Er-3.1	-2.2	-3.6	-13.5
25	IFBA-Gd-4	-1.8	-4.0	-14.9
25.1	IFBA-Gd-4.1	-1.9	-4.0	-14.9
26	IFBA-Hf-1	-2.5	-4.2	-15.4
26.1	IFBA-Hf-1.1	-2.5	-4.2	-15.3
27	IFBA-Er-1	-2.5	-4.1	-14.9
27.1	IFBA-Er-1.1	-2.5	-4.1	-14.9
30	HOMO-Gd-3	-1.8	-4.0	-15.5
30.1	HOMO-Gd-3.1	-1.8	-4.0	-15.5
37	HOMO-Hf-1	-2.6	-4.4	-19.0
37.1	HOMO-Hf-1.1	-2.6	-4.4	-18.8
46	HOMO-Er-1	-2.6	-4.1	-14.8
46.1	HOMO-Er-1.1	-2.6	-4.1	-14.9

V. Summary and Conclusions

In this task, reactivity feedback coefficients of fertile free Pu containing fuels were evaluated and compared with those of conventional UO_2 fuel.

The objective of this task was to investigate the potential of different burnable poison materials and geometrical arrangements to improve the reactivity coefficients of fertile free fuels. The main design challenges of FFF include:

- Positive Moderator Temperature Coefficient (MTC)
- Significantly reduced Doppler Coefficient (DC)
- Significantly reduced Soluble Boron Worth (BW)

The calculations were performed with BOXER computer code on the fuel assembly level for the fuel composition corresponding to 18 months fuel cycle length (determined and reported in Task 2 of the current project).

Two reservations must be made regarding applicability of the results presented in this report.

1. All reactivity coefficients have large sensitivity to the soluble boron concentration in the coolant. The calculations in this task were performed by "guessing" soluble boron concentration to approximate the real conditions. The boron concentration in the actual core may be different. Therefore, values obtained in this task may serve only as a guideline for comparing different fuel options on the consistent basis.
2. The used computation methods themselves may introduce significant uncertainties in evaluation of FFF reactivity coefficients as concluded from the series of computational benchmarks for various Fertile Free Fuel unit cells reported in Reference 5. Although, the most important findings in the current research task were verified by Monte Carlo neutronic simulations and found to be in good agreement with BOXER results.

The results of calculations, performed in this task, can be summarized as follows:

MTC:

1. FFF for a No BP case shows small and positive MTC at BOL.
2. WABA-Gd shows a negative MTC of a reasonable value close to a standard LWR core.
3. IFBA-Gd and HOMO-Gd cases show unacceptably large and positive MTC values.
4. Hf and Er BP materials show a potential to improve MTC, where all Er designs seem more efficient in "correcting" the MTC value.
5. For all BP materials and geometries simultaneous burnout of Pu and BP results in acceptable MTC values at MOL and EOL time-points.

DC:

1. As expected, the DC of Pu loaded FFF is reduced to ~1.0 pcm/°C as compared with ~2.0pcm/°C for conventional All-U fuel.
2. No dramatic influence of BP on DC is found, with exception of a modest improvement for Er cases (up to -1.6 pcm/°C)
3. DC value is becoming more negative with burnup reaching -1.5 – 2.0 at EOL.

BW:

1. The well-known effect of BW reduction was also observed. In comparison with the reference All-U fuel, the BW is reduced by approximately a factor of 2 to 3 due to the presence of Pu.
2. Hf and Er in Homogeneous and IFBA configurations show modest potential of increasing the BW. Otherwise, addition of Gd burnable poison slightly reduces the BW.
3. The effect of increasing BW towards the fuel EOL as a result of fissile isotopes depletion was found to be much more pronounced in FFF than in All-U fuel, which may potentially cause power peaking problem in FFF core.

In conclusion, Pu loaded FFF showed potential feasibility to be used in existing PWRs. All FFF problems may be significantly mitigated through the correct choice of BP material and configuration. Based on the performed analysis, it was found that a combination of BP materials and geometries may be required to meet all FFF design goals. The use of enriched (in most effective isotope) burnable poisons, such as Er-167 and Gd-157, will further improve the BP effectiveness and reduce the fuel cycle length penalty associated with their use. However, these findings can be confirmed only by performing a full core 3-dimensional neutronic analysis.

The final result of the next stage of this research will be the choice of acceptable FFF core fraction with appropriate mix of BP designs. This result will be obtained and verified by a full core 3-dimansional simulation and fuel cycle analysis.

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