

10-2004

# Dissolution, Reactor, and Environmental Behavior of ZrO<sub>2</sub>-MgO Inert Fuel Matrix: Neutronic Evaluation of ZrO<sub>2</sub>-MgO Inert Fuels

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**Dissolution, Reactor, and Environmental Behavior of ZrO<sub>2</sub>-  
MgO Inert Fuel Matrix**

**Neutronic Evaluation of MgO-ZrO<sub>2</sub> Inert Fuels**

Progress Report

Prepared by Reactor Analysis Group

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October 2004

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# Task 2: Determination of Pu Loading

## I. Introduction

Second task of the BGU part of “Dissolution, Reactor, and Environmental Behavior of ZrO<sub>2</sub>-MgO Inert Fuel Matrix” project aims at evaluation of the fertile free fuel matrix composition effect on the fuel reactivity and corresponding reactivity limited burnup. Fertile free fuel with different MgO to ZrO<sub>2</sub> ratio in the matrix will require different PuO<sub>2</sub> loading in order to assure certain fuel cycle length. This is due to the fact that absorption cross section of Zr is slightly higher than that of Mg, although absorption in both of these elements is small compared to Pu. Therefore, the resulting effect on criticality is marginal as pointed out in the Progress Report on Task 1 of the current project [1].

This progress report summarizes results of the calculations performed on Task 2 of the BGU program. The scope of current task includes two objectives:

- Determination of Pu loading necessary to achieve industry standard fuel cycle lengths of 12, 18, and 24 calendar months using the reference fuel matrix composition with 1:1 volume ratio of MgO and ZrO<sub>2</sub> components.
- Quantitative evaluation of the matrix composition effect on Pu loading required to maintain mentioned reference fuel cycle lengths.

## II. Analysis Methodology

All calculations were performed with BOXER[2] computer code in typical PWR fuel assembly geometry and operating conditions. The fuel assembly geometry and operating conditions assumed in this analysis are identical to those used for the benchmarking of BOXER code reported in Task 1 Progress Report [1].

The calculations were performed for the fuel assembly with reflective boundary conditions; that is in infinite medium.

Two different Pu isotopic compositions were considered in order to capture the effect of using Pu from old Light Water Reactors spent fuel with low burnup and long decay time versus Pu from advanced LWR spent fuel with high burnup and relatively short time after discharge. Pu239 fraction in low burnup fuel is higher but long cooling period reduces the fraction of fissile Pu241, which decays to Am241 with half-life of about 14 years. Therefore, the effects of these two phenomena on the fuel reactivity are expected to be mutually compensating to some extent. The two Pu vectors considered and basic assumptions used to obtain these vectors are summarized in Table 1.

**Table 1. Pu Isotopic Vector and Calculation Assumptions Summary**

<i>Pu Vector Composition</i>	PWR-50	PWR-33
UO <sub>2</sub> Initial Enrichment, %	4.2	3.2
Discharge fuel Burnup, MWd/kg	50.0	33.0
Decay time after discharge, Years	10	25
Pu-238, wt. %	3.18	1.35
Pu-239, wt. %	56.35	62.56
Pu-240, wt. %	26.62	26.53
Pu-241, wt. %	8.02	4.30
Pu-242, wt. %	5.83	5.25

The discharge fuel burnup and fuel cycle length were obtained by applying Linear Reactivity Model (LRM) [3] to the results of 2-dimensional fuel assembly burnup calculations.

The basic assumptions of LRM are

- Equal power share between different fuel batches within the core
- Linear dependence of fuel reactivity on burnup

The former assumption is not necessarily true in most realistic cases. However, the effect of unequal power share on discharge burnup estimation is typically small [3].

The latter assumption holds approximately for conventional UO<sub>2</sub> fuel. In the case of Pu – fertile free fuel, however, the dependence of batch reactivity on burnup is clearly non linear, as illustrated by Figure 1. Therefore, the LRM estimation of the fuel discharge burnup in 3-batch core simply as 1.5×BU<sub>1</sub> introduces significant uncertainty into calculations. Here, BU<sub>1</sub> is the burnup of single batch core at which the corrected for leakage core reactivity becomes zero.

In order to eliminate such an uncertainty in calculation of discharge fuel burnup by straight forward LRM, we fit the calculated reactivity versus burnup data for Pu fertile free fuel to 3<sup>rd</sup> order polynomial function using Least Square Fit algorithm.

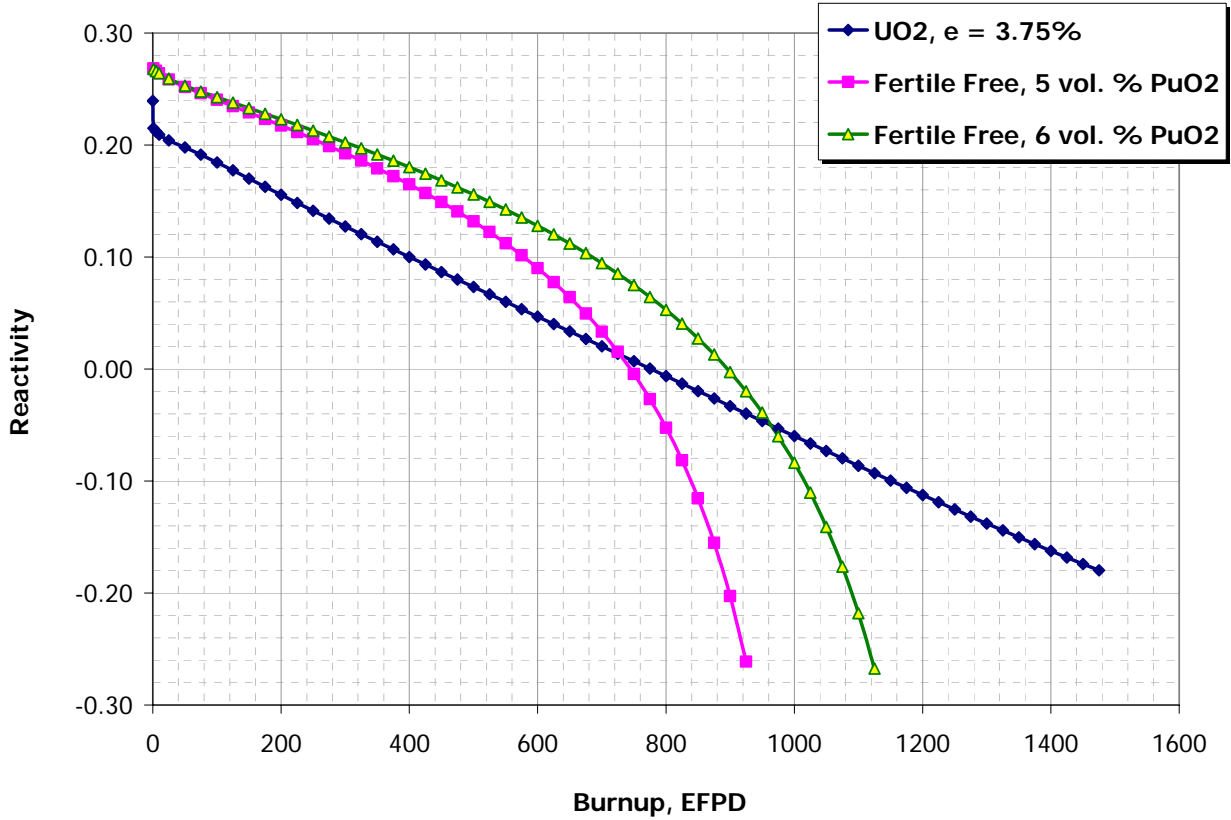
$$\rho(BU) = A_0 + A_1 \times BU + A_2 \times BU^2 + A_3 \times BU^3 \quad (1)$$

Then, using the same LRM assumption of equal power sharing between all fuel batches in the core, we postulate that average core (corrected for leakage) reactivity becomes zero at the end of each cycle (EOC). Therefore, the burnup accumulated by each batch in one cycle ( $BU^{CYCLE}$ ) can be found from the following relation:

$$\rho_{EOC}^{CORE} = \frac{\rho_{EOC}^{Batch1} + \rho_{EOC}^{Batch2} + \rho_{EOC}^{Batch3}}{3} = \frac{\rho(BU^{CYCLE}) + \rho(2 \times BU^{CYCLE}) + \rho(3 \times BU^{CYCLE})}{3} = 0$$

where the discharge fuel burnup is the EOC burnup of the 3<sup>rd</sup> batch or

$$BU^{DISCHARGE} = 3 \times BU^{CYCLE}.$$



**Figure 1. Reactivity vs. burnup curves for conventional UO<sub>2</sub> and fertile free fuel.**

The neutron leakage effect was taken into account by assuming the leakage reactivity worth of 3%  $\Delta\rho$ ; namely, assuming 1.03 be the average core criticality value at the end of cycle. This leakage reactivity worth is typical for PWRs with conventional UO<sub>2</sub> fuel employing “low leakage” fuel management schemes. However, in the Pu loaded fertile free core, the neutron leakage rate is expected to be higher due to the harder neutron energy spectrum. The neutron spectrum effect on leakage reactivity is also evaluated and discussed in more details in the following section.

The number of Effective Full Power Days (EFPD) in standard 12, 18, and 24 calendar month cycles was calculated assuming 90% capacity factor and 30 days long refueling outage period.

$$\text{EFPD} = \left[ \frac{365.25}{12} \times \text{Cycle Length (Calendar Months)} - 30 \right] \times 0.90$$

The obtained, in such manner, number of EFPD in 12, 18, and 24 month cycles is 905, 1398, and 1891 days respectively.

### III. Results

#### III.1 Determination of Pu Loading for Standard Fuel Cycle Lengths

In order to determine Pu loading required to achieve established cycle lengths, we performed a series of fuel assembly burnup calculations. The PuO<sub>2</sub> content in the fuel was varied between 4 and 15 volume %. The remaining fuel volume was occupied with ZrO<sub>2</sub> – MgO mixture in 1:1 volume ratio. The results of these criticality calculations are presented in Figures 2.a and 2.b for PWR-33 and PWR-50 Pu vectors correspondingly.

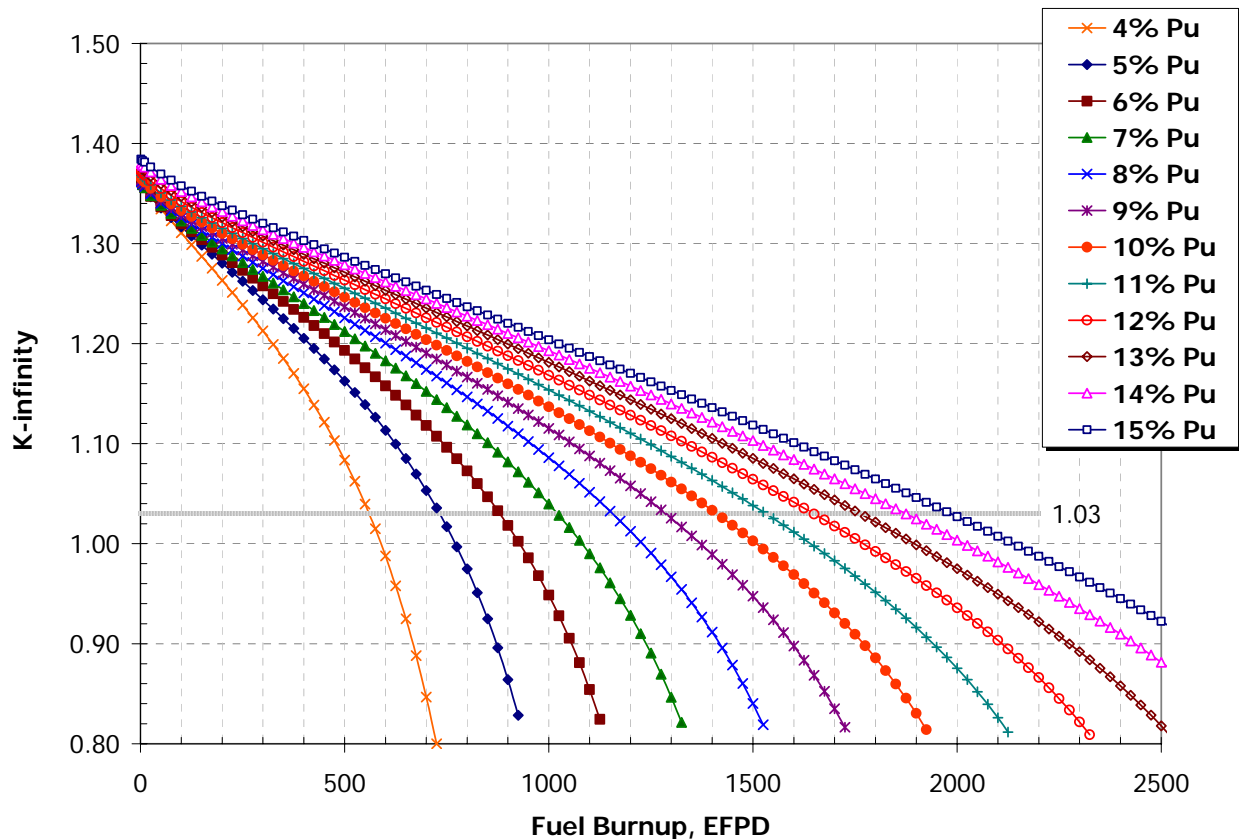
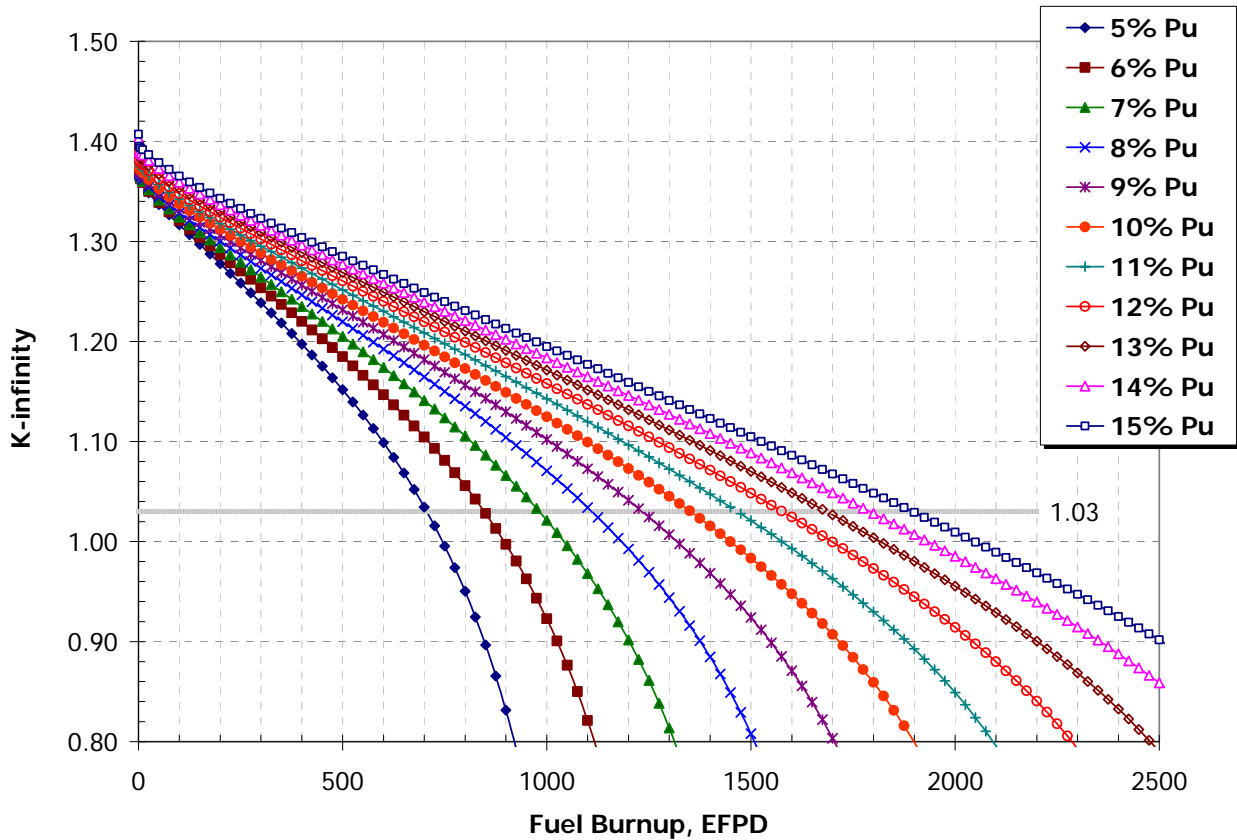


Figure 2.a. Criticality curves for PWR-33 Pu vector.



**Figure 2.b. Criticality curves for PWR-50 Pu vector.**

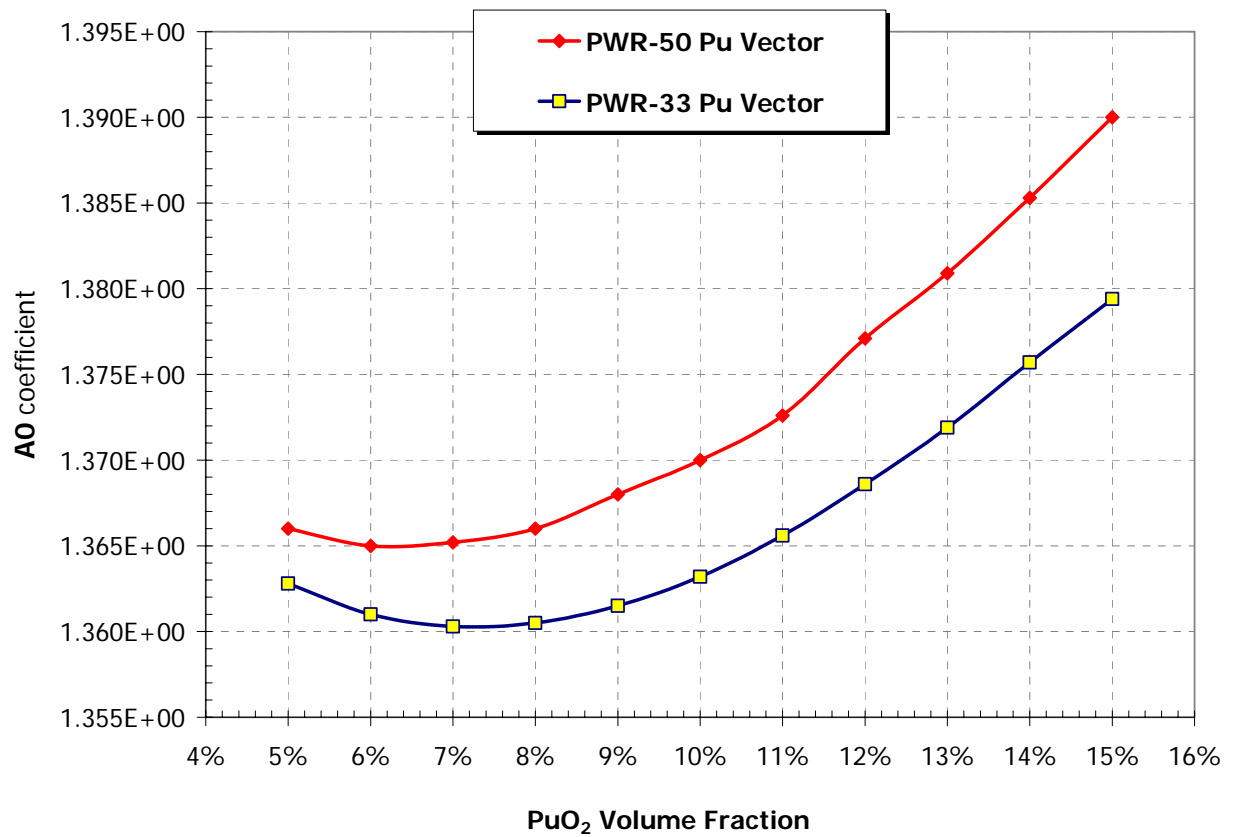
Table 2 reports reactivity limited single batch core burnup  $BU_1$  for all calculated cases assuming 3%  $\Delta\rho$  leakage reactivity. PWR-33 Pu isotopic vector always results in higher single batch burnup than that for the PWR-50 vector. The difference in  $BU_1$  between the two Pu isotopic compositions for comparable Pu loadings ranges from about 20 days for 5%  $PuO_2$  to over 90 days for 15%  $PuO_2$ . As mentioned earlier, this difference is the result of slightly higher fissile Pu fraction in PWR-33 isotopic vector.

According to the analysis methodology, the discharge fuel burnup was calculated by fitting the reactivity versus burnup data for each Pu loading case to a 3<sup>rd</sup> order polynomial function. Figures 3.a, 3.b, 3.c, and 3.d report correspondingly coefficients  $A_0$ ,  $A_1$ ,  $A_2$ , and  $A_3$  as a function of  $PuO_2$  content.

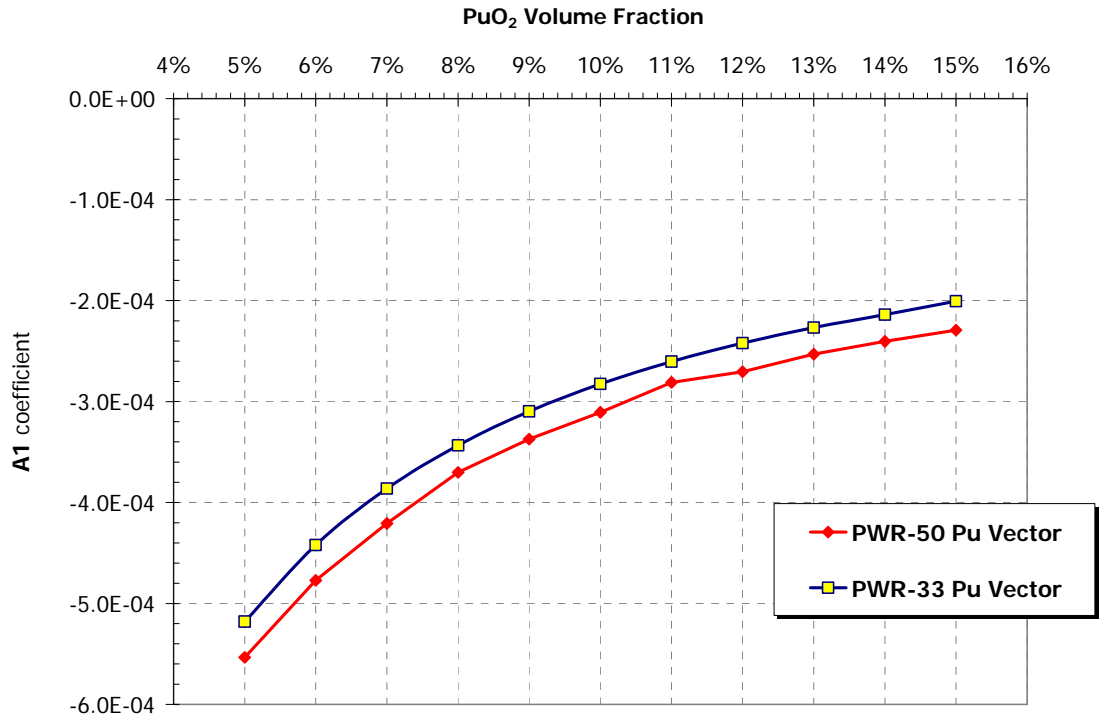


**Table 2. Single Batch Burnup (EFPD) as a Function of Pu Loading**

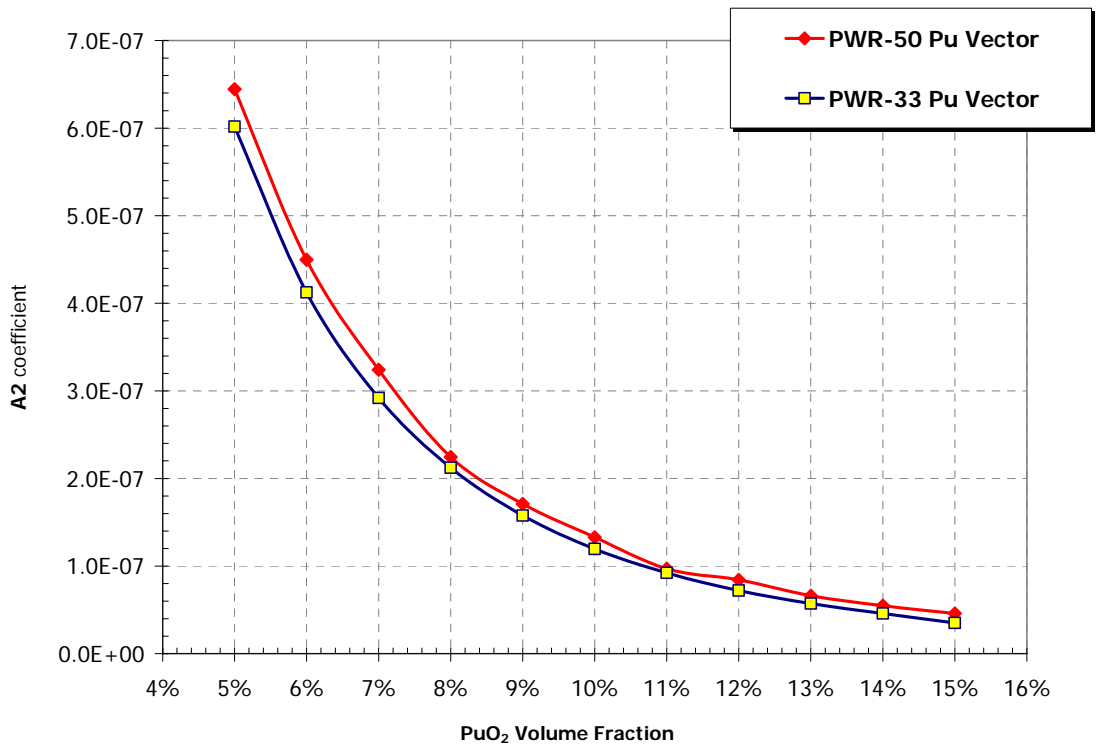
PuO <sub>2</sub> Loading, vol. %	Initial Pu Composition	
	PWR-33	PWR-50
5%	729	706
6%	876	847
7%	1017	981
8%	1154	1110
9%	1285	1234
10%	1411	1352
11%	1533	1467
12%	1652	1577
13%	1767	1685
14%	1880	1791
15%	1990	1895



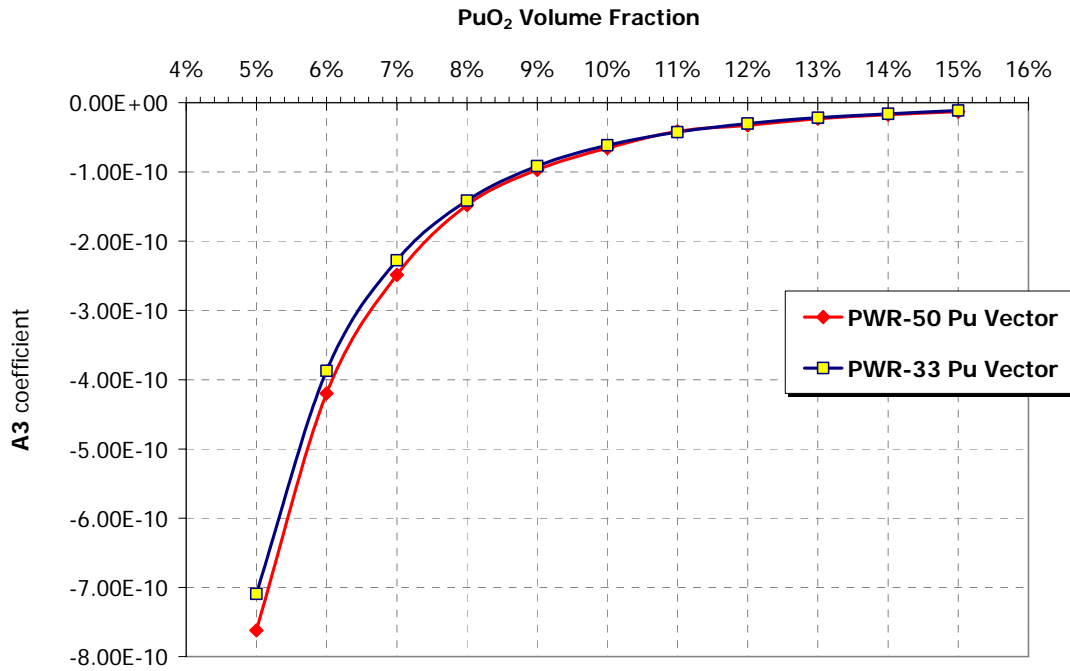
**Figure 3.a. A<sub>0</sub> Polynomial Coefficient**



**Figure 3.b. A<sub>1</sub> Polynomial Coefficient**



**Figure 3.c. A<sub>2</sub> Polynomial Coefficient**



**Figure 3.d. A<sub>3</sub> Polynomial Coefficient**

Coefficient  $A_0$  (in Eq. 1) represents the beginning of life fuel criticality. It has relatively weak dependence on Pu loading since it is primarily a function Pu  $\eta$ -factor value ( $v\Sigma_f / \Sigma_a$ ) for a given Pu isotopic vector. An increase of  $A_0$  with Pu content (7-15% PuO<sub>2</sub>) could be explained by reduction in neutron absorption in fuel matrix due to the hardening of the spectrum and reduction in matrix atoms concentration.  $A_0$  decrease with Pu content for the low Pu loadings is a result of more thermalized spectrum and corresponding change in  $\eta(\text{Pu})$ .

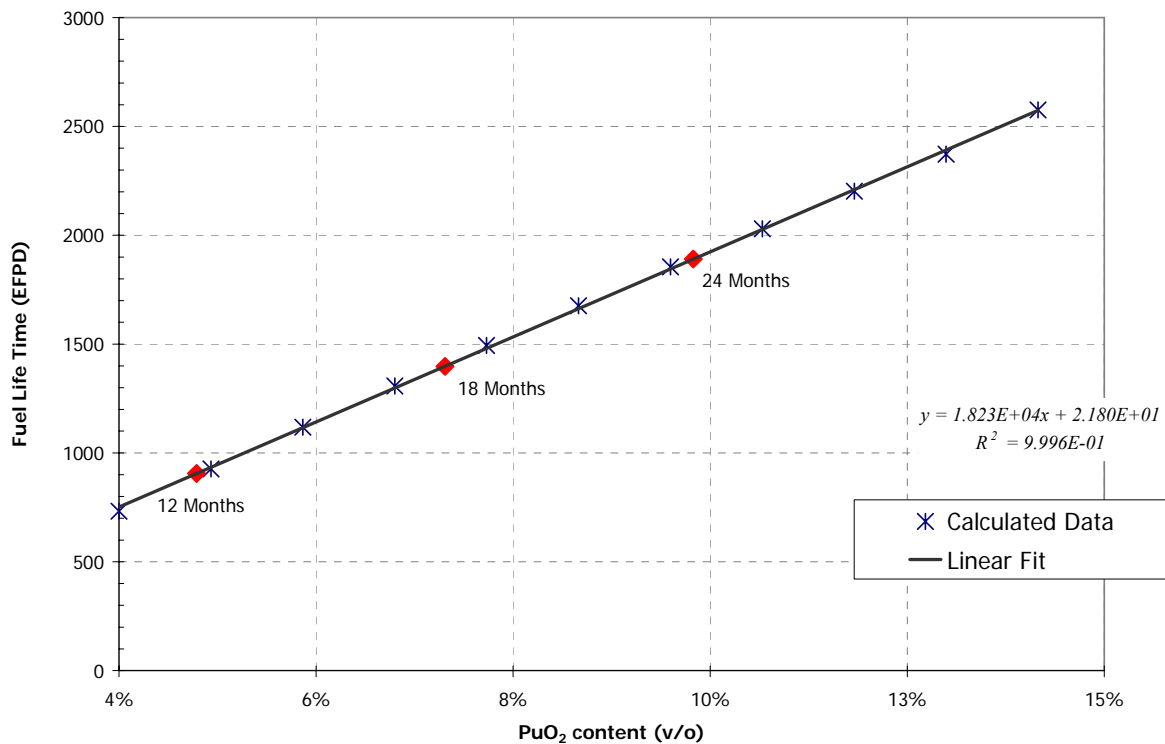
Coefficient  $A_1$  is a measure of overall slope of the criticality curves. The slope decreases monotonically with Pu content resulting in higher  $\text{BU}_1$  for higher Pu loadings.

Coefficient  $A_2$  and  $A_3$  are responsible for the curvature of the criticality vs. burnup lines. As can be observed from Figures 2.a, 2.b, the criticality lines become more linear for the higher Pu loadings within the analyzed range of cycle lengths. Therefore,  $A_2$  and  $A_3$  coefficients approach zero as Pu loading increases (Figures 3.c and 3.d).

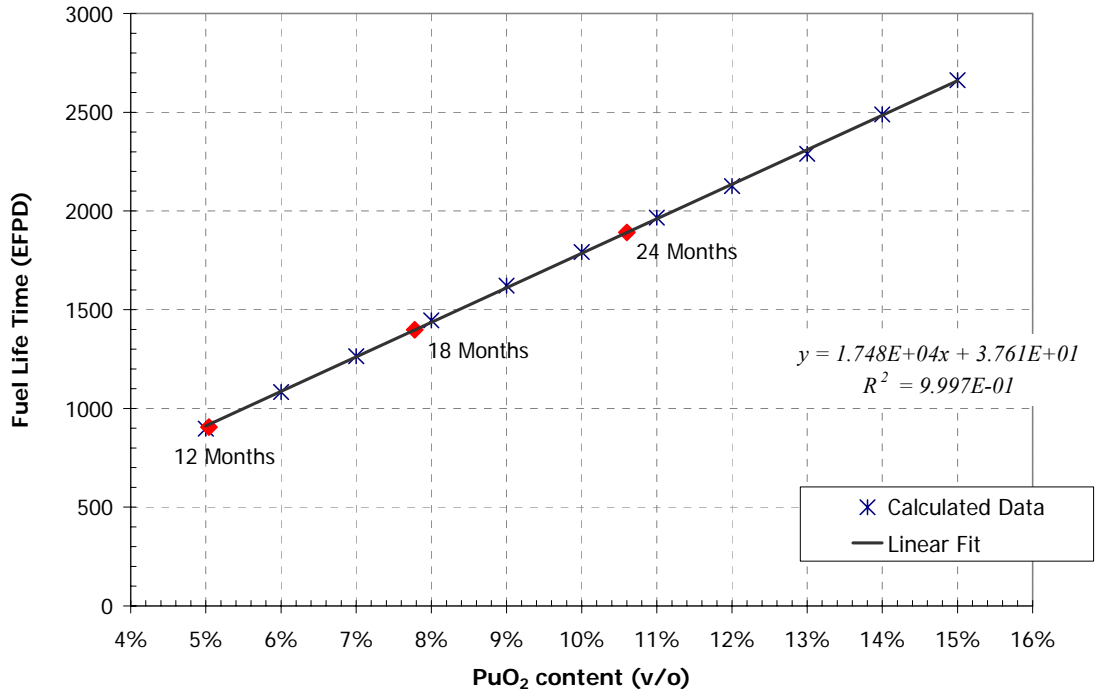
It should also be noted however, that criticality lines have almost identical shape and slope if plotted against burnup in terms of MWd/kg of initial Heavy Metal (iHM).

The calculated values of discharge burnup assuming 3-batch fuel management and 3% leakage reactivity are plotted against initial Pu loading in Figures 4.a and 4.b. The results indicate markedly linear dependence of achievable fuel cycle length and corresponding discharge burnup on the initial Pu loading for the entire range of interest.

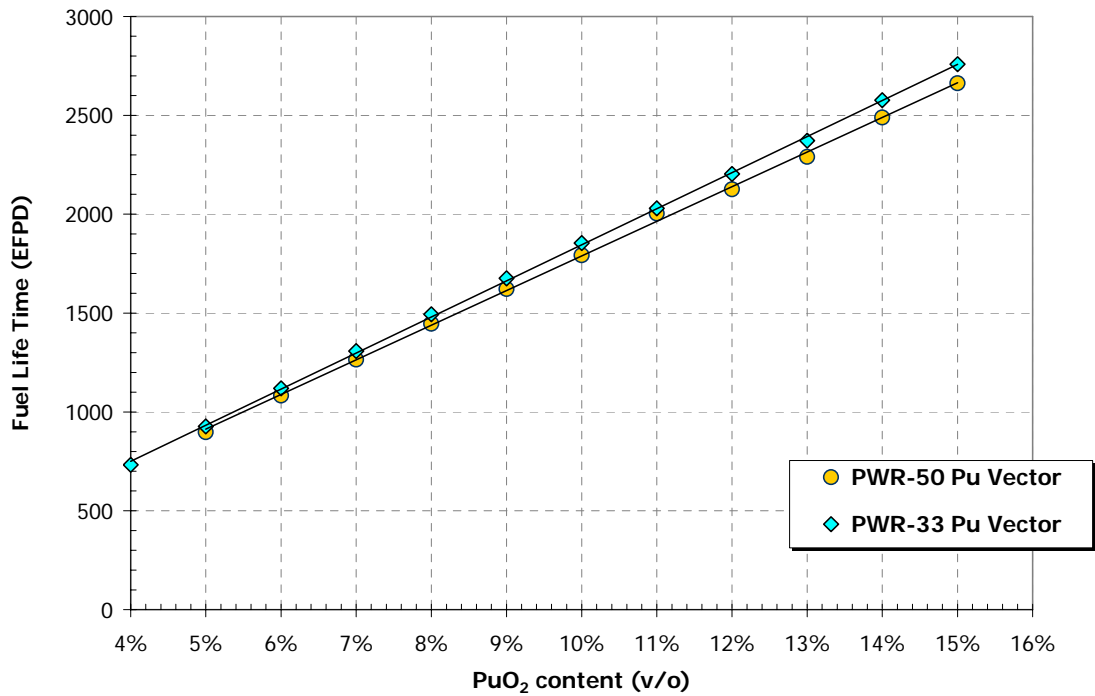
Figure 5 compares achievable discharge burnup versus Pu loading for the two different Pu vector compositions. The difference in discharge burnup is relatively small and ranges from 30 to about 60 EFPD in the fuel cycle lengths range of interest. Therefore, it can be concluded that the Pu composition has generally minor effect on fertile free fuel criticality and achievable discharge burnup. As mentioned earlier, this is partially due to the mutually canceling effects of higher Pu239 fraction but lower Pu241 fraction in PWR-33 as compared to PWR-50 grade plutonium.



**Figure 4.a. Fuel Discharge Burnup vs. Pu Loading (PWR-33 Pu Vector).**



**Figure 4.b. Fuel Discharge Burnup vs. Pu Loading (PWR-50 Pu Vector).**



**Figure 5. Comparison of Discharge Burnup vs. Pu Loading Data for PWR-33 and PWR-50 Grade Plutonium**

Table 3 shows the results of the calculation of initial Pu content required to achieve 12, 18, and 24 months fuel cycle length. These PuO<sub>2</sub> volume % values are used as reference cases for the investigation of cycle length sensitivity to fuel matrix composition.

**Table 3. Pu Loading Requirements for Standard Fuel Cycle Lengths**

Cycle Length, Calendar Months	Cycle Burnup, EFPD	Discharge Burnup, EFPD	PuO <sub>2</sub> loading, vol. % (PWR-33 Pu)	PuO <sub>2</sub> loading, vol. % (PWR-50 Pu)
12	302	905	4.84	5.04
18	466	1398	7.55	7.78
24	630	1891	10.25	10.60

### ***III.2 Linear Reactivity Model Applicability***

#### *Leakage Reactivity*

Pu loadings required to achieve the standard fuel cycle lengths were determined assuming typical PWR leakage reactivity worth value of  $\rho_{\text{Leakage}} = 0.03$ . However, fertile free Pu containing fuel is known to have somewhat harder neutron spectrum than typical UO<sub>2</sub> fuel increasing the leakage from the core.

The leakage from a finite reactor core is roughly proportional to its surface to volume ratio (S/V) and to the average neutron migration length. Assuming a flat with “drooping ends” core power shape the leakage reactivity defect is given by [3]:

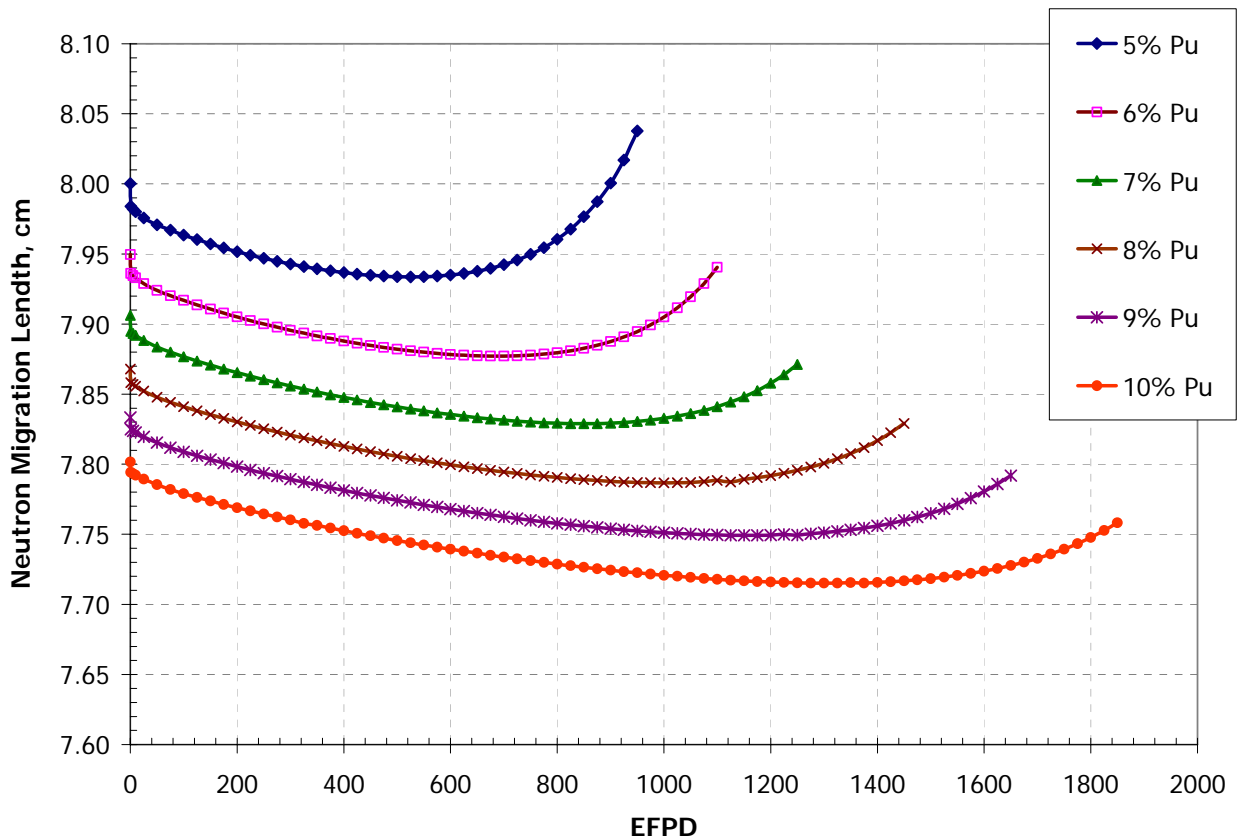
$$\rho_{\text{leakage}} = \frac{S}{V} \times \frac{M}{4}$$

Meaning that all neutrons, less than ¼ of a migration length away from the system boundary, will leak out. For a PWR of typical dimensions and migration length typical of UO<sub>2</sub> fuel (about 7.3 cm for 4.5% enriched UO<sub>2</sub>), the above formula yields 0.032 leakage reactivity worth. The neutron migration length for the Pu containing fertile free fuel is slightly higher than that of a

typical UO<sub>2</sub> fuel. In the low leakage core fuel management schemes, “once” burnt fuel assemblies are typically placed at the core periphery. At the end of the cycle, the neutron migration length of “once” burnt fuel varies with Pu content from roughly 7.8 cm for 10 vol. % PuO<sub>2</sub> to about 8 cm for 5 vol. % PuO<sub>2</sub> in the matrix (Figure 6). As a result, the leakage reactivity of fertile free fuel can be as high as 0.037.

Table 4 shows sensitivity of the Pu loadings to the uncertainty of leakage reactivity estimation. Consideration of larger neutron migration length in fertile free fuel than in UO<sub>2</sub> fuel results in an increase in required PuO<sub>2</sub> loading, which ranges from 0.05 to 0.17 vol. %.

The correct leakage reactivity effect can be assessed only based on the full core 3-dimensional neutronic simulation.



**Figure 6. Neutron Migration Length**

**Table 4. Effect of Leakage Reactivity**

	Cycle Length		
	12	18	24
$\rho_{LEAKAGE}$	<i>PWR - 33</i>		
0.030	4.89%	7.55%	10.25%
0.037	4.94%	7.65%	10.40%
$\Delta \text{PuO}_2$ loading	0.05%	0.10%	0.15%
	<i>PWR - 50</i>		
0.030	5.04%	7.78%	10.60%
0.037	5.09%	7.89%	10.77%
$\Delta \text{PuO}_2$ loading	0.05%	0.11%	0.17%

### Modified vs. Standard Linear Reactivity Model

Figure 6 presents the dependence of discharge fuel burnup on the initial Pu content estimated on the basis of two different LRM approaches. The first one adopts a classical LRM approximation where the fuel reactivity is a linear function of burnup. Then, the discharge fuel burnup ( $BU_d$ ) is given by:

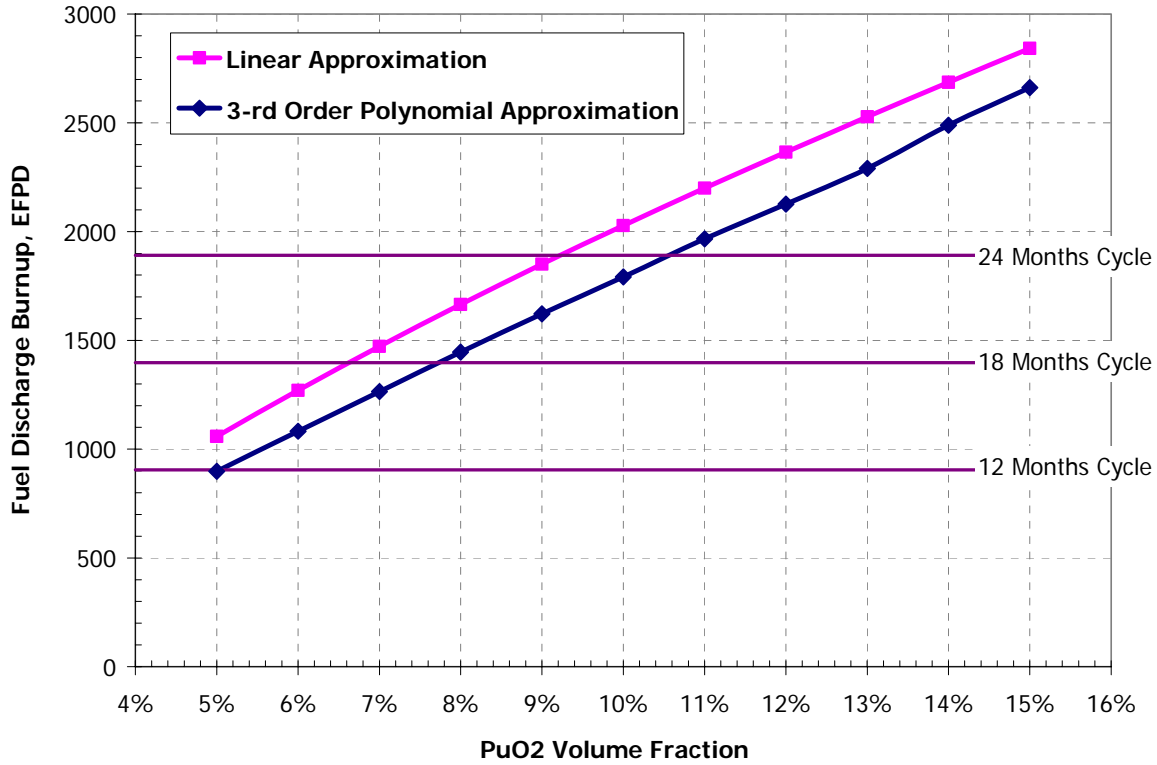
$$BU_d = BU_1 \times \frac{2n}{n+1}$$

where  $BU_1$  is the single batch core burnup and  $n$  is the number of batches in the core.

The second approach, used in this work and described in Section II, approximates fuel reactivity dependence on burnup by the 3<sup>rd</sup> order polynomial function. Such an approach describes the fuel reactivity behavior more accurately and therefore, it provides more accurate estimation of  $BU_d$  than the classical approach. The differences in discharge burnup estimation between the two LRM approaches may range from 130 to 180 EFPD (Figure 6).

It should also be noted that in both calculation approaches, the fuel reactivity is not power weighted. Therefore, adequate discharge burnup estimation may be obtained only by performing a full core 3-dimensional analysis.





**Figure 6. Linear vs. Polynomial Approximation Effect on Discharge Burnup Estimation**

### *III.3 Sensitivity of Fuel Cycle Length to Matrix Composition*

The list of calculated in this task cases is presented in Table 5. The case number nomenclature consists of 3 digits. The first digit represents the initial Pu isotopic vector used (PWR-33 or PWR-50), second – the reference fuel cycle length (12, 18, or 24 months), third - ZrO<sub>2</sub> volume fraction in the fuel matrix ranging from 30 to 70 vol. %.

Table 6 reports the results of discharge burnup calculations for all fuel compositions from Table 5. The discharge burnups were obtained using the same assumptions as in Section III.2; namely, 3<sup>rd</sup> order polynomial function fit for criticality curves and 0.03 leakage reactivity.

Figures 7.a and 7.b show the difference in discharge fuel burnup between various fuel matrix compositions and the reference one (1:1 ratio of ZrO<sub>2</sub> to MgO) for PWR-33 and PWR-50 Pu vectors respectively. The discharge burnup values exhibit a weak dependence on the fuel matrix

composition. The deviation from the reference discharge burnup is from  $\pm 3$  days for 12 month cycle to about  $\pm 7$  days for 24 month cycle for considered fuel matrix compositions (Table 5).

The initial Pu isotopic composition has almost no effect on the relative discharge burnup changes as a result of the changes in fuel matrix composition.

Figures 8.a and 8.b report the relative changes in PuO<sub>2</sub> loading necessary to maintain the reference fuel burnup for PWR-33 and PWR-50 initial Pu vectors respectively.

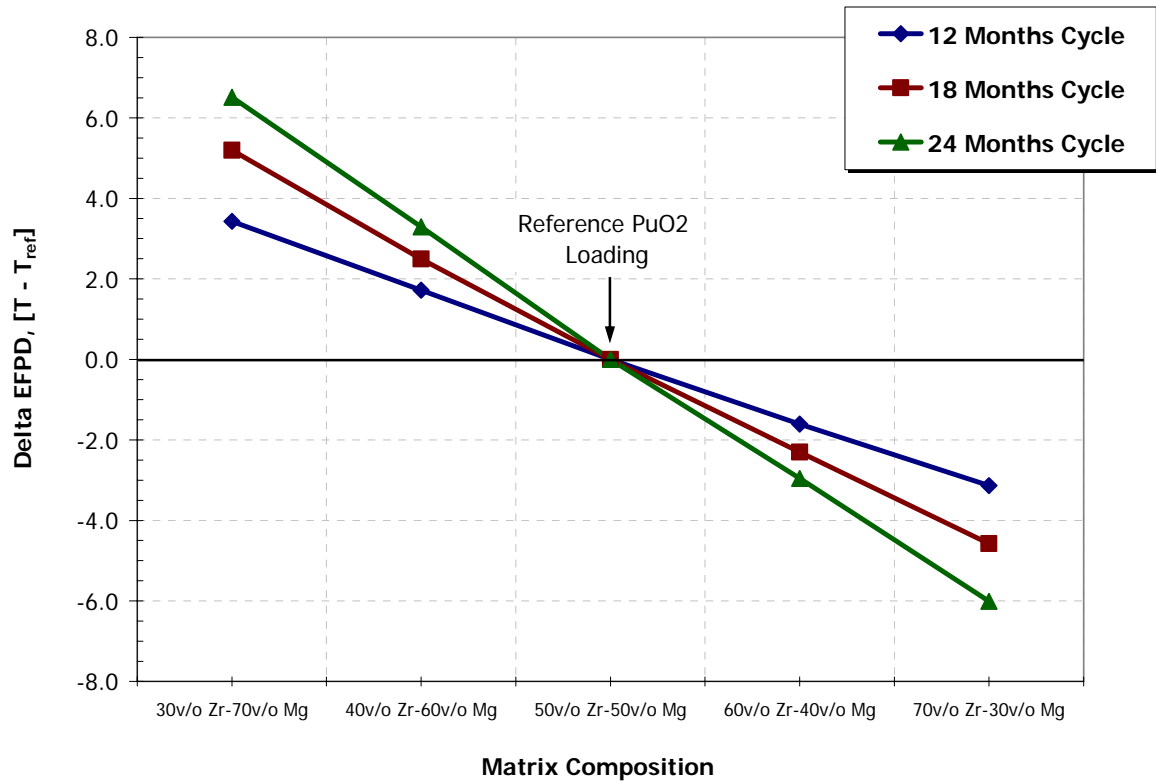
The spread in required Pu loading is from about  $\pm 0.02$  volume % PuO<sub>2</sub> for 12 month cycle to about  $\pm 0.04$  volume % PuO<sub>2</sub> for 24 month cycle. Matrices with low Zr content require less Pu loading necessary to maintain the reference fuel cycle length due to the lower neutron absorption in Mg than in Zr. However, the changes in achievable fuel burnup or corresponding changes in Pu loading requirements are relatively small.

**Table 5. List of Calculated Cases**

Case	PuO <sub>2</sub> loading, vol. %	Pu Vector	ZrO <sub>2</sub> in Fuel vol. %	MgO in Fuel vol. %	ZrO <sub>2</sub> in Matrix vol. %	MgO in Matrix vol. %
1.1.1	5.04	PWR-50	28.49	71.51	30.0	70.0
1.1.2	5.04	PWR-50	37.98	62.02	40.0	60.0
1.1.3	5.04	PWR-50	47.48	52.52	50.0	50.0
1.1.4	5.04	PWR-50	56.98	43.02	60.0	40.0
1.1.5	5.04	PWR-50	66.47	33.53	70.0	30.0
1.2.1	7.78	PWR-50	27.67	72.33	30.0	70.0
1.2.2	7.78	PWR-50	36.89	63.11	40.0	60.0
1.2.3	7.78	PWR-50	46.11	53.89	50.0	50.0
1.2.4	7.78	PWR-50	55.33	44.67	60.0	40.0
1.2.5	7.78	PWR-50	64.55	35.45	70.0	30.0
1.3.1	10.60	PWR-50	26.82	73.18	30.0	70.0
1.3.2	10.60	PWR-50	35.76	64.24	40.0	60.0
1.3.3	10.60	PWR-50	44.70	55.30	50.0	50.0
1.3.4	10.60	PWR-50	53.64	46.36	60.0	40.0
1.3.5	10.60	PWR-50	62.58	37.42	70.0	30.0
2.1.1	4.88	PWR-33	28.54	71.46	30.0	70.0
2.1.2	4.88	PWR-33	38.05	61.95	40.0	60.0
2.1.3	4.88	PWR-33	47.56	52.44	50.0	50.0
2.1.4	4.88	PWR-33	57.07	42.93	60.0	40.0
2.1.5	4.88	PWR-33	66.58	33.42	70.0	30.0
2.2.1	7.57	PWR-33	27.73	72.27	30.0	70.0
2.2.2	7.57	PWR-33	36.97	63.03	40.0	60.0
2.2.3	7.57	PWR-33	46.22	53.79	50.0	50.0
2.2.4	7.57	PWR-33	55.46	44.54	60.0	40.0
2.2.5	7.57	PWR-33	64.70	35.30	70.0	30.0
3.3.1	10.26	PWR-33	26.92	73.08	30.0	70.0
3.3.2	10.26	PWR-33	35.90	64.10	40.0	60.0
3.3.3	10.26	PWR-33	44.87	55.13	50.0	50.0
3.3.4	10.26	PWR-33	53.84	46.16	60.0	40.0
3.3.5	10.26	PWR-33	62.82	37.18	70.0	30.0

**Table 6. Sensitivity of Discharge Burnup (EFPD) to Matrix Composition**

	ZrO <sub>2</sub> vol. % in Matrix				
	<i>PWR-50</i>				
Cycle Length	30	40	50	60	70
12 Months Cycle	909.4	907.6	906.0	904.3	902.7
18 Months Cycle	1409.8	1407.3	1404.8	1402.5	1400.2
24 Months Cycle	1902.1	1898.8	1895.4	1892.4	1889.3
	<i>PWR-50</i>				
12 Months Cycle	899.5	897.8	896.1	894.5	893.0
18 Months Cycle	1416.0	1413.3	1410.8	1408.5	1406.3
24 Months Cycle	1907.9	1904.7	1901.4	1898.4	1895.3



**Figure 6.a. Cycle Length Sensitivity to Fuel Matrix Composition (PWR-33 Pu)**

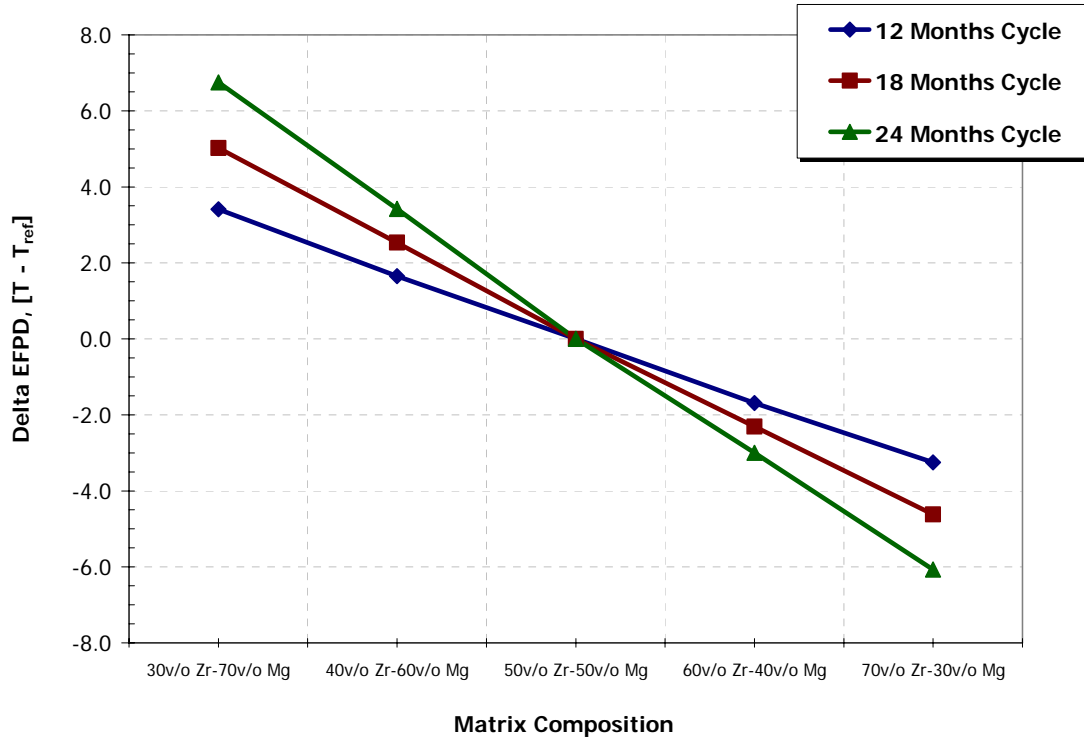


Figure 6.b Cycle Length Sensitivity to Fuel Matrix Composition (PWR-50 Pu)

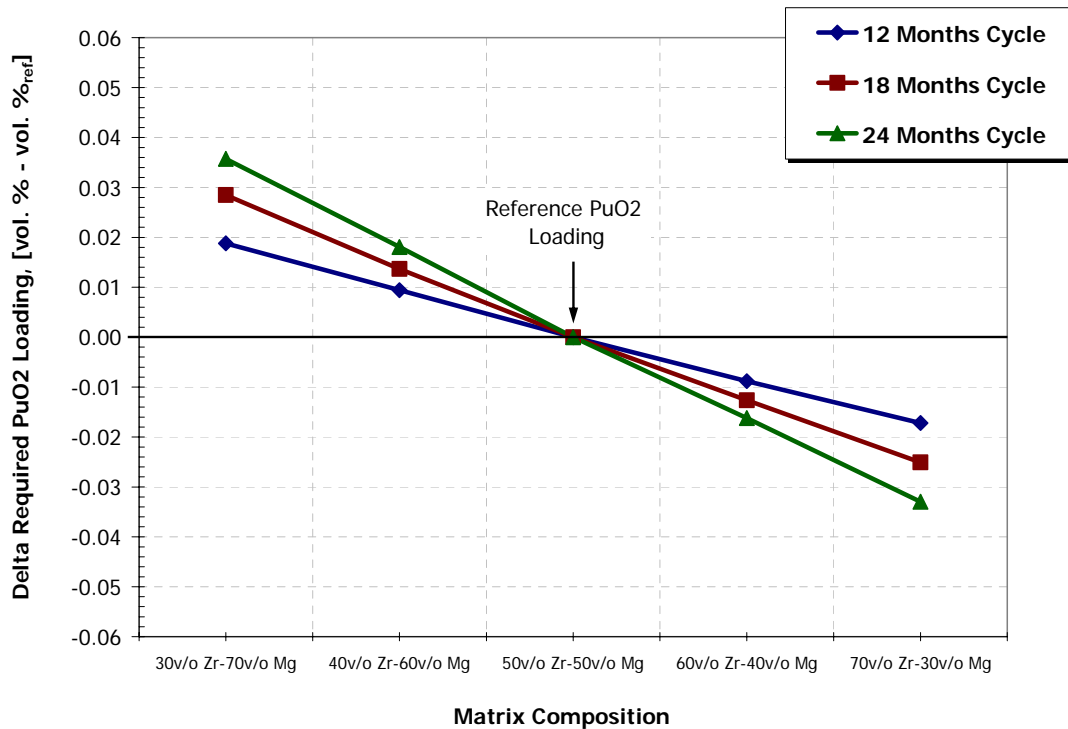
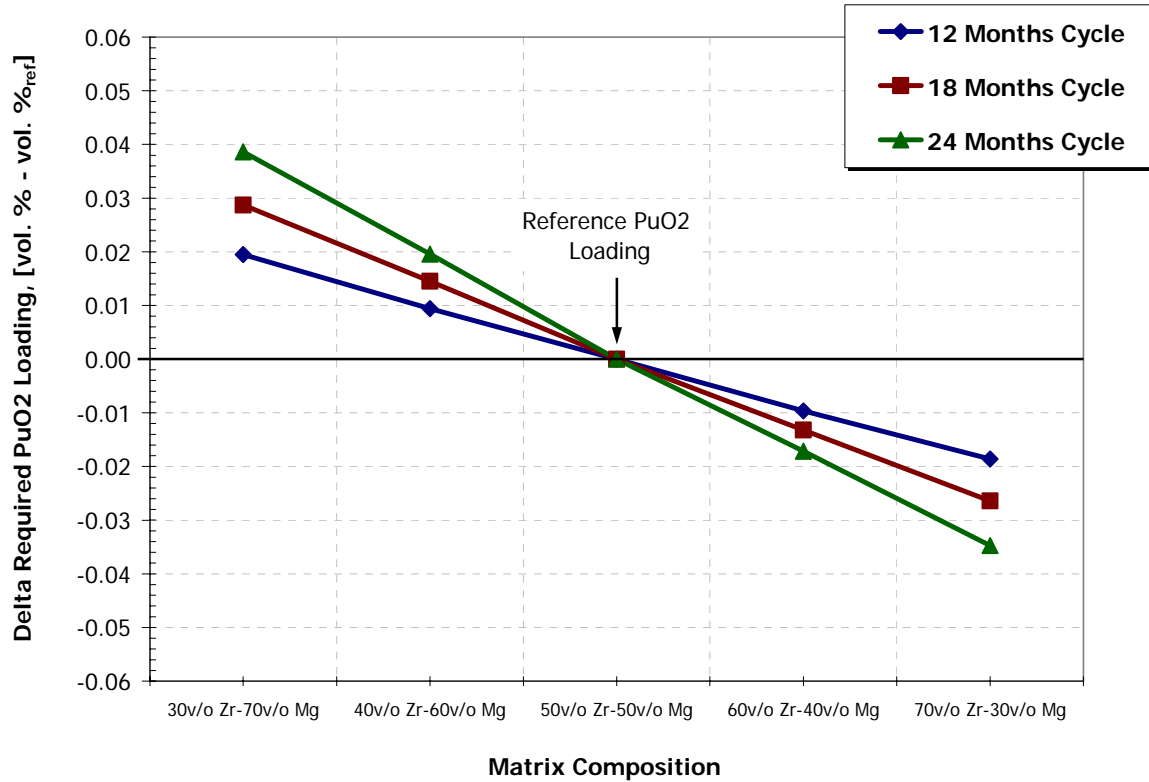


Figure 7.a. Sensitivity of Pu Loading Requirements to Fuel Matrix Composition (PWR-33 Pu)



**Figure 7.b Sensitivity of Pu Loading Requirements to Fuel Matrix Composition (PWR-50 Pu)**

#### IV. Summary and Conclusions

In this task, we determined Pu loading necessary to achieve industry standard fuel cycle lengths of 12, 18, and 24 months. Additionally, we investigated the achievable fuel burnup sensitivity to the composition of fertile free MgO – ZrO<sub>2</sub> matrix.

In order to account for the non-linear shape of the criticality as a function of burnup curves, modified Linear Reactivity Model was applied to the results of 2-dimensional fuel assembly burnup calculations in order to estimate the discharge fuel burnup. The reactivity dependence on burnup was described by the 3<sup>rd</sup> order polynomial function instead of conventional linear dependence assumption. Such LRM modification was found to be important. The error in discharge burnup estimation by the simple LRM versus more accurate polynomial description approach may reach up to 180 EFPD.

In the current analysis, we used typical PWR with  $\text{UO}_2$  fuel leakage reactivity worth value of 0.03. However, the leakage reactivity worth was estimated to be somewhat higher than in a typical  $\text{UO}_2$  fuel case and dependant on Pu loading. The uncertainty in leakage reactivity estimation may result in an increase of required  $\text{PuO}_2$  loading by up to 0.17 volume %. The leakage effect in fertile free cores can be correctly evaluated only in 3-dimensional full core neutronic simulation.

All calculations in current analysis were performed for two Pu isotopic vectors: from low burnup LWR fuel with long decay time (PWR-33) and high burnup LWR fuel with short decay time (PWR-50). The difference in estimated discharge burnup between the two considered Pu vectors ranges from 30 to about 60 EFPD in the fuel cycle lengths range of interest. Therefore, we concluded that the Pu composition has generally minor effect on fertile free fuel criticality and achievable discharge burnup. This is partially due to the mutually canceling effects of higher Pu239 fraction but lower Pu241 fraction in PWR-33 as compared to PWR-50 grade plutonium.

The achievable fuel burnup exhibits extremely weak dependence on the fuel matrix composition. This is due to the low absorption in both Zirconia and Magnesia. Zr is slightly more neutron absorbing material than Mg. Therefore, an increase in  $\text{ZrO}_2$  fraction in the matrix results in a decrease in discharge fuel burnup and corresponding increase in required Pu loading. Variation of  $\text{ZrO}_2$  volume fraction from 30 to 70% results in up to  $\pm 8$  EFPD deviation from the discharge burnup of the reference 50%  $\text{ZrO}_2$  – 50% MgO fuel matrix composition. This range of differences in discharge burnup values translates into almost  $\pm 0.04$  volume % range in  $\text{PuO}_2$  loadings required to achieve the reference fuel cycle length.

## References

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