Multiple Recycling of Plutonium in Advanced PWRs

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Summary

In this paper, the influence of the moderator-to-fuel ratio in MOX fueled PWRs on the moderator void coefficient, the fuel temperature coefficient, the moderator temperature coefficient, the boron reactivity worth, the critical boron concentration, the mean neutron generation time and the effective delayed neutron fraction has been assessed. Increasing the moderator-to-fuel ratio to values larger than three, gives a moderator void coefficient sufficiently large to recycle the plutonium at least four times.

Scenario studies show that four times recycling of plutonium in PWRs reduces the plutonium mass produced with a factor of three compared with a reference once-through reactor park, but that the americium and curium production triple. If the minor actinides and the remaining plutonium after four times recycling are disposed of, the reduction of the radiotoxicity reaches only a factor of two. This factor increases to five at the maximum when the plutonium is further recycled. Recycling of americium and curium is needed to further reduce the radiotoxicity of the spent fuel.

Introduction

The slow-down of fast reactor development urges for another solution to get rid of the plutonium produced in light water reactors (LWRs). One possibility is to recycle this plutonium in LWRs before disposition or further use in fast reactors. Multiple recycling of plutonium in (U/Pu) MOX fuel in present day LWRs is limited. At each recycling, the fraction of fissile plutonium isotopes decreases, which requires an ever increasing plutonium weight fraction. After two recyclings, this fraction becomes that high that the moderator void coefficient (MVC) may become positive, which is not allowed of course. Increasing the MF ratio reduces the neutron capture rate in the resonance energy region, which translates into a lower plutonium weight fraction and a negative MVC. The maximum number of times plutonium can be recycled depends on the MF ratio, the target burnup and the blending ratio at the reprocessing plant. Present day technology requires the spent MOX fuel being mixed with a three times larger amount of spent UO_2 fuel to limit the α emission and the heat generation during the reprocessing.

This paper shows the results of a study on the influence of the MF ratio in PWRs on the safety and the reactor physics aspects of MOX recycling. Various reactivity coefficients and kinetic parameters, as well as the plutonium consumption rate and the associated production rate of the minor actinides are calculated for four plutonium recycling steps with the MF ratio as a parameter varied between two (standard PWR) and four. The basic reactor design is based on a French N4 PWR. More results may be found in (Kloosterman, 1998).

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Calculational procedures

The calculations have been performed with the OCTOPUS burnup and criticality code system (Kloosterman et al. 1996), applying the BONAMI-NITAWL-XSDNRPM codes for the resonance shielding and neutron spectrum calculations, respectively, and the ORIGEN-S code for the burnup calculations. The data libraries for both the spectrum calculations and the burnup calculations were based on the JEF2.2 and EAF4.1 nuclear data libraries.

The reactivity coefficients and kinetic parameters have been calculated at a number of branchings during the burnup. The fuel temperature coefficient (FTC) has been calculated by increasing the fuel temperature from 930 K till 1030 K, the moderator temperature coefficient (MTC) by increasing the moderator temperature from 586 till 596 K, and the MVC by reducing the moderator density to one percent of its nominal value. The BRW has been calculated by increasing the boron concentration with 100 ppm. Furthermore, an adjoint spectrum calculation has been performed to calculate the contribution of each nuclide to the FTC by means of first-order perturbation theory and to calculate the mean neutron generation time Λ and the effective delayed neutron fraction β_{eff}. All reactivity coefficients have been calculated as $\alpha = \delta k/k$.

The calculations have been performed for five values of the MF ratio, which has been increased from 2 to 4 by reducing the fuel pin diameter. To limit the heat flux at the rod outer surface, the irradiation time as well as the linear power have been reduced proportional to the fuel pin diameter.

Recycling scheme

For each MF ratio, the plutonium has been recycled four times. First, the plutonium in the spent UO_2 fuel of the standard PWR has been mixed with UO_2 made of depleted uranium to fabricate the MOX fuel for the advanced PWR. Subsequently, the spent MOX fuel unloaded from this reactor is blended with a three times larger amount of spent UO_2 fuel to recover the plutonium needed for the fresh MOX fuel of the next recycling. The sensitivity of the results to this blending ratio has not been investigated. The recycling scheme is shown in figure 1.

The fuel enrichment has been determined by the following reactivity adjustment procedure. The French N4 reactor operates in five batches each with a burnup of 9.5 GWd/tHM. When the reactivity decreases linearly as a function of burnup, the k_∞ at the end of the third fuel batch equals at first order the k_∞ of the reactor core at the end of the equilibrium cycle. This value can be determined from reactor core calculations ran to determine the UO_2 fuel enrichment. Given the UO_2 fuel enrichment of 4^w% ,(CEA, 1992), the k_∞ at the end of the third fuel batch equals 1.057, and this value has been used to determine the plutonium densities in the MOX fuels.

The critical boron concentration (CBC) has been determined such to obtain a flat k_∞ curve during the third fuel batch. Assuming that the reactivity decreases linearly as a function of time, the CBC just compensates for the buildup of fission products and the depletion of fissile material. For UO_2 fuel in the standard PWR (MF=2), this concentration equals 872 ppm at the beginning of each batch. Of course, the needed plutonium density in the MOX fuel and the CBC depend on each other and have been determined iteratively.

Results

The plutonium weight fraction in the fuel is given in table 1 as a function of the MF ratio and of the recycle number. Increasing the MF ratio reduces the resonance capture rate, which translates into a lower plutonium fraction in the fuel. With increasing recycle number, the fraction of fissile

plutonium isotopes decreases and the plutonium weight fraction in the fuel has to be enlarged again to meet the reactivity requirements.

Table 1: *The plutonium weight fraction (^{w}%) in the fuel (heavy metal).*

MF ratio	Recycle number 1	2	3	4
2.0	10.0	13.0	15.0	16.6
2.5	7.7	11.5	14.1	16.2
3.0	6.5	10.1	12.8	15.1
4.0	5.9	8.7	11.1	13.5

The number of times plutonium can be recycled is limited because of the MVC which may become very small and even positive for fuel types with a low MF ratio. This is shown in table 2. Together with the rather large uncertainty margin of about 5000 pcm (OECD, 1997), the number of recyclings is probably limited to two times for the MF=2 fuel, three times for the MF=2.5 fuel and at least four times for the fuels with higher MF ratios.

Table 2: *The moderator void coefficient (MVC) of the fuel (pcm/99%).*

MF ratio	Fuel type	Recycle number 1	2	3	4
2.0	UO2	-52700			
2.0	MOX	-21000	-11300	-5810	-2130
2.5	MOX	-35800	-22400	-14800	-9650
3.0	MOX	-44300	-32000	-23900	-18000
4.0	MOX	-51900	-43200	-36500	-30700

The BRW is shown in table 3. Its value decreases from -7.7 pcm/ppm for the UO_2 fuel to -2.4 pcm/ppm for the MOX fuel due to the much harder neutron spectrum in the MOX fuel. Increasing the MF ratio softens the neutron spectrum, which gives larger values in magnitude for the BRW. The increasing plutonium weight fraction with increasing recycle number reduces the BRW. Increasing the MF ratio reduces the neutron capture rate in the resonance range, and hence the conversion rate of fertile uranium and plutonium isotopes to fissile ones also reduces. Consequently, the neutron spectrum softens considerably as a function of burnup, which leads to a large variation of the BRW during the burnup.

Table 3: *The boron reactivity worth (BRW) of the fuel (pcm/ppm).*

MF ratio	Fuel type	Recycle number 1	2	3	4
2.0	UO2	-7.7			
2.0	MOX	-2.4	-2.0	-1.8	-1.7
3.0	MOX	-6.1	-4.2	-3.9	-3.5
4.0	MOX	-9.6	-7.5	-6.4	-5.7

The CBC [ppm] is roughly equal to the reactivity loss during the burnup [pcm] divided by the BRW [pcm/ppm]. For the first recycle, the CBC decreases from 1193 ppm for the fuel with MF=2,

to 626 ppm for the MF=4 fuel. The CBC does not depend very much on the recycle number, due to the fact that both the BRW and the reactivity loss decrease with increasing recycle number. The latter is due to the increasing conversion ratio of fertile to fissile plutonium isotopes. For the fourth recycle, the CBC equals 1357 ppm for the MF=2 fuel and 510 ppm for the MF=4 case.

The fuel temperature coefficient (FTC) of a UO_2 fueled reactor increases as a function of burnup due to the buildup of ^{240}Pu. For a MOX fueled reactor, the contribution of each nuclide to the FTC is rather constant as a function of burnup. The magnitude of the FTC of MOX fuel exceeds that of UO_2 fuel due to the larger neutron absorption rate in the resonance region by the even plutonium isotopes. This is seen from the contributions of the individual nuclides to the FTC in figure 2. Increasing the MF ratio decreases the magnitude of the FTC due to a better thermalization of the spectrum and a corresponding lower resonance absorption rate.

The moderator temperature coefficient (MTC) increases in magnitude for MOX fueled reactors. The MTC of a UO_2 fueled reactor decreases from -20 pcm/K at BOL to -70 pcm/K at EOL mainly due to the linearly decreasing boron concentration as a function of burnup. For the MOX fuel with the same MF ratio, the MTC ranges from -40 pcm/K at BOL to -70 pcm/K at EOL. Although a negative MTC is required, its magnitude should not be too large to limit the power excursion in case of moderator cooling accidents. The magnitude of the MTC for reactors with MF>3 decreases again due to the increasing BRW.

There are several kinetic parameters important for the safe control of a reactor. The most important ones are the mean neutron generation time Λ and the effective delayed neutron fraction β_{eff}. For UO_2 fuel in a standard PWR, the first parameter equals $1.8{\cdot}10^{-5}$ s. However, for MOX fuel, the neutron spectrum is much harder and the Λ decreases to about $5{\cdot}10^{-6}$ s for the first recycle. Increasing the MF ratio gives larger values for the Λ varying between $1.9{\cdot}10^{-5}$ s for the first recycle number to $1.1{\cdot}10^{-5}$ s for the fourth.

The β_{eff} for the UO_2 fueled reactor decreases from 0.7% at BOL to 0.5% at EOL, due to the burnup of U-235 and the buildup of Pu-239. The latter nuclide has a much smaller delayed neutron fraction. For the MOX fueled reactors, the β_{eff} equals about 0.4% and is almost constant as a function of burnup.

Scenario Studies

A reactor park operating at equilibrium, i.e. such that the plutonium in the spent fuel of one reactor is completely recycled in another, consists of reactors loaded with fresh UO_2 fuel and of reactors loaded with MOX fuel made of recycled plutonium. The composition of such reactor parks (the fraction of reactors containing fresh UO_2 fuel, the fraction containing MOX fuel of the first recycle, the fraction containing MOX fuel of the second recycle, etc.) are shown in table 4. About 80% of the reactors are loaded with UO_2 fuel. This fraction decreases slightly with increasing MF ratio, which is due to two opposite effects. At the one hand, the needed plutonium fraction in the MOX fuel decreases with increasing MF ratio, which means that less UO_2 reactors are needed to fuel the MOX reactors, while at the other, the plutonium consumption in the MOX reactors increases slightly with increasing MF ratio, which means that more UO_2 fueled reactors are needed.

For an electricity production of 100 GW_ea , about 102 reactors are needed of 1450 MW_e each with an availability factor of nearly 70%. The yearly produced actinide masses are shown in table 5 for the reactor parks shown in table 4 and for a once-through reactor park containing solely UO_2 fueled reactors as a reference. In table 5, it is assumed that all actinides are completely sent to the waste (see column "Losses"), except for the plutonium which is recovered with a loss fraction

of 0.12% (the minimum achievable nowadays). The fourth column shows the plutonium mass recycled each time. After four recyclings, the remaining unloaded plutonium can be sent to the waste or stored for further use.

The sixth column in table 5 shows the ratio of the actinide masses produced by the advanced reactor park and by the UO_2 fueled once-through park (REFR scenario). Clearly, plutonium recycling gives a considerable reduction of the plutonium mass produced (factors of 0.36 and 0.31) at the expense of a two to four times higher americium and curium production.

In figure 3, the radiotoxicity curves of the reference once-through scenario, and of the recycling scenarios are shown. The plutonium after four times recycling is assumed to be sent to the waste repository (curve "losses+invent") or assumed to be stored for further use (curve "losses"). Clearly, the remaining plutonium after four times recycling contributes significantly to the radiotoxicity produced by the reactor park. It is therefore recommended to continue the recycling of this plutonium, e.g. in fast reactors. However, even in such a case, the reduction of the radiotoxicity does not exceed a factor of five due to the accumulation of americium and curium. Recycling these two nuclides is a necessity to further reduce the radiotoxicity of the spent fuel produced.

Conclusions

The number of times plutonium can be recycled in LWRs is limited due to the degrading of the plutonium and the accompanied increase of the plutonium density in the MOX fuel. For plutonium densities too large, the moderator void coefficient becomes very small. The maximum number plutonium can be recycled depends on the moderator-to-fuel (MF) ratio of the fuel and increases from two times for a MF ratio of 2 to at least four times for MF=4. Enhancing the moderation in a MOX fueled PWR also increases the boron reactivity worth to values comparable to that of UO_2 fueled reactors. The same holds for the mean neutron generation time. The effective delayed neutron fraction of MOX fuel is about 0.4% compared to 0.7% of fresh UO_2 fuel.

In a reactor park with four times recycling of plutonium in PWRs with enhanced moderation, the plutonium mass reduction may reach a factor of three at the expense of a three times larger production of americium and curium. The resulting reduction of the radiotoxicity reaches a factor of two in case the remaining plutonium after four times recycling is disposed of, and a factor of five at the maximum in case the remaining plutonium is further recycled. A further reduction of the radiotoxicity can be achieved by recycling americium and curium in advanced, possibly accelerator driven, reactors.

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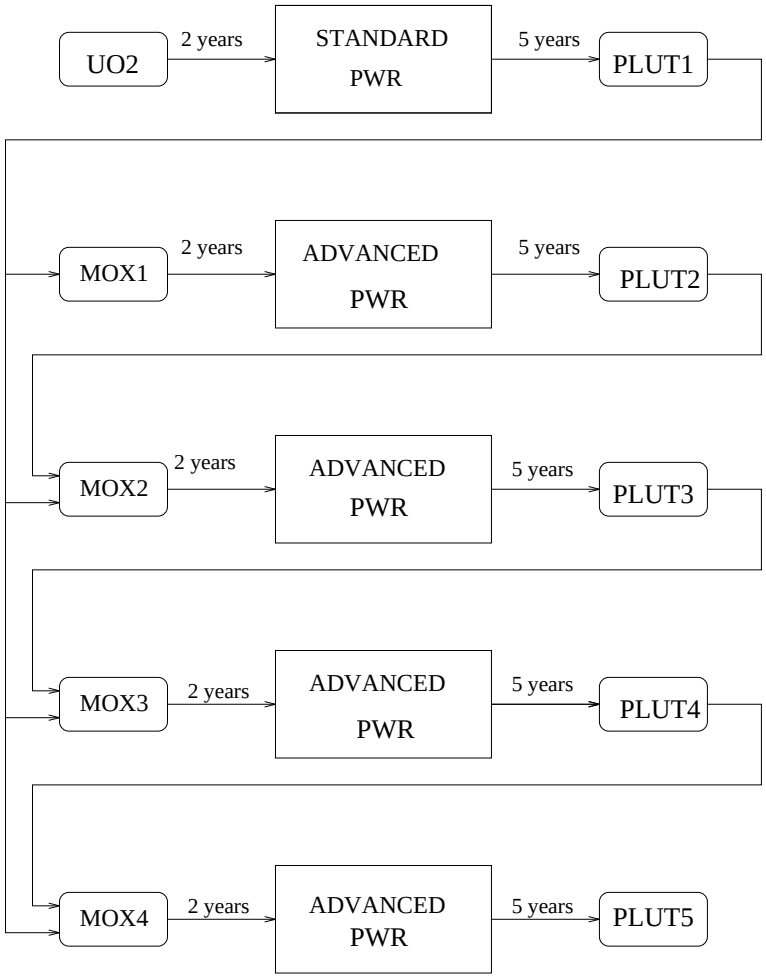

Figure 1: *A schematic representation of the recycling calculations. The standard PWR has a MF ratio of two, the advanced PWRs all have MF ratios of 2, 2.5, 3, 3.5 or 4. A blending ratio of three has been used in the calculations, which means that each unloaded MOX assembly is mixed with three spent UO_2 assemblies.*

Table 4: *The composition of a reactor park in equilibrium.*

		Fraction of reactors			
Fuel Cycle	UO_2	MOX 1	MOX 2	MOX 3	MOX 4
MF20	0.82	0.054	0.045	0.040	0.037
MF30	0.80	0.071	0.053	0.043	0.037
MF40	0.79	0.073	0.057	0.046	0.038

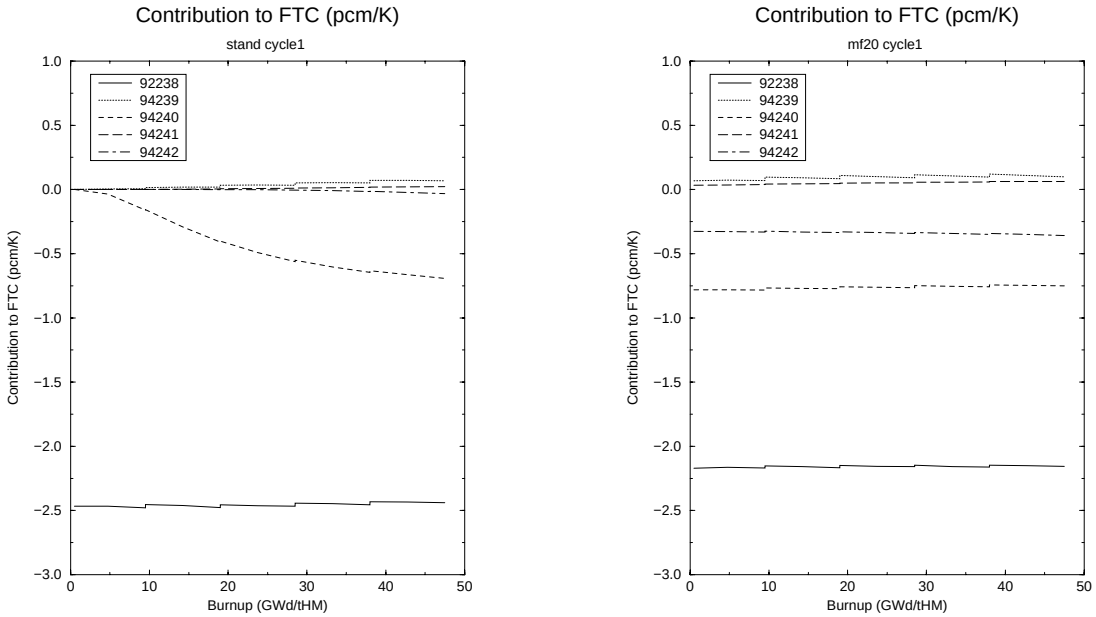

Figure 2: *The contributions of the individual nuclides to the FTC for the UO_2 fuel (left figure) and for the MOX fuel with MF=2 as a function of burnup.*

Table 5: *The yearly produced actinide masses in a 100 GWe reactor park consisting of UO_2 fueled reactors in once-through mode (REFR) or of UO_2 fueled reactors with four times plutonium recycling in MOX fueled reactors with the MF ratio indicated.*

Scenario	Actinide	Losses (kg/a)	Recycled mass (kg/a)	Total mass (kg/a)	Mass ratio MF/REFR
REFR	Np	1433	0	1433	
REFR	Pu	26490	0	26490	
REFR	Am	1433	0	1433	
REFR	Cm	164	0	164	
REFR	Sum	29520	0	29520	
MF30	Np	1206	0	1206	0.84
MF30	Pu	65	9412	9477	0.36
MF30	Am	4072	0	4072	2.84
MF30	Cm	607	0	607	3.70
MF30	Sum	5950	9412	15362	0.52
MF40	Np	1185	0	1185	0.83
MF40	Pu	59	8277	8336	0.31
MF40	Am	3678	0	3678	2.57
MF40	Cm	573	0	573	3.49
MF40	Sum	5495	8277	13772	0.47

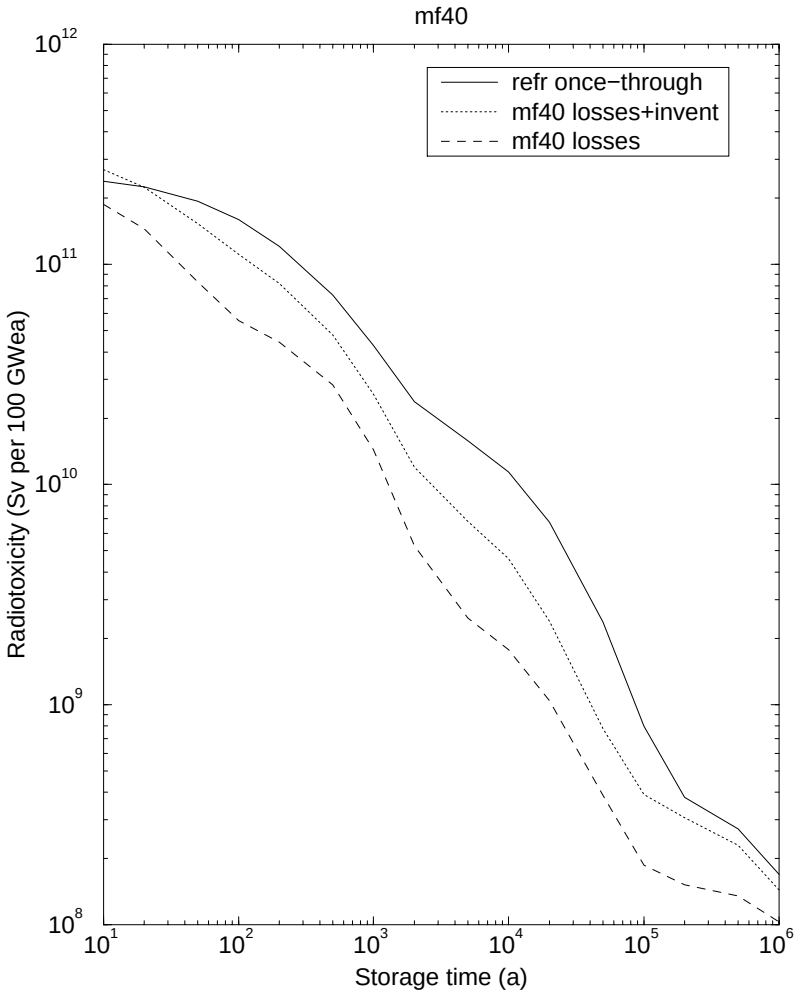

Figure 3: *A comparison of the radiotoxicity of the reactorpark consisting of UO_2 fueled reactors in once-through mode (refr), and of the reactorpark with plutonium recycling in MOX fueled reactors with MF=4. The plutonium after four times recycling is assumed to be disposed of (curve "losses+invent") or to be stored for further use (curve "losses").*