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Criticality safety of spent nuclear fuel assemblies from the transmutation of minor actinides in fast reactors

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Abstract

Criticality safety concerning the packaging of spent nuclear fuel assemblies from the transmutation of ^{237}Np and ^{241}Am in a fast reactor are investigated and compared to cases of spent fuels from commercial nuclear power stations. The investigation is based on the neutron multiplication factor calculated using the fresh and spent fuel compositions. Fuels based on a fresh composition with high amount of fissile material and low amounts of ^{237}Np and ^{241}Am resemble the fast reactor fuel, with the multiplication factor decreasing when calculated using the spent fuel compositions. However, the multiplication factor increases for targets based on a fresh composition without fissile content and high amounts of ^{237}Np and ^{241}Am, although it remains below 1 for up to 90 GWd/t burnup.

1. Introduction

Criticality safety associated with the packaging of spent nuclear fuels is a challenging issue for the scientific and legislative communities involved in efforts to prevent criticality accidents (IAEA, 2000). Safety issues associated with criticality accidents are assessed through appropriate criticality calculations usually performed on the assumption that the spent nuclear fuel is represented by its fresh composition. This is a simple approach, rendering unnecessary any knowledge of the fuel irradiation history. However, it overlooks any possible decrease in the fuel reactivity due to the evolution in its composition and the formation of actinides and fission products during irradiation in the reactor. Some of these nuclides are strong neutron absorbers, responsible for the decrease in the reactivity of the spent fuel. Therefore, the inclusion in criticality calculations of these nuclides may result in a considerable improvement regarding criticality safety of packaging the spent fuel.

Emerging nuclear fuel cycles consider partition and transmutation (P&T) as an option to reduce the radiotoxicity of the high level radioactive waste (HLW) (Koch, 1986). In P&T, some long-lived radiotoxic nuclides are partitioned from HLW and added to fuels in order to be transmuted to shorter lived or more stable nuclides, through irradiation in a neutron field (Koch et al., 1997). This work assesses criticality issues associated with the packaging of spent nuclear fuel from a P&T fuel cycle involving the transmutation of ^{237}Np and ^{241}Am in a fast reactor and compares them to those of commercial spent nuclear fuels from existing nuclear power stations.

2. Materials and methods

The required criticality analysis was based on the determination of the neutron multiplication factor (k_{eff}) which is a key parameter for criticality safety. Criticality calculations were carried out for a range of spent nuclear fuels from commercial and transmutation fuel cycles on the basis of their composition at the beginning and end of irradiation (BOI and EOI).

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2.1. Nuclear fuels considered

- *Transmutation fuels*: regarding the transmutation of ^{237}Np and ^{241}Am in a fast breeder reactor (FBR), fresh fuels with low and high concentrations of these nuclides have been considered, resembling the homogeneous and heterogeneous recycling concepts respectively. In the former, the nuclides of interest are homogeneously mixed in a fast reactor fresh fuel of the form $U_{0.745}Pu_{0.245}$ – 1% ^{241}Am or – 1% ^{237}Np (cases T1 and T2). Alternatively, in the heterogeneous recycling concept, large amounts of the nuclides of interest are added in special oxide targets of the form $U_{0.60}$ – 40% ^{237}Np and $U_{0.50}$ – 25% ^{237}Np 25% ^{241}Am (cases T3 and T4).
- *Commercial fuels*: from nuclear reactor power stations were considered as reference cases for comparison purposes: PWR-UO_2 (case R1); PWR-MOX (case R2); fast breeder reactor FBR-MOX (case R3).

The cases considered are summarized in Table 1, indicating the fresh fuel compositions, the burnup of the fuels at EOI and the content in fissile material.

2.2. Computational methodology

The neutron multiplication factor was calculated using the MCNP-4C code (Briesmeister, 2000). The calculation was performed for an infinite array of cells in unborated water, containing typical spent fuel assemblies for the fuel–reactor combination considered. The spacing between the assemblies in the infinite array was 10 mm. The physical description of the fuel assemblies considered in the study is given in Table 2 (Knief, 1992). Each calculation was based on 1 million neutron histories: 550 generations were simulated, with 2000 neutron histories each, skipping the first 50 generations before averaging. The k_{eff} was calculated on the basis of both the fresh and spent fuel compositions. The standard deviation associated with the calculation of k_{eff} ranged between 0.3% and 0.6% for k_{eff} values of 1.5 and 0.5, respectively. The fresh fuel composition corresponds to the inventory at BOI. The spent fuel composition used corresponds to the inventory in actinides and fission products at EOI. Depletion calculations are required to simulate the evolution in fuel composition during reactor operation.

Table 2
Physical description and irradiation history of the fuel assemblies considered

	PWR assemblies	LMFBR assemblies
Assembly		
Type	16 × 16 pins Square	271 pins Hexagonal
Pin pitch (mm)	13	9.7
Fuel pin		
Active length (m)	3.81	1
Pellet diameter (mm)	8.2	7
Clad material	Zr	Stainless steel
Clad thickness (mm)	0.64	0.7
Irradiation in reactor		
Power level (MW)	37.5	123
Burnup at EOI (GWd/t)		
Case R1	50	–
Case R2	40	–
Cases R3, T1, T2	–	100
Cases T3, T4	–	50

The calculations were performed using the ORIGEN-2 code (Croff, 1980) for the reactor power levels and target burnup values mentioned in Table 2. Then, selected isotopes of each spent fuel at the end of irradiation were used as an input to determine the k_{eff} value in each case.

The nuclides of interest used for the criticality analysis, have been chosen on the basis of their reactivity worth and their nuclear and chemical stability (Neuber, 1998). The following isotopes were considered in the analysis:

- Actinides: ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm.
- Fission products: ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{113}Cd, ^{133}Cs, ^{135}Cs, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{147}Pm, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{153}Eu, ^{155}Eu, ^{155}Gd.

The importance of these nuclides stems from their influence on the neutron multiplication factor, due to their cross-sections for nuclear reactions. The fissile isotopes ^{235}U, ^{239}Pu and ^{241}Pu have a significant positive reactivity worth due to their fission cross-section. The Cm isotopes

Table 1
Content of U, total Pu and fissile Pu (^{239}Pu and ^{241}Pu) in fuel at beginning and end of irradiation (BOI and EOI)

Case	Reactor-fuel	Burnup (GWd/t)	U (wt%)	Pu (wt%)	Fissile material	
					BOI (wt%)	EOI (wt%)
R1	PWR-U	50	100 (U)	–	3.5 (U^{235})	1.2 (U^{235} and Pu)
R2	PWR-MOX	40	95 (U_N)	5	3 (Pu)	1.4 (Pu)
R3	LMFBR-MOX	100	75 (U_D)	25	17.5 (Pu)	12.8 (Pu)
T1	LMFBR-MOX 1%Am	100	74.5 (U_D)	24.5	17 (Pu)	12.8 (Pu)
T2	LMFBR-MOX 1%Np	100	74.5 (U_D)	24.5	17 (Pu)	12.9 (Pu)
T3	UO_2-target 40%Np	50	60 (U_D)	–	–	1.3 (Pu)
T4	UO_2-target 25%Np 25%Am	50	50 (U_D)	–	–	1.2 (Pu)

U, enriched in 3.5% ^{235}U; U_N, natural U (0.7% ^{235}U); U_D, depleted U (0.2% ^{235}U).

are of particular interest due to their high neutron emission and also in addition any possible influence on the k_{eff} of arrays of spent fuel. Increased amounts of ^{242}Cm and ^{244}Cm are produced during reactor operation when plutonium and particularly ^{241}Am are present in the fresh MOX fuels. These fuels emit at the end of irradiation up to two orders of magnitude more neutrons than the commercial MOX fuels considered (Nicolaou and Koch, 1994). The fission product ^{149}Sm has a negative reactivity worth due to its high thermal cross section for neutron capture (41,000 barns).

3. Results

The reactivity worth of the actinides and fission products mentioned in Section 2.2 is shown in Figs. 1 and 2, respectively. The reactivity worth of the nuclides in Figs. 1 and 2 is relative to the highest value in each of the figures. The figures refer to the fast reactor fuel UPu – 1%^{241}Am at the end of irradiation with final burnup values of 40, 60 and 100 GWd/t. The reactivity worth ($\Delta\rho$) has been defined as

$$\Delta\rho = (1/k_{\text{eff}})(\Delta k/k_i) \quad \text{and} \quad \Delta k = k_{\text{eff}} - k_i$$

where k_{eff} refers to the model calculation which includes the actinide and fission product nuclides of interest, and k_i is the k_{eff} of the model without nuclide i.

The positive reactivity worth due to the fissile nuclides ^{235}U, ^{239}Pu and ^{241}Pu is clearly demonstrated in Fig. 1. The nuclide ^{238}U exhibits a strong negative reactivity worth due to its high amount present in the fuel and neutron capture. The other actinides have an insignificant negative reactivity worth. The fission products generally exhibit a small negative reactivity worth (Fig. 2). The exception is ^{149}Sm with a rather prominent negative reactivity worth due to its high thermal cross section for neutron capture (41,000 barns).

The change in k_{eff} when calculated on the basis of the fresh and spent fuel compositions is shown in Fig. 3 for the different fuels considered in the study. The different fuel types have been clustered in three distinct families which comprise:

1. the two fast reactor transmutation fuels $U_{0.745}Pu_{0.245}$ with 1% ^{241}Am or ^{237}Np and the fuel from the reference fast reactor case FBR–MOX ($U_{0.75}Pu_{0.25}$);
2. the two transmutation targets from the heterogeneous recycling concept, namely $U_{0.60}$ – 40% ^{237}Np and $U_{0.50}$ – 25% ^{237}Np 25% ^{241}Am; and,
3. the reference cases for thermal reactor fuels PWR-UO_2 and – MOX UPu.

The criterion that influences the clustering of the fuels is the content in fissile material both at BOI and EOI (Table 1). Fuels with comparable amounts of fissile material have been clustered together, while k_{eff} increases with the fissile material content.

Within the first group, the change of k_{eff} for the transmutation fuels resembles the reference case of the fast reactor fuel (Fig. 3). The neutron multiplication factor decreases when it is calculated using the actinide and fission product composition at EOI instead of the fresh fuel composition. The k_{eff} is higher for the UPu – 1% ^{237}Np fuel due to the formation of fissile Pu during reactor operation, from neutron capture reactions in ^{237}Np present in the fresh fuel, according to the reaction

$$^{237}\text{Np}\ (n,\gamma)^{238}\text{Np} \underset{\tau_{1/2}=50.8\text{h}}{\overset{\beta^-}{\longrightarrow}} {}^{238}\text{Pu}\ (n,\gamma)^{239}\text{Pu} \qquad (1)$$

The transmutation targets in the second group, exhibit a different trend in the k_{eff} change when computed using the compositions at BOI and EOI. The negligible fissile content (~0.01% in ^{235}U) at BOI results in $k_{\text{eff}} \ll 1$ ($k_{\text{eff}} \approx 0.6$). However, k_{eff} increases to about 0.8 when the composition

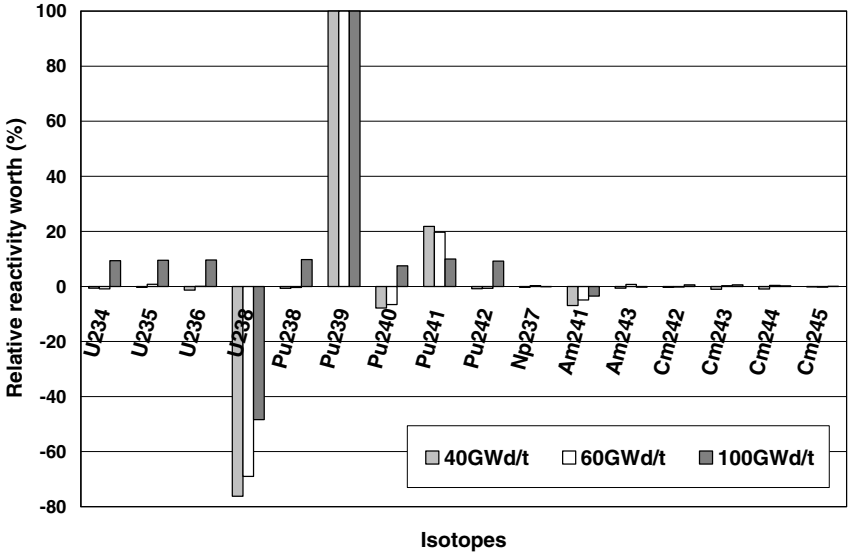

Fig. 1. Reactivity worth of actinides at the end of irradiation.

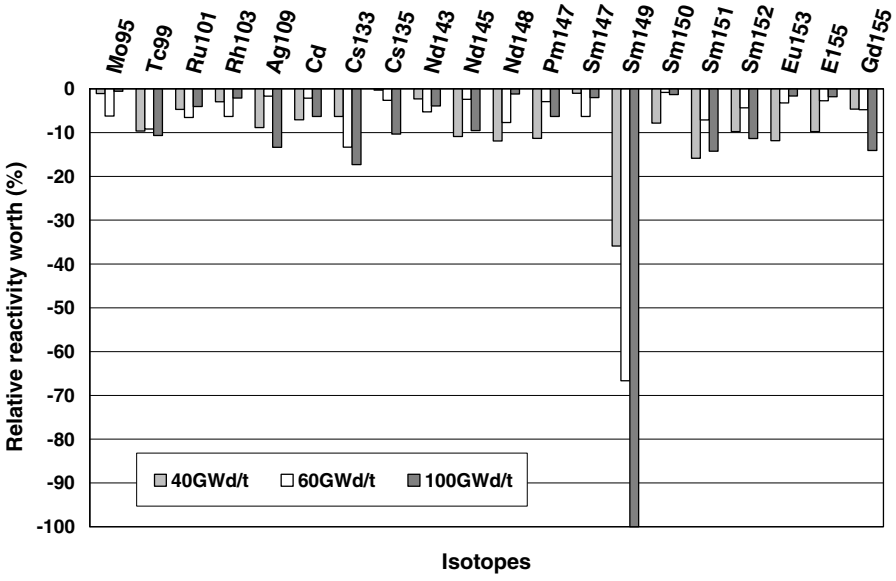

Fig. 2. Reactivity worth of fission products at the end of irradiation.

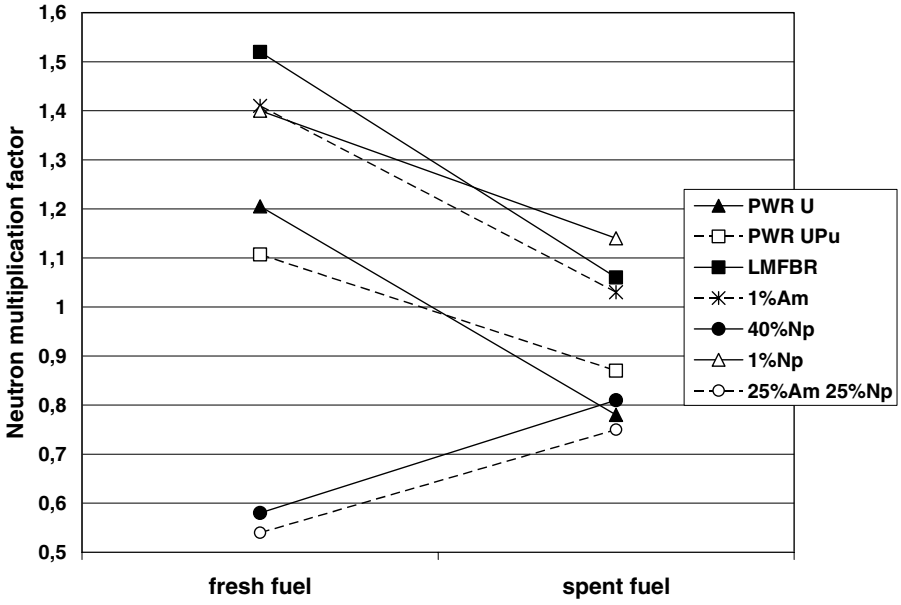

Fig. 3. Change of k_{eff} when calculated using the fresh and spent fuel compositions.

at EOI is considered in the computation; nevertheless, $k_{eff} < 1$ up to a burnup of 90 GWd/t. The increase in k_{eff} is attributed to the formation of fissile Pu during reactor operation, from neutron capture reactions in ^{237}Np present in high amounts in the two fresh targets. The neutron multiplication factor remains below one due to the negative reactivity of ^{149}Sm. The difference in the two transmutation fuels is due to the different amounts of fissile material present at EOI (Table 1).

The third group contains the fuels from the reference reactor cases of PWR-UO_2 and – MOX UPu. These fuels were clustered together between the first and second groups, having less fissile content than the former and less than the latter. The neutron multiplication factor decreases when it is calculated on the basis of the spent fuel composition (Fig. 3).

The effect of cooling of the spent fuel on the neutron multiplication factor was investigated. The k_{eff} is now calculated on the basis of the actinides and fission products in the transmutation spent fuel UPu – 1% ^{237}Np with 5 years cooling. The neutron multiplication factor is now 1.07 instead of the 1.14 calculated when the inventory at EOI was used. The decrease in k_{eff} is attributed to the increase of ^{155}Gd with cooling, from the decay of ^{155}Eu (half life 4.96 years). The fission product ^{155}Gd has a high thermal cross section for neutron capture (61,000 barns) resulting in a strong negative reactivity worth.

4. Conclusions

Criticality issues associated with the storage of spent nuclear fuel from the transmutation of ^{237}Np and ^{241}Am in a fast reactor have been investigated. The studies focussed on the neutron multiplication factor, computed using the compositions at BOI and EOI. The outcome of the investigation was compared to fuels from existing nuclear power stations.

The fuels based on a fast reactor fuel composition with the addition of 1% ^{237}Np or ^{241}Am resemble a commercial fast breeder reactor fuel. The multiplication factor decreases when the spent fuel compositions are used in the calculation of k_{eff}. The fuels based on depleted U with high amounts of ^{237}Np and ^{241}Am show a low value for k_{eff} when calculated using the composition at BOI. However, due to the formation of Pu during reactor operation, the multiplication factor increases when computed on the basis of the compositions at EOI.

The fissile isotopes ^{235}U, ^{239}Pu and ^{241}Pu exhibit a strong positive reactivity worth while the fertile ^{238}U a strong negative reactivity worth. Concerning the fission products, ^{149}Sm shows a rather prominent negative reactivity worth. The use of the actinide and fission product inventory at 5 years after EOI, reduces further the multiplication factor due to the increase of ^{155}Gd from the decay of ^{155}Eu.

The investigation demonstrates that the inclusion in such calculations of the minor actinides and fission products contained in the spent fuel may prove a cost down step with a considerable improvement in effectiveness of packaging such spent fuels.

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