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Energy Procedia 71 (2015) 237 – 243

Energy Procedia

The Fourth International Symposium on Innovative Nuclear Energy Systems, INES-4

Validation of ^{241}Am Capture Cross Section through Integral Test using Criticality Data of Light-water Moderated MOX Cores

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Abstract

In the development of JENDL-4.0, the thermal and resonance capture cross section of ^{241}Am was increased by about 8-9% to reduce the overestimation of criticality observed in the MOX-Fueled light-water-moderated cores using ^{241}Am-rich plutonium-fuel. The revised data gives the better results of criticality prediction for those cores than that with JENDL-3.3. The Pu ageing problem was also investigated using the revised data. This problem is that the calculated k-effs for critical MOX cores with different experiment dates showed the tendency of increase with time. The change of k-eff in seven years was about 0.2%dk/k with JENDL-3.3 and it was reduced to about 0.1%dk/k with JENDL-4.0. These results show that the revision of ^{241}Am capture cross section seemed to be successful, although there still remains the tendency of k-eff increase with time. On the other hand, the recent measurement of thermal and resonance capture cross section of ^{241}Am indicates that the data of JENDL-3.3 should be increased by about 18%, that is about 8% greater than that of JENDL-4.0.

In the present study, the possibility of further revision of ^{241}Am capture cross section was studied through the integral test for Pu ageing problem by reflecting the recent experimental data. Three criticality data of a light-water moderated MOX core with different experiment date (7 years interval at maximum) have been analyzed with JENDL-3.3 and 4.0 libraries. In addition, the calculations with the number density of ^{241}Am multiplied by 1.18 for JENDL-3.3 and by 1.08 for JENDL-4.0 were also conducted to simulate the increase of ^{241}Am capture cross section suggested by the recent measurement. The results show that the increase tendency of k-eff as a function of time observed in the calculations with JENDL-4.0 is not found in those with modified ^{241}Am number density. This means that the improvement of ^{241}Am capture cross section will be achieved by employing the recent experimental data.

Selection and peer-review under responsibility of the Tokyo Institute of Technology

Keywords: ^{241}Am capture cross section; Integral test; Criticality; MOX cores

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Selection and peer-review under responsibility of the Tokyo Institute of Technology
doi:10.1016/j.egypro.2014.11.875

1. Introduction

The critical size of a MOX core increases with time, since ^{241}Pu decays to ^{241}Am in the MOX fuel. We refer to this effect as the Pu ageing effect. Previous studies for the thermal MOX critical experiments with CEA's EOLE and JAEA's TCA, show that the multiplication factors for the critical cores increase with time[1-3]. This would be caused by the underestimation of ^{241}Am capture cross section, since the experimental results of ^{241}Am capture cross section show wide variation. The evaluated capture cross section of ^{241}Am in the JENDL has been increased to reduce the overestimation of criticality as shown in Table 1[4-7]. As seen in the table, the thermal capture cross section in the latest evaluated library, JENDL-4.0[8], increased by about 8-9% from that in the JENDL-3.3. However, the recent experiment[9] of thermal and resonance capture cross section measurements of ^{241}Am indicates that the data of JENDL-3.3 should be increased by about 18%, that is about 8% greater than that of JENDL-4.0.

In the present study, the possibility of further revision of ^{241}Am capture cross section was studied through the integral test using TCA experimental data for Pu ageing effect.

Table 1 Status of ^{241}Am Thermal Capture cross section

Reference	Data (b)	Issued	Comments
Mughabghab[5]	587 ± 12*	1983	Recommendation
Shinohara et al.[6]	854 ± 58	1997	Experiment
Fioni et al.[7]	696 ± 48	2001	Experiment
JENDL-3.2	600	1989	Evaluation
JENDL-3.3	640	2002	Evaluation
JENDL-4.0	684	2010	Evaluation

* Error of cross section

2. Critical Experiments at TCA

A series of critical experiments for light-water moderated MOX cores was performed using the Tank-Type Critical Assembly (TCA)[10-12]. The core, denoted as 2.42Pu, was constructed to investigate the change of criticality as a function of time. The core was composed of 3.0 wt% enriched PuO_2-UO_2 fuel rods and light water. The light water acted as a moderator and also as a reflector. The moderator-to-fuel volume ratio (V_m/V_f) of the core was 2.42. The core was constructed in the core tank by vertically positioning the fuel rods into square lattices and feeding water from the bottom of the tank. The specifications of the fuel rod and the core are shown in Tables 2 and Figs. 1 and 2.

Since the criticality is controlled by the height of light water, the change of critical water level with time was measured in the experiments. The measurements were performed over 7 years (in 1972 to 1978). During this experiment period of 7 years, the number density of ^{241}Am increased from about $1x10^{-6}$ to $9x10^{-6}$ (10^{24} atom/cm^3).

The measured results are shown in Table 3. In the criticality calculation, the critical water levels at 20°C were employed which were evaluated from the critical water levels at measured water temperature using the temperature coefficient of reactivity. As seen in the table, the critical water level increases with time, i.e., the increase of ^{241}Am, or the decrease of ^{241}Pu.

The experimental error in criticality was evaluated in Ref.[10] at about 0.5 %dk/k, and it is larger than the change of k-eff in 7 years. However, this error includes systematic errors mainly due to the uncertainties in fuel rod characteristics and lattice pitch. Such systematic errors would affect on the absolute value of k-eff, but not on its trend investigated here. Then, we have estimated the random error using the past experimental data for the reproducibility of a uranium critical core of TCA. It is estimated as 0.1%dk/k at maximum. Other errors, such as the error in temperature correction of critical water level (estimated at about 0.02%dk/k) and the error in half life of ^{241}Pu (estimated at 0.003%dk/k or less) are negligible.

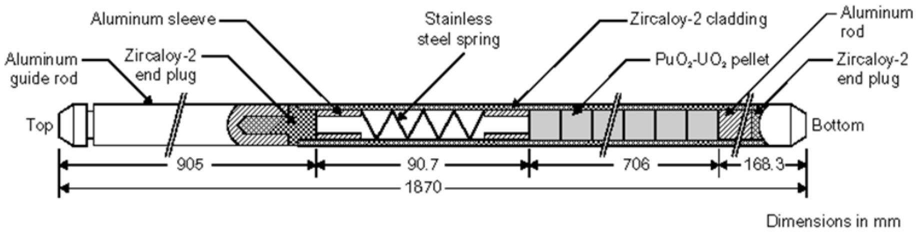

Fig. 1 Specifications of Fuel Rod

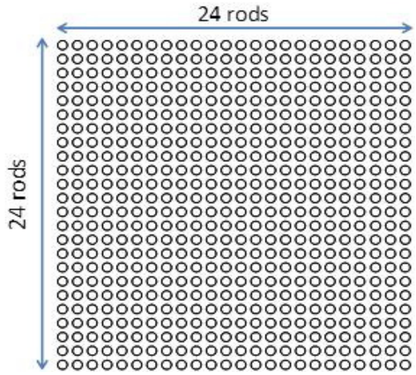

Fig. 2 Configuration of 2.42Pu core (Lattice pitch =1.825cm)

Table 2 Specifications of MOX pellet

Parameter	Value
PuO_2 enrich. (wt%)	3.01 ± 0.05
Uranium	Natural
Pu isotope (wt%)	(19-Aug-71)
^{238}Pu	0.494
^{239}Pu	68.18
^{240}Pu	22.02
^{241}Pu	7.26
^{242}Pu	2.04
^{241}Am (16-Aug-71)	530 ppm in PuO_2
Pellet density (g/cm^3)	6.056 ± 0.076
Impurity content	0.90 (+0.09, -0.12) ppm equivalent boron concentration in PuO_2-UO_2
Oxygen/Metal atom ratio	2.04
Pellet	
Diameter (mm)	10.65
Density (g/cm^3)	6.056 ± 0.076
Stack length (mm)	706 ± 3
Cladding	
Material	Zircaloy-2
Inner diameter (mm)	10.83 ± 0.06
Thickness (mm)	0.70 ± 0.07

Table 3 Criticality data of 2.42Pu core

Run #	Date	Elapsed time (y)	Hc (cm)	Temperature (°C)	Hc at 20°C (cm)
5111	07-Jun-72	0.80	53.29	19.6	53.30
5749	16-May-75	3.74	58.30	16.1	58.36
6378	24-Apr-79	7.68	65.18	15.2	65.27

Hc : Critical water level

3. Criticality Calculation

The effective neutron multiplication factor for each critical core was calculated using a continuous energy Monte Carlo code, MVP[13]. The Monte Carlo calculations were made on the full arrangement of each core in the tank, including both the fuel lattices under and over the critical water levels. The cross section libraries based on the JENDL-3.3[14] and 4.0 nuclear data libraries were used, respectively. In the calculation, the change of atomic number densities of ^{241}Pu, ^{241}Am, ^{238}Pu and ^{234}U in MOX fuel were evaluated using the following relations:

^{241}Pu → ^{241}Am (β-decay, half life = 14.35 y),

The atomic number densities at the date of Pu composition assay was used as the initial values, which were quoted from Ref. [10]. Note that the number densities of ^{238}Pu and ^{234}U also vary with time by ^{238}Pu → ^{234}U (α-decay, half life = 87.74 y), but this change is negligible, since the half life of ^{238}Pu is relatively long in comparison with the experiment periods of 7 years and the change of those number densities are very small.

The calculated results are plotted as a function of time in Fig. 3. Error bars in the figure show a standard deviation of the multiplication factor in Monte Carlo calculation. As seen in the figure, the effective multiplication factors (k-effs) are smaller than unity and increase with time. The bias (difference from unity) of JENDL-4.0 is smaller than that of JENDL-3.3. This bias difference is mainly caused by the improvement of capture cross sections of ^{238}U, ^{238}Pu, ^{239}Pu and ^{241}Am in thermal energy range for JENDL-4.0 (see Appendix A. for detail). The increase tendency of k-eff is improved for JENDL-4.0, although the tendency of increase with time still observed.

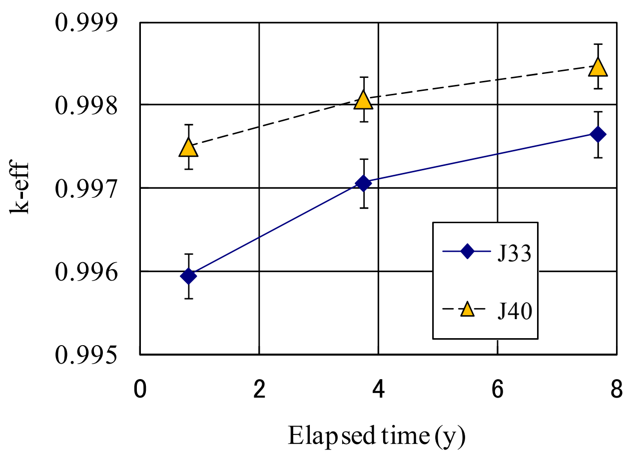

Fig. 3 Change of Effective Multiplication Factor (k-eff) as a Function of Time
(J33:JENDL-3.3, J40:JENDL-4.0)

4. Modification of ^{241}Am Number Density

In 2012, Schillebeeckx showed the experimental results of thermal and resonance capture cross section of ^{241}Am and indicated that the cross section in JENDL-3.3 should be increased by about 18%, that is about 8% greater than that in JENDL-4.0[9].

To investigate this effect of ^{241}Am capture cross section change, the number density of ^{241}Am was changed in this study. Because that the number density of ^{241}Am is quite low and the capture reaction is mainly occurs in thermal energy range, the self-shielding effect in ^{241}Am capture cross section will be negligible. Therefore, the change of cross section can be simulated by the change of number density in this case.

The k-effs calculated using JENDL-3.3 with the number density of ^{241}Am multiplied by 1.18 are shown in Fig. 4 and those using JENDL-4.0 with the number density of ^{241}Am multiplied by 1.08 in Fig. 5. It can be seen from those figures that the increase tendency with time almost disappears using the modified number density of ^{241}Am, although the variation range of k-effs with the modified number density in Fig.4 is larger than the statistical error of Monte Carlo calculation.

In Fig.5, the variation range of k-eff with the modified number density is in the statistical error of the calculation. Therefore, the increase of ^{241}Am thermal and resonance capture cross section in JENDL-4.0 by about 8% will resolve the problem caused by the Pu ageing effect observed in the thermal MOX cores.

The cause of different behavior of keffs with the modified number density in Figs.4 and 5 is not clear in the present study that employs the number density modification to simulate the cross section change. Further study that directly modifies the thermal capture cross section will be necessary to understand the difference.

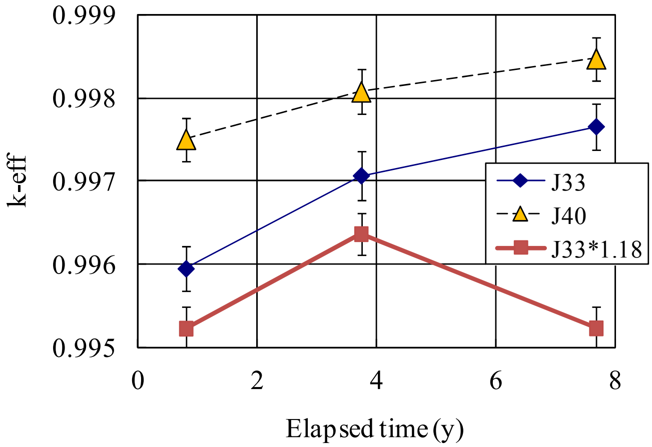

Fig. 4 Effect of Modified Number Density of ^{241}Am in JENDL-3.3
(J33*1.18: JENDL-3.3 with the number density of ^{241}Am multiplied by 1.18)

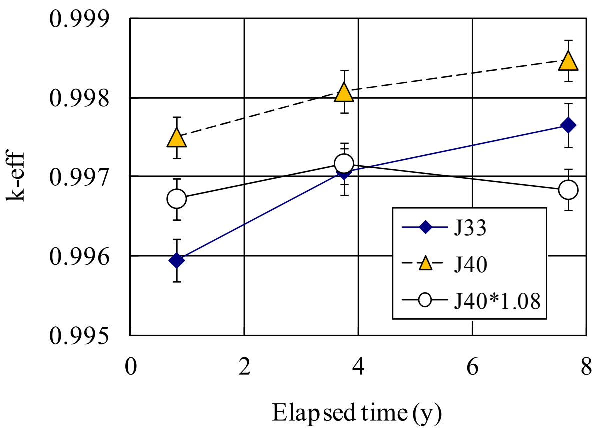

Fig. 5 Effect of Modified Number Density of ^{241}Am in JENDL-4.0
(J40*1.08: JENDL-4.0 with the number density of ^{241}Am multiplied by 1.08)

5. Conclusion

The possibility of further revision of ^{241}Am capture cross section was studied through the integral test for Pu ageing problem by reflecting the recent experimental data.

Three criticality data of a light-water moderated MOX core with different experiment date (7 years interval at maximum) have been analyzed with JENDL-3.3 and 4.0. In addition, the calculations with the number density of ^{241}Am multiplied by 1.18 for JENDL-3.3 and by 1.08 for JENDL-4.0 were also conducted to simulate the increase of ^{241}Am capture cross section suggested by the recent measurement.

The results shows that the increase tendency of k-eff as a function of time observed in the calculations with JENDL-3.3 and 4.0 is not found in those with modified ^{241}Am number density. This means that the improvement of ^{241}Am capture cross section will be achieved by employing the recent experimental data.

Appendix A. Components of the bias difference between JENDLE-4.0 and 3.3

In order to investigate the components of the bias difference discussed in Chap. 3, the change of k-eff was calculated for 2.42Pu core (Run5111) by replacing the cross section of a heavy nuclide from JENDL-4.0 to JENDL-3.3 while the other nuclides were those of JENDL-4.0. In the calculation, the same method and model were used as described in Chap. 3, and all heavy nuclides, i.e. ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am, were replaced, respectively. The comparisons of thermal capture cross sections for those nuclides are shown in Table A-1.

Figure A-1 shows the change of k-eff by replacing the cross section from JENDL-4.0 to JENDL-3.3. As seen in the figure, ^{238}U, ^{238}Pu, ^{239}Pu and ^{241}Am show the significant effect on the k-eff change. The nuclide ^{238}U shows the largest effect on the bias difference because of the highest number of nuclide although the change of thermal capture cross section is very small as shown in Table A-1.

Table A-1 Comparisons of thermal capture cross sections for heavy nuclides[8]

Nuclide	JENDL-3.3	JENDL-4.0
U235	98.71	98.71
U238	2.718	2.683
Pu238	540.2	412.9
Pu239	270.7	271.5
Pu240	289.3	289.3
Pu241	361.6	363.1
Pu242	18.77	19.89
Am241	639.5	684.3

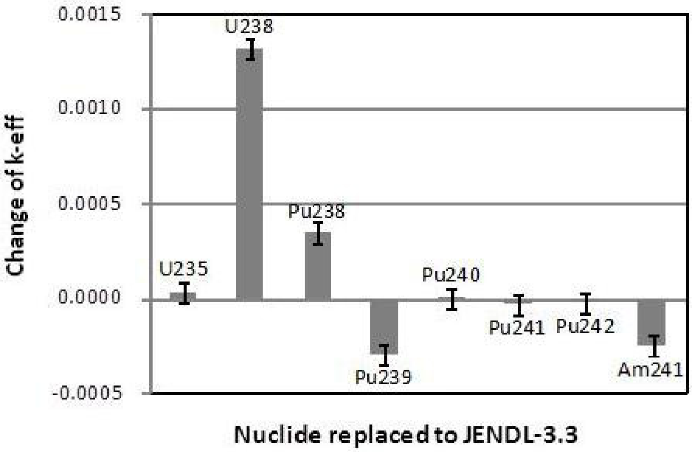

Fig. A-1 Change of k-eff by replacing the cross section from JENDL-4.0 to JENDL-3.3.

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