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Analysis of Criticality Change with Time for MOX Cores

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In plutonium-uranium mixed oxide (MOX) fuel, the composition changes with time due to the decay of ^{241}Pu (half life of 14.35 y) and build up of ^{241}Am. This change reduces the criticality of a MOX core with time because of the decrease in fissions and the increase in neutron captures. At the Japan Atomic Energy Research Institute (JAERI), a series of critical experiments for MOX cores had been performed, and criticality data as a function of time were obtained for about 7 years to investigate the effect of fuel composition change. We have analyzed the criticality change for those cores with a Monte Carlo code, MVP, employing the Japanese Evaluated Nuclear Data Libraries, JENDL-3.2 and 3.3.

The effective multiplication factors for the TCA critical cores showed the dependence on time, that is, they increased with time. This indicates that there exists some errors in cross sections of ^{241}Am and/or ^{241}Pu.

KEYWORDS: MOX, ^{241}Pu decay, ^{241}Am build up, critical experiments, criticality analysis, Monte Carlo code, MVP, JENDL-3.2, JENDL-3.3

1. Introduction

In MOX fuel, the composition changes with time due to the decay of ^{241}Pu and build up of ^{241}Am. This causes the decrease of fissions and the increase of neutron captures, and hence the critical size of the core increases with time. If the neutron cross sections of ^{241}Pu and/or ^{241}Am are inaccurate, the effective neutron multiplication factors, calculated using those cross sections, for the critical cores at different experiment date would vary depending on the date. Actually, the numerical analysis for the French MOX critical experiments, EPICURE and MISTRAL, in which the same MOX fuel rods were used with a total experimental period of about 7 years, showed that the multiplication factor for the critical cores increases with time.[1] This indicates that there exists some uncertainties in the cross sections of ^{241}Pu and/or ^{241}Am. However, those French experiments were performed with the different core configurations, and then the effect of core configuration difference is also included in the calculation. To investigate the effect of fuel composition change alone, it is desirable to use a series of experiments continued over a long period with the same core configuration.

At JAERI, a series of critical experiments for MOX cores had been performed, and criticality data with the same core configurations as a function of time over about 7 years were obtained to investigate the effect of the decay of ^{241}Pu and build up of ^{241}Am.[2,3] The present paper describes the analysis of criticality change for those cores performed with a Monte Carlo code, MVP, employing the Japanese Evaluated Nuclear Data Libraries, JENDL-3.2 and 3.3.

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2. Experiments

A series of critical experiments for light-water moderated MOX cores was performed using the Tank-Type Critical Assembly (TCA) at JAERI.[2,4] Four experimental cores, denoted as 2.42Pu, 2.98Pu, 4.24Pu and 5.55Pu, were 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 ratios (V_m/V_f) of the cores were 2.42, 2.98, 4.24, and 5.55, respectively. Each core was constructed in the core tank of TCA by vertically positioning the fuel rods into square lattices and feeding water from the bottom of the tank. The specifications of the fuel rods and the cores are shown in Tables 1, 2 and Figure 1.

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 1×10^{-6} to 9×10^{-6} (10^{24} atom/cm^3).

The measured results are shown in Figure 2. The abscissa axis is the elapsed time from the date of plutonium isotope composition assay (19-Aug-71). As seen in the figure, the critical water level increases with the increase of ^{241}Am, or the decrease of ^{241}Pu.

Table 1 Specifications of Fuel Rod

PuO_2 enrichment (wt%)	3.01 ± 0.05 [a)]
Uranium	Natural
Pu isotope (wt%)	
^{238}Pu	0.494 [b)]
^{239}Pu	68.18
^{240}Pu	22.02
^{241}Pu	7.26
^{242}Pu	2.04
Americium ^{241}Am	530 ppm [c)] in PuO_2
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

a) PuO_2/(PuO_2+UO_2),
b) Pu isotope composition was assayed on 19-Aug-71,
c) ^{241}Am was assayed on 16-Aug-71.

Table 2 Core configurations

ID	L_p (cm)	V_m/V_f	Rod #	H_c (cm)	Date
2.42Pu	1.825	2.42	24×24	53.30	07-Jun-72
2.98Pu	1.956	2.98	22×22	56.88	22-Nov-73
4.24Pu	2.225	4.24	21×21	53.41	14-Apr-72
5.55Pu	2.474	5.55	23×23	53.50	06-Jun-73

L_p: Lattice pitch, V_m/V_f: Moderator to fuel volume fraction, Hc: Critical water level, Date: Date of H_c measurement.

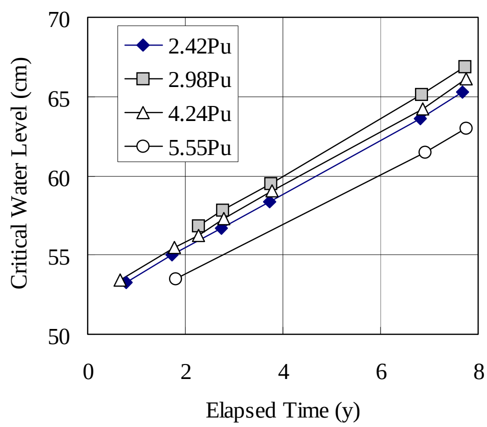

Fig.2 Critical Water Level as a Function of Time

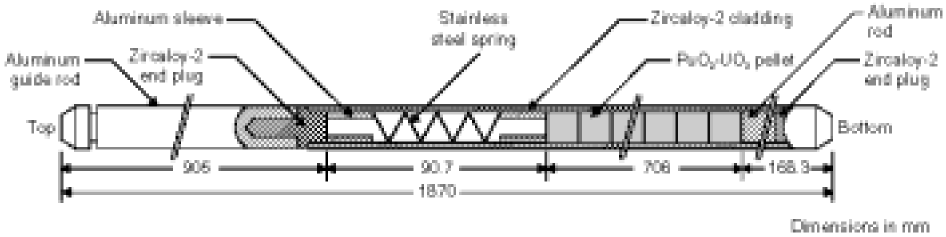

Fig.1 Specifications of Fuel Rod

3. Criticality Calculation

The effective neutron multiplication factor for each critical core was calculated using a continuous energy Monte Carlo code, MVP.[5] 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 and 3.2 nuclear data were used.[6,7] 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 $\rightarrow$ ^{241}Am (β-decay, half life = 14.35 y),

^{238}Pu $\rightarrow$ ^{234}U (α-decay, half life = 87.74 y).

The atomic number densities at the date of Pu composition assay was used as the initial values, which were quoted from Ref. [4].

The calculated results are plotted as a function of time in Figure 3. Error bars in the figure show a standard deviation of the multiplication factor in Monte Carlo calculation. Since all the calculations were performed for the cores at critical state, the effective multiplication factors should be unity (1.0). However, most of them are smaller than 1.0, and increase with time. The difference between the calculated value and 1.0, which is called a “bias”, would be caused by the ambiguity in the cross sections of major actinides (^{239}Pu and ^{235,238}U) and /or by the systematic error of the experiment. If the bias is constant for any time, we can correct the calculated results easily. But, as shown in the figure, the bias varies with time. The effective multiplication factor increases about 0.2 to 0.3 %dk/k in 7 years. This change is apparently larger than a standard deviation in the Monte Carlo calculation, which is less than 0.05 %dk/k. This indicates that there exists some errors in cross sections of ^{241}Am and/or ^{241}Pu. Note that the number densities of ^{238}Pu and ^{234}U also vary with time, but it is negligible, since the initial amount of ^{238}Pu is very small, and its half life (87.74y) is relatively long in comparison with the experiment periods of 7 years.

4. Discussion

4.1 Experimental Error

The experimental error in critical height measurement was evaluated in Ref.[4] at about 0.5 %dk/k, and it is larger than the change of multiplication factors in 7 years. However, this error includes the systematic errors mainly due to the uncertainties in fuel rod characteristics and lattice pitch. Such systematic errors would affect on the absolute value of the multiplication factor, or the “bias”, 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. Although this

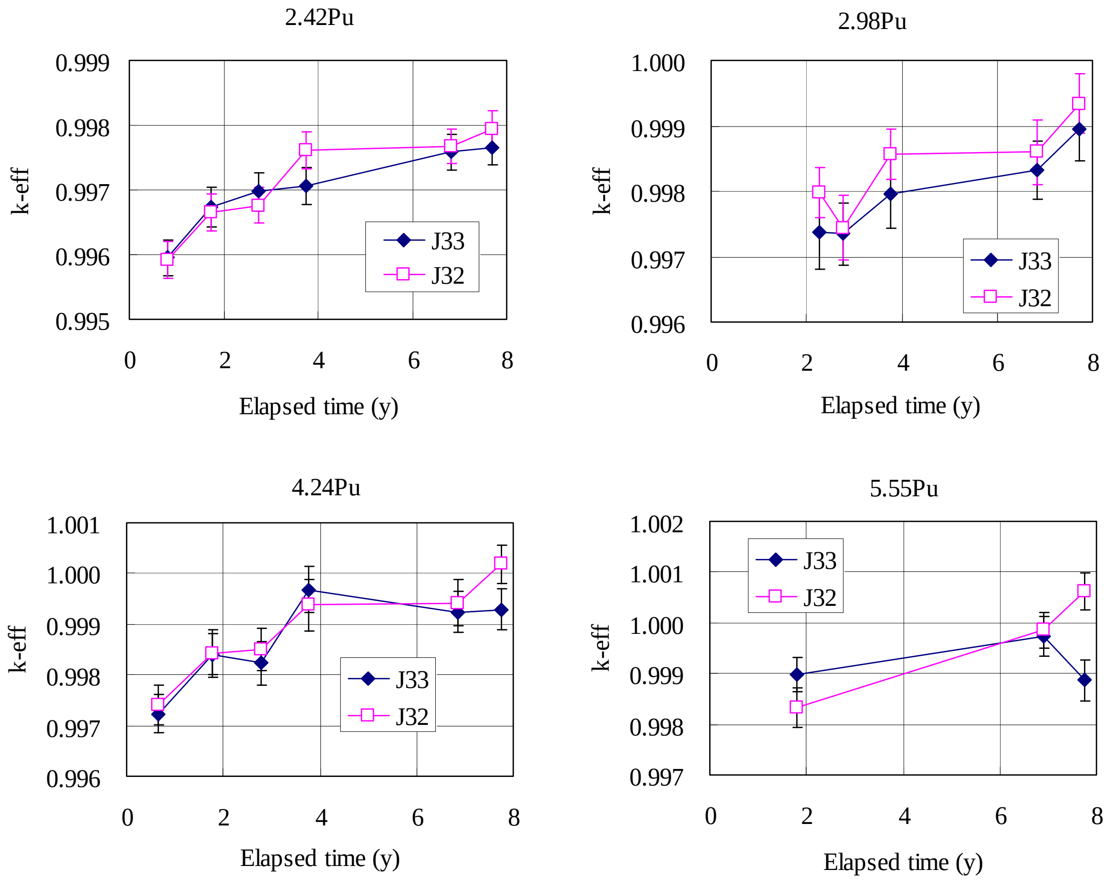

Fig.3 Change of Effective Multiplication Factor (k-eff) as a Function of Time

error is almost compatible with the change of multiplication factors, we can conclude that the trend of increase is significant one.

4.2 Library Effect

As seen in Fig.3, the effective multiplication factors using JENDL-3.2 and 3.3 almost agree with each other within a standard deviation. For the dependence on time, although it is very difficult to say, the calculation with JENDL-3.3 seems to show less dependence in comparison with that with JENDL-3.2. This difference would be caused by the improvement in the thermal capture cross section of ^{241}Am. Since the experimental data for thermal capture cross section of ^{241}Am varied widely, the accuracy of the evaluated cross section would not be high enough.[8] For the JENDL-3.3 library, the thermal cross section of ^{241}Am became about 6% larger than that of JENDL-3.2. On the other hand, the variations of experimental and evaluated values for the fission and capture cross sections of ^{241}Pu are relatively small. Thus, the probable cause of the time dependence of calculated criticality for MOX cores is the uncertainty in the capture cross section of ^{241}Am. However, it is necessary to obtain the calculated results with a much smaller standard deviation for the further investigation.

5. Conclusion

The analysis of criticality change with time for MOX cores has been performed to investigate the effect of fuel composition change due to the decay of ^{241}Pu and build up of ^{241}Am. The change of criticality for MOX cores over 7 years was measured at TCA, and they were analyzed with a Monte Carlo code, MVP, employing the Japanese Evaluated Nuclear Data Libraries, JENDL-3.2 and 3.3.

The calculated effective multiplication factors for the critical cores showed the dependence on time, that is, they increased with time. This indicates that there exists some errors in cross sections of ^{241}Am and/or ^{241}Pu. Although JENDL-3.3, in comparison with JENDL-3.2, slightly improves the time dependence of criticality by revising the thermal capture cross section of ^{241}Am, the calculation still shows the dependence. Further investigations are necessary to resolve this problem.

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References

1) T. Yamamoto, Y. Iwata, M. Ueji et al., "Analysis of MISTRAL experiments with JENDL-3.2," J. Nucl. Sci. Technol., Supplement 2, p.1061 (2002).
2) H. Tsuruta, I Kobayashi, T. Suzaki et al., "Critical sizes of light-water moderated UO_2 and PuO_2-UO_2 lattices," JAERI 1254, Japan Atomic Energy Research Institute (1978).
3) H. Sasajima, T. Matsumoto, R. Yumoto et al., "Experiment and analysis on reactivity decrease due to ^{241}Pu decay in light-water moderated PuO_2-UO_2 lattices," PNCT831-80-01, 86, Power Reactor and Nuclear Fuel Development Corporation (1980).
4) T. Yamamoto, "Critical arrays of mixed plutonium-uranium fuel rods with water-to-fuel volume ratios ranging from 2.4 to 5.6," MIX-COMP-THERM-004, International Handbook of Evaluated Criticality Safety Benchmark Experiments, NEA/NSC/DOC/(95)03, Volume VI, Nuclear Energy Agency, Organization for Economic Corporation and Development (1999).
5) T. Mori, M. Nakagawa, M. Sasaki, "Vectorization of continuous energy Monte Carlo Method for neutron transport calculation," J. Nucl. Sci. Technol., **29**, 325 (1992).
6) K. Shibata, T. Kawano, T. Nakagawa et al., "Japanese Evaluated Nuclear Data Library Version 3 Revision-3: JENDL-3.3," J. Nucl. Sci. Technol., **39**, 1125 (2002).
7) T. Nakagawa et al., "Japanese Evaluated Nuclear Data Library Version 3 Revision 2," J. Nucl. Sci. Technol., **32**, 1259 (1995).
8) T. Nakagawa, O. Iwamoto and A. Hasegawa, "Improvement of evaluated neutron nuclear data for ^{237}Np and ^{241}Am," JAERI-Research 201-059, Japan Atomic Energy Research Institute (2002).