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SHORT NOTE

Analysis of Curium Isotopes in Mixed Oxide Fuel Irradiated in Fast Reactor

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Curium is one of the key elements in recycling and transmutation of minor actinides (MA) because of its high radiotoxicity and difficulty of transmutation. In order to make isotopic analysis of curium in heavily irradiated fuel, the isolation technique of curium was developed by adopting anion exchange chromatography in nitric acid-methanol mixed media. The technique was successfully applied to the analysis of curium in mixed oxide (MOX) fuel irradiated in the experimental fast reactor "JOYO". The transmutation behaviors of curium in fast reactor are discussed on the basis of observed isotopic ratio of curium.

1. Isolation of Curium from Irradiated MOX Fuel

Mixed oxide fuel, initial plutonium content and uranium enrichment of 29.97 and 8.3 wt%, respectively, were irradiated up to 120 GWd/t in "JOYO". The isotopic analysis of curium, uranium, plutonium, and neodymium were performed to examine the transmutation behavior of curium and the burnup measurement.[1)]

Anion exchange separation with nitric acid-methanol mixed medium[2)] was selected and optimized for establishment of the curium isolation technique from irradiated fuel. The procedure for curium isolation from irradiated MOX fuel has been decided as shown in **Fig. 1** considering the easiness of the handing the highly radioactive sample.[1,3)]

The experimental conditions were as follows. The Am/Cm/FPs fraction was separated from irradiated fuel as an 8 M (mol/dm^3) nitric acid solution by the method reported in Ref. 1). A glass column (ϕ5 mm$\times$10 cm) packed with anion exchange resin (Bio-Rad 1$\times$4 200–400 mesh) was prepared for curium isolation. The mixed solution of 8 M nitric acid and methanol (1:9) containing samples was used as loading solution, and a mixed solution of 0.013 M nitric acid and methanol (1:9) was used as eluent.

Figure 2 shows an example of chromatogram, and alpha spectra of the Am/Cm/FPs fraction and curium fraction were shown in **Fig. 3**. High efficiency for the mutual separation of curium and americium was attained and most fission products were eluted before curium elution. However europium, which have high γ-radioactivity caused from its relative high fission yield, was mixed to curium fraction in one step chromatographic separation. The further removal of europium from the curium fraction by repeating the chromatographic separation was necessary to reduce the γ-radioactivity of the curium sample subjected to isotopic analysis.

2. Isotopic Composition of Curium Formed in MOX Fuel Irradiated in JOYO

The result of these determined curium contents were shown in **Table 1**. A very small amount of ^{247}Cm was detected and its content was less than 1×10^{-7} at%.

Figure 4 shows the contents of curium and each isotope in all of specimens as a function of burnup. The content of curium shows to become equilibrium above burnup of 100 GWd/t except ^{247}Cm, which shows to decrease with an increase in burnup. This behavior should be inquired into more details with taking account of the error of determined values and difference of neutron spectrum condition caused by loaded position.

3. Evaluation of Transmutation Behavior of Curium

In order to evaluate the transmutation behavior of curium quantitatively, effective half life T_{ei} of each curium isotope in the fast reactor was evaluated using the observed isotopic composition of curium in Sample No. 4 whose axial loaded position was center of core region. Each T_{ei} indicates the half life of each curium isotope under the conditions which these isotopes are loaded to the reactor individually. The T_{ei} was defined as,

$$T_{ei} = \ln(2)/(\sigma_{ei}\times\phi+\lambda_i), \quad (1)$$

where T_{ei}: Effective half life of nuclide i
σ_{ei}: Effective 1 group total (capture and fission) cross section of nuclide i
ϕ: 1 group neutron flux at irradiated position[4)]
λ_i: Decay constant of nuclide i.

Effective half life of each curium nuclide was determined by iteration procedure to adjust the calculated isotopic composition of curium with the observed values within 1% discrepancy. Calculations were carried out using ORIGEN2[5)] code using 1 group flux[4)] and effective cross section library[6)] prepared for "JOYO" as initial values.

Table 2 shows evaluated effective half life of each curium isotope in "JOYO". It is remarkable that evaluated effective half life of ^{245}Cm and ^{246}Cm were much smaller than prepared value for JOYO.[6)] In addition, the evaluated effective half life falls in narrow range, 1–7 years.

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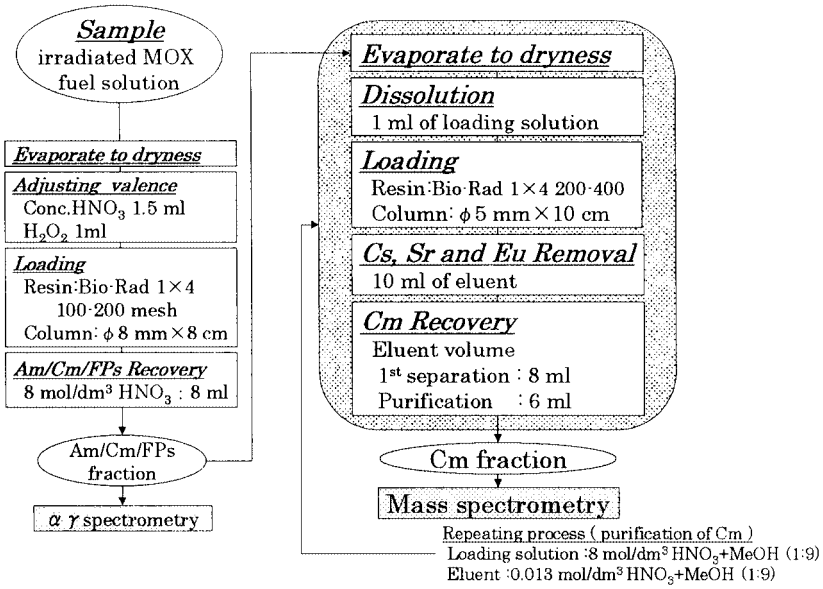


Fig. 1 Flow sheet of curium isolation from irradiated MOX fuel

Table 1 Results of isotope analysis of curium

Sample No.	Axial loaded position[a] (mm)	Burnup (GWd/t)	Isotopic composition (at%)					Total curium in MOX (at%)
			^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	
1[b]	+250	66.5	29.3	69.1	1.51	0.017	0.00045	1.43×10^{-3}
2[b]	−250	82.6	22.4	75.0	2.61	0.042	0.00061	3.99×10^{-3}
3	0	105.5	28.9	68.9	2.18	0.040	0.0017	3.79×10^{-3}
4[b]	0	113.6	29.0	68.8	2.22	0.043	0.0011	3.17×10^{-3}
5	0	119.7	28.6	69.0	2.31	0.046	0.00078	3.07×10^{-3}

[a] Distance from core center
[b] Loaded to the same fuel pin

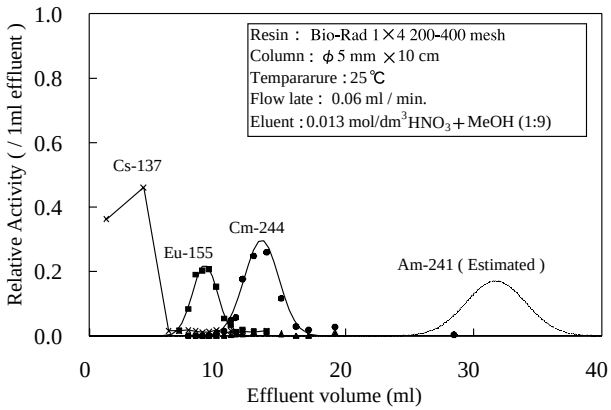


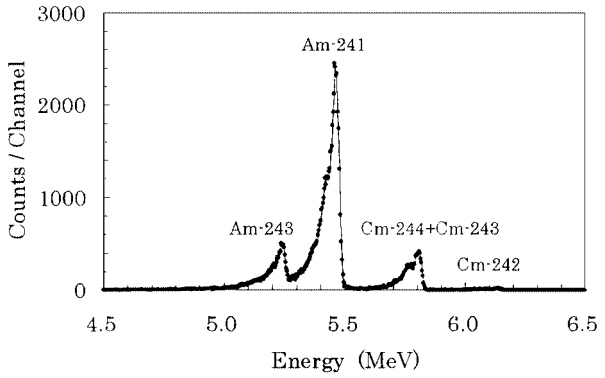
Fig. 2 Chromatogram of present technique

Table 2 Evaluated effective half life of curium isotopes

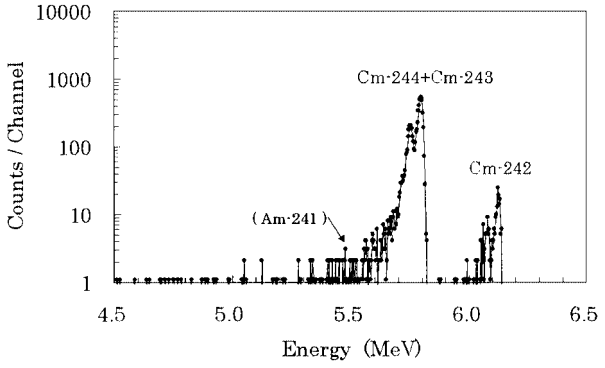
Nuclide	Total cross section (barn)		Effective half life (year)	
	Evaluated value	JOYO[6]	Evaluated value	JOYO[6]
^{243}Cm	2.79	2.78	2.18	2.18
^{244}Cm	1.22	1.06	4.15	4.61
^{245}Cm	6.30	2.50	1.04	2.63
^{246}Cm	0.99	0.62	6.64	10.6
^{247}Cm	1.37	2.44	4.82	2.69

4. Conclusion

The analytical technique of curium isotopes was established and successfully applied to the analysis of isotopic composition of curium that was formed in the mixed oxide (MOX) fuel irradiated in the experimental fast reactor "JOYO". The content of curium shows to become equilibrium above burnup of 100 GWd/t except ^{247}Cm. Very small amount of ^{247}Cm was observed. The effective half life of each curium isotope was evaluated from analyzed value.



(a) Loaded sample



(b) Curium fraction

Fig. 3 Alpha spectrums of loaded sample and curium fraction of preliminary test

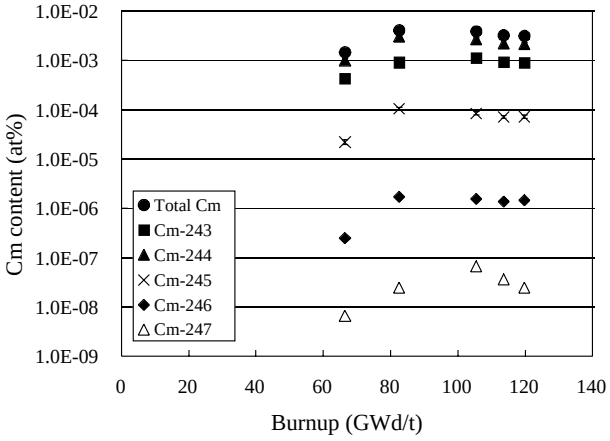


Fig. 4 Curium content changes as a function of burnup

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