Proceedings of the Korean Nuclear Society Spring Meeting
Cheju, Korea, May 1996

Transmutation of Am-241, 243 and Cm-244 in a Conventional Pressurized Water Reactor

Duck Joon Koh, Mynug Chan Lee and Woo Tae Jeong
Korea Atomic Energy Research Institute

Boris P. Kochurov
Institute of Theoretical and Experimental Physics, Russia

Abstract

The feasibility study on burning Am-241, 243 and Cm-244 nuclides in a conventional PWR (Pressurized Water Reactor) was carried out by using the TRIFON code that was developed by the Institute of Theoretical and Experimental Physics in Russia in 1992. TRIFON code uses updated ABBN Russian nuclear cross section library. The reference reactor is the Korea nuclear power plant unit 8 (YGN 2). The burning effect of Am-241, 243 and Cm-244 nuclides was studied with UO_2 (3.5 w/o)fuel assembly and MOX (4.44 w/o) fuel assembly. The loaded mass ratio of Am-241,243 and Cm-244 nuclides was obtained from the mass ratio of Am-241,243 and Cm-244 nuclides in 10 year cooling spent fuel with average discharge burnup of 33 GWD/MTU. The effective transmutation rates of Am-241, 243 and Cm-244 nuclides in UO_2 fuel assembly were found to be higher than those in MOX fuel assembly. The result from TRIFON code was compared to that from CASMO-3/NEM-3D code system. For more reliable calculation of transmutation for MA(Minor Actinides) more sophisticated decay chain scheme of MA should be investigated and nuclear cross section library of MA should be considerably improved.

1. Introduction

Nuclear transmutation of long-lived radionuclides into short-lived radionuclides is an attractive option which may alleviate the burden of geologic disposal. If these radionuclides get into the environment as the result of an accident or in violation of spent fuel storage regulations, the consequences may be extremely severe. The leakage accident of high-level nuclear waste shall be lead to drastic changes in the strategy of handling spent fuel. Before Chernobyl and TMI nuclear accident, main attention was paid to providing safe storage of the nuclear waste , in the recent years it turned to research for technical means of efficient treatment of the most dangerous radionuclides by converting them in reactors with the help of neutrons (or in accelerators) into nuclides that are either stable or have short half lives. In order to investigate the feasibility of transmutation of Am and Cm isotopes in a conventional Pressurized Water Reactor (PWR), we used TRIFON code for the

calculation of number density as a function of burnup of MA isotopes. TRIFON code was developed as an universal code for the calculation of detailed space and energy distribution of neutrons in a multi- region reactor cell with nuclear chain transformation depending on time variable. This code uses updated ABBN Russian nuclear cross section library. Performance of this library was reported to be equivalent to that of ENDF-B/VI and JENDL-3.2 nuclear cross section libraries[1]

2. Characteristics of TRIFON Code

TRIFON code is based on the utilization of a library of microscopic nuclear data as follows :

- multigroup(26 group) microscopic cross sections
- parameters of resonances and effective resonances
- scattering matrices for neutron thermalization and tables giving the dependence of absorption and fission cross sections on energy.

The data for chain transformations(channels for capture, fissions, decay, (n,2n) reactions) are included in the library of TRIFON. This code was tested by comparisons with the calculation results of reaction rates for TRX cells by other Monte-Carlo codes (MCU, WIMS) and by comparisons with the calculation results of conversion ratios, k_∞, neutron density ratios and neutron flux ratios for critical experiments in heavy water critical assemblies and graphite-moderated , light water cooled critical assemblies(RBMK). Neutron transport in every group is evaluated by CPM in a multi-region cylindrical geometry and by S_n method in a r-z geometry. Neutron transport equation is solved for space-energy distribution of neutrons. The source of neutrons in every group in epithermal region consists of two parts. The first part is due to neutrons scattered from the upper group and the second part is due to fissions corresponding to fission spectrum with the normalization of space and energy distribution to unity. Space distribution of generated neutrons by fission reactions is estimated according to a pre-calculated distribution of fissions by thermal neutrons. This is a weak supposition since in a cell of small dimensions fast neutrons have large mean free paths, the solution weakly depends on their space distribution and this problem can be improved by iteration procedure.

3. Calculational Methods

The main purpose of this calculation is to investigate the transmutation rates of Am-241, Am-243 and Cm-244 at the optimal loading patterns which are found by a trial and error technique. For this purpose, several loading methods were tested such as single-element loading method and mixed-element loading method with UO_2 fuel and with MOX fuel, respectively. In a single-element loading method, each element among Am-241, Am-243 and Cm-244 was assumed to be separately loaded into UO_2 and into MOX fuels, respectively. In a mixed-element loading method, we loaded a mixed compound of Am-241, Am-243 and Cm-244 into UO_2 and MOX fuels, respectively. The loading mass for Am-241, 243 and Cm-244 in the single-element loading was 2523 g, 1225 g and 348 g, respectively. The total loaded mass of Am-241, 243 and Cm-244 in the mixed-element loading was 4.096 kg. These masses are equivalent to the masses of Am-241, 243 and Cm-244 in a spent

fuel assembly with averaged discharge burnup of 33,000(MWD/MTU) and 10 year cooling before reprocessing. In all cases of these calculations, Am-241, 243 and Cm-244 remained in MOX and UO_2 fuel assemblies without reprocessing and refabrication during 3 cycles.

In a transmutation analysis of MA's with MOX fuel, we performed the color set calculation. The color set consists of 1 MOX fuel assembly and 3 UO2 fuel assemblies[2]. In the determination of the content of Pu in MOX fuel we adopted the SIEMENS' design data for MOX fuel - 4.44 w/o Pu(total) in a MOX fuel assembly is equivalent to 3.5 w/o U-235 in UO2 fuel assembly[3].

4. Results

4.1 Single -Element Loading Method

4.1.1 MOX Fuel Assembly(case 1)

Table 2-1 shows that the computed results of transmutation of Am-241, Am-243 and Cm-244 when each isotope was loaded separately in MOX fuel assembly. As shown in Table 2-1, Am-241 and Am-243 were accumulated with cycles. The buildup of Am-241 and Am-243 are thought to be caused by the neutron capture of Pu-239 in MOX fuel assembly due to the hardened neutron spectrum[4]. Such buildup can be deduced from the comparision of nuclear cross sections. Table 2-2 and Table 2-3 show the microscopic cross sections which are produced by the TRIFON code in the calculation process. As suggested in Table 2-2 and Table 2-3, the microscopic capture cross sections of Pu-240 which after neutron capture becomes Am-241 through β-decay was gradually increased with burnup and these values are greater by a few hundreds times than the microscopic capture and fission cross sections of Am-241. The mass of Cm-244 has decreased about 38.2 % during 3 cycles.

Table 2-1. Transmutation rates of Am-241, Am-243 and Cm-244 loaded separately into MOX fuel.

Isotopes	Cycle #	BOC Mass(g)	EOC Mass(g)	Burn Up (Mwd/kgU)	Transmutation Rate (%)
Am-241	4	2523	2343	11.2	-7.1
	5	2343	4378	12.2	+86.9
	6	4378	8159	11.4	+86.4
	Total	2523	8159	34.8	+223.4
Am-243	4	1225	848	11.2	-30.8
	5	848	1025	12.2	+20.9
	6	1025	2326	11.4	+126.9
	Total	1225	2326	34.8	+89.9
Cm-244	4	348	300	11.2	-13.8
	5	300	255	12.2	-15.0
	6	255	215	11.4	-15.7
	Total	348	215	34.8	-38.2

* Transmutation rate = { EOC Mass(g) - BOC Mass(g) }/BOC Mass(g) x 100

Table 2-2. Microscopic capture, fission cross sections and capture to fission ratios of Pu-240

	σ_c^1	σ_c^2	σ_f^1	σ_f^2	α^1	α^2
4 cycle	0.00493	0.00890	0.00083	0.00083	5.930	10.72
5 cycle	0.00740	0.01506	0.00146	0.00146	5.069	10.32
6 cycle	0.00924	0.02061	0.00195	0.00195	4.738	10.57

* 1 : epithermal neutron group
  2 : thermal neutron group , $\alpha = \sigma_c / \sigma_f$

Table 2-3. Microscopic capture, fission cross sections and capture to fission ratios of Am-241

	σ_c^1	σ_c^2	σ_f^1	σ_f^2	α^1	α^2
4 cycle	0.00059	0.00080	0.00001	0.00002	59.0	40.0
5 cycle	0.00106	0.00148	0.00003	0.00003	35.3	49.3
6 cycle	0.00191	0.00276	0.00005	0.00005	38.3	55.2

4.1.2 UO_2 Fuel Assembly(case 2)

Table 3 shows that the computed results of transmutation of Am-241, Am-243 and Cm-244 in UO_2 fuel assembly, respectively. The calculation results of case 2 are much different from those of Case 1. The buildup phenomena of target nuclides did not occur in Case 2 calculation. The main reason of this diminished buildup phenomena of Am-241 and Am-243 are that the precursor isotopes of Am-241 and Am-243 shall be less contained in UO_2 fuel than in MOX fuel. Moreover, the neutron spectrum in the UO_2 fuel assembly is much softer than that in the MOX fuel assembly.

Table 3. Transmutation rates of Am-241, Am-243 and Cm-244 loaded separately into UO_2 fuel.

Isotopes	Cycle #	BOC Mass(g)	EOC Mass(g)	Burn Up (Mwd/kgU)	Transmutation Rate (%)
Am-241	4	2523	1428	11.2	-43.4
	5	1428	575	12.2	-59.7
	6	575	243	11.4	-57.7
	Total	2523	243	34.8	-90.4
Am-243	4	1225	854	11.2	-30.3
	5	854	452	12.2	-47.1
	6	452	295	11.4	-35.0
	Total	1225	202	34.8	-75.9
Cm-244	4	348	302	11.2	-13.2
	5	302	253	12.2	-16.2
	6	253	220	11.4	-13.0
	Total	348	220	34.8	-36.8

4.2 Mixed Loading Method

4.2.1 MOX Fuel Assembly(Case-3)

Table 4 shows the calculation results of mixed Am-241, 243 and Cm-244 in MOX fuel assembly. All Am-241, 243 and Cm-244 nuclides were buildup in the calculation results of case-3. The main reasons of these buildup phenomena were caused by following facts. The first fact is the build up of precursor nuclide of Am-241, 243 and Cm-244 due to Pu-239 in MOX fuel assembly. The second fact is due to the difference of magnitude of capture and fission cross sections Am-241, 243 and Cm-244.

Table 4 . The calculated results of mixed burning effect of case-3

	Mass of 4 cycle (g)		Mass of 5 cycle (g)		Mass of 6 cycle (g)		Transmutation rate (%) (period = 3 cycles)
	BOC	EOC	BOC	EOC	BOC	EOC	
Am-241	2523.0	2162.2	←	4072.8	←	7884.4	+ 212.5
Am-243	1225.0	829.2	←	940.2	←	2173.8	+ 77.5
Cm-244	348.0	679.4	←	895.2	←	1351.3	+ 288.3
Total	4096.0	3670.8	←	5908.2	←	11,409.5	+ 178.6
k_∞	1.486	1.283	←	1.235	←	1.077	-

4.2.2 UO_2 Fuel Assembly(Case-4)

Table 5 shows the calculation results of mixed burning effect of Am-241, 243 and Cm-244 when Am-241, 243 and Cm-244 were homogeneously mixed and loaded in UO_2 fuel assemblies. According to the results, total transmutation rate was 64.1% during 36 months. The annual transmutation rate is equivalent to 21.36 %. The transmuted mass of Am-241 and Am-243 have increased, but the transmuted mass of Cm-244 has decreased to 2.7 times compared to initial mass. This phenomena was caused by the difference of microscopic capture and fission cross sections of Am-241, An-243 and Cm-244. The microscopic capture cross sections of Am-241 and Am-243 are much higher than the microscopic fission cross section of Am-241 and Am-243. The difference of capture and fission cross sections of each nuclide leads to the buildup of Cm-244.

Table 5. The calculated results of mixed burning effect in UO_2 fuel assembly(Case-4).

	Mass of 4 cycle (g)		Mass of 5 cycle (g)		Mass of 6 cycle (g)		Transmutation rate (%) (period = 3 cycles)
	BOC	EOC	BOC	EOC	BOC	EOC	
Am-241	2523.0	1367.0	←	646.5	←	244.4	- 90.3
Am-243	1225.0	808.8	←	506.5	←	296.0	- 75.8
Cm-244	348.0	688.1	←	874.0	←	929.6	+ 167.1
Total	4096.0	2863.9	←	2027.0	←	1470.0	- 64.1
k_∞	1.439	1.287	←	1.195	←	1.077	-

* Transmutation rate (%) = {mass of 6 cycle(BOC) - mass of 4 cycle(BOC)}x 100/ mass of 4 cycle (BOC)

5. Discussion and Conclusion.

When transuranium nuclides are mixed homogeneously to target fuels, initial excess reactivity and burnup reactivity swing become remarkably small, and high conversion ratio is obtained. Because long-lived radioactive MA such as Am and Cm make instead of burnable poison at the initial state, and they are gradually transmuted to fissile materials with burnup. The calculation results show that the MA nuclides are not waste , but incinerated usefully as poison and burning fuels.

Table 6 and Table 7 show the total transmutation rate calculated by CASMO/NEM and TRIFON code systems for Case-3 and Case-4, respectively. The calculation results from CASMO/NEM and TRIFON in Case-3 show disagreement. However, the calculation results in Case-4 show a good agreement. The effective transmutation rates of Case 4 (UO_2 fuel assembly and mixed loading method) is superior than that of the other three Cases(case 1~3) for Am-241, Am-243 and Cm-244. Especially, the generation of americium and curium isotope from plutonium isotope is significant in MOX assembly. It can be concluded that, even disregarding the inherent difficulties associated with the buildup and decay process of MA, transmutation of Am-241 and Am-243 in PWR-MOX thermal reactors does not lead to a significant reduction but significant buildup of their mass in MOX fuel.

To get a considerable reduction of the radiotoxicity due to MA , high thermal and fast neutron flux is required. The superiority of recycling MA elements in fast reactors, in a multiple recycling scheme, had been showed already in Reference 5. Thermal reactors are more efficient in transmuting MA elements in other ones, and the benefit on waste activity is not obvious. However,

as most present reactors are LWRs, it is of prime importance for electricity utilities, fuel manufacturers and decision makers to know more about the capabilities of LWRs to recycle plutonium and MA[6]. For more reliable calculation of transmutation for MA, more sophisticated decay chain scheme of MA should be investigated and nuclear cross section library of MA should be considerably improved.

Above all things, it should be investigated that the cost evaluation and comparision of the fuel cycle between MA recycling in PWR with a dedicated system is inevitable for wise judgement of a useful transmutation system.

Table 6. Comparison of the Results of CASMO-3/NEM-3D and TRIFON for Case-3 Study.

Actinide Element	Burn-up (Mwd/kgU)	Total transmutation rate (%) (CASMO-3)/ (NEM-3D)	Total transmutation rate(%) (TRIFON)	Burn-up time
Am-241	34.8	- 72.0 %	+ 212.5 %	36 months
Am-243	34.8	- 18.8 %	+ 77.5 %	36 months
Cm-244	34.8	+169.3 %	+ 288.3 %	36 months

Table 7. Comparison of the Results of CASMO-3/NEM-3D and TRIFON for Case-4 Study.

Actinide Element	Burn-up (Mwd/kgU)	Total transmutation rate(%) (CASMO-3)/ (NEM-3D)	Total transmutation rate(%) (TRIFON)	Burn-up time
Am-241	34.8	- 92.0 %	- 90.3 %	36 months
Am-243	34.8	- 61.8 %	- 75.8 %	36 months
Cm-244	34.8	+145.5 %	+ 167.1 %	36 months

References

1. B. P Kochurov and A. Yu. Kwaratzhely., " Computer code TRIFON," VANT, ser : PHTYAR (Russia), 1985.
2. M.C Lee et al., " Feasibility Study on Transmutation of MA in Conventional Fission Power Reactors," 3rd OECD/NEA Information Exchange Mtg. on P-T, Cadarache, Dec 12-14,1994.
3. Expert Group in OECD/NEA, " Plutonium Fuel- An Assessment," OECD,1989.
4. Hiroshi AKIE, Keisuke OKUMURA and Hideki TAKANO, " Improvement on Burnup Chain Model and Group Cross Section Library in The SRAC System," JAERI-1323, Jan., 1992.
5. Th. Maldague and J. Journet at al, "Reduction of Nuclear Waste Toxicity by Actinide Recycling in Fast and Thermal Reactors", SAFEWASTE Conference, Avignon, June 1993.
6. Th. Maldague, S. Pilate and A. Renard, " Core Physics Aspects and Possible Loading for Actinide Recycling in LWRs," International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles France, Sep., 11-14,1995.