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Concentrated recycling of MAs is evaluated for these fuels, where additional enrichment is determined to compensate for reactivity penalty due to MAs.

Fuel material balance, transmutation rate, reactivity coefficients, and other fuel cycle effects are discussed.

It is concluded that, though transmutation rate is highest in UO₂ fuel, MOX fuel is most suitable as MA-carrier, because of its minimum fuel cycle effects.

TRANSMUTATION OF MINOR ACTINIDES IN BOILING WATER REACTORS

K. Hida¹, Y. Ando¹ and R. Yoshioka²

¹ Nuclear Engineering Laboratory, Toshiba Corporation
4-1, Ukishima-cho, Kawasaki-ku, Kawasaki 210, JAPAN

² Isogo Engineering Center, Toshiba Corporation
8, Shinsugita-cho, Isogo-ku, Yokohama 235, JAPAN

ABSTRACT

Transmutation of minor actinides (MAs) is investigated for two types of BWR fuel (UO₂ and MOX) and for an HCR fuel. Irradiation calculation for pure MAs shows that, while transmutation by fission is scarce, half-lives of principal daughter actinides are much shorter than those of parent MAs, indicating potential transmutation of MAs in these reactors. Concentrated recycling of MAs is evaluated for these fuels, where additional enrichment is determined to compensate for reactivity penalty due to MAs. Fuel material balance, transmutation rate, reactivity coefficients, and other fuel cycle effects are discussed. It is concluded that, though transmutation rate is highest in UO₂ fuel, MOX fuel is most suitable as MA-carrier, because of its minimum fuel cycle effects.

1. INTRODUCTION

Transmutation of minor actinides (MAs) is important to reduce long-term hazard for geological disposal of

high level wastes (HLWs). Since fission-to-capture cross section ratios of MAs are larger for higher energy neutrons, it seems natural to recycle them in fast reactors to be converted into fission products (FPs). However, it can be expected to transmute MAs into other actinides of shorter half-lives by means of neutron capture. In this sense, recycling MAs in thermal reactors is worth studying, taking advantage of large neutron capture cross sections and high thermal neutron flux level.

This study evaluates potential transmutation of MAs in current BWR and high conversion BWR (HCR).¹ Two types of fuel are considered for BWR: uranium-oxide (UO₂) and mixed-oxide (MOX) fuels. UO₂ fuel can be expected to be efficient in MA transmutation, because of its high thermal neutron flux level. On the contrary, MOX fuel is favored, because the increased radiation dose rate due to MAs can be covered by that due to plutonium isotopes. One of the purposes is to compare these two types of fuel as MA-carrier, from the viewpoint of overall fuel cycle effects. For HCR, highly-enriched MOX-type fuel is evaluated, to study neutron spectrum dependence of MA transmutation.

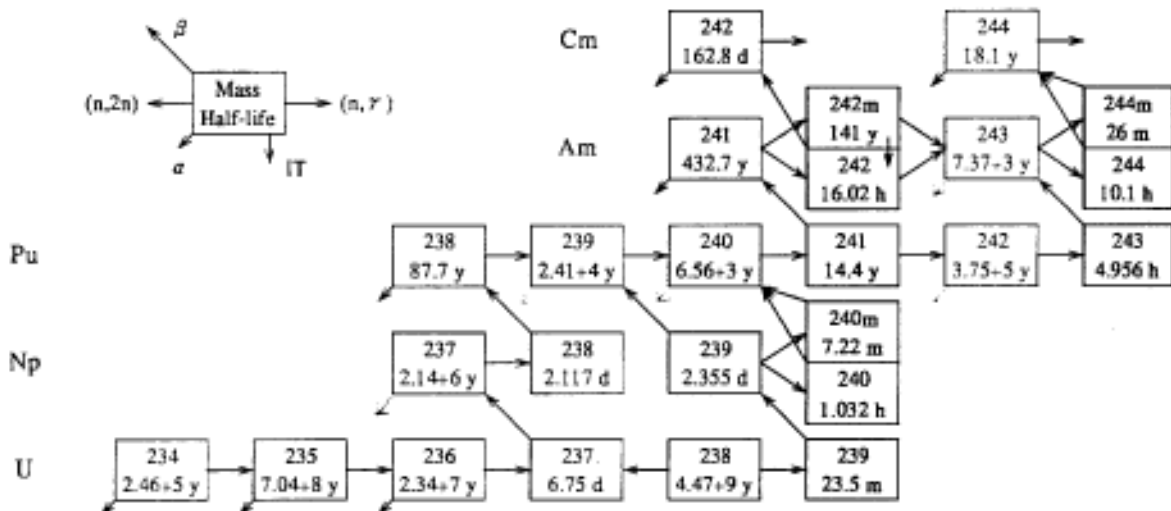


Fig. 1. Actinide transmutation chain employed in this study.

Table 1 Isotopic composition of spent UO₂ fuel

Minor actinides	(wt%)	Plutonium Isotopes	(wt%)
Np-237	0.0588	Pu-238	0.028
Am-241	0.0233	-239	0.473
-243	0.0142	-240	0.265
Cm-242	0.00002	-241	0.111
-244	0.0047	-242	0.068

Initial U-235 enrichment = 4.0 wt%.
 Discharge burnup = 45 GWd/t.
 Cooling time = 3 yr.

2. CALCULATIONAL ASSUMPTIONS

Isotopic composition of spent UO₂ fuel, whose initial U-235 enrichment is 4.0 wt%, is given in Table 1. Np-237, Am-241 and Am-243 account for the majority of MAs, which are investigated in this study. Curium isotopes are assumed not to be recycled but to be disposed as HLWs, because they are very short-lived and are less produced than other MAs. If curium isotopes are recycled, fuel fabrication and transportation would be seriously influenced by their great neutron emission from spontaneous fission and (α, n) reaction.

Depletion calculations for BWR fuels are made with the lattice physics code TGBLA,² while for HCR fuel the HELIOS.HX code³ is used. Figure 1 shows actinide transmutation chain employed in this study. Nuclear data are generated from ENDF-IV or ENDF-V for uranium and plutonium isotopes, and from JENDL-3 for other actinides.

Initial enrichments for reference UO₂, MOX and HCR fuels are given in Table 2, which are determined to

achieve a discharge burnup of 45 GWd/t. The same burnup is assumed for all fuel types, to eliminate its great influence on MA transmutation. Since current BWR fuels are implemented with natural uranium blanket on top and bottom ends, enrichments are given in Table 2 for internal enriched zones. Plutonium isotopic vector enriched to MOX and HCR fuels is that recovered from spent UO₂ fuel after three-years' cooling, as given in Table 1.

3. TRANSMUTATION PRODUCTS OF MINOR ACTINIDES

It is important to reveal which nuclides are produced by burning MAs. If depletion calculation is made for MAs mixed up with fuel materials, transmutation products of MAs could not be distinguished from those of fuel materials. Thus, irradiation of pure MA is convenient for this purpose.

To do this, depletion calculation by TGBLA or HELIOS.HX is made for reference fuels that are free from MAs, and multi-group cross sections are collapsed into one-group cross sections. They, in turn, are fed to the point depletion code ORIGEN-2.⁴

Irradiation calculation for pure Np-237, Am-241 or Am-243 is made with different neutron spectra simulating UO₂, MOX and HCR fuels, until neutron fluence accumulates up to that for burnup of 45 GWd/t. Isotopic compositions after irradiation are shown in Fig. 2.

Transmutation rates for all these MAs are highest in UO₂ fuel, while difference is small between MOX and HCR fuels. Harder neutron spectrum in MOX fuel, due to large absorption cross sections of plutonium isotopes, reduces neutron capture cross sections of MAs. It also reduces fission cross sections of fissile materials, which, in turn, raises neutron flux level to generate the same power density as UO₂ fuel. However, increase in neutron flux level is smaller than expected, because fissile

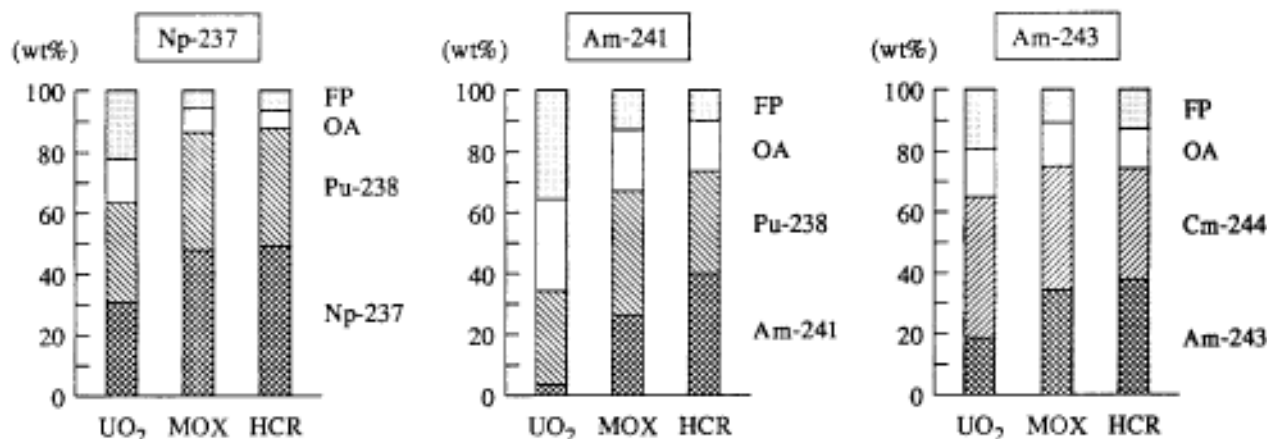


Fig. 2. Transmutation products of pure MAs. OA denotes other actinides than specified.

enrichment is increased to maintain reactivity. As a consequence, the reaction rates for MAs are smaller in MOX fuel than in UO_2 fuel. For HCR fuel, increase in neutron flux level cancels out decrease in neutron capture cross sections of MAs, resulting in nearly equal transmutation rates to MOX fuel.

Transmutation products are also shown in Fig. 2, in which FP denotes the amount of MAs that have transmuted into FPs, including fissions of those actinides which had been produced by successive neutron captures. The fission rate is highest, not in HCR fuel of hardest neutron spectrum, but in UO_2 fuel. Note that both Np-237 and Am-241 transmute mainly into Pu-238, whose half-life is much shorter than those of the parent MAs. Similarly, Am-243 transmutes mainly into Cm-244, which has very short half-life.

Considering short half-lives of principal daughter actinides for the three MAs mentioned above, thermal recycling is effective to reduce long-term hazard of HLWs. While recycling MAs in UO_2 fuel is more efficient than in MOX and HCR fuels for this purpose, the conclusion is still open until overall fuel cycle effects are evaluated.

4. CONCENTRATED RECYCLING

Figure 3 shows material flow for MA transmutation fuel cycle. Multiple recycling is not considered in this study. Np-237, Am-241 and Am-243, recovered from spent UO_2 fuel by partitioning, are recycled into MA-burning reactor. They are uniformly added to all the fuel rods within the fuel assembly. Initial MA content per fuel for MA-burning reactor is assumed to be 1, 5 or 10 times that recovered from spent reference UO_2 fuel given in Table 1. Concentrated recycling is favored to minimize MA proliferation in fuel cycle, while nuclear performances would limit the maximum MA content per fuel.

4-1. Fuel material balance

Adding MAs to the fuel reduces k_{∞} throughout the

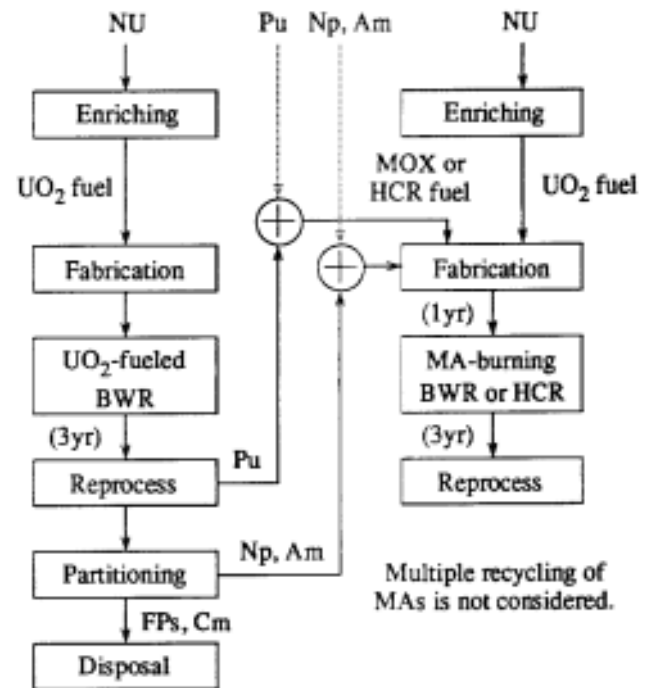


Fig. 3. Material flow for MA transmutation fuel cycle. Broken arrows indicate material feeds from other UO_2 -fueled BWRs if necessary. Time lag is given in parenthesis.

life. Reactivity penalty is evaluated at the core-averaged burnup for end-of-cycle (EOC), which is given in Table 2. It is smaller for fuel type of harder neutron spectrum, because of smaller neutron capture cross sections of MAs. In all fuel types, the penalty increases almost linearly with initial MA content, implying that resonance self-shielding effect is small for the MA concentration under study.

To compensate for this reactivity penalty, additional enrichment is determined to preserve EOC k_{∞} . As given in Table 2, additional enrichment is highest in MOX fuel, because reactivity worth is lower for fissile plutonium

Table 2 Initial and discharged fuel material balance

Fuel type	UO_2				MOX				HCR			
	Ref.	1	5	10	Ref.	1	5	10	Ref.	1	5	10
No. of MA-supplying BWRs												
Reactivity penalty (% Δk)	0.0	0.74	3.44	6.54	0.0	0.47	2.30	4.42	0.0	0.30	1.44	2.77
Initial (wt%)												
U-235	4.000	4.096	4.521	5.153	0.181	0.181	0.178	0.174	0.174	0.174	0.172	0.169
Pu-fiss.	0.0	0.0	0.0	0.0	5.779	5.957	6.740	7.647	8.300	8.391	8.768	9.190
Discharged (wt%)												
U-235	0.703	0.768	1.064	1.523	0.091	0.093	0.098	0.103	0.102	0.103	0.104	0.105
Pu-fiss.	0.584	0.605	0.684	0.773	3.241	3.393	4.052	4.817	6.851	6.920	7.204	7.523
Fissile consumed (wt%)	2.713	2.723	2.773	2.857	2.628	2.652	2.768	2.901	1.521	1.542	1.632	1.731

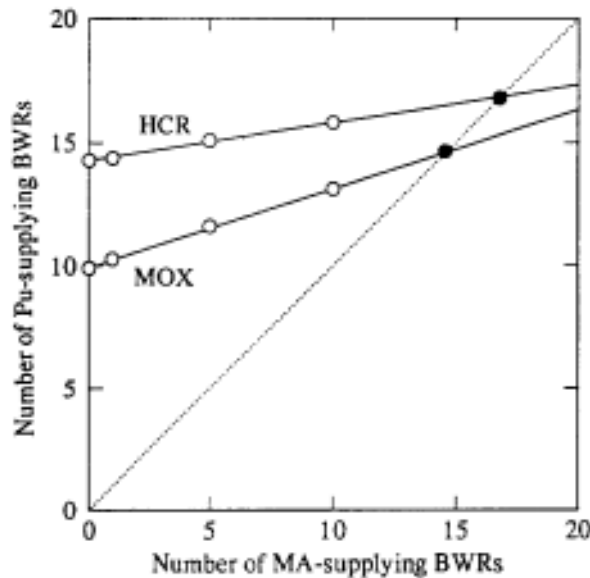


Fig. 4. Balance between plutonium and MAs. Solid and dashed lines represent reactivity and material balances, respectively. Open circles are taken from Table 2. At closed circles, both reactivity and material balances are maintained.

isotopes than for U-235 under thermal neutron spectrum.

The amount of fissile materials consumed during irradiation is also given in Table 2. Its increase with initial MA content is smaller than initial enrichment increase. Since constant discharge burnup consumes the same amount of fissile materials, no additional fissile material takes part in energy production. As a result, more fissile materials remain in spent fuel of higher ini-

tial enrichment, and hence recycling fissile materials recovered from spent fuels improves resource utilization.

Since plutonium is fed from UO₂-fueled BWRs by reprocess, the material balance between plutonium and MAs is important. Figure 4 shows the relation between the numbers of plutonium-supplying BWRs and of MA-supplying BWRs. Open circles represent the reactivity balance taken from Table 2, which can be approximated linearly as indicated by solid lines. When these lines intersect with the dashed line, the simple material balance, reactivity penalty due to MAs can be compensated for by plutonium recovered from BWRs of the same number as MA-supplying BWRs. This balance is found at 15 BWRs for MOX fuel and 17 BWRs for HCR fuel, as indicated by closed circles in Fig. 4.

4-2. Transmutation rate

MA concentration in spent fuels is given in Table 3, from which transmutation rate is calculated as the initial content less net residual. Since MAs remaining in spent MA-bearing fuel come not only from initial MAs but also from fuel materials, the latter contribution must be eliminated to obtain the net residual.

The amount of MAs originated from fuel materials is calculated by depleting MA-free fuels, whose initial fissile enrichments are those given in Table 2. MA concentration in spent these fuels is given in parenthesis in Table 3. Compared with reference fuels, more Np-237 is produced in UO₂ fuel, and more americium isotopes are produced in MOX and HCR fuels, since their productions are dependent on U-235 and plutonium enrichments, respectively. Note, in addition, that neutron absorption by Pu-241 competes with its beta-decay. Therefore,

Table 3 MA concentration in spent fuels (wt%)

Fuel type	UO ₂				MOX				HCR			
	N.M.B. *	Ref.	1	5	10	Ref.	1	5	10	Ref.	1	5
Np-237	0.0588	0.0782	0.1659	0.2965	0.0141	0.0437	0.1707	0.3456	0.0236	0.0528	0.1750	0.3382
**		(0.0592	0.0608	0.0626)		(0.0141	0.0141	0.0141)		(0.0236	0.0234	0.0233)
Am-241	0.0233	0.0247	0.0316	0.0443	0.3100	0.3289	0.4134	0.5221	0.4924	0.5069	0.5684	0.6477
**		(0.0234	0.0235	0.0236)		(0.3225	0.3774	0.4413)		(0.4982	0.5223	0.5500)
-243	0.0142	0.0166	0.0281	0.0474	0.1578	0.1646	0.1942	0.2340	0.1995	0.2055	0.2311	0.2657
**		(0.0136	0.0115	0.0092)		(0.1592	0.1649	0.1708)		(0.2000	0.2022	0.2043)
Cm-242	0.00002	0.00003	0.00006	0.00010	0.00024	0.00026	0.00032	0.00039	0.00027	0.00028	0.00033	0.00038
**		(0.00002	0.00002	0.00002)		(0.00025	0.00026	0.00028)		(0.00027	0.00028	0.00028)
-244	0.0047	0.0113	0.0373	0.0690	0.0673	0.0725	0.0922	0.1146	0.0767	0.0808	0.0969	0.1158
**		(0.0045	0.0036	0.0026)		(0.0669	0.0651	0.0630)		(0.0762	0.0740	0.0715)

* Number of MA-supplying BWRs.

** Figures in parentheses are MA concentrations originated from fuel materials.

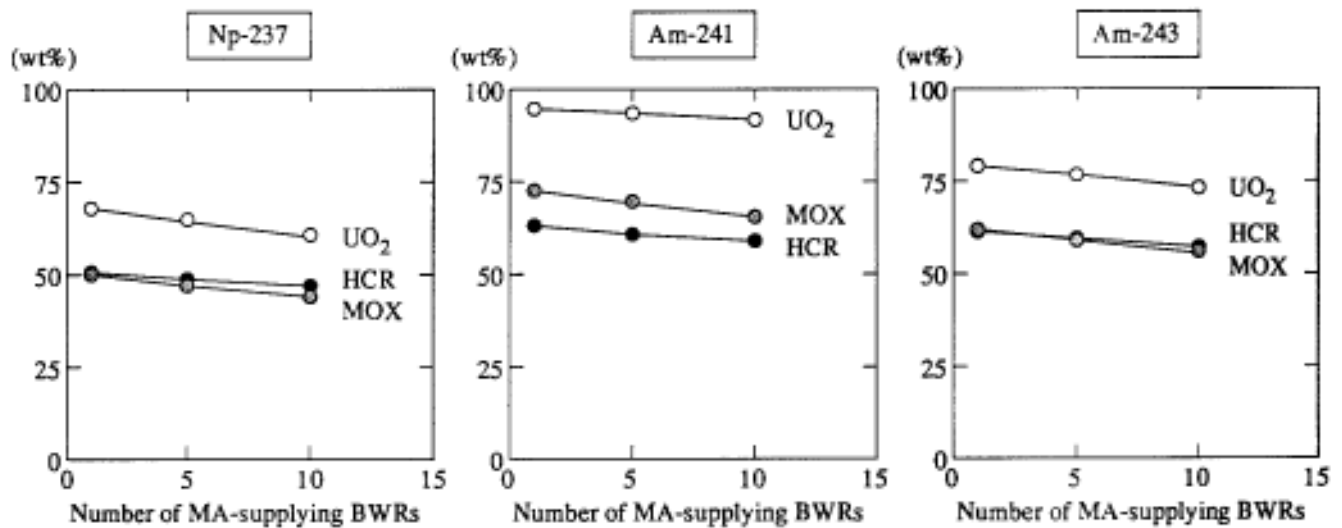


Fig. 5. Transmutation rate of MAs by concentrated recycling.

neutron absorption cross section of Pu-241, reduced by harder neutron spectrum, accelerates Am-241 production but decelerates Am-243 production in MOX and HCR fuels. These effects are more pronounced in MOX fuel, because neutron spectrum change due to additional enrichment is more enhanced.

Transmutation rate, thus calculated, is shown in Fig. 5. Similar trend is observed as in Fig. 2: Transmutation rates for all these MAs are appreciably higher in UO₂ fuel than in MOX and HCR fuels. Note again that hardest neutron spectrum in HCR fuel transmutes least MAs. In all fuel types, transmutation rate decreases with initial MA content, since increased enrichment reduces neutron capture cross sections of MAs and neutron flux level.

4-3. Reactivity coefficients

Reactivity coefficients evaluated at EOC burnup are summarized in Table 4. In UO₂ fuel, void coefficient becomes more negative, and reactivity difference

between cold and hot operating conditions (cold-hot swing) increases with initial MA content; both are unfavorable features for BWR. In MOX and HCR fuels, however, magnitudes of void coefficient and cold-hot swing decrease. Change in Doppler coefficient is small in all fuel types.

Void coefficient depends on fissile enrichment rather than MA content. For UO₂ and MOX fuels, k_{∞} decreases with void increase, since decrease in resonance escape probability overcomes increase in thermal neutron utilization. However, void dependence of k_{∞} becomes smaller or even positive for HCR fuel, because eta-value of Pu-241 increases with spectrum hardening due to void increase. This positive dependence of eta-value on spectrum hardening, in turn, makes void coefficient less negative for MA-bearing MOX and HCR fuels, since additional fissile enrichment shifts neutron spectrum harder.

In reference fuels, magnitude of reactivity coefficients is larger in MOX fuel than in UO₂ fuel. Therefore, some changes of fuel lattice design, such as increase in

Table 4 EOC reactivity coefficients

Fuel type	UO ₂			MOX			HCR					
	Ref.	1	5	10	Ref.	1	5	10	Ref.	1	5	10
No. of MA-supplying BWRs												
Void coef. (ϵ)	-6.60	-6.71	-7.08	-7.15	-8.37	-8.24	-7.60	-6.75	-2.95	-2.71	-1.78	-0.70
Doppler coef. ($10^{-5} d k/k/C$)	-1.78	-1.77	-1.71	-1.66	-1.76	-1.75	-1.69	-1.63	-2.10	-2.08	-1.99	-1.90
Cold-hot swing* ($\% d k/k$)	+4.41	+4.63	+5.36	+6.14	+5.89	+5.87	+5.74	+5.51	+4.49	+4.30	+3.56	+2.74

* Reactivity difference between cold and hot operating conditions.

moderator-to-fuel ratio, are desirable in MOX fuel to be compatible with UO_2 fuel in a BWR core. Table 4 suggests that enrichment increase in MA-bearing MOX fuel has the potential to minimize lattice changes for compatibility. However, in HCR fuel, less negative void coefficient is prohibitive, which tends to be positive.

4-4. Other fuel cycle effects

As is already discussed, recycling MAs increases Pu-238 production. This is most pronounced in UO_2 fuel, because Pu-238 comes not only from MAs but also from plutonium isotopes via Cm-242 in MOX and HCR fuels. Increased neutron emission from (α, n) reaction, in Pu-238 rich plutonium, influences fuel fabrication and transportation, when plutonium recovered from spent MA-bearing fuels is recycled.

Recycling Np-237 also increases Pu-236 and U-232 productions, whose daughters are known to emit high energy gamma-rays.⁵ Their concentrations in spent fuels are shown in Fig. 6. Concentration of Pu-236 is smaller than that of U-232, because accumulated Pu-236 decays into U-232 during cooling time. Therefore, U-232 increase more influences uranium recycling than Pu-236 increase does plutonium recycling.

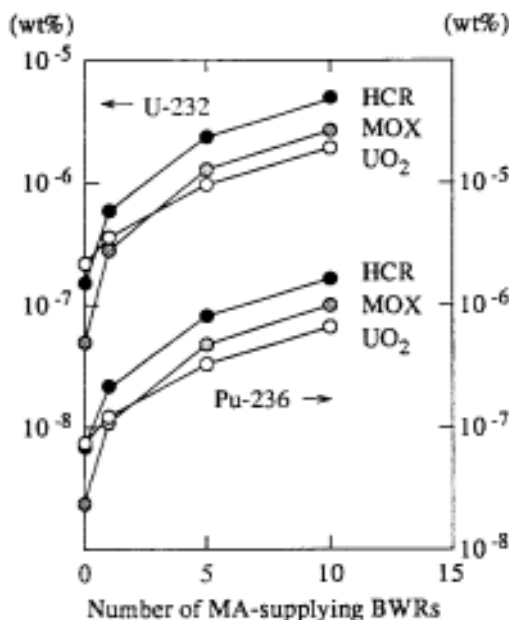


Fig. 6. U-232 and Pu-236 concentrations in spent fuels.

5. SUMMARY AND CONCLUSION

Transmutation of MAs has been investigated for two types of BWR fuel (UO_2 and MOX) and for an HCR fuel.

Irradiation calculation for pure MAs has shown which nuclides are produced by burning MAs in BWR and HCR cores. While transmutation by fission is scarce, Np-237 and Am-241 are transmuted mainly into Pu-238, and Am-243 into Cm-244. Since these daughter actinides are very short-lived, recycling MAs in BWR and HCR is shown to have the potential to reduce long-term hazard of HLWs.

Fuel cycle consideration in concentrated recycling has shown that, though transmutation rate is highest in UO_2 fuel, MOX fuel is most suitable as MA-carrier for the following reasons:

1. Increased radiation dose rate due to MAs could be covered by that due to plutonium isotopes, thus minimizing influence on fuel fabrication and transportation.
2. Fissile enrichment increase in MA-bearing MOX fuel reduces magnitude of reactivity coefficients, to be compatible with UO_2 fuel with minimum change of fuel lattice design. Too less negative void coefficient in HCR fuel is prohibited.
3. Increase in Pu-238 and U-232 productions may influence plutonium and uranium recycling from spent MA-bearing UO_2 fuel.
4. Transmutation rates of MAs are nearly equal between MOX and HCR fuels.

Although evaluation is made on the basis of specific fuels in this study, it is of more practical importance to discuss MA transmutation for total power generating system. Americium production from plutonium isotopes must be consistently taken into account, because Am-241 is generated from beta-decay of Pu-241 even if plutonium is not recycled but stored in a fuel cycle facility.

REFERENCES

1. R. Yoshioka, "Study and View on High Conversion Light Water Reactors", p.65, Atomic Energy Society of Japan (1991) (in Japanese).
2. M. Yamamoto, *et al.*, "Proc. ANS Topl. Mtg. Physics and Shielding", Chicago, Sep. 17-19, 1984, Vol. 1, p.364 (1984).
3. M. Yamamoto, *et al.*, Nucl. Technol. **80**, 240 (1988).
4. A.G. Croff, ORNL-5621 (1980).
5. K. Hida, *et al.*, Nucl. Technol. **75**, 148 (1986).