Behavior of Solid Fission Products in Irradiated Fuel

Ungsup Song, Yanghong Jung, Heemoon Kim, Byungok Yoo, Dosik Kim, Yongsun Choo, Kwonpyo Hong
Irradiated Materials Examination Facility, Korea Atomic Energy Research Institute, Ducjin, Yusung, Taijeon, Korea,305-353 (nussong@kaeri.re.kr)

1. Introduction

Many fission products are generated by fission events in UO_2 fuel under irradiation in nuclear reactor. Concentration of each fission product is changed by conditions of neutron energy spectrum, fissile material, critical thermal power, irradiation period and cooling time[1]. Volatile materials such as Cs and I, the fission products, degrade nuclear fuel rod by the decrease of thermal conductivity in pellet and the stress corrosion cracking in cladding. Metal fission products (white inclusion) make pellet be swelled and decrease volume of pellet by densification[2]. It seems that metal fission products are filled in the pore in pellet and placed between UO_2 lattices as interstitial. In addition, metal oxide state may change structural lattice volume.

Considering behavior of fission products mentioned above, concentration of them is important.

Fission products could be classified as bellows[3];

- solid solution in matrix : Sr, Zr, Nb, Y, La, Ce, Pr, Nd, Pm, Sm
- metal precipitates : Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sb, Te
- oxide precipitates : Ba, Zr, Nb, Mo, (Rb, Cs, Te)
- volatile and gases : Kr, Xe, Br, I, (Rb, Cs, Te)

2. Experimental

EPMA test were performed to obtain concentration of several fission products from three different spent fuels.

- UO2 spent fuel with 3.5% of enrichment and 45,000 MWd/tU of burnup in PWR
- U3Si-Al dispersed fuel with 121 kW/m of linear power and 63% of burnup in HANARO research reactor.
- DUPIC (Direct Use of Spent PWR Fuel in CANDU Reactors) fuel with 61 kW/m of linear power and 1,770 MWd/tU of burnup in HANARO research reactor.

Summary of metal precipitates in each fuel and figures from secondary electron image are shown in Table.1. Metal precipitates were observed in only DUPIC fuel. the composition of metal precipitates in DUPIC is Mo-47.34 at.%, Ru-46 at.% and Pd+Rh-6.65 at.% and has ε-phase (tetragonal structure) with a = 3.883 of lattice parameter and v = 593 unit cell volume as consistency with references.[4]

3. Results and Discussion

The results of EPMA are shown in Table.2. To get the reliability of EPMA, test with simulated fuel(known composition) was performed and compared with chemical quantitative test.

Neodymium distributed in UO_2 matrix is standard for burnup estimation. its concentration agreed well with the simulated fuel made with ORIGEN-2 code calculation. it means that ORIGEN-2 code predicted well composition of fission products in spent fuel.

U_3Si-Al dispersed fuel with 20% of enrichment, fuel of HANARO research reactor, showed high concentration of Nd by 1.908 wt.%. In this result, data of Nd would be useful for the fuel behavior and performance research.

Concentration of Nd in DUPIC shown in Table.2 is in error then, that in spent fuel is same as that in simulated fuel.

Concentrations of other elements except Nd show large error due to irregular migration under manufacturing and irradiation as well as local sampling surface with non-homogenius distribution as specimen. So, in the case, errors occur in comparison with chemical analysis.

4. Conclusion

Three different spent fuels were analyzed by EPMA. Metal precipitates were observed in only DUPIC fuel. the composition of metal precipitates in DUPIC is Mo-47.34 at.%, Ru-46 at.% and Pd+Rh-6.65 at.%.

Neodymium distributed in UO_2 matrix is standard for burnup estimation. its concentration agreed well with the simulated fuel made with ORIGEN-2 code calculation. it means that ORIGEN-2 code predicted well composition of fission products in spent fuel.

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3. H.KLEYKAMP "The Chemical State of the Fission Products in Oxide Fuels:, J.Nucl.Mater.(1985),221-246
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Table. 1 Concentration of three spent fuel types

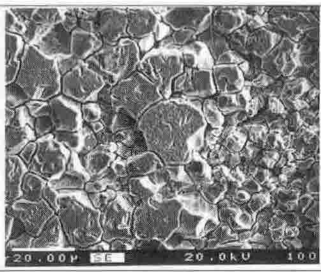
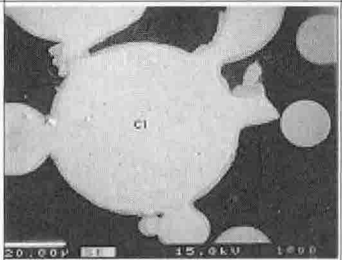
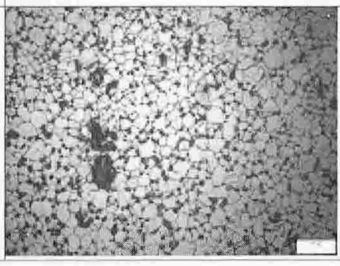
specimen	SEM pics.	Matrix concentration wt%		Metal precipitates
		non-irrad.	irrad.	
3.5 % enriched UO_2 [PWR with 45,000 MWd/tU]		U: 88.532 O: 11.882	U: 81.892 O: 12.605	Not observed.
U3Si-Al (fuel of HANARO research reactor)	 point :C1	wt% U: 97.023 Si: 3.742 at% U: 75.277 Si: 24.607	wt% U: 96.120 Si: 3.851 at% U: 74.252 Si: 25.748	Not observed.
DUPIC fuel		U: 84.535 O: 11.847	U: 80.334 O: 13.543	< Ø 2 um Observed. conc. wt% Mo : 45.0 Ru : 45.6 Rh : 4.25 Pd : 1.68

Table. 2 Comparison of between chemical analysis and EPMA test (wt%)

fission products	Simfuel		irrad. UO_2 fuel	U3Si-Al	DUPIC fuel
	chemical	EPMA			
Nd	0.480	0.476	0.446	1.908	0.42
Sr	0.076	0.084	-	0.431	0.01
Y	0.050	0.052	0.055	-	0.02
Zr	0.397	0.367	-	1.88	0.19
Pd	0.028	0.187	0.314	0.299	0.02
Ru	0.239	0.269	0.385	0.963	0.22
Rh	0.073	0.049	-	0.131	0.06
Ba	0.203	0.218	0.374	0.560	-
La	0.155	0.143	0.129	0.318	-
Mo	0.449	0.392	0.319	1.816	0.27
Ce	0.233	0.278	0.42	1.05	0.64