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Analysis of a Metallic Precipitates in the Irradiated Fuel using EPMA

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1. Introduction

Metallic precipitates in the irradiated fuels affect the fuel's electric and thermal conductivities, and a large quantity of them changes the fuel performance, especially for high burn-up fuels[1].

Therefore, characterizing the metallic precipitates in a post irradiation examination is very important[2]. In this paper, a characterization procedure and a manner for improving the measurement accuracy by EPMA (Electron probe Micro Analyzer) were established with a well informed simulated fuel which was applied to characterizing the fission products and metallic precipitates of the simulated fuel irradiated at HANARO

2. Specimen preparation and Results

2.1 Specimen preparation

To characterize the concentration of the metal precipitates from the irradiated fuel, the radiation activity of a specimen is required to be below $3.7x10^{10}$Bq for protecting the operator and damage to the EPMA. Although the specimen size has to be minimized, its volume could be managed by the manipulator in a hot cell. After cutting the specimen to a manageable size by the manipulator, the specimen was hot mounted with conducting resin at 150 ℃ and 0.6 Mpa. Too thin a specimen is liable to be broken during the hot mount, so its thickness was cut to be about 5 mm at first. And finally, a specimen of 2.5 x 5 x 1.5 mm^3 (WxLxD) was fabricated by repeating the mounting and cutting several times for decreasing the radiation with a manipulator. After polishing the specimen, it was etched by an immersion method.

2.2. EPMA

EPMA(Electron Probe Micro-Analyzer, SX-50R, CAMECA, Paris, France) used in this experiment can treat a irradiated nuclear fuel by a special shielding of the specimen holder and is specifically shielded with lead and tungsten to permit the analysis of an irradiated nuclear fuel. The maximum radiation activity in this EPMA is allowed to be below to $3.7x10^{10}$Bq. The condition of EPMA was 20 kV of an electron acceleration potential and 20 nA of a beam current.

2.3 Metallic Precipitates of an Un-irradiated fuel

To compare the results of the analysis by the EPMA and a wet chemical analysis were carried out[3]. Table 1 shows these results. Column (a) in Table 1 presents the result of the wet chemical analysis. They agreed well with the initial concentration of the added chemicals as well as the results by the EPMA of column (b) in Table 1. The EPMA measurement is carried out by scanning the small area with a TV mode at a magnification of 20,000.

In the case of the fix mode, error can occur from eliminating the precipitates extracted at the grain boundaries or the vacancies, so a large area was scanned. After the set up of the characterizing conditions such as applying the voltage and current, characterizing by the EPMA was accomplished for the metallic precipitates of the simulated fuel

Table 1 Comparison of the concentrations between wet chemical analysis and EPMA (unit: wt.%).

Chemical elements	Chemical analysis, (a)	EPMA analysis, (b)	Concen, of ppt. (c)
$Zr(ZrO_2)$	0.397	0.367	0.108
$Mo(MoO_3)$	0.449	0.392	**49.521**
$Ru(RuO_2)$	0.239	0.269	**32.153**
Pd(PdO)	0.028	0.187	**3.682**
$Ba(BaCO_3$	0.203	0.218	-
$La(La_2O_3)$	0.155	0.143	0.09
$Ce(CeO_2)$	0.233	0.278	0.035
$Nd(Nd_2O_3)$	0.480	0.476	0.055
Sr(SrO)	0.076	0.084	-
$Y(Y_2O_3)$	0.050	0.052	-
$Rh(Rh_2O_3)$	0.073	0.049	**3.232**
U			11.48

Column (c) in Table 2 shows these results. The concentrations of the components of the metallic precipitates such as Mo, Ru, Rh and Pd are relatively lower than those of the oxide precipitates such as Ba, Zr and Mo in Table 2, which is suggests that the metallic precipitates are produced less than the oxides

The result of the quantitative analysis of the metallic precipitate with 10 points were Mo-49.512 wt%, Ru-32.153 wt%, Pd-3.682 wt%, Rh-3.232 wt% and U-11.48 wt% respectively

Representing them in an atomic percent by excluding the uranium, they were Mo-53.89 at.%, Ru-37.40 at.% and Pd+Rh-8.71 at.%. From this, the

metallic precipitates in the un-irradiated simulated fuel seems to have a hexagonal structure of the ε-phase[4, 5]

2.4 Metallic Precipitates of an irradiated fuel

Figure 1 shows metallic precipitates in the irradiated SIMFUEL and their concentration. Figure 2 shows the quantitative analysis result of metallic precipitate about at 2.5 ㎛ with a beam size of 1 ㎛ by every 0.5 ㎛. Measured point #3 does not contained uranium, so this point may stand for a typical concentration of the metallic precipitates of irradiated SIMFUEL. Table 2 shows the concentrations of precipitates both at grain boundary and intra grain. There was no difference among the adjacent ones as shown in Table 2. But their concentrations are presented somewhat differently according to their position in the radius direction.

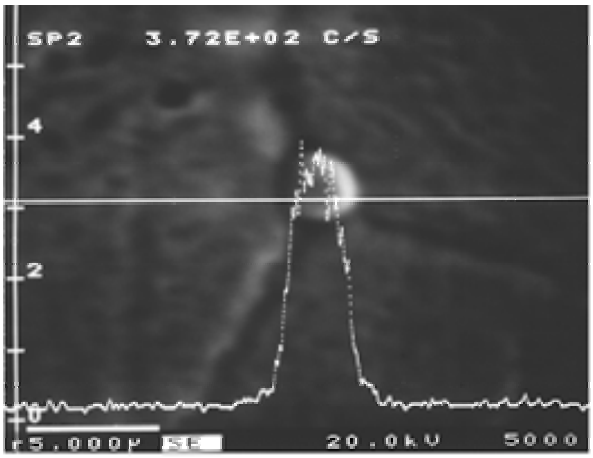

Figure 1 Metallic precipitates in the irradiated SIMFUEL.

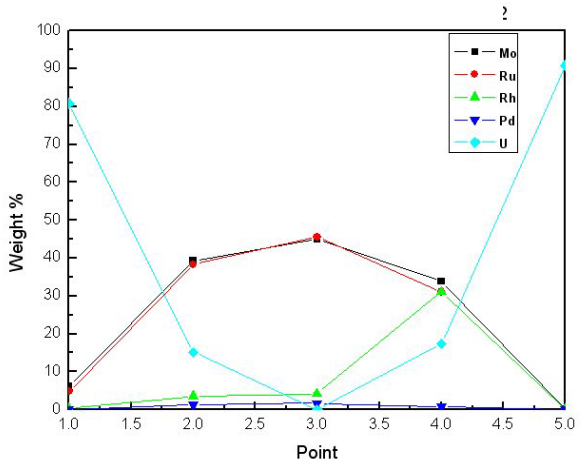

Figure 2 Concentration of a Metallic precipitates contains in the irradiated SIMFUEL.

Concentrations of the metallic precipitates in Table 2 are Mo-47.34 at.%, Ru-46 at.% and Pd+Rh-6.65 at.% in atomic percent. This ratio corresponds to tetragonal structure of ε-phase [4, 5].

Table 2 Concentrations of precipitates on grain boundaries and intra grains (wt.%).

element	Intra grain	Grain boundary
Mo	45.000	42.490
Ru	45.590	45.210
Rh	4.239	4.246
Pd	1.675	1.806
U	0	1.319
Total	96.634	95.172

3. Conclusion

Metallic precipitates of the SIMFUEL were characterized by EPMA. It was irradiated at HANARO at the conditions of a 61 kW/m maximum linear, 53 kW/m average linear power and a 1,770 MWd/tU average burn-up. To establish the procedure of an accurate measurement by EPMA, The measurement was accomplished with simulated fuel which was fabricated with chemicals instead of fission products. Those results were also compared with the results of a wet chemical analysis. The size of metallic precipitate of the fresh simulated fuel was about 1 ㎛, and their concentrations were 49.512 wt% of Mo, 32.153 wt% of Ru, 3.682wt% of Pd and 3.232 wt% of Rh.

The size of the metallic precipitates in the irradiated SIMFUEL was observed at 2～2.5 ㎛ in diameter, and they appeared to be 47.34 at.% of Mo, 46 at.% of Ru and 6.65 at.% of Pd+Ph.

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