The release behavior of volatile corrosive fission products (I, Cs) from SIMFUEL

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1. Introduction

Fission products are one of the important factors that affect fuel's behaviors during irradiation. The longer the fuel burns up in the reactor, more important the fission products become[1]. The high burnup fuel contains many fission products, including many types of solid solutions and metallic/oxide precipitates. In the development of new nuclear fuel, the release of volatile corrosive fission products plays an important role in pellet-cladding interaction. But, few data on the release behavior of volatile corrosive fission products (I, Cs) from have been published so far.

The volatile corrosive fission products (I, Cs) are likely to exist in the form of complex compounds. It is difficult to measure the radioactivity of fission gas. We carried out the chemical experiment method, not measurement of radioactivity of fission gas.

In this study, a SIMFUEL (simulated for high burnup fuel) was made with natural elements to simulate a spent fuel corresponding to a declared burnup. The objective of this study is to know the release behaviors of volatile corrosive fission products (I, Cs) from SIMFUEL.

2. Experimental

2.1 Specimen preparation

Polycrystalline SIMFUEL was prepared. This SIMFUEL was a simulated spent fuel with natural elements corresponding to fission products. To make the SIMFUEL with a given burnup (27,300MWd/t U), all the compositions and amounts were obtained by using the ORIGEN-2 code. The SIMFUEL in this study was fabricated with 13 dominant compositions, as shown in Table 1.

Table 1. Composition of the SIMFUEL(27,300MWd/tU) based on calculation

Nuclide	Wt%	Nuclide	Wt%
Mo	0.300	Y	0.044
Ru	0.285	Zr	0.330
Rh	0.039	La	0.113
Pd	0.107	Ce	0.773
Te	0.043	Nd	0.513
Ba	0.130	U	97.237
Sr	0.085		

The SIMFUEL specimens were made by a pelletizing process (powder mixing, green packing, then a sintering). For the experiment, they were cut into 2×2×2 mm ($8mm^3$) cubes. Grain size was shown in Fig. 1. Some precipitates were observed as dots in the figure. The properties of the SIMFUEL in this study are shown in Table 1 including the UO_2 sample.

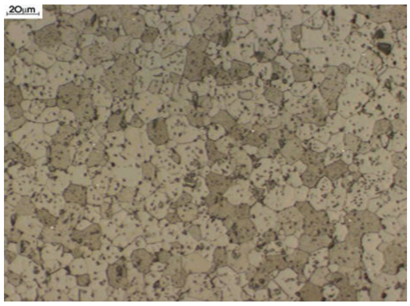

Fig. 1. Microstructure of the SIMFUEL

It was irradiated I the HANARO research reactor for up to a burnup of 0.1 MWd/t U. After an irradiation, the specimen was cooled for 10-11 days to reduce the radiation exposure. We used about 300mg(three cubes) of the SIMFUEL for each irradiation and annealing test[2].

2.2 Apparatus of the post annealing experiments

A sketch of the experimental apparatus is shown in Fig. 2. The maximum annealing temperature was about 1650℃. In this experiment, the temperature in the furnace was raised to 1500℃. During the annealing with a low oxygen potential, the temperature was held for 10h at 1500℃. Helium gas flowed through the system to transport the released fission gases into the trap system. The helium gas was mixed with hydrogen gas in order to control its oxygen potential of the gas[3].

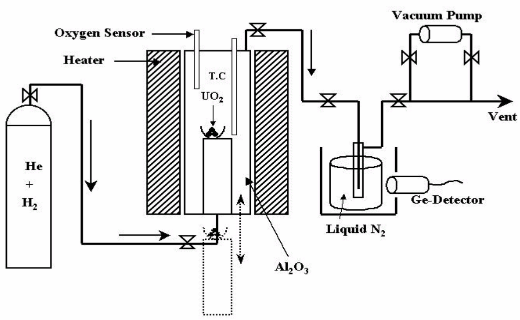

Fig. 2. Schematic of the annealing apparatus

2.3 Qualitative analysis

The volatile corrosive fission products (I, Cs) are likely to exist in the form of complex compounds. It is difficult to measure the radioactivity of fission gas with the method we used. So, the experiment is carried out using dilute nitric acid.

To carry out qualitative analysis, the surface of an Alumina crucible was dissolved. Then the surface of the SIMFUEL was dissolved in that solution for 10 seconds. A measurement confirmed that I-131 and Cs-137 were exist at the surface of the specimen.

2.4 Step by step experiment for quantitative analysis

The surface area changes according to the surface of the SIMFUEL piece. Analyzing the solution formed by dissolving the surface of the SIMFUEL showed the nuclide exists at the surface of the SIMFUEL. In this experiment, the uranium's radioactivity was very low enough to be undetectable. In this case, strong gamma concentration is an important factor to analyze the nuclide in solution.

Because of annealing at 1500℃ for 10h, the concentration gradient existed inside the SIMFUEL by the diffusion of fission products. Assuming that the concentration gradient changes exponentially at the of SIMFUEL surface, we can analogize the concentration of fission products. Fig. 4 shows the results of analysis based on U-238. Because of the fission products released by diffusion, the inside concentration is higher than the outside. According to the change in radioactivity of I-131 and I-132 by location, their relative radioactivity can be obtained. The measured radioactivity at the surface is the sum of the absorbed Iodine and the dissolved Iodine inside. About 70~80% of Iodine's radioactivity is generated at the surface. And based on La-140, about 55~65% of the Iodine is generated from the absorbed Iodine.

The change in radioactivity of Cs-137 has a lower slope than that of Iodine, which shows the Cs-137 diffuse more slowly than Iodine. About 80% of Cs-137 is generated from the absorbed Cs-137 at surface. And based on La-140, about 70~80% of the Cesium is generated from the absorbed cesium compound.

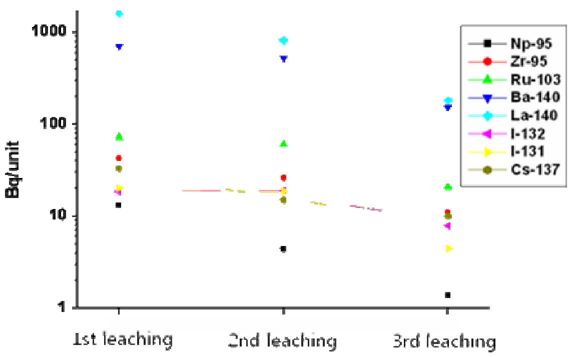

Fig. 3. The results of the step by step leaching experiment

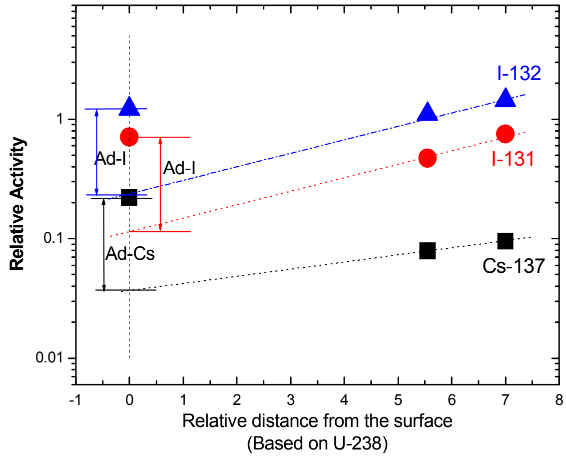

Fig. 4. The relative radioactivity of fission products by location from the surface

3. Conclusion

Specimens of a SIMFUEL(27,300MWd/t U) were prepared to measure the release behavior of volatile fission products (I, Cs). After the specimens were irradiated in the HANARO research reactor, annealing tests were carried out. The release behavior of I and Cs was proved by a chemical experiment. About 60~80% of the Iodine and Cs is exist at surface.

Acknowledgment

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