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Post-irradiation Examination of Dry Process Fuel

In Ha Jung, Ho Jin Ryu, Je Sun Moon, Yang Hong Jung,
Byoung Ok Yoo, Kee-Chan Song, and Myung-Seung Yang

Korea Atomic Energy Research Institute
P.O. Box 105, Yuseong-gu, Daejeon 305-353, Korea

ABSTRACT

Post-irradiation examination of dry process fuels were performed. Irradiated fuels were discharged from the 2nd, 3rd and 4th irradiation tests, which performed at different irradiation conditions. Secondary emission microscope and electron probe microscope analyzer were used to assess the microstructures and the concentration profiles of fission products. In a pellet, any remarkable trend in concentration of fission product was not found because of its low burn-up. Precipitates were observed at the inside of grains as well as at grain boundaries. Their sizes measured at the center and surface of the irradiated pellet were different. Concentrations of Mo and Ru decreased near the surface of the pellet. It might be resulted from the oxidation tendency of both Mo and Ru. However, the concentrations of both nuclides in the fuel matrix increased at the surface compared to the center. Fuel performance evaluation code modified from FEMAXI-V made resonable estimation on the maximum grain size in the range of burn-ups of this study.

1. INTRODUCTION

The electrical utilities are continuously striving to reduce the fuel cycle cost and the amount of spent fuel to be reprocessed or stored. Dry process fuel is based upon dry thermal and mechanical processes to directly fabricate the "dirty-fuel" from spent PWR (Pressurized Water Reactor) fuel material, without separating any fissile material and stable fission products.

The fissile material that typically remains in spent PWR fuel exceeds the natural uranium fissile content of 0.711 wt%. The neutron economy of a CANDU reactor is sufficient to allow dry process fuel that can be used in a CANDU reactor which is originally designed for natural uranium fuel, even though the neutron-absorbing fission products remain as impurities in the dry process fuel[1].

The OREOX (Oxidation and Reduction of Oxide fuel) process is the key feature in the dry process fuel fabrication. The repeated processes of oxidation and reduction cause crystallographic transformations in the pellet matrix, which make the pellets break into a fine powder. Once

the powder is conditioned to improve its sinterability, the rest of the dry process fuel fabrication process is much the same as the conventional fuel manufacturing process that utilizes the powder-pellet route. Because the fuel material remains highly radioactive throughout the process, all processing must be done in a shielded facility[2].

2. EXPERIMENTAL

All specimens for PIE were irradiated at HANARO. All of them were fabricated from PWR spent fuel in dry process[3]. Summary of PIE specimens presented at Table 1. Du-2 is a fuel pellet of 0.72% U-235 and 0.56% Pu. Initial grain size was 3.5 ㎛ and its density was 10.07 g/cm^3. It was irradiated high linear element power rate of 61 kW/m and burn-up of 1,770 MWd/t during 28 full power days. DU-34 is irradiated 2 cycle at HANARO during 219.6 full power days with maximum linear element power rate of 34 kW/m and 25 kW/m in average. Calculated burn-up by ORIGEN estimated as 6,700 MWd/t. DU-4 irradiated for 111.6 full power days with maximum linear element power rate of 36 kW/m and 31 kW/m in average.

SEM (Secondary Emission Microscope), EPMA (Electron Probe Microscope Analyze) were used to assess their microstructure and concentration profile. Concentration profile was also observ by gamma-scanning to radial direction.

3. RESULTS AND DISCUSSION

Figure 1 and 2 show the estimated centerline temperatures and grain sizes by modified FEMAXI-V code. The grain sizes at the center of the fuels of DU-2, DU-34 and DU-4 are predicted about 10 times, 1.5 times and 1.3 times comparing to their initial grain sizes respectively. The PIE results on Fig. 3 showed similar outcome with calculated ones as 11 times, 1.7 times and 1.5 respectively. The applied code for this dry process fuel was modified from FEMAXI-V at the respect of thermal and mechanical properties. From this, grain sizes are dominantly affected by the centerline temperature during irradiation, and modified FEMAXI code is useful at the examined range of burn-up.

Figure 4 is the overview of microstructure of DU-2. About 900 ㎛ void at the center, 27 × 150 ㎛ columnar grain near the void are observed. These microstructure is corresponded to 7,600 MWd/MtU burn-up of CANDU fuel [4]. Overall apparence of fuel pellet looked sound. Figure 5 and Fig. 6 show the microstructure near the void and fractured surface of DU-2 on the same position. Small micro bubble on the grain and gas channel on the grain boundary, precipitates about 3.5 ㎛ size are observed. Gas channel is frequently found at the high burn-up fuel pellet. Diffused micro bubble to the grain boundary link together in large quantity and escape to out of grain[5]. In this fuel pellet, these are likely arisen from the thermal release for high centerline temperature[6].

Figure 7 and Fig. 8 represent the overall view of DU-34 and DU-4, and small windows below them show the microstructures at the center and surface, respectively. Grain at the center of DU-34 is grown about 1.5 times compared to that of the surface. For DU-4, it is about 1.7 times comparing to the initial grain size. On DU-4 - irradiated at higher linear element

power rate - micro bubble, fission gas coalescence at grain boundaries, and precipitates at grain boundaries as well as inside of grains are observed. Since fission gas diffusion depends largely on the temperature, much gas bubbles are likely to move to their grain boundaries. Unlikely to DU-2, because not inter-linkage of bubbles at grain boundaries are arisen, the state of fuel pellet during irradiation might be just at the beginning of fission gas release.

The fractional fission gas release of DU-2 element determined by puncturing and analysed by chemical analysis. Even though gas pressure in the cladding and released gas amount are valuable for PIE data in the respect of fuel performance, no suitable equipment was retained. The isotopic composition of the plenum gas in the element gives a Xe/Kr ratio of 11.9. This ratio is intermediate between the values for gas resulting from the fission of ^{235}U (Xe/Kr = 7) and the fission of ^{239}Pu (Xe/Kr = 19) [7], confirming that Pu fission made an important contribution to the burn-up. This is for the dry process fuel characteristic, i.e., no separation any materials from the PWR fuel. Raw material of this fuel has 8.4 mg/g-fuel of Pu initially.

Radial γ-scanning for DU-2 is performed from the center of the fuel to surface. On the whole, no tendency in concentration of fission products of the fuel pellet is observed as shown in Fig. 9. However, for Cs-137 as shown at Fig. 10, lower concentration exhibited at the center of the fuel and then increased gradually to the direction of the surface. This is also from the high temperature of the fuel during irradiation at HANARO.

Finally, precipitates arisen on the DU-2 fuel pellet are observed and analysed by EPMA. The size of precipitates and composition according to the position of the fuel pellet, identity of the precipitates in chemical composition between grain boundary and inside of the grain were assessed. Size of precipitate close to the void represented 3.5 ㎛, 2.0 ㎛ at the columnar grain, 1.5 ㎛ at large equiaxed grain, and about 0.4 ㎛ at the surface of the fuel pellet. Large size of precipitates as this fuel are hardly to be seen at the fuels from commercial nuclear power plant. Figure 11 and 12 show the precipitates observed at center and near the surface of the fuel pellet. White line on the picture represents relative intensity of molybdenum around precipitate.

Table 2 shows the chemical composition of precipitates of both at grain boundary and inside of the grain. Both of them are consistent together as a whole. The dominant chemical compositions of the precipitates are Mo, Ru, Rh and Pd as shown at Fig. 13. However, their chemical ratio are exhibited a little differently by position to position. Concentrations of Mo and Rh are decreased to the direction of surface. This is because of the higher oxygen potential near the surface resulting from the higher fission rate of uranium oxide. Excess oxygen makes metallic Mo and Rh to their metallic oxide In that case metallic Mo and Rh are more likely to be in metallic oxides [8]. Figure 14 is the concentration profile of molybdenum in the fuel matrix, where the concentration of molybdenum slightly increased at the surface comparing to center. Therefore, a little much amount of molybdenum is dissolved at the surface of the fuel matrix and they expected to be in oxides. Similar results have been observed at the reference [8].

4. CONCLUSION

Modified code from FEMAXI-V can well estimate the maximum grain size and microstructure of dry process fuel in the range of irradiated bun-up. From the results of concentration profile

of fission products, no significant tendency was found. This might be from low burn-up of the the fuel.

Precipitates were observed inside of the grains as well as at the grain boundaries. Their sizes were different according to the position. Chemical compositions of both precipitates inside of grains and grain boundaries were almost similar together. But, the sizes increased to center. Its size was about 3.5 ㎛. Main components of the precipitate were Mo, Ru, Rh and Pd. Concentrations of Mo and Ru decreased near the surface of the pellet. However, the concentrations in matrix of both nuclides increased to surface. It was deducted to oxidation tendency of both Mo and Ru.

ACKNOWLEDGEMENT

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Table 1 Summary of PIE specimens

구분	연료봉 제원	소결체 제원	조사조건	선출력	연소도
DU-2	· 연료봉 1, 2 & 3: SF소결체 각 5개 · 피복관: Zr-4 · 두께: 0.66 mm · 내경: 10.8 mm · 길이: 199 mm · 갭: 125 ㎛	· SF 소결체: U-235 0.72% Pu 0.56% · 길이: 10.0 mm · 직경: 10.6 mm · 밀도: 10.07 (93.4%) · 입도: 3.5㎛	· 하나로, OR4 (22MW)	· 최대: 61kW/m · 평균: 53kW/m	· 평균연소도: 1,770 MWd/t
DU-34	· 연료봉 1: 3차 및 4차 조사 연료봉 · 피복관: Zr-4 · 두께: 0.66 mm · 갭: 125 ㎛	· SF 소결체: U-235 1.06% Pu 0.51% · 길이: 11.5 mm · 직경: 10.6 mm · 밀도: 10.3 (95.5%) · 입도: 9.5㎛	· 하나로, OR4 & 5 (24MW)	· 최대: 34kW/m · 평균: 25kW/m	· 평균연소도: 6,700 MWd/t
DU-4	연료봉 : SF소결체 5개 · 피복관: Zr-4 · 두께: 0.66 mm · 갭: 125 ㎛	· SF 소결체: U-235 1.06% Pu 0.51% · 길이: 11.5 mm · 직경: 10.6 mm · 밀도: 10.3 (95.5%) · 입도: 14㎛	· 하나로, OR5 (24MW)	· 최대: 36kW/m · 평균: 31kW/m	· 평균연소도: 3,300 MWd/t

Table 2 Chemical composition of precipitates inside of grain and grain boundary

	Zr	Sr	Y	Mo	Ru	Rh	Pd	Cd	U	Total
inside of grain	0.056	0.056	0.108	45	45.59	4.239	1.675	0	0	96.63
grain boundary	0.066	0.009	0.02	42.49	45.21	4.246	1.806	0	1.319	95.17

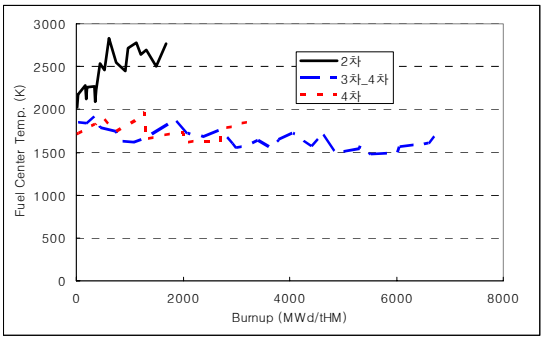

Fig. 1 Estimated centerline temperature.

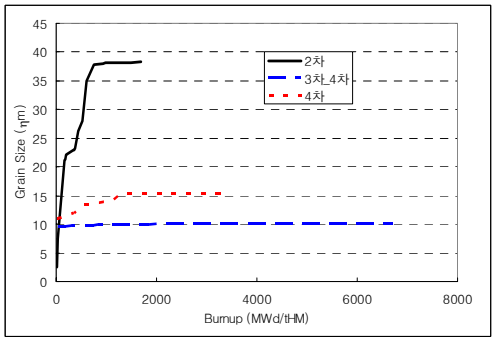

Fig. 2 Estimated maximum grain size.

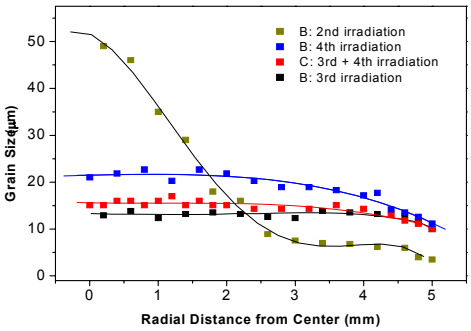

Fig. 3 Observed grain size to radial direction from center.

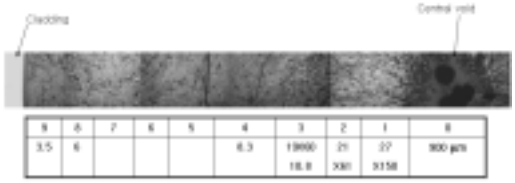

Fig. 4 overview of microstructure of DU-2.

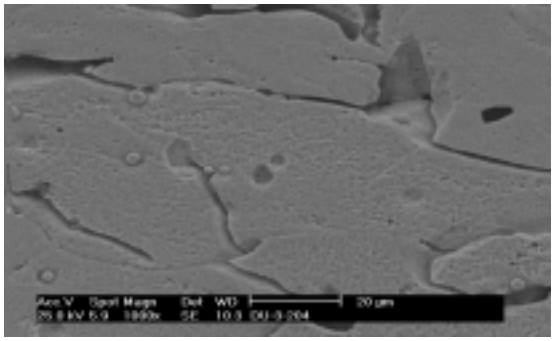

Fig. 5 Microstructure near the void of DU-2.

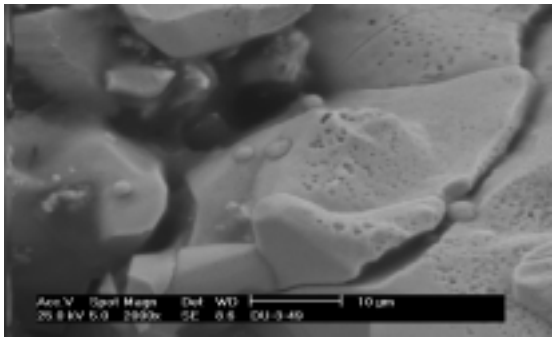

Fig. 6 Fractured surface near the void of DU-2.

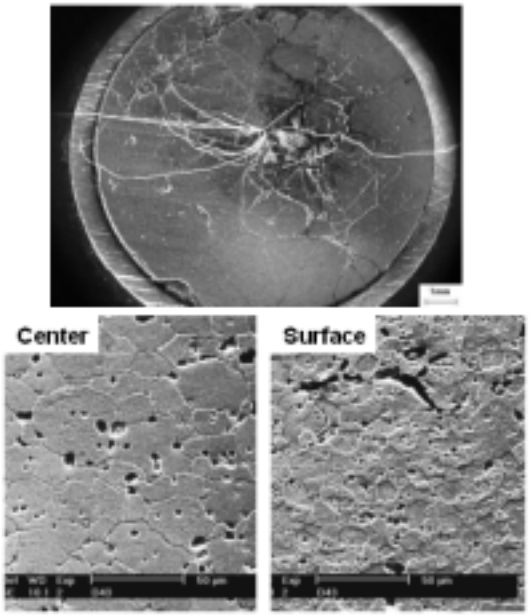

Fig. 7 Microstructure of DU-34; burn-up: 6,700 MWd/tHM, LER: 34 kW/m (Max), 25 kW/m (Ave).

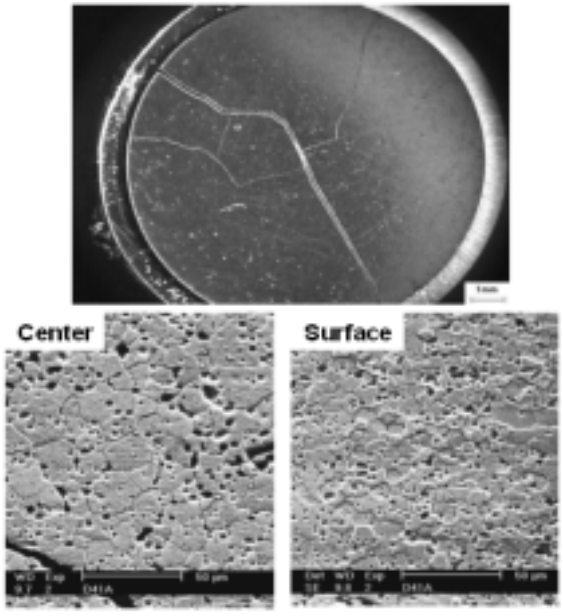

Fig. 8 Microstructure of DU-4; burn-up: 3,300 MWd/tHM, LER: 36 kW/m (Max), 32 kW/m (Ave).

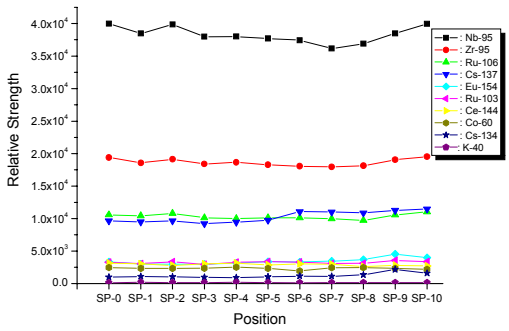

Fig. 9 Concentration profile of fission products from center(sp-0) to surface(sp-10).

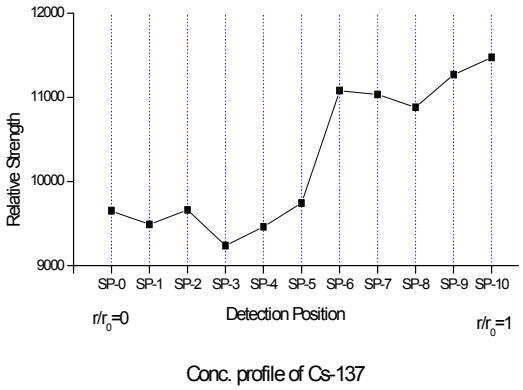

Fig. 10 Concentration profile of Cs-137 by γ-scanning.

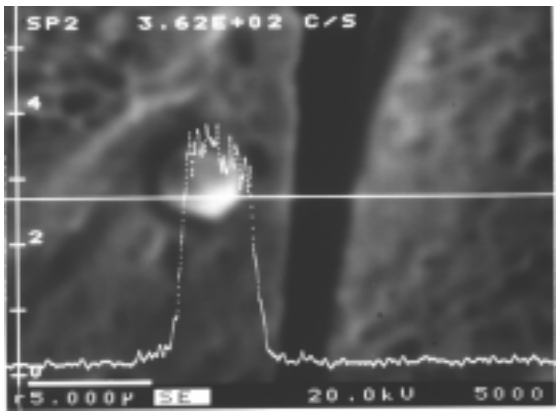

Fig. 11 Precipitate near the void(3.5㎛).

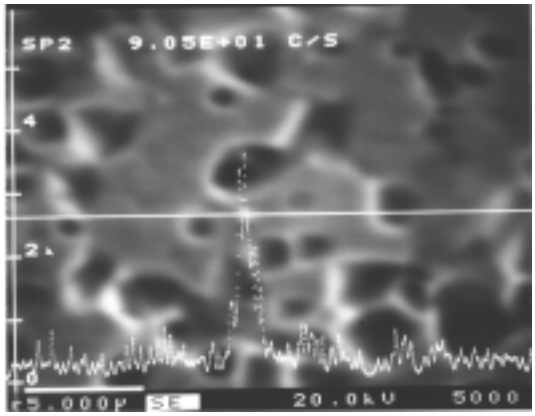

Fig. 12 Precipitate at surface(0.4㎛).

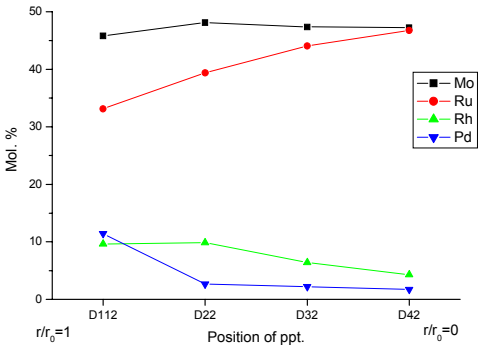

Fig. 13 Chemical compositions of precipitate.

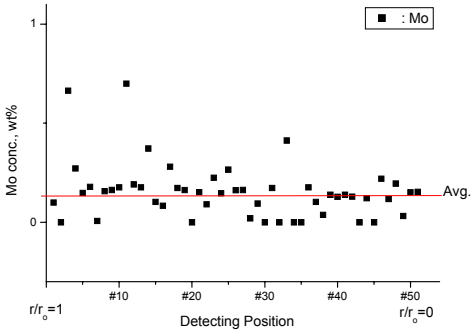

Fig. 14 Concentration profile of Mo in fuel matrix.