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Fabrication method for large grain-sized UO_2 fuel pellets

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1. Introduction

To reduce the fuel cycle costs and the total mass of spent LWR fuels, it is necessary to extend the fuel discharged burn-up. Research on fuel pellets focuses on the modification of microstructure to significantly reduce Pellet-Cladding Interaction (PCI) and Fission Gas Release (FGR) to increase the high burn-up safety margins on LWRs. [1]

Large grain-sized UO_2 fuel pellets can be the most promising candidate for high or very-high burn-up fuels because they can reduce the PCI by enhancing the fuel dislocational creep behavior [2], and FGR [3]. Moreover, large grain-sized pellet can increase the critical degree of burn-up for High Burn-up Structure (HBS) formation. [4]

This paper deals with the fabrication process for large-grained UO_2 pellets. In commercial fuel pellets fabrication process, the defective UO_2 pellets are recycled in the form of U_3O_8 powder by oxidizing the defective pellets. This paper shows that the recycled U_3O_8 power morphology can be significantly enhanced by only modifying the oxidizing temperature. In addition, this U_3O_8 powder can effectively promote a grain growth of UO_2 pellets.

2. Experimental

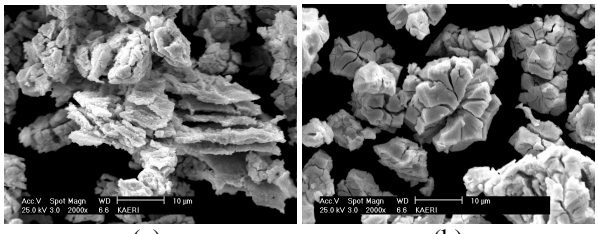
The UO_2 fuel pellets were prepared by sintering the ADU-UO_2 green pellet via conventional sintering process. In order to investigate the dependence of U_3O_8 powder property on the oxidizing temperature, the sintered UO_2 pellets are oxidized at different temperatures of 325 and 450 °C in air, respectively. The U_3O_8 powders morphology was examined by SEM and their BET surface areas were measured by BET surface area analyzer.

Two groups of U_3O_8 powder were prepared. First one is as-received un-doped U_3O_8 and other is Al doped U_3O_8 powders. 3~10wt% of those U_3O_8 powders were added to the UO_2 powder, which was produced through the Dry-Conversion (IDR-DC) process. Powder mixtures were mixed with a tumbling mixer. The powder mixture was pressed into green pellets at 300 MPa. The green pellets were sintered at 1730 °C for 4 h in flowing H_2+1.5%CO_2 gas.

The sintered density of UO_2 pellets was measured by the water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1250 °C in carbon dioxide gas in order to examine grain boundaries. The pore and grain structure were examined by an optical microscope and grain size was determined by the linear intercept method.

3. Results

Fig. 1 shows the SEM micrographs of the U_3O_8 powders which were obtained by oxidizing the UO_2 pellets at 325 and 450 °C, respectively. During the oxidation, UO_2 pellets were spontaneously pulverized to U_3O_8 powder because the lattice volume of U_3O_8 is about 30% larger than that of UO_2. The U_3O_8 powder which was oxidized at 450 °C (Fig. 1(b)) is angular in shape and many cracks developed on the surface. In a commercial UO_2 pellet fabrication process, the defective pellets are oxidized at about 450 °C. And this pop-cone-like-shaped U_3O_8 is typical one. When the UO_2 pellets are oxidized at low temperature of 325 °C, however, the U_3O_8 powder morphology is remarkably changed. Several thin U_3O_8 plates are piled up and form a fault plane-like shape. The surface of thin U_3O_8 plate is uneven and fine columnar shape grains can be observed. The BET surface area was measured to be 1.50 m^2/g for U_3O_8 powder oxidized at 325 °C and 0.66 m^2/g for that at 450 °C. These results reveal that oxidation sequence of UO_2 pellet at low temperature of 325 °C is quite different to that at 450 °C and the BET surface area of U_3O_8 powder is significantly increased when UO_2 pellets are oxidized at low temperature.



(a) (b)

Fig. 1. The SEM images for U_3O_8 powders which are obtained by oxidizing the UO_2 pellets at (a) 325°C and (b) 450°C.

The U_3O_8 powders are added in a quantity of 3, 5, 10wt% to the UO_2 powder, and then sintered at 1730°C for 4h in H_2-1.5%CO_2 gas. About 10 wt% of U_3O_8 powder is usually added to the UO_2 powder in the commercial process. Fig. 2 shows the pellet density changes according to the un-doped U_3O_8 powder

addition. The pellet density is decreased with the U_3O_8 addition in both cases. However, the density of pellet is increased when the low temperature oxidizing U_3O_8 powder is added. In addition, the density drop with U_3O_8 addition is mitigated in the pellet which contains low temperature oxidizing U_3O_8 powder.

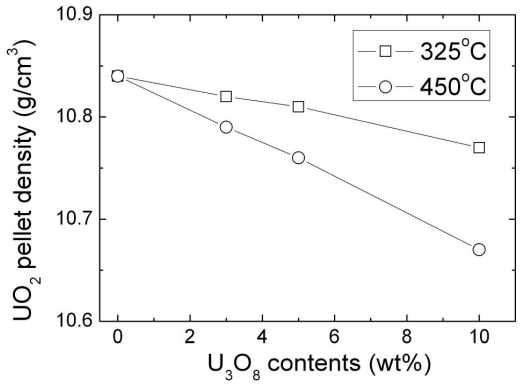
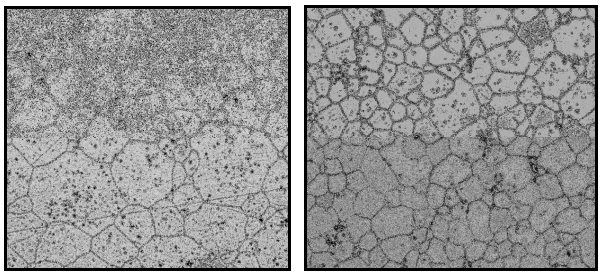


Fig. 2. The UO_2 pellet density changes according to the U_3O_8 powders contents.

The U_3O_8 powder effect on the grain growth of the UO_2 pellets was examined. Small amount of Al_2O_3 powders were mixed with U_3O_8 powders oxidized at 325°C and 450°C, respectively. These powers were added in a quantity of 5 wt% to the UO_2 powder, and then sintered at 1730°C for 4 h in H_2-1.5%CO_2 gas. Fig. 3 shows the grain structures of the 40 ppm Al-doped UO_2 pellets. It is known that Al doping promotes a grain growth of UO_2 pellets. It can be seen that the grains are enlarged by Al doping in both pellets. However, grain enlarging effect is quite different from each other even though the same amount of Al is doped in both pellets. In the case of 325°C oxidized U_3O_8 containing pellet, grains are grown from 10.5 to 16.6 μm on average by a 40ppm Al doping. On the other hand, in the 450°C oxidized U_3O_8 containing pellet, grains are grown from 9.4 to 11.2 μm on average.



(a) (b)

Fig. 3. The grain morphologies of 5wt% of U_3O_8 containing Al-doped UO_2 pellets. (a) 325°C (b) 450°C

The pore structures of the UO_2 pellets are slightly different from each other. Pore distribution and its size are homogeneous in Fig. 3(a). Whereas, grapes-like pore clusters are formed on some grain boundaries in Fig. 3(b). These grapes-like pore clusters might be obstacles to a grain boundary migration, so grain growth can be impeded.

4. Summary

The BET surface area of recycled-U_3O_8 powder can be increased by lowering the oxidation temperature of defective UO_2 pellets. The U_3O_8 powder having higher BET surface area is more effective to mitigate the density drop of UO_2 pellets when the U_3O_8 powders are added. Moreover, U_3O_8 powder having high BET surface area effectively promotes a grain growth of UO_2 pellets.

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