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Manufacturing of nuclear fuel pellets by using re-oxidized U_3O_8 powder

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1. Introduction

In a commercial UO_2 fuel pellets manufacturing process, defective UO_2 pellets which do not meet the fuel specifications of density and diameter may be produced. Defective UO_2 pellets should be reused in manufacturing new UO_2 pellets. It is a common recycling method that defective UO_2 pellets are oxidized in air at about 450 °C to make U_3O_8 powder and then added to UO_2 powder [1]. Because the recycled U_3O_8 powder has very low sintering activity compared to the UO_2 powder, it tends to decrease the density of UO_2 pellets. Other pellet properties such as grain size and open porosity are a little degraded by the addition of the U_3O_8 powder. So the amount of the recycled U_3O_8 powder is generally not higher than 10 wt% in UO_2 powder. There has been a series of work to deal with the heat treatments of the recycled U_3O_8 powder in order to improve its sintering activity [2].

This paper deals with the recycling process of defective UO_2 pellets or scraps. The defective UO_2 pellets are oxidized to U_3O_8 powders at conventional temperature of 350°C and 450°C in air. Those powders are pressed into green pellets and then sintered at 1700°C in H_2 flowing gas. Sintered pellets are re-oxidized to U_3O_8 at 450°C in air. Re-oxidized powders are added to virgin UO_2 powders to fabricate UO_2 pellets. This paper shows that the re-oxidized U_3O_8 powder sizes and the BET surface areas are greatly dependent on the sintered density of UO_2 pellets before oxidation. The re-oxidized U_3O_8 powders having a large BET surface area significantly promote a grain growth of UO_2 pellets.

2. Experimental

The defective UO_2 fuel pellets were oxidized at 350°C and 450°C in air, respectively. Those U_3O_8 powders were pressed into green pellets. The compressive pressures were 1, 2, 3 ton/cm^2, respectively. The green pellets were sintered at 1700°C for 4h in H_2 gas. Re-oxidized U_3O_8 powders were prepared by re-oxidation of above sintered pellets at 450°C. The re-oxidized U_3O_8 powders morphology were 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. 5wt% of those U_3O_8 powders was added to the UO_2 powder, which was produced through the ADU process. In Al-doped powders, the final cation weight ratio of Al/U was 40 ppm. Powder mixtures were mixed with a tumbling mixer. The powder mixture was pressed into green pellets at 3 ton/cm^2. The green pellets were sintered at 1730°C for 4 h in flowing H_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 1290°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 pellet densities which were fabricated by sintering the U_3O_8 powders. It can be seen that sintered densities depend on the oxidation temperatures of defective pellets and the pressures for green pellets compaction. The low oxidation temperature and high compaction pressure resulted in higher density of the sintered pellets.

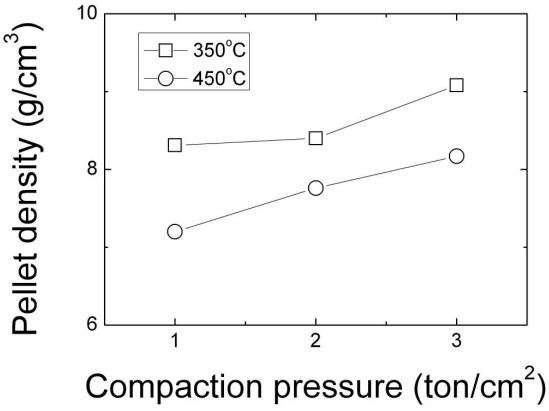

Fig.1. The sintered pellet density dependence on the compaction pressure of green pellets.

The sintered pellets were re-oxidized at 450°C in air. Fig. 2 shows the BET surface area of the re-oxidized U_3O_8 powder according to the density of sintered pellets. The BET surface area has a tendency to increase with a decreasing sintered pellet density. The BET surface area

of U_3O_8 powder which obtained by an oxidation of defective UO_2 pellet at 450°C is also shown in this figure (left-lower part). The BET surface area of the re-oxidized U_3O_8 powder can be significantly increased when the sintered pellet before oxidation has low density.

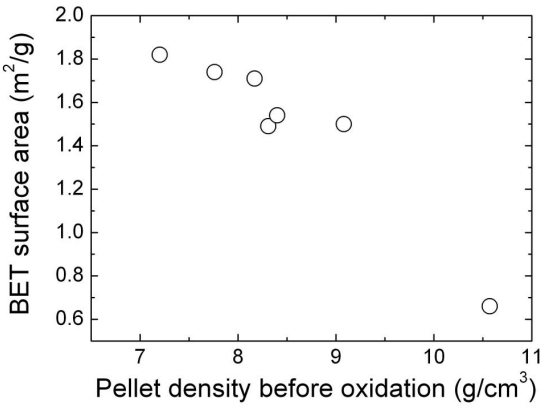

Fig. 2. The BET surface area changes of re-oxidized U_3O_8 powders according to the pellet density

It is known that powder having larger BET surface area has more sintering activity compared to that having lower BET surface area. The effect of the re-oxidized U_3O_8 powder property on the grain growth of UO_2 pellet has been examined. Fig. 3 shows the grain size distribution for the UO_2 pellets in which 5wt% of un-doped and Al-doped U_3O_8 were contained.

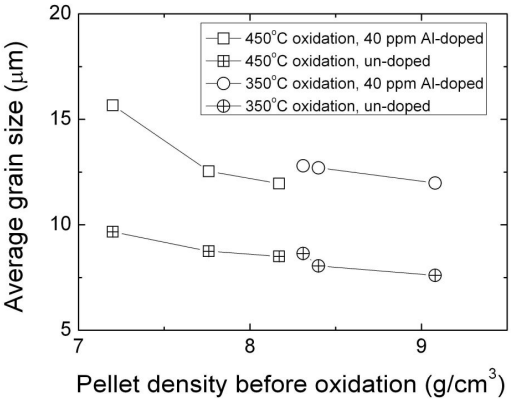

Fig. 3. The re-oxidized U_3O_8 powder effects on the grain size of UO_2 pellets.

The average grain size of UO_2 pellets are increased with decreasing density of sintered pellet before oxidation. This result is coincident with our expectation from the measured BET surface areas of different U_3O_8 powders. That is, the U_3O_8 powder having large BET surface area promotes the grain growth of UO_2 pellets more effectively. Especially, the grain size of 40ppm Al-doped UO_2 pellets are enlarged to about 16 μm which is about 2 times larger than the conventionally fabricated UO_2 pellets. Fig. 4 shows the photos of the grain structures of the 5wt% of U_3O_8 and 40ppm Al doped-UO_2 pellets. The BET surface areas of U_3O_8 are 1.82m^2/g for Fig. 4(a) and 1.49m^2/g for Fig. 4(b), respectively. It can be clearly seen that pellet of Fig.4 (a) has more sound pore structure and larger grain size.

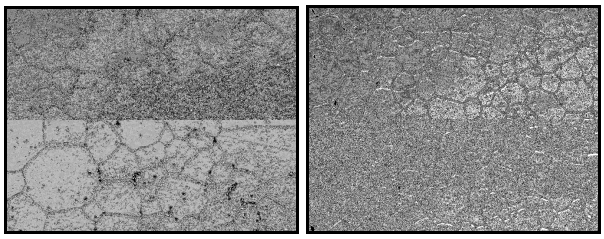

Fig. 4. The grain structures of 5wt% of U_3O_8 and 40 ppm Al containing UO_2 pellets. The BET surface area of U_3O_8 are (a) 1.82m^2/g and (b) 1.49m^2/g.

4. Summary

By sintering the U_3O_8 powders from defective pellets or scrap into UO_2 pellets and then re-oxidizing it into U_3O_8 powders, the U_3O_8 powder morphology and its BET surface area can be controlled. The BET surface area of the re-oxidized U_3O_8 powder is increased with decreasing the sintered pellet density before oxidation. The U_3O_8 powder having higher BET surface area acts as grain-size-enlarging promoter of UO_2 pellet more effectively.

Acknowledgements

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