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Characteristics of Simulated Spent Oxide Fuel Powder Prepared by Oxidation and Reduction Treatment

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1. Introduction

DUPIC (Direct use of spent PWR fuel in CANDU reactor) is a fuel recycling technology to fabricate CANDU (Canadian Deuterium Uranium) fuel directly from spent PWR (Pressurized Water Reactor) fuel material using a dry process. The key requirement for DUPIC fuel fabrication is to produce sinterable powder from spent PWR fuel pellet as a starting material. An oxidation and reduction treatment (OREOX : Oxidation and Reduction of Oxide fuels) was chosen to convert a spent fuel pellet into a sinterable powder. This treatment causes crystallographic transformations in the pellet matrix, causing the pellets to break into a fine powder.

In the present study, the preparation and characterization of the powder suitable for refabrication into a CANDU fuel pellet was carried out using simulated spent oxide fuel (SIMFUEL : simulated fuel) as a surrogate for actual spent PWR fuel. SIMFUEL is an unirradiated UO_2 fuel blended with stable chemical additives to simulate composition of an irradiated UO_2 fuel. The characteristics and sinterability of the powder are dependent on the details of the process used to prepare it. In this study, the characteristics of OREOX treated powder and pellets were investigated in terms of OREOX treating cycle and attrition milling time.

2. Experimental Method

The 11 oxide powder as a substitute for stable fission products such as Nd, Zr, Ce etc., was added into the UO_2 powder for preparing simulated pellets. The powder mixture was pressed into green pellets at 300 MPa and sintered at 1700°C for 6 hours in an atmosphere of Ar-4%H_2.

The simulated pellet was treated by the OREOX process to produce a fine powder. The oxidation was performed at 450°C in air and the reduction at 700°C in Ar-4%H_2. Two kinds of powders, 1 cycle-OREOX powder and 3 cycles-OREOX powder, were prepared for the experiment. The 1 cycle-OREOX powder was produced by only one cycle of oxidation and reduction, whereas the 3 cycles-OREOX powder was made by 3 cycles of oxidation and reduction.

The OREOX powder was milled using an attrition mill. The milling time varied from 15 to 120 minutes. After milling, the powder was pressed into green pellets at a pressure range of 100 to 350 MPa. Green pellets were sintered at 1700°C for 6 hours in an atmosphere of Ar-4%H_2.

The mean particle size and distribution of the powder was measured using a laser particle size analyzer (Malvern, UK). The specific surface area was determined by the BET method. The morphology of powder was observed by a scanning electron microscope (SEM). The densities of green pellets and sintered pellets were measured by geometric dimension and water immersion.

3. Results and Discussion

3.1 Characteristics of OREOX treated UO_2 Powder

The particle size and specific surface area of the OREOX treated UO_2 powder which was oxidized at 400°C and reduced at 600°C are summarized in Table 1. The particle size decreases with the number of oxidation and reduction cycle, while the specific surface area increases. The sintered density increases with the number of oxidation and reduction cycle, but reaching to only a final density of 9.58 g/cm^3 (87% of theoretical density). This would be due to the poor sinterability of the powder produced by only OREOX process. In this experiment, it is concluded that a milling process should be adopted for further improvement in powder sinterability.

3.2 Characteristics of OREOX treated SIMFUEL Powder

Fig.1 shows the particle size distributions of the OREOX treated and the milled SIMFUEL powders. The particle size distribution shows a typical Gaussian form changing from a narrow range shape to a wide range shape with increasing the number of cycles of OREOX treatment. The particle size of 3 cycles-OREOX powder ranges from 48.7μm to 0.5μm. The milled powder shows a broad distribution in a submicron-sized range. Therefore, milling of the OREOX powder was found to make the powder finer and more sinterable for pellet formation.

Fig. 2 shows the variations of green and sintered densities of pellets as a function of milling time. The green pellets were pressed at 300 MPa and sintered at 1700°C. The green density of 1 cycle-OREOX powder is higher than that of 3 cycles-OREOX powder. But green density of 3 cycles-OREOX powder increases from 6.14 to 6.47 g/cm^3 with increasing milling time. However, for 1 cycle-OREOX powder, green densities are almost constant in the whole range of milling time. The sintered densities of 1 cycle-OREOX powder greatly increase from 10.01 to 10.25 g/cm^3 (92.9 to 95.1% of T.D.) with increasing milling time as shown in the figure. But, for 3 cycles-OREOX powder, they range from 10.28 to 10.40 g/cm^3 (95.4 to 96.5% of T.D.) regardless milling time.

4. Conclusions

In the case of the 3 cycles-OREOX powder, good pellets were obtained at the conditions of 15 minutes milling, pressing at lower pressures than 300 MPa. Good pellets with a high sintered density were also produced from the 120 minutes milled powder after 1 cycle-OREOX treatment.

ACKNOWLEDGMENTS

This work has been carried out under the Nuclear R&D Program supported by the Ministry of Science and Technology of Korea.

TABLE 1: Characteristics of UO_2 powder produced by OREOX process

No. of cycles	Particle size (um)	Specific surface area (m^2/g)	Green density (g/cm^3)	Sintered density (g/cm^3)
Cycle 1	5.70	0.3579	6.588	8.115
Cycle 2	4.10	0.7447	6.279	9.048
Cycle 3	3.15	0.9695	6.034	9.585

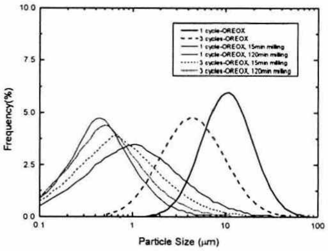

Fig.1 The particle size distributions of the cyclic OREOX powder with milling time

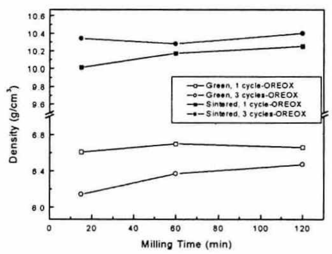

Fig.2 Effects of milling time on the sintered and green densities