

## A Study on the Sinterability Enhancement of Dry Recycling Nuclear Fuel Pellets by Using Simulated Spent Fuel

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### 1. Introduction

Burnup of nuclear fuel was continuously increased to enhance economy of nuclear power plants. In 1960's, burnup ranged 20,000~30,000 MWd/tU. In 1990's, it reached 60,000 MWd/tU[1]. A lot of high burnup spent PWR fuel will be discharged in the future.

In this study, to study fabrication characteristics of dry recycling nuclear fuel from spent PWR fuel with high burnup of 60,000 MWd/tU, the fission products of spent PWR fuel was analyzed by ORIGEN-2 code. Simulated spent PWR fuel pellets were fabricated by using  $\text{UO}_2$  powder added by the simulated fission products.

The simulated dry-recycling-fuel pellets were fabricated by dry recycling fuel fabrication flow including 3 cycle treated OREOX(Oxidation and Reduction of Oxide fuel) process[2, 3]. A small amount of dopant such as  $\text{TiO}_2$ [4],  $\text{Nb}_2\text{O}_5$ [5],  $\text{Li}_2\text{O}$ [6] are added to enhance sinterability of the powder.

### 2. Simulated Spent PWR Fuel

Fission products were analyzed by ORIGEN-2 code based on spent PWR fuel with burnup of 60,000 MWd/tU. Simulated spent PWR fuel pellets were fabricated by the general fabrication process for  $\text{UO}_2$  nuclear fuel mixed with the simulated fission products. The average density of the sintered pellets was  $10.26 \text{ g/cm}^3$ .

### 3. Simulated Dry Recycling Nuclear Fuel

The simulated spent PWR fuel pellets were treated through the three cyclic OREOX process, where the oxidation temperature was  $500 \text{ }^\circ\text{C}$ , and the reduction temperature was  $700 \text{ }^\circ\text{C}$ . The OREOX-treated powder was milled by a vertical attritor at the speed of 150 rpm for 15 minutes. The milled powder was mixed with zinc stearate to be suitable for compacting. 0.07~0.2 wt% of dopant such as  $\text{TiO}_2$ ,  $\text{Nb}_2\text{O}_5$ ,  $\text{Li}_2\text{O}$  are added to each prepared powder.

Green pellets were fabricated by the final compaction process with a pressure of 200 MPa. Zinc stearate was removed during heating of the green pellets for 2 hours at  $800 \text{ }^\circ\text{C}$  in an Ar-4% $\text{H}_2$  atmosphere. The sintering process was performed at  $1700 \text{ }^\circ\text{C}$  for 6 hours in an Ar-4% $\text{H}_2$  atmosphere.

### 3.1 Characteristics of Powder

Average particle size, surface area, the apparent density and the tap density of the powders were measured after the OREOX process and the milling process. For the OREOX-treated powder, average particle size was  $8.3 \text{ } \mu\text{m}$ . Surface area was  $4.57 \text{ m}^2/\text{g}$ . The apparent density was  $0.7 \text{ g/cm}^3$ . And the tap density was  $1.35 \text{ g/cm}^3$ . After milling process, average particle size was  $0.6 \text{ } \mu\text{m}$ . Surface area was  $6.08 \text{ m}^2/\text{g}$ . The apparent density was  $1.72 \text{ g/cm}^3$ . And the tap density was  $2.69 \text{ g/cm}^3$ . Particle size decreased, therefore surface area increased by the milling process. The densities increased to become about 3 times higher than those of the OREOX-treated powders.

### 3.2 Characteristics of Pellets

Geometric density of the green pellet was calculated by measuring the weight, length and diameter of the pellet. The density ranged from  $5.79$  to  $5.83 \text{ g/cm}^3$ . The sintered densities of the pellets without dopant ranged from  $10.04 \text{ g/cm}^3$  (94.3 % of T.D.) to  $10.32 \text{ g/cm}^3$  (96.9 % of T.D.), and the average grain size ranged from  $3.4$  to  $3.8 \text{ } \mu\text{m}$ . Both sintered density and grain size did not reach minimum criteria for CANDU fuel specification.

The sintered densities of the pellets doped with  $\text{TiO}_2$  ranged from  $10.42$  to  $10.35 \text{ g/cm}^3$ , and the average grain size ranged from  $8.8$  to  $9.7 \text{ } \mu\text{m}$ . The densities of the pellets doped with  $\text{Nb}_2\text{O}_5$  ranged from  $10.46$  to  $10.32 \text{ g/cm}^3$ , and the grain size ranged from  $7.3$  to  $9.9 \text{ } \mu\text{m}$ . The densities of the pellets doped with  $\text{Li}_2\text{O}$  ranged from  $10.37$  to  $10.28 \text{ g/cm}^3$ , and the grain size ranged from  $9.4$  to  $12.2 \text{ } \mu\text{m}$ . Sintered density increased dominantly by doping  $\text{TiO}_2$  and  $\text{Nb}_2\text{O}_5$ . All of the dopants helped to increase grain size largely.

Then, visual inspection was conducted to check for any surface defect for all the fabricated pellets. A lot of surface defects were found in the pellets doped with volatile  $\text{Li}_2\text{O}$ . As a result of experiment, a small amount of  $\text{TiO}_2$  or  $\text{Nb}_2\text{O}_5$  will be helpful to enhance the sinterability of dry recycling nuclear fuel pellets.

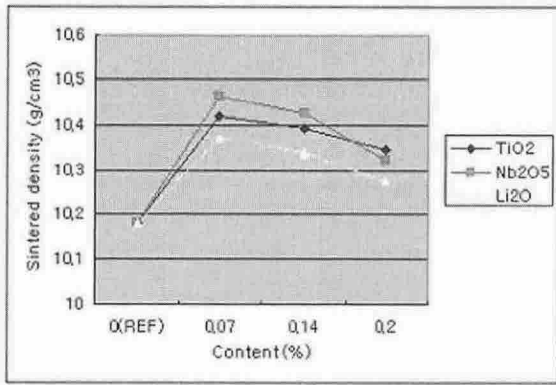


Figure 1. Density of sintered pellets.

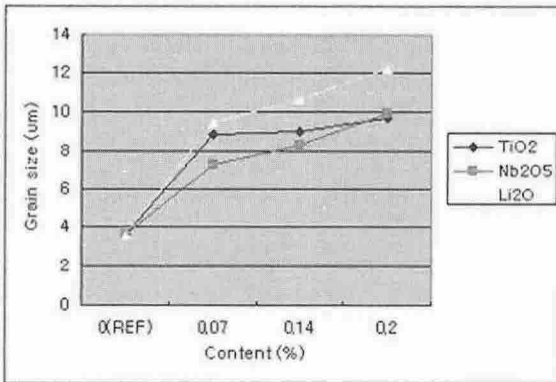
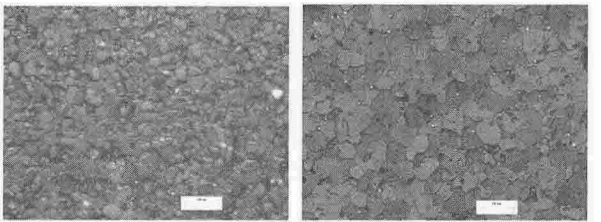


Figure 2. Grain size of sintered pellets.



(a) Not doped (b) Doped with 0.07 wt% of TiO<sub>2</sub>  
Figure 3. Microstructure of sintered pellets.

#### 4. Conclusion

To study fabrication characteristics of dry recycling nuclear fuel from spent PWR fuel with high burnup of 60,000 MWd/tU, simulated spent PWR fuel pellets were fabricated by using UO<sub>2</sub> powder added by the simulated fission products. The simulated dry recycling fuel pellets were fabricated by dry recycling fuel fabrication flow including 3 cycle treated OREOX process. A small amount of dopant such as TiO<sub>2</sub>, Nb<sub>2</sub>O<sub>5</sub>, Li<sub>2</sub>O are added to enhance sinterability of the powder. The results are as follows.

- Both sintered density and grain size of the pellets without dopant did not satisfy the criteria of CANDU fuel specification.
- Sintered density increased greatly by doping TiO<sub>2</sub> and Nb<sub>2</sub>O<sub>5</sub>.

- All of the dopants increased grain size largely.
- A lot of surface defects were found in the pellets doped with Li<sub>2</sub>O.
- Consequently, a small amount addition of TiO<sub>2</sub> or Nb<sub>2</sub>O<sub>5</sub> is effective to enhance the sinterability of dry recycling nuclear fuel pellets from spent PWR fuel of high burnup.

#### REFERENCES

- [1] W. K. Kim, etc., A Study on the Fabrication of Dry Recycling Nuclear Fuel Pellets by Using Simulated Spent PWR Fuel of High Burnup, Proceedings of KNS Spring Meeting, 2004.
- [2] M. S. Yang, etc., A Study on the Direct Use of Spent PWR Fuel in CANDU Reactors, KAERI/RR-2022/99, 2000.
- [3] W. K. Kim, etc., Fabrication Characteristics of DUPIC Fuel Pellets at DFDF, KAERI/TR-2005/2002, 2002.
- [4] K. W. Song, etc., Effect of TiO<sub>2</sub> on sintering behavior of mixed UO<sub>2</sub> and U<sub>3</sub>O<sub>8</sub> powder compacts, J. of KNS, Vol. 31, No. 5, pp.455-464, 1999.
- [5] K. W. Song, etc., Effects of Nb<sub>2</sub>O<sub>5</sub> addition on grain growth and densification in UO<sub>2</sub> pellets under reducing and/or oxidizing atmospheres, J. of Nuclear Materials 209, pp.280-285, 1994.
- [6] S. H. Kim, etc., Variation of Sintering Behavior of UO<sub>2</sub> Compact with the Addition of Li<sub>2</sub>O Powder, Proceedings of KNS Spring Meeting, 2002.