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Effect of Two-Step Sintering on Densification and Grain Growth of Uranium Dioxide

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1. Introduction

The microstructure of high burn-up structure is generally characterized by very small grain of about 300 nm in diameter, intergranular pores of 1-2 μm size and high porosity of 6-26 %.[1] Recently, HBS-analogue bulk nano-crystalline oxide fuel was proposed for a new concept of an innovative materials option for increasing fission gas retention, plasticity, and radiation-tolerance.[2] Even though a number of considerations must be taken to prove the postulation and its in-pile properties, HBS-analogue bulk pellets can be used as good samples for various out-of-pile tests to understand the properties of HBS.

Spino et al.[2] reported that HBS-analogue bulk nano-crystalline pellet was fabricated by using nano-sized YSZ (yttria stabilized zirconia) powder and PMMA bead as a pore former. They also reported that nano-sized UO_2 powder of 9-36 nm in diameter could be produced by a organic precursor decomposition method.[3]

However, nano-sized powder usually has more difficulties to be dealt with in powder metallurgical processing than commercial powders. Thus, we tried to find a way to fabricate HBS-analogue nano-crystalline UO_2 pellet with commercial ADU-UO_2 powder of about 0.1 μm size. In this study, we applied two-step sintering technique to densifying ADU-UO_2 compact without final-stage grain-growth.

2. Experimental Procedures

ADU route UO_2 powder was used for a sample preparation. UO_2 powder was compacted in a single acting press with the die-wall lubrication of zinc stearate. Two-step sintering was conducted at various temperatures for 20 h in H_2 atmosphere. The temperature range of the first step was from 1250 °C to 1500 °C and that of the second step was from 1200 °C to 1400 °C. The sintered density was measured by a water immersion method. Microstructures were observed using a scanning electron microscopy.

3. Results

It is well known that the densification and the grain growth simultaneously occur during sintering of ceramics. Figure 1 shows a typical trajectory of grain size and sintered density during sintering of UO_2. Generally, grain growth is significantly accelerated at a final-stage of sintering (over 93 %TD). This is the reason why it is not easy to obtain nano-crystalline sintered pellet even if we started with nano-sized powder.

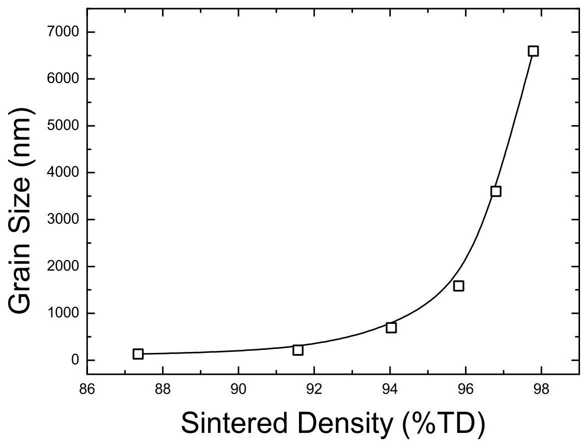

Fig. 1. Typical trajectory of grain size and sintered density during sintering of UO_2.

However, Chen et al.[4] suggested that two-step sintering enables us to densify nano-crystalline bulk pellet without final-stage grain growth. At certain condition, densification can occur with almost no grain growth and it is called 'microstructure freezing'.

Two-step sintering successfully prohibited grain growth of UO_2 during final stage of sintering. The UO_2 pellet can be densified up to 94 %TD with the grain size of less than 400 nm. The microstructures of two-step sintered UO_2 pellets were compared with those of irradiated PWR HBS and previous study[2] in Fig. 2. In this stage, it seems that HBS could be simulated by two-step sintered UO_2 in an aspect of grain size. Further efforts to simulate pore and porosity will be continued.

4. Conclusion

Two-step sintering method enables us to densify UO_2 pellet up to ~ 94%TD with very small grain size of about 300-400 nm. Porosity and pore size can be controlled with smaller uniform sized pore former through the further study. High Burn-up Structure might be simulated in an aspect of microstructure such as grain size and porosity by using two-step sintering and adding uniform sized polymer pore former. HBS-analogue UO_2 Pellet might give us good out-of-pile test samples for understanding the fuel properties of RIM (HBS) region.

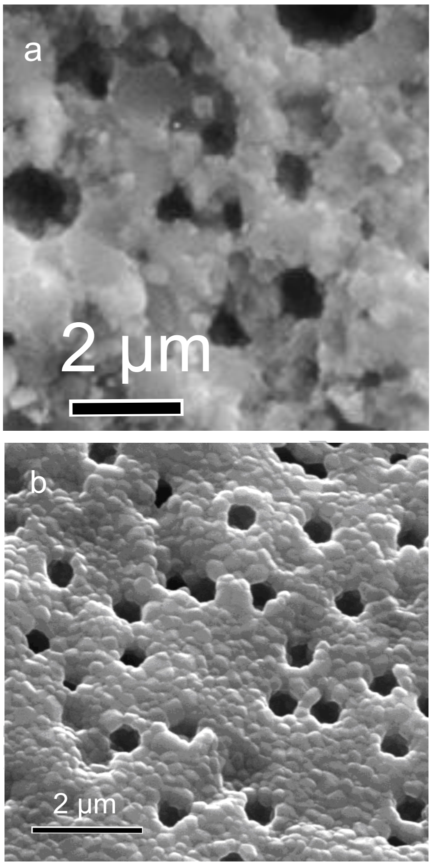

Fig. 2. Microstructures of (a) HBS region in irradiated PWR UO_2 pellet and (b) HBS-analogue yttria-stabilized zirconia (YSZ) pellet.

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