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In-Situ Observation of Sintering Shrinkage of UO_2 Compacts Derived from Different Powder Routes

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1. Introduction

Uranium dioxide is widely used as nuclear fuel pellets in light water reactors. UO_2 fuel pellets are manufactured by a conventional powder metallurgical process. UO_2 powder is compacted into green pellets under 3-5 ton/cm^2 and then sintered at 1700-1780 °C in H_2 atmosphere.

The in-reactor performance of UO_2 pellet is closely related with the characteristics of sintered pellets such as density, pore structure, and grain size. During sintering of UO_2, those properties are influenced by various sintering variables such as powder size, O/U ratio, sintering atmosphere, temperature, time, and heating rate.

The properties of sintered UO_2 pellets also depend on UO_2 powder manufacturing routes. It is well known that quite different sintering characteristics of UO_2 are shown according to the UO_2 powder manufacturing routes.[1-3] Commercially, UO_2 powders are derived from ADU (Ammonium Diuranate) route, AUC (Ammonium Uranyl Carbonate) route, or IDR (Integrated Dry Routes or Dry Conversion).

It would be important to understand the different sintering characteristics of UO_2 powders according to the powder routes, when it comes to designing a new sintering process or choosing a sintering additive for new fuel pellet like PCI (Pellet Cladding Interaction) remedy pellet.

In this paper, we have investigated the initial and intermediate sintering shrinkage of UO_2 from different powder routes by in-situ observation of green samples during H_2 atmosphere sintering. Effect of powder characteristics of three different UO_2 powders on the initial and intermediate sintering were closely reviewed including crystal structure, powder size, specific surface area, primary crystal size, and O/U ratio.

2. Experimental

UO_2 powders of three different powder routes – ADU, AUC and IDR – were used for sample preparation. UO_2 powders were mixed with a 0.3 wt% of zinc stearate in a tumbling mixer for 30 min. The compaction was conducted in a double acting mechanical press. To control the green density precisely, the same amount of powder was loaded into the compaction mold. The diameter and height of green pellets were controlled to be about 10.3 mm and 8.5 mm, respectively. The green densities of the three kinds of powder compacts were in the range of 51.5 %TD to 51.6 %TD.

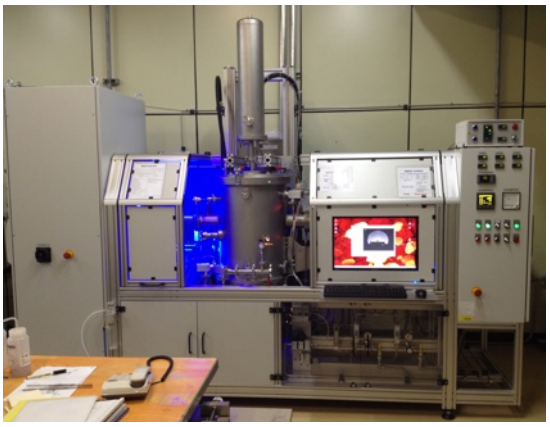

Fig. 1. Thermo-optical measurement system, TOM-AC.

Green pellets were observed in-situ during heating up to 1700 °C in a H_2 atmosphere by using Thermo-optical measuring system (TOM, Fraunhofer ISC) as shown in Fig. 1. In-situ images of green pellet were recorded by TOM. The diameter and height of samples were measured from in-situ images according to the sample temperature.

The sintered density was measured by a water immersion method. Microstructures were observed using an optical microscope after polishing the cross-section of the sintered pellet up to a 1 μm diamond polish.

Characteristics of three kinds of raw powders were investigated by using SEM, XRD and BET surface area measurement.

3. Results

Typical powder morphologies of UO_2 from different powder routes were shown in Fig. 2. ADU-UO_2 and IDR-UO_2 powders appeared to be softly agglomerated and irregular shaped. AUC-UO_2 powder had a round shape and appeared to be more hardly agglomerated. It should be noted that the size of primary crystal or particle is much smaller than that of agglomerates in all three kinds of UO_2 powders.

It appears to be little differences in the final sintered density and grain size according to the powder routes after sintering at 1700 °C for 4 h in H_2 atmosphere. However, a big difference was observed in densification kinetics at the initial and intermediate sintering stages.

Figure 3 shows the linear shrinkages of green pellets compacted with three kinds of UO_2 powders during the H_2 atmosphere sintering. The onset temperatures of

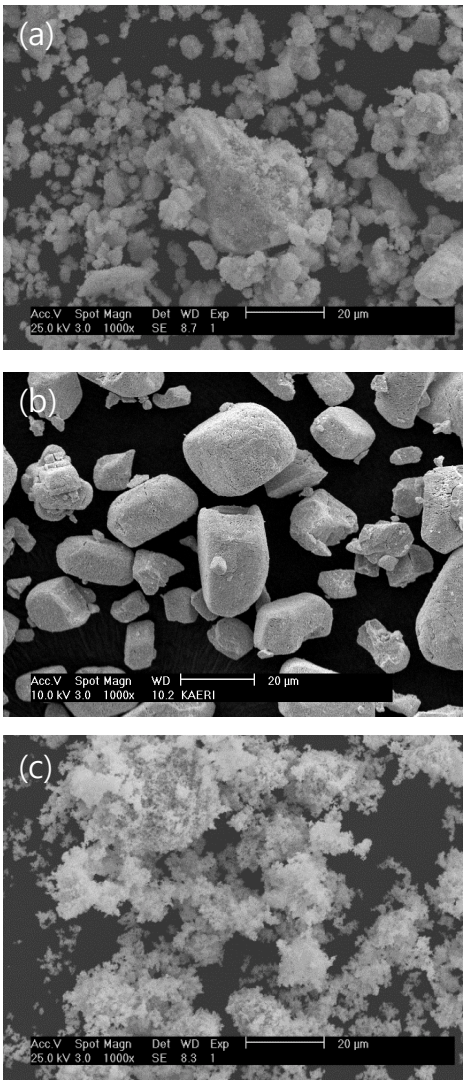

Fig. 2. SEM images of the different routes of UO_2 powders; (a) ADU-UO_2, (b) AUC-UO_2, and (c) IDR-UO_2.

densification for ADU and AUC-UO_2 appeared to be about 200 °C lower than that of IDR-UO_2. IDR-UO_2 shows the smaller linear shrinkage than those of ADU and AUC-UO_2 at a certain temperature. It would be attributed to slower neck formation and growth among particles.

Neck growth rate corresponds to the densification rate in the initial sintering stage. Densification rate is proportional to atomic mobility and driving force for materials transport. The atomic mobility depends on the diffusivity which is mostly affected by O/U ratio in pure UO_2 system. The difference in O/U ratio among three kinds of UO_2 powders appears not to be enough to explain the above retardation of the shrinkage rate in IDR-UO_2.

Different sintering behaviors might be attributed to the difference in primary crystal size of those three UO_2 powders. The driving force of sintering is inversely proportional to the powder size. The primary particle size of IDR-UO_2 powder might be larger than those of ADU- and AUC UO_2 powders. It could be confirmed by the results of high magnification SEM observation

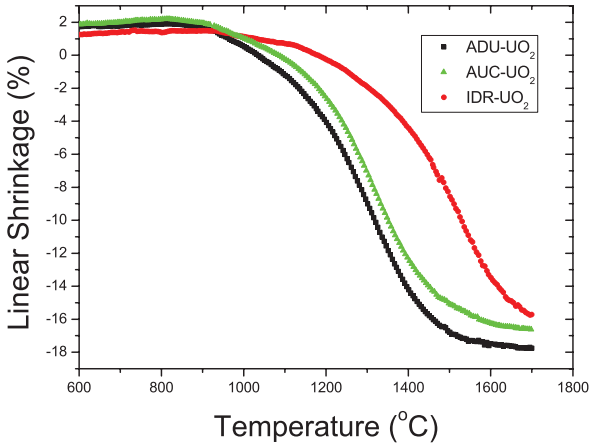

Fig. 3. Sintering Shrinkage of UO_2 compacts from the different powder routes.

and specific surface area measurement for the three kinds of UO_2 powders.

4. Conclusions

In-situ observations on the shrinkage of green pellets with precisely controlled dimensions were carefully conducted by using TOM during H_2 atmosphere sintering. The shrinkage retardation in IDR-UO_2 might be attributed to the larger primary particle size of IDR-UO_2 than those of ADU- and AUC- UO_2 powders.

ACKNOWLEDGMENTS

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