Grain-shaped UO_{2+x} seeds: preparation method & their effects on the microstructure of UO_2 pellets

Yong Woo Lee, Kun Woo Song, Ki Won Kang, Youn Ho Jung
Advanced LWR Fuel Development, Korea Atomic Energy Research Institute,
P.O. Box 105 Yusong, Daejon, 305-600, South Korea

1. Introduction

It has been known that a uranium dioxide (UO_2) pellet with large grains reduces the amount of fission gas released during irradiation[1], and thus the large-grained pellet is recognized as desirable at high burnup. The fabrication of large-grained UO_2 pellets has been investigated so widely that many fabrication methods have been developed.

Recently, it has been known that UO_2 seeds (particles) are prepared by crushing and milling defective UO_2 pellets and have a beneficial effect on the grain size of UO_2 pellet when they are added to UO_2 powder[2]. This kind of UO_2 seeds inevitably has a wide size distribution and especially has a considerable proportion of very fine sized seeds that prohibit grain growth during sintering.

This paper describes a new method of making UO_{2+x} seeds and their effects on microstructure development under conventional hydrogen sintering conditions.

2. Experimental Procedures

The overall experiment consists of making UO_{2+x} seeds and fabricating UO_2 pellets. At first, UO_2 pellets were fabricated to have a wide range of the average grain size; 14㎛, 8㎛, 6㎛, 4㎛. The 8㎛-grain sized pellet was sintered at 1700℃ for 4 h in hydrogen gas, the larger grain was at 1900℃, and the small grain size was at lower temperatures. These four kinds of pellets were slightly oxidized to become various UO_{2+x} seeds in the specially designed equipment shown in Fig. 1. This equipment consists of a vertical tube furnace, a perforated container, a vibrator, an air flowing tube, a receiving jar.

When UO_2 pellets put in the perforated container were heat treated in air, the grain boundary of UO_2 pellet was predominantly attacked (oxidized) rather than the grain interior. The oxidation of UO_2 to U_3O_8 causes a volume expansion of about 30%, which is too large to be accommodated in the pellet. So cracks develop along the grain boundary and grain-shaped particles (seeds) can be separated from the pellet. Each separated grain-shaped particle consists of a single grain or many grains. The perforated container is designed to screen large particles, which continue to be subdivided by oxidation until grain-shaped parcels are small enough to pass the perforated container. The perforated container was vibrated to help the separated seeds to pass. The separated UO_{2+x} seeds are received in a jar set at the bottom of the furnace. The jar was kept at room temperature to prohibit further oxidation of UO_{2+x} seeds.

The four kinds of UO_2 pellets, depending on the grain size, were oxidized at 320~450℃. The produced UO_{2+x} seeds were analyzed in terms of SEM morphology, size and O/U ratio.

The UO_{2+x} seeds were added to UO_2 powder and then mixed for 1h with a tumbling mixer. The powder mixtures contain the amounts of UO_{2+x} seeds ranging from 2wt% to 8wt%. The powder mixture was pressed under 300 MPa to form a green pellet and then sintered at 1700℃ for 4h in hydrogen gas. The UO_2 pellet was examined in terms of grain size. In order to examine the resintering behavior, the UO_2 pellet was heat-treated at 1700℃ for 24h in Ar-H_2 gas.

3. Results and Discussion

The UO_{2+x} seeds prepared from the four kinds of UO_2 pellets are shown in Fig. 2. The polyhedral shape of UO_{2+x} seeds is identical to that of the grain. The sizes of UO_{2+x} seeds are also similar to the grain size of pre-oxidized UO_2 pellets, so the size of a seed is mainly determined by the grain size of the pre-oxidized UO_2 pellets. However, it is found that part of the UO_{2+x} seeds is composed of multi grains. These multi-grained seeds seem to be easily formed as the grain size of the pre-oxidized UO_2 pellet decreases. This is probably ascribed to the fact that a small-grained pellet, which includes such a large area of the grain boundary, needs much more energy in separating all grain boundaries than a large-grained pellet.

The oxygen to uranium ratio of UO_{2+x} seeds was assessed from the weight gain occurring by the oxidation of the UO_{2+x} seed to U_3O_8. The values are in the range between 2.2 and 2.3, and they tend to become larger as the size of seeds decreases. In addition, the SEM morphology reveals some

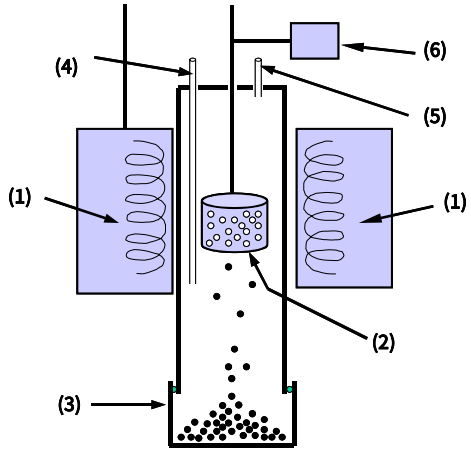

Fig. 1. An equipment for preparing UO_{2+x} seeds.
(1) vertical furnace, (2) perforated container, (3) receiving container, (4)&(5) air flowing tube, (6) vibrator .

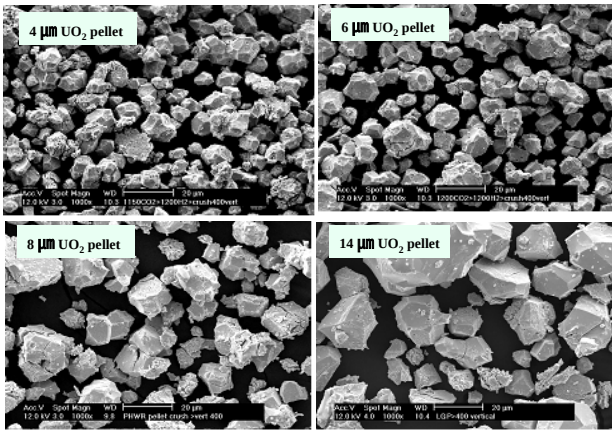

Fig. 2. UO_{2+x} seeds produced from various grain-sized UO_2 pellets.

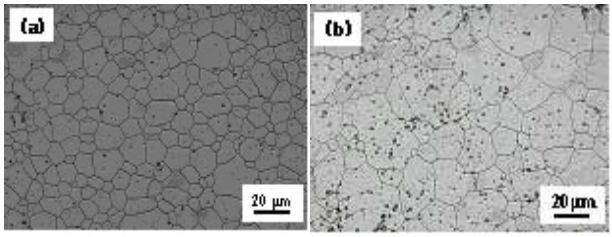

Fig. 3. Grain structure of UO_2 pellets.
(a) normal pellet, (b) seeded pellet .

fine cracks on the surface, which developed by the U_3O_8 formation. So it is believed that the surface of the UO_{2+x} seed contains U_3O_8 phase and the interior contains UO_{2+x} phase.

The UO_{2+x} seeds were added to UO_2 powder and then UO_2 pellets were fabricated through the conventional sintering process. The pellet microstructure is shown in Fig. 3. Large grains are mainly formed, with some regions of small grains. The size of large grains increases with the size of the added UO_{2+x} seeds. The average grain size determined by the linear intercept method is 14㎛ when the 5wt% of 6㎛-seeds is added to UO_2 powder.

The results of the resintering test indicate that the seeded UO_2 pellet with large grains is thermally more stable than the normal UO_2 pellet.

4. Conclusion

The grain-shaped UO_{2+x} seeds are fabricated by oxidizing the grain boundary of UO_2 pellets. This developed technique uses a perforated container that can detach the separated UO_{2+x} seeds from the oxidizing UO_2 pellet. The size of the UO_{2+x} seeds is quite dependent on the grain size of pre-oxidized UO_2 pellet, so the control of seed size can be easily achieved by varying the grain size of pre-oxidized UO_2 pellet.

The UO_2 pellet fabricated by adding the UO_{2+x} seeds has the grain structure in which large grains are mainly formed with some regions of small grains. The average grain size of UO_2 pellet is about 14㎛, and the seeded UO_2 pellet is thermally more stable than the normal (small-grained) UO_2 pellet.

Acknowledgement

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References

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