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Fabrication of UO_{2+x} single crystal rods

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1. Introduction

This paper deals with the reduction of pellet-sized U_3O_8 to UO_2 at 1300°C, and we find out that columnar and equiaxed grains, which are similar to the as-cast microstructure [1] resulting from solidification, are formed in the reduced UO_2 pellet. This paper also describes the grain structure evolution during the reduction experiments in various reducing gases with different oxygen partial pressures. The single crystal rods could be obtained by heat-treating the as-cast like structured UO_2 pellets

2. Experimental

The U_3O_8 powder was obtained by oxidation of ADU-UO_2 powder at 400°C in air. The U_3O_8 phase formation was identified by X-ray diffraction. The green pellet samples of U_3O_8 were prepared by pressing the U_3O_8 powder under a pressure of 3 t/cm^2. Upon heating of the green pellet to 1300°C at a rate of 5K/min in air, it was sintered to a dense U_3O_{8-x} pellet. When the temperature of the specimen reached 1300°C, the isothermal reduction of the U_3O_{8-x} pellets was conducted in the reducing gas atmospheres of H_2, Ar and CO_2, respectively. The microstructure of the UO_2 pellet produced by the TGA experiments was examined by an optical microscope and SEM. The polished section of the sintered pellets and the mechanically fractured sample were prepared for an optical microscope and SEM, respectively. The rod-type UO_2 single crystals are obtained by annealing the reduced UO_2 pellets. The separated single crystal rods were examined by SEM.

3. Results

The U_3O_8 green pellet was heated to 1300°C at a rate of 5K/min in air and dwelled for a few minutes. And then it was isothermally reduced in a H_2 atmosphere. Fig. 1 shows the SEM micrograph of the grain morphology at the fractured surface of a reduced sample pellet. The photo of the center shows the complete microstructure of the pellet. The grain structure of the reduced pellet is quite different from that of the normal grain pellet. At the periphery of the pellet, the layer consisting of equiaxed grains is formed. Inside of this layer, the grains grow into the pellet center and form a columnar grain structure. In the center, fine and equiaxed grains are developed. These overall features closely resemble the as-cast structure formed during the solidification of liquid under thermal gradient. That is, the as-cast structure develops during a solid-state reduction of the U_3O_{8-x} pellet.

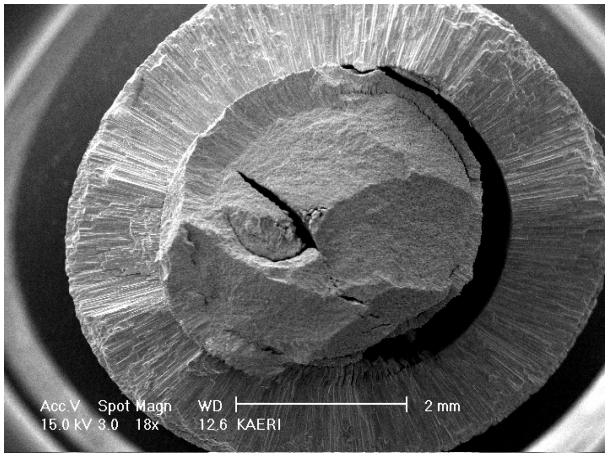

Fig. 1. The SEM images from the fractured surface of pellet obtained by reducing the U_3O_8 sintered pellet at 1300°C in H_2 atmosphere.

In order to examine the influence of the oxygen potential of the reducing gas atmospheres on microstructure evolution of U_3O_8 pellet, three different gases, H_2, Ar and CO_2, have been used for the reducing gases. Fig. 2 shows the optical micrographs of the reduced pellets. It can be readily seen that the equiaxed grains at the surface and the columnar grains at the interior are quite different depending on the reducing gases. In the pellet reduced in H_2, the equiaxed grain is very small compared to those in the other gases. The equiaxed grain size seems to be consistent with the width of columnar grains, which suggests that the UO_2 grains are formed in a random orientation at the surface and then certain favorably-oriented grains at the UO_2/U_3O_8 interface begin to grow inward. The columnar grains grow directionally and the interface between two adjacent columnar grains is almost flat. The columnar grain size is small in H_2, while the columnar grain size of the pellets reduced in CO_2 and Ar becomes large up to 100 μm wide and 1000 μm long.

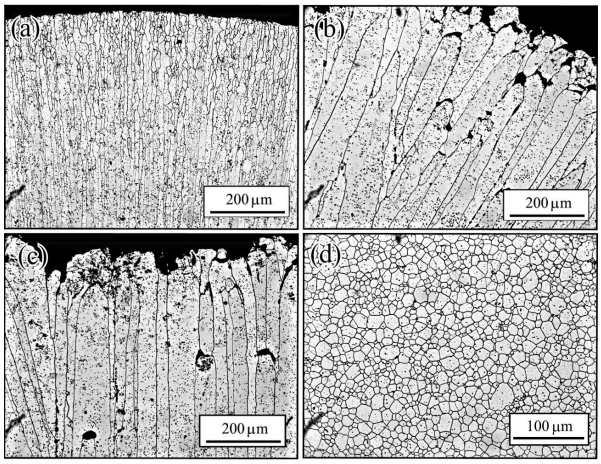

Fig. 2. Optical microstructure at the chill and columnar zone for reduced pellets.
(a) H_2 (b) Ar (c) CO_2 (d) normal UO_2 pellet

Fig. 3 shows the rod shape UO_2 single crystals obtained by heat treating the reduced UO_2 pellet in H_2 atmosphere. The grains in columnar zone are separated into individual single crystal bars. The clusters of equiaxed grains are also shown.

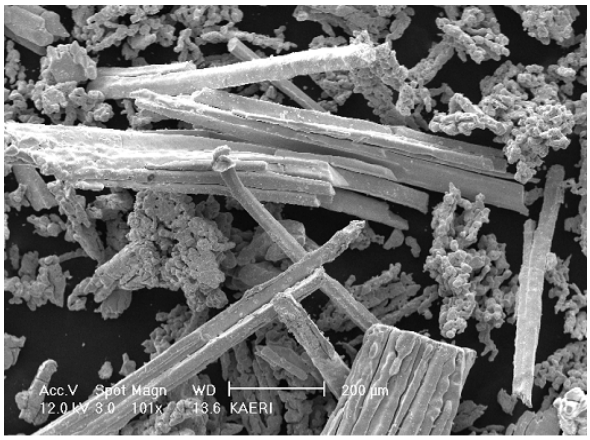

Fig. 3. SEM image or rod shape UO_2 single crystals

3. Conclusion

The experimental result shows that it possible to tailor the grain size or grain texture of the UO_2 pellets simply by changing the oxygen partial pressure of the reducing gas atmosphere during the reduction of U_3O_8 pellets. This method has a potential for applications such as the fabrication of single crystals UO_2 or textured UO_2 pellets etc.[2-4].

Acknowledgements

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