Microstructures of Columnar Grains in Uranium Dioxide

By Tadashi Kubota*

Microstructures of columnar grains of uranium dioxide grown under the temperature gradient — sintered pellets, swaged compacts and vibratory compacts — below the melting point have been studied by optical and electron micrography.

The main results obtained are as follows:

1. The dominant mechanism in columnar grain growth varies with porosity. The columnar grains in high density sintered pellets of 97% theoretical density are considered to be grown by the migration of grain boundary. As the porosity increases, the migration of large voids plays a dominant role in the columnar grain growth.

2. In addition to lenticular voids, voids or gaps along the grain boundary of columnar grains are observed in porous specimens.

3. As for the powder compacts of the same composition and different compact densities, the size and number of lenticular voids increase with decreasing density.

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I. Introduction

The attention of many researchers has been directed to the mechanism of the columnar grain growth in uranium dioxide under a certain temperature gradient.

The interpretations of the growth mechanism may be classified into two categories: One is due to the solidification after melting down, and the other is a directional grain growth at the temperatures below the melting point. Eichenberg et al. (1) have found the columnar grain growth in irradiated specimens and suggested the existence of a molten core during irradiation which formed a typical solidification pattern when cooled down. Robertson et al. (2) reported the presence of columnar grains in the temperature range below the melting point. McEwan and Lawson (3) proposed that columnar grains growth has been established satisfactorily.

In this report, the author applied a temperature gradient to uranium dioxide specimens, including sintered pellets, swaged compacts and vibratory compacts, by using the center-line heating method, and studied the microstructure of columnar grains formed at temperatures below the melting point for the purpose of obtaining more detailed information on the growth mechanism.

II. Experiments

The temperature gradient in the specimen was produced by heating a tungsten wire inserted along the center-line axis of the specimen with an alternative electric current of 50~200 amperes in a vacuum of $1.0 \times 10^{-4}$ mmHg. The principle and experimental procedure of the apparatus is essentially the same as employed by McEwan and Lawson (3). Details of the apparatus were described in a previous report (5).

UO2 powder was supplied from the Mitsubishi Metal Mining Company. For swaged compact specimens, swaging-grade high temperature-fired UO2 powder was used, while a mixture of 35wt% of swaging-grade UO2 powder (7) fired at high temperature and 65wt% of fused UO2 (3~20 mesh), and swaging grade high temperature-fired UO2 were used for vibratory compact specimens. The outline of the specimen history is summarized in Table 1.

The reasoning for the evidence that the columnar grain growth is induced in this experiment at a temperature below the melting point can be explained as follows:

Since the specimen is heated longitudinally, the molten UO2 tends to flow down to the outside through the clearance between UO2 and tungsten wire, leaving a void at the central part of the specimen. When there is no clearance between UO2 and tungsten, as in the case of a swaged compact, the molten UO2 is kept in the inside of the specimen. However, since tungsten molecules vaporized from the wire at high temperatures are mixed with the molten UO2 by the concurrent flow of the molten UO2 liquid, the molten region is distinguishable from the solid region.

Electron micrography was made on the fractured surface by use of the vacuum-evaporated carbon replica film. The unevenness of the surface structure of pores was discerned by the shadow contrasting technique.

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Table 1 Summary of the Specimens*1, *2.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Starting UO₂ Powder</th>
<th>Density of pellet &amp; compact (percentage of the theoretical density)</th>
<th>Outer diameter of UO₂ (cm)</th>
<th>Length of specimen (cm)</th>
<th>Estimated temperature at the central part of UO₂ (°C)</th>
<th>Measured temperature at the outer surface (°C)</th>
<th>Temperature gradient (°C/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1 Sintered pellet</td>
<td>Ceramic grade powder</td>
<td>97</td>
<td>2.5</td>
<td>7.5</td>
<td>2500</td>
<td>1100</td>
<td>1200</td>
</tr>
<tr>
<td>#2 Sintered pellet</td>
<td>Ceramic grade powder</td>
<td>92</td>
<td>3.5</td>
<td>7.5</td>
<td>2500</td>
<td>1100</td>
<td>1200</td>
</tr>
<tr>
<td>#3 Swaged compact</td>
<td>Swaging grade high-fired powder</td>
<td>90</td>
<td>1.9</td>
<td>7.0</td>
<td>2500</td>
<td>1000</td>
<td>1750</td>
</tr>
<tr>
<td>#4 Vibratory compact</td>
<td>Mixture of swaging grade high-fired 36wt% arc-fused powder(3-10mesh) 65wt%</td>
<td>88</td>
<td>1.9</td>
<td>7.0</td>
<td>2500</td>
<td>1100</td>
<td>1650</td>
</tr>
<tr>
<td>#5 Vibratory compact</td>
<td>Swaging grade high-fired powder</td>
<td>75</td>
<td>1.9</td>
<td>7.0</td>
<td>2500</td>
<td>1000</td>
<td>1750</td>
</tr>
</tbody>
</table>

*1 All specimens are center-line heated in a vacuum (10⁻⁴ mmHg) at about 800°C for two hours and then center-line heated at the peak temperature in purified argon atmosphere for 3 hours.

*2 Tungsten heating wire of 0.2 mm in diameter was inserted at the center-line of the specimen.

III. Results

1. General features of columnar grains

   (1) Sintered pellet

   There is a difference in microstructures of columnar grain between a high density pellet (87% T.D.) and one with a comparatively low density (92% T.D.). Photo. 1 shows an optical micrograph of columnar grains formed in the high-density pellet. The shape of columnar grains is rugged alike an expanded shape of equiaxed grains along the direction of thermal gradient. Lenticular voids are not observed in this photograph.

   As shown in Photo. 2, several different features are observed in the low density pellet. Large lenticular voids 50~80 µ in average major diameter, as have been reported either in the in-pile test or the out-pile test by many other researchers, are observed, behind which single crystalline columnar grains are formed. Voids are present in a perpendicular direction to the thermal gradient. Gaps between those single crystalline columnar grains and neighbouring grains are also observed in many cases. This is considered to be caused by the shrinkage
resulting from the densification of UO₂. Moreover, the
gaps disappear and form a dotted line structure behind
the lenticular pores. Some parts of porosities, composed
of lamination cracks and closed pores, are considered to
have contributed to the formation of voids along grain
boundaries of the columnar grains and also to the for-
mation of lenticular voids (Photo. 3).

(2) Swaged compacts
The cold swaged compact of 90 percent theoretical
density resembles in the microstructure of columnar
grains the low density pellets (Photo. 4). There is some
difference in the number and size of lenticular voids
between the two. As shown in Photo. 5, the voids
between the powder particles are combined together by
sintering phenomena, which is the origin for generating
lenticular voids. In the case of the low-density pellets,
lamination cracks and other smaller voids becomes parents
for generating lenticular voids. If such low-density
pellets having smaller voids or cracks are prepared, the
lenticular voids in columnar grains would be smaller than
those observed in this experiment, and also the number
of the lenticular voids would increase. The number of
the lenticular voids and their total volume are considered
to be slightly higher in the case of the swaged compacts
than in the case of the low-density pellets. The dif-
ference is due to the larger porosity of the compacts than
the low-density pellets. The size of the lenticular voids
in the swaged compacts is not necessarily smaller than
that of the low-density sintered pellet. This may be due
to the difference in size of internal voids contributing to
the formation of the lenticular voids.

(3) Vibratory compacts
The cross section of a center-line heated vibratory compact specimen, which is prepared from a mixture of 35 wt% of swaging grade UO\textsubscript{2} powder fired at high temperature and 65 wt% of arc-fused UO\textsubscript{2} powder (3~20 mesh) is shown in Photo. 6. The particle distribution suitable for high-density compaction, which is explained in detail by Brayer et al.\textsuperscript{(8)} reveals a characteristic in the microstructure of columnar grains in the vibratory compacts. There is no grain growth in the large fused UO\textsubscript{2} particles even above the temperature where the initial columnar grain growth can initiate, but the columnar grain growth occurs in the regions between large particles, where small powder particles are assembled together. In the case of swaged compacts, the localization of columnar grain growth is not observed, probably because the particle size is limited to be below 150 mesh. Also, in the case of vibratory compacts, lenticular voids and gaps along grain boundaries are observed in the region of columnar grains (Photo. 7). The size of the lenticular voids is larger than that of that in the swaged compacts. This may be due to the size of initial voids, resulting from larger porosity.

Although the density of the vibratory compacts is 88\% of the theoretical density and is not so much different from that of the cold swaged UO\textsubscript{2} compacts, the density of the smaller particle region is calculated to be 65\% of the theoretical density considering the volumes occupied by larger particles with 100\% theoretical density.

Photo. 8 shows columnar grains of a vibratory compact specimen of swaging grade UO\textsubscript{2} powder fired at temperature. The particle distribution of the swaging grade UO\textsubscript{2} powder is not suitable for dense vibratory compaction and the density of the compacts obtained is 75\% of the theoretical density. The compact specimen of the swaging grade UO\textsubscript{2} powder was employed in order to compare the microstructure of its columnar grains to that of the cold swaged compacts. The average size of lenticular pores is about 80 \mu, which is about twice as large as that in the swaged compacts. Moreover, the number of lenticular voids is much larger than that in the swaged compacts. The lenticular voids are arranged in a row one after another and the shape of a columnar grain behind a pore

is deformed by the following pore.

(2) Micro-pores in columnar grains

Small fragments were sampled from the columnar grains of swaged compact and the structure of micro-pores was investigated by electron micrography.

Photos. 9 and 10 show a typical pore structure in the columnar grains. The presence of tails of the pore is to be noticed. From the standpoint of the sublimation process as a means to analyze the pore migration, the tails are considered to be located at the lower temperature side. Crystal growth steps, as reported already in a previous paper (9), are also observed in the inner surface of the pore (Photo. 9).

Photo. 10 shows a much clearer tailing structure, in which the traces of a very slight tailing structure are seen. The most adequate interpretation of these three kinds of pores is as follows: The pores in Photo. 9 is in a very early stage of columnar grain growth and the lenticular pores are in the final stage, although the movement of the pores has not been confirmed since the electron micrographs were taken after the center-line heating and the position of the pores was not distinguished.

If it is possible to sample the fragment in optical micrographic magnitude, the movement of pores might be distinguished.

IV. Summary and Discussion

The growth of columnar grains has not been observed in the region of non-porous large crystals as in large particles of vibratory compacts. In the case of high-density sintered pellets, rugged columnar grains are formed, but large lenticular voids and single crystalline columnar grains are not observed. The grain growth mechanism of this type may probably be the grain boundary migration induced by the temperature gradient, as proposed by Suzuki (10) or by Robert et al (11). Of course, it may be predicted that lenticular pores of the electron micrographic size are moving and single crystalline columnar grains are formed behind the lenticular pores, but such columnar grains are too small to be discerned. Anyway, as compared with the grain boundary migration, the role of lenticular pores is not so effective for the growth mechanism of columnar grains in high-density sintered pellets. Two types of columnar grains are present in relatively low-density pellets (92% T.D).

In these pellets, large lenticular voids and single crystalline columnar grains formed behind the voids have been observed. The migration mechanism of lenticular voids or pores may probably be a combined form of the vaporization-condensation process across the voids and the diffusion in the void surface. These lenticular voids are considered to arise from the cracks or voids having already existed in the specimen in the as-manufactured state. The size of the lenticular voids has a close relation to the size of cracks or voids in the as-manufactured state and also to porosity. In high porous specimens such as the vibratory compact of swaging grade UO₂ powder fired at high temperature, almost no rugged type columnar grains are grown and the movement of the lenticular voids plays a dominant role in the growth mechanism of columnar grains.

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