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Fabrication of $BaTiO_3$ Single Crystals by Using the Exaggerated Grain Growth Method

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Abstract

The exaggerated grain growth which can occur during the sintering of $BaTiO_3$ was implemented for the fabrication of single crystals. A very small amount of SiO_2 was placed on top of a $BaTiO_3$ powder compact to form seed grains composed of twin lamellae. The seed grains continued to grow without limit and single crystals of up to a few centimetres in size were obtained. © 1997 Elsevier Science Ltd. All rights reserved

1 Introduction

In our previous investigation[1] on the exaggerated grain growth of $BaTiO_3$, it has been shown that the inhomogeneous presence of a small amount of SiO_2 liquid gives rise to an exceptional grain growth. As shown again in Fig. 1, a few grains were observed to grow to an enormous size of up to a few millimetres. For this specimen, a small amount of SiO_2 (1 mol%) was placed on top of the $BaTiO_3$ powder compact and then heat-treated at 1350°C. The formation of (111) twin lamellae (double twin) intersecting at two faceted surfaces was observed to be greatly enhanced by the SiO_2. As a consequence, the twin-plane re-entrant edge (TPRE) growth mechanism has been suggested to play a major role in this exaggerated grain growth. Note that the re-entrant edges provide perpetual ledge sites for easier growth.

An interesting question that can be raised from Fig. 1 is: what can take place if a larger powder compact which contains only a limited number of seed grains is heat-treated for a longer time? The aim of this communication is to report the answer to that question. The results will have practical significance, since $BaTiO_3$ single crystals of a few centimetres are very easily obtained.

2 Experimental

All the details of the experimental procedure are identical to those reported earlier.[1] The physical and chemical characteristics of the $BaTiO_3$ powder used have also been reported previously.[2] The average particle size and the Ba/Ti ratio determined by the manufacturer were 1·5 μm and 0·999, respectively. The SiO_2 impurity content of the powder used was 0·01 wt%. The powder was gently compacted into cubic specimens (2 × 2 × 2 cm) and then pressed hydrostatically at 250 MPa. In parallel, a slurry of SiO_2 (99·5%, Junsei Chemical Co., Ltd, Tokyo, Japan) powder was prepared using ethyl alcohol. By using a pipette, a minimum quantity of slurry was dropped on the centre of the top surface of the green $BaTiO_3$ compact. After drying, the compacts were sintered at 1370°C in air for up to 80 h.

Figure 2 shows the overall shape of the specimen after sintering for 40 h. The single crystals, i.e. the regions swept by the seed crystals, are clearly discerned from the appearance of the specimen because they are rather dark and brilliant compared to the remaining, polycrystalline, region. Due to a very limited quantity of SiO_2 the number of grains with (111) twin lamellae which can act as the seeds for fast growth was minimized and consequently these can grow without impinging on each other. For this particular specimen, three or four seeds are believed to have formed and each one has grown to about 0·5 cm in size.

In Fig. 3, some single crystals obtained by this method are shown. They were obtained after heat-treatment for 80 h, under conditions where the number of seeds was limited to one or two,

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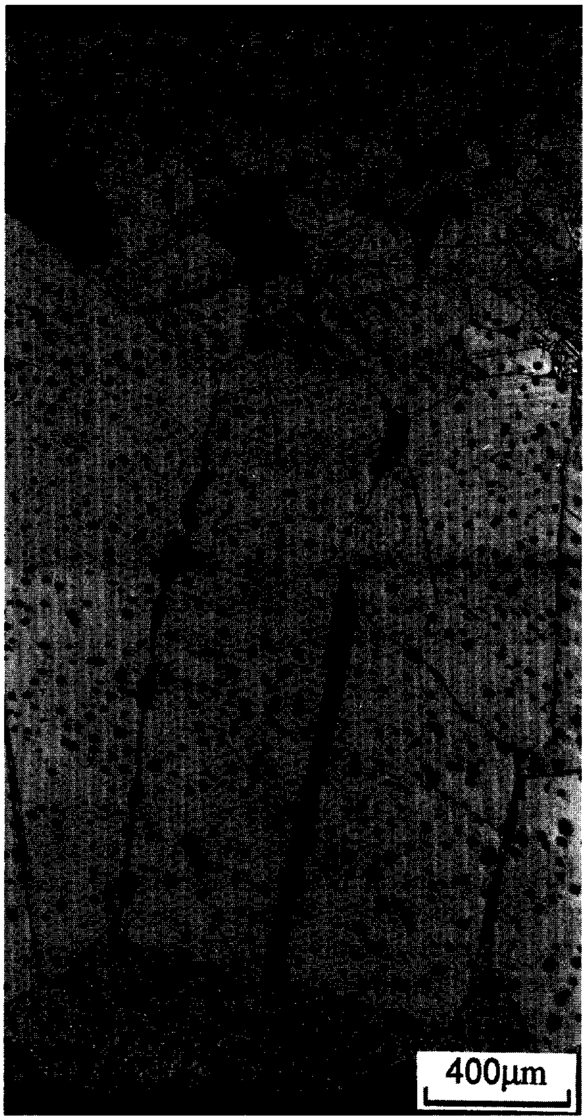

Fig. 1. Microstructure of the $BaTiO_3$ specimen. The SiO_2 slurry was dropped on top of the green $BaTiO_3$ compact and then heat-treated at 1350°C for 8 h (From Ref. 1).

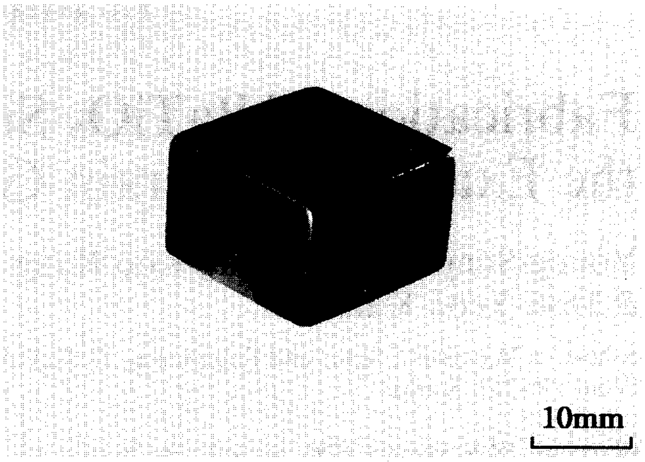

Fig. 2. Overall shape of the $BaTiO_3$ specimen. The minimum amount of SiO_2 slurry was dropped on to the compact and then heat-treated at 1370°C for 40 h. Note the single crystals grown from the top of the specimen.

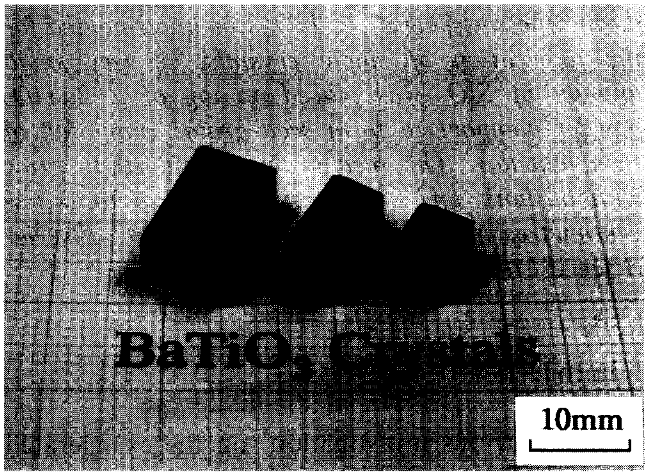

Fig. 3. The $BaTiO_3$ single crystals obtained after heat-treatment at 1370°C for 80 h. They have been cut from the specimen and machined.

crystals of up to 1·5 cm in size were produced. In this respect, control of the number of seeds is critical for the preparation of large crystals. By dropping the minimum of SiO_2 onto a very limited region of the $BaTiO_3$ powder compact, the number of seed grains having twin lamellae could be minimized. From the size of the crystals obtained, the growth rate in this experiment is expected to be higher than about 200 μm/h. As can be expected from Fig. 1, many pores were trapped inside the single crystals and consequently these were not transparent. Twin lamellae were also present without exception in each crystal.

It has been reported[3] that $BaTiO_3$ single crystals can be fabricated by making a sandwich type specimen: the seed single crystals are contacted to a polycrystalline fine-grained sample and bonded under pressure. It is found, however, that the growth of a single crystal in the sandwich method is only possible when the heat-treatment temperature is lower than 1300°C. At higher temperature, coarse grains develop in the fine-grained matrix and thus the material transfer to the single crystal becomes negligible. The growth rate in that experiment is around 30 μm/h, which is significantly lower than that observed in the present experiment. The high temperature adopted in this experiment is expected to be one of the reasons for such a fast growth rate. Note, however, that the growth mechanism is also different as has been explained in detail in our previous report.[1] Various properties of the single crystals shown in Fig. 3 as well as the fabrication of transparent single crystals without residual pores are being investigated.

Acknowledgements

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