Crystal Growth of High-Tc Superconducting Oxides from the High-Temperature Solutions(Abstracts of Doctoral Dissertations,Annual Report (from April 1995 to March 1996))

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Abstract

Chap. I. General Introduction

The high-Tc superconducting oxides are complex crystal structures are multicomponent compounds which shows incongruent melting. There are several difficulties in growing single crystals of high quality in practice. In this study, the growth of the 123 compounds ($RBa_2Cu_3O_x$:R=Y and rare earth elements) was chosen to be a model case for high-temperature solution growth.

In this chapter, the outline, the aim and the background of the study were described with special reference to the concepts of crystal growth.

Chap. II. In situ Observation Experiments

In situ observation of the crystallization and the dissolution were carried out by using high temperature optical microscopy, which is made up of an optical microscope and an infared heating furnace. The whole process of melting and crystllization was recorded by a time lapse video recorder using a CCD camera. The error of the temperature measurement of a sample was estimated to be about ± 2°C. This method was proved to be effective both in constructing the phase diagram of a complex system and in confirming the peritectic reaction.

The aims of experiments were (1) to determine the pseudo-binary phase diagram of the "$SmBa_2Cu_3O_x$"–"BaO+CuO" systems in air , in oxygen and in argon mixed with oxygen, respectively, (2) to observe how the $SmBa_2Cu_3O_x$ crystals nucleate and grow from the solution, (3) to observe the peritectic reaction of Y_2BaCuO_5 + Liquid $\rightleftharpoons$ $YBa_2Cu_3O_x$.

The results of these experiments give us an important clue how to grow a high quality single crystal of high-Tc superconducting oxide from the high-temprature solutions.

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Chap. III. Crystal Growth by Self-seeded Solution Growth Method

Growth experiments of the 123 crystal were tried using a self-seeding technique, which controls the number of seeds by dissolving the crystals until only one crystal is left.

The sample pellet of 2g was put on a cleaved plate ($20 \times 20 \times 0.5mm^3$) of an MgO single crystal, which was set on the aluminum plate in the electrical furnace. It was possible to observe the liquid surface by a telescope, when the sample was melted to be droplet on an MgO at high temperature up to 1000°C.

As a result of this self-seeding experiment, only one crystal which has the dimension of $10 \times 10 \times 0.5mm^3$ was successfully obtained from the starting material of 2g. The surface of the grown crystal was so flat and smooth that the elementary growth steps were observed by using an Atomic Force Microscope.

Chap. IV. Crystal Growth by the Trvelling-Solvent Floating-Zone Method

Single crystal of $SmBa_2Cu_3O_x$ have been grown successfully by the travelling-solvent floating-zone (TSFZ) method. It has been possible to keep the stable molten-zone and continuing growing crystal by newly applying low oxygen pressure atmosphere.

The optimun composition of the solvent was determined to be 30mol% (123 phase) from the liquidus line of the "$SmBa_2Cu_3O_x$"–"$Ba_7Cu_{18}O_{25}$" system in argon mixed with oxygen at a oxygen partial pressure of 0.3×10^{-2} atm. The growth experiments were carried out in the mixed gas at a pressure of $0.3\text{-}0.7 \times 10^{-2}$ atm. The growth rate was in the range of 0.7-1.0mm/h.

As a result of this experimrnt, the size of the grown boules was 6mm in diameter × 40mm in length. The grown crystals were identified to be $SmBa_2Cu_3O_x$ phase by X-ray powder diffraction and Energy Dispersive X-ray Analysis (EDXA). The size of the grain was about 1×1 mm^2 × 5mm in length along the c-axis. The orientation of the 123 phase was confirmed by using a polarizing optical microscope, after twinning patterns were brought about by annealing in O_2 atmosphere at 400°C for about 100 hours.

Superconducting transition temperature of a crystal annealed in oxygen was confirmed to occur at 94K in magnetization measurement down to 77K.

Chap. V. Summary

In this chapter, the results of this study are summarized.