

Growth of $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ single crystals by travelling solvent floating zone method

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Abstract

Superconducting characteristics have been studied from the viewpoint of the annealing condition on single crystals of the $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ system grown by the travelling solvent floating zone (TSFZ) method. The possibility that Ce substitution to the Re site is discussed.

Key words: $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ single crystal, TSFZ, high-temperature Ar anneal

1. Introduction

In $\text{YBa}_2\text{Cu}_3\text{O}_y$ the superconducting transition temperature T_c is above 90K¹⁾. It has been well established that the layered-perovskite structure of $\text{YBa}_2\text{Cu}_3\text{O}_y$ is retained when Y is replaced by most of the rare earth elements. While a sample including the Ce ions, which are easily oxidized by valence state of +4, tend to form another phase from a structure with defects. However, some reports insist that Ce is substituted to the Re site^{2,3)}. C.R.Fincher et al. reported that the effect of Ce substitution on T_c is the same as that of Pr. On the other hand, S.Y.Xiong et al. suggested that $\text{Pr}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ polycrystal was created by solid state reaction. From this points we must go on to an even more detailed examination of the concept of substitution effect. In this paper we report the growth of $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ single crystals with various Ce contents x and discuss the Ce substitution to the Re site by the annealing conditions such as temperature and atmosphere.

2. Experimental

Growth of single crystals of $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ was performed by the TSFZ method. The composition of the feed rod and solvent was $(\text{Nd}+\text{Ce}):\text{Ba}:\text{Cu}=1:2:3$ and $(\text{Nd}+\text{Ce}):\text{Ba}:\text{Cu}=1.0:7.6:21.6$, respectively. The growth rate was in the range of 0.3-0.4mm/h. During the growth, a mixture gas of Ar and O_2 in the ratio of 1000:1 was introduced into the sample space. Many small single crystals were included in the obtained rod and were picked up from it. Typical crystal size was $2\times 2\times 0.5$ mm³. The characterization of the obtained single crystals was carried out by the X-ray back Laue method and the X-ray powder diffraction analysis using a X-ray diffractometer. Grown crystals have been also characterized by a superconducting quantum interference device (SQUID) magnetometer. T_c value is defined as the onset temperature of the diamagnetic signal.

3. Results and discussion

According to Energy-Dispersive X-ray Spectroscopy (EDX) analysis the Ce contents in all samples were less than that in the feed rod ($x=0.2, 0.3$). Ce concentration in the growth crystal is about 10-11%. The re-

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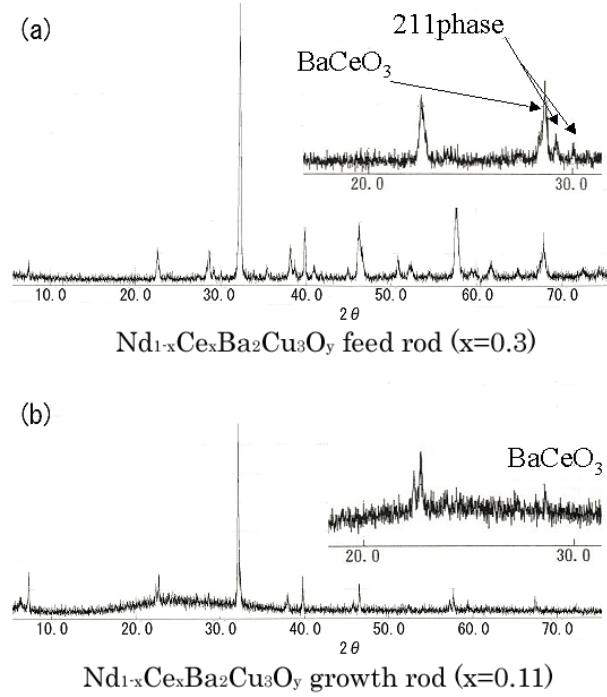


Fig. 1. X-ray diffraction patterns of $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ for (a) feed rod ($x=0.3$) and (b) growth rod ($x=0.11$).

sult of about 10% in Ce concentration is consistent with the previous report³⁾. According to the X-ray powder diffraction analysis, the as-grown single crystal ($x=0.11$) was confirmed to have a tetragonal structure with $a=3.90\text{\AA}$ and $c=11.86-88\text{\AA}$. Figure 1 shows the x-ray diffraction patterns of feed rod ((a) $x=0.3$) and growth rod ((b) $x=0.11$). The peak at 28.9° was attributed to (110) of BaCeO_3 while the 29.2° and 30.1° peaks were assigned as one by the 211 phase⁴⁾. The intensity of these peaks was quite small in the growth rod. Because of the existence of about 10% of the Ce ions by the EDX analysis and the result of the X-ray diffraction, Ce has the possibility to be substituted to the Re site in the $\text{ReBa}_2\text{Cu}_3\text{O}_y$ structure. To realize the superconductivity, post annealing was necessary and various annealing conditions were adopted. The samples are annealed at high temperatures in Ar atmosphere and then at 300°C in O_2 for 1 week to introduce fully the oxygen into the sample. Figure 2 shows the temperature dependences of the magnetization in the single crystal and polycrystalline samples, annealed at several temperatures in Ar and O_2 . The remarkable decrease of T_c for polycrystal is observed which originates from the off-stoichiometry from the $\text{ReBa}_2\text{Cu}_3\text{O}_y$ composition. The $\text{NdBa}_2\text{Cu}_3\text{O}_y$ crystal annealed at 900°C has the best quality because of its sharp transition. The transition width is quite large in the $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ sample obtained by an-

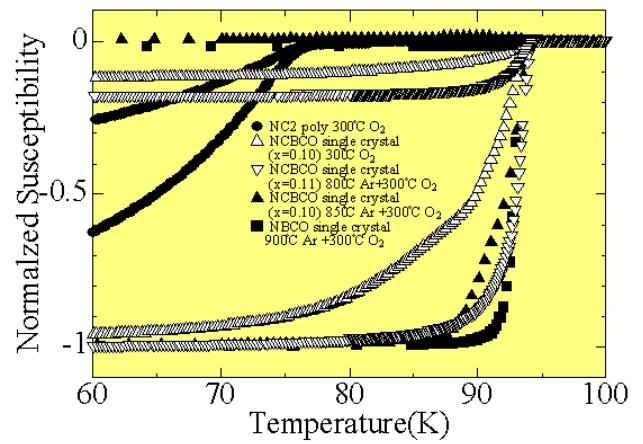


Fig. 2. Temperature dependence of the magnetization of the $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ polycrystal ($x=0.2$) and single crystal ($x=0.0, 0.1, 0.11$).

nealing at 800°C - 850°C although T_c is the same. In contrast, the transition of the O_2 -annealed sample at 300°C is much lower than that of Ar-annealed sample. This result indicates that the contamination of the Nd ion with large atomic size at the Ba site is strongly suppressed by the high temperature Ar anneal. This is also observed in the $\text{Nd}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ single crystals⁵⁾.

4. Summary

In summary, we have tried the growth of $\text{Nd}_{1-x}\text{Ce}_x\text{Ba}_2\text{Cu}_3\text{O}_y$ single crystals with various Ce concentrations by the TSFZ method under 0.1% O_2 in Ar atmosphere and annealed the obtained single crystals at various temperatures and atmosphere. It succeeded in the growth of the single crystal that the precipitation of BaCuO_3 and 211 phase was strongly suppressed. It becomes possible to create the single crystal that substituted Ce to R site by TSFZ method. The experimental results of the magnetization indicated that the high temperature Ar-anneal at $800 - 850^\circ\text{C}$ was quite effective to obtain the sample with sharp superconducting transition in the growth single crystal. T_c was similar to 94K of $\text{NdBa}_2\text{Cu}_3\text{O}_y$ system.

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