Low temperature liquid phase epitaxy of YBa₂Cu₃O_y films by the molten KOH method

Shuhei Funaki, Yasuji Yamada, Ryota Okunishi, Yugo Miyachi

Shimane Univ. 1060 Nishikawatsu-cho, Matsue, 690-8504, Japan

Abstract

In this paper, we endeavored to fabricate the high performance REBa₂Cu₃O_y (RE123; RE: rare earth elements) films by feasible simple process which is liquid phase epitaxy method using molten KOH flux at low temperature. Obtained films of Y123 fabricated in N₂ atmosphere showed bi-axial orientation of 123 single phase completely above 550°C. Furthermore, the Y123 film fabricated at 650°C showed sharp transition and critical temperature achieved over 90 K.

1. Introduction

It is well known that the RE-Ba-Cu-O superconducting material has anisotropic physical properties due to layered crystal structure with alternating two-dimensional CuO₂ planes, and the critical current density (J_c) along the CuO₂ planes (// ab-plane) is superior to that of along the c-axis direction. Furthermore, in the case of REBa₂Cu₃O_v (RE123; RE: rare earth elements), the increase of misorientation angle between two grains causes serious degradation of J_c , even for the c-axis oriented film [1]. However, RE123 superconductor needs a high growth temperature during bi-axial oriented film fabrication that causes degradation of superconducting properties due to impurity diffusion from substrate. Moreover, for the achievement of high critical current (I_c) , a particular technique for the fabrication of thick RE123 film is needed. From these results, In order to apply any superconducting devices, lower temperature and higher rate fabrication are required.

As example of high growth rate fabrication methods, it is well known the Liquid phase growth process such as top-seeded solution growth (TSSG) and liquid phase epitaxy (LPE). For single crystalline bulk crystals and LPE films of RE123, BaO-CuO flux (so-called self-flux) was used as a growth flux. Although the BaO-CuO flux provides a high growth rate more than 10 µm/min, the growth temperature needs approximately 1000°C. Recently, we achieved the synthesis of bi-axial oriented REBa₂Cu₄O₈ (RE124) epitaxial films on NdGaO₃ (NGO) single crystalline substrate fabricated by molten KOH method with low growth temperature of ~650°C in air [2]. From P-T diagram of Y-Ba-Cu-O superconducting material, Y124 decomposed into Y123+CuO under low $P(O_2)$ [3]. In this investigation, in order to establish the fabrication method of Y123 film with lower temperature and higher growth rate, we fabricated the Y123 films on NGO substrate by molten KOH method in reducing atmosphere, and also discussed

the grown phases, orientation and superconducting properties.

2. Experimental procedure

Yttrium oxide, barium carbonate and copper oxide powders were used as starting materials, and potassium hydroxide (KOH) was used as a solvent. These starting materials were weighed with a total weight of 10 g with a molar ratio of Y : Ba : Cu = 1 : 2 : 3, and then mixed and put into alumina crucible with KOH of 100wt% to the raw materials. The NGO (001) single crystalline substrate was also put into the mixture. Then the mixture was heat-treated using a tube furnace at 500~700°C for 12 hours in N₂ flow. The obtained samples were washed in distilled water and ethanol using ultrasonic cleaning to eliminate the KOH and K₂CO₃.

Phase identification and orientation were measured by X-ray diffraction (XRD) pattern with a CuK α source. Grain size and surface morphology were obtained by scanning electron microscope (SEM). Superconducting properties was measured by a standard four-probe method.

3. Results and discussion

Fig. 1(a) shows XRD $2\theta - \theta$ pattern of films grown fabricated at 500~700°C. It is revealed that the *c*-axis oriented Y123 phase grew on NGO (001) substrate, although the







Fig. 2 XRD $2\theta \chi - \phi$ pattern (ϕ -scan) of Y123 films and NGO substrate

precipitation of Y211 and Cu₂O phase were contained above 700°C. Fig. 1(b) shows in-plane orientation of Y123 phase in relationship to NGO substrate. We can found that the Y123 phase grew with in-plane alignment such as Y123 [110] // NGO [100]. From these results, we succeed fabrication of bi-axial oriented Y123 films by molten KOH flux with low temperature of 550°C.

 ρ -*T* curve of Y123 films fabricated at 650°C is shown in Fig. 3. Obtained film showed sharp transition and critical temperature (T_c) achieved over 90 K, which is compare favorably with conventional Y123 superconductor.



Fig. 3 *ρ*-*T* curve of Y123 film fabricated at 650°C

3. Conclusions

We fabricated the Y123 films on NGO substrate by molten KOH method in reducing atmosphere, and also discussed the grown phases, orientation and superconducting properties. Y123 film showed a bi-axial orientation on NdGaO₃ (001) with 45° rotations in *ab*-plane direction. Moreover, from ρ -*T* curve, Y123 film fabricated at 650°C showed sharp transition and T_c achieved over 90.4 K. Finally, it is considered that the molten KOH method is suitable for the production of Y123 film at low temperature with high crystalline perfection and superconducting properties.

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