Experimental study in the development of HTS NMR probe

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Abstract. We investigated the effect of introducing artificial pinning centers (APCs) into $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films to improve the critical current density (J_c) and reduce the microwave surface resistance (R_s) in a high dc magnetic field. Introduction of the APC was realized by dispersing $BaHfO_3$(BHO) particulates in the YBCO thin films. Four types of sintered YBCO ceramics with x wt % (x = 0.5, 1.0, 2.0) BHO were used as laser targets in order to fabricate YBCO thin films with various amounts of BHO and investigate the influence of the BHO doping concentration. J_c was measured by a four probe method. We measured J_c in a magnetic field which was applied parallel to the c-axis (J_c(p)), and J_c in a magnetic field which was applied perpendicular to the c-axis (J_c(n)). The R_s was calculated using a correlating equation of J_c and R_s. The J_c(p) at 5 T and 20 K of 1.5 wt% BHO doped YBCO thin film was the largest at 3.4 MA/cm^2. The J_c(n) of 1.5 wt% BHO doped YBCO thin film was 2.5 MA/cm^2, which is comparable to non-doped. The R_s(n) and R_s(p) of all YBCO thin films was less than 25 μΩ at 500 MHz. Therefore, we clarified the possibility that the most effective way to fabricate thin film for NMR coil application is by doping YBCO with BHO 1.5 wt%.

1. Introduction

High resolution nuclear magnetic resonance (NMR) spectroscopy is an important technique for determining molecular structure and physical properties of liquid samples [1]. A major drawback is its low signal-to-noise ratio (S/N) compared with other spectroscopic methods. (S/N) can be improved by raising the magnetic field strength, lowering the temperature of the sample, or improving the performance of the radio frequency (RF) probe and electronics [2]. RF probes can be improved in three general ways: First, reducing the volume of the detection coil, the signal per nuclear spin can be increased [3]. Second, by cooling the coils and preamplifiers, the noise can be reduced by up to a factor of four [4]. Finally, coils built from low-loss materials such as superconductors can improve the sensitivity [5]. In particular, the R_s of high temperature superconductivity (HTS) thin film is smaller than that of the conducting metals usually used in the microwave frequency region [6], and improvement of sensitivity in nuclear magnetic resonance (NMR) analysis can be expected from the characteristics of the low R_s under a high magnetic field. However, when NMR pick-up coils are applied to high magnetic fields, such as 11.7 T in the case of a 500 MHz NMR, the J_c of HTS films reduce rapidly according to the applied magnetic fields. Furthermore, the pick-up coils are necessary to generate a radio frequency during the excitation phase of a pulsed NMR experiment. Due to these factors, superconducting of NMR pick-up coils are not yet wide spred.

In particular, because the magnetic field is applied perpendicular to the c-axis, it is necessary to improve the J_c and R_s for NMR detection coil applications. It has been reported that to improve the J_c at high magnetic fields, high temperature superconductivity (HTS) thin film doped with $BaMO_3$ (M = Metal, Zr,

Sn) as APCs are effective [7]-[8]. Recently, H. Tobita et al reported that GdBCO coated conductors doped with BHO exhibit superior properties as APC material [9]. Additionally, Y. Ichino et al reported that a J_c of 2 MA/cm^2 at 20 K, 10 T was obtained by doping a 3.7 vol% of BHO to SmBCO films. In this study, we will clarify the doping amount of BHO suitable for NMR pick-up coils by evaluating the J_c of YBCO films doped with BHO at a low temperature and high magnetic field, targeting an increase in J_c and a decrease in R_s for YBCO thin films [10].

2. Experiment

Non-doped and BHO doped YBCO thin films with a thickness of 200 nm were deposited by using a pulsed-laser-deposition (PLD) method which used a KrF excimer laser (λ = 248 nm) on MgO (100) single crystalline substrates. The laser energy and repetition rate were 180 mJ and 1 Hz, respectively. We used commercial 1.5, 2.0 and 3.0 wt% BHO doped YBCO targets to fabricate BHO doped YBCO thin films. For comparison, we also used a non-doped YBCO target. The holder was heated by a SiC heater, and a substrate was attached directly to the holder and was heated by heat conduction. The holder was rotated to form homogeneous films. In this study, all films were deposited at approximately 720 °C with an oxygen pressure of 250 mTorr. The deposited thin films were then annealed at 430 °C with an oxygen pressure of 2000 Pa for 1 hour. For the analysis of the transport properties, UV photolithography and dry etching were used in order to obtain 2 mm long and 100 μm wide strip patterns. Au was deposited in order to reduce contact resistance to the electrode. The patterned samples were then lodged on a sample holder and mounted in cryostat endued with a 5 T superconducting magnet. The standard 1 μV/cm criterion was used to evaluate critical current values from the characteristics collected in the four probe configuration. In this study, since we could not use the 1 μV/cm standard due to influence of noise from the characteristics of *I-V*. We defined the critical current values by using the standard 1 μV/cm^2 after averaging the data from 10 experiments. The R_s values were calculated using a correlation equation with J_c and R_s [11-12].

$$R_s\ [\mathrm{m\Omega}] = 1.8 \times J_c^{-1}\ [\mathrm{MA/cm^2}] \text{ at } 21.8\ \mathrm{GHz} \quad (1)$$

The crystalline of the films was evaluated by X-ray diffraction (XRD) to obtain the $\theta/2\theta$ value and ϕ-scans. The $\delta\omega$ was the full-width half-maximum (FWHM) of the rocking curve of the (005) peak of the films, and the $\delta\phi$ was the FWHM of the ϕ-scan of the (102) plane of the thin films.

3. Result & Discussion

Figure 1 (a) shows the $\theta/2\theta$ results of the XRD measurement of the YBCO superconducting thin films. Both the non-doped and BHO doped samples showed strong YBCO (00l) peaks. Thus, the observations of the YBCO phase suggest that the YBCO films grew with c-axis orientation on the MgO substrate. Figure 1(b) shows X-ray ϕ-scan diffraction patterns of the (102) face of the YBCO. Each of the YBCO (102) peaks was clearly detected at 90° intervals, indicating that thin films have a four-hold symmetry on the MgO substrate. Thus, all thin films were revealed to be epitaxially grown cube-on-cube biaxially oriented thin films. Table 1 shows the $\delta\omega$, $\delta\phi$ and T_c of the non-doped YBCO and BHO doped YBCO thin films. For all BHO doped YBCO thin films other than 3 wt% doped thin film, the value of $\delta\omega$ was less than 1.6 and the value of $\delta\phi$ was less than 1.8. Therefore, no effect on the crystallinity in 2.0 wt% or less for BHO doped YBCO was observed.

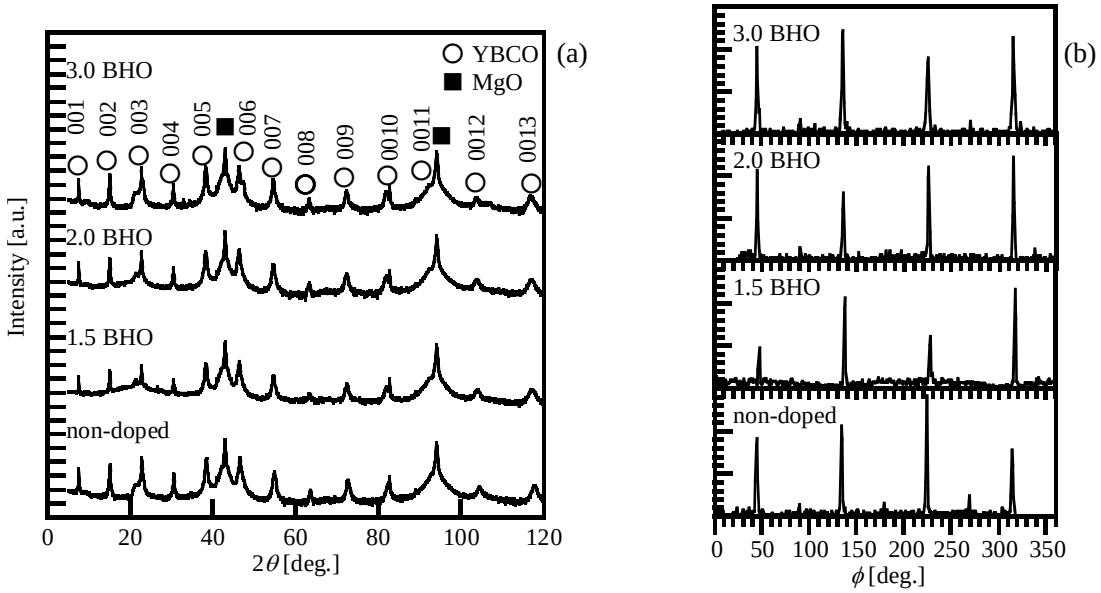

Figure 1. (a): X-ray θ-2θ pattern collected on the non-dope and BHO1.5, BHO2.0 and BHO3.0. (b): φ-scan of the non-dope and BHO1.5, BHO2.0 and BHO3.0.

Table 1. APC material and composition of the thin film. T_c, $\delta\omega$ and $\delta\phi$ of the each thin films.

Sample name	Super conductor	APC material	Additive amount [wt%]	$\delta\omega$	$\delta\phi$	T_c [K]
non-doped	$YBa_2Cu_3O_{7-\delta}$	–	–	1.62	1.79	86.9
BHO1.5	$YBa_2Cu_3O_{7-\delta}$	$BaHfO_3$	1.5	1.34	1.79	82.3
BHO2.0	$YBa_2Cu_3O_{7-\delta}$	$BaHfO_3$	2.0	1.54	1.71	81
BHO3.0	$YBa_2Cu_3O_{7-\delta}$	$BaHfO_3$	3.0	2.23	2.57	83

Figure 2 (a) shows the J_c(n) and J_c(p) measured at 20 K. The dotted lines represent the J_c fitting data calculated though formulas (1), (2) and (3). These fitting data could be explained by using the two-fluid model for high frequency and low magnetic field limits [13]. The R_s is given as (1) in a low magnetic field and (2) in a high magnetic field [14]. Where λ_L is the London penetration depth, B is the dc magnetic field, Φ_0 is the flux quantum, and $\hat{\mu}$ is the dynamic mobility. The η is the viscous drag coefficient which is related to the motion of the magnetic flux.

$$R_s(B) = R_s(0\mathrm{T}) + \frac{B\Phi_0}{2\lambda_L \eta_{\mathrm{low}}(B,T,d)}, \quad \mathrm{B} << \frac{\omega\mu_0{\lambda_L}^2}{\Phi_0\hat{\mu}} \quad (2) \qquad R_s(B) = \sqrt{\frac{\omega\mu_0\Phi_0 B}{2\eta_{\mathrm{high}}(B,T,d)}}, \quad \mathrm{B} >> \frac{\omega\mu_0{\lambda_L}^2}{\Phi_0\hat{\mu}} \quad (3)$$

Non-doped typically showed J_c = 6.75 MA/cm^2 in zero magnetic field. The J_c of the YBCO thin films doped with BHO were comparable J_c of non-doped in zero magnetic field. However, the YBCO doped with BHO 1.5 wt% was 7.4 % greater than the non-doped YBCO. Improvement of the J_c and R_s by APCs under a zero magnetic field was reported by Matsumoto et al. and Mukaida et al [1-2]. In particular, the J_c(p) of BHO1.5 was 4.0 MA/cm^2 in 5 T, 41 % greater than that of the non-doped. It was possible to improve the J_c(p) by doping the BHO. The J_c(n) of BHO1.5 was 3.4 MA/cm^2, and was almost the same for the non-doped.

In addition, it is necessary to generate a magnetic field of B1 = 3.69×10^{-3} T for the 500 MHz NMR pick-up coil. It is assumed that for the application of NMR to a one-turn coil 1mm in width and 1 μm thick, 18.3 A of current will flow through the coil in order to product B1, J_c(p) 2 MA/cm^2 critical current density is required at 11.7 T. Thus, it was revealved from the fitting data that the J_c(p) at 10 T of BHO1.5 was 2.0 MA/cm^2, and 1.5wt% BHO doped YBCO is the most effective in the application of NMR . Figure 2.(b) shows the R_s(n) and R_s(p) calculated from a correlating equation of J_c. The dotted line is the fitting data of R_s under a weak magnetic field calculated by using equation (1) and under a strong magnetic field calculated by using equation (2). The R_s(p) and R_s(n) of BHO1.5 was two orders of magnitude lower than the R_s of copper, and is thus effective for NMR pick-up coils.

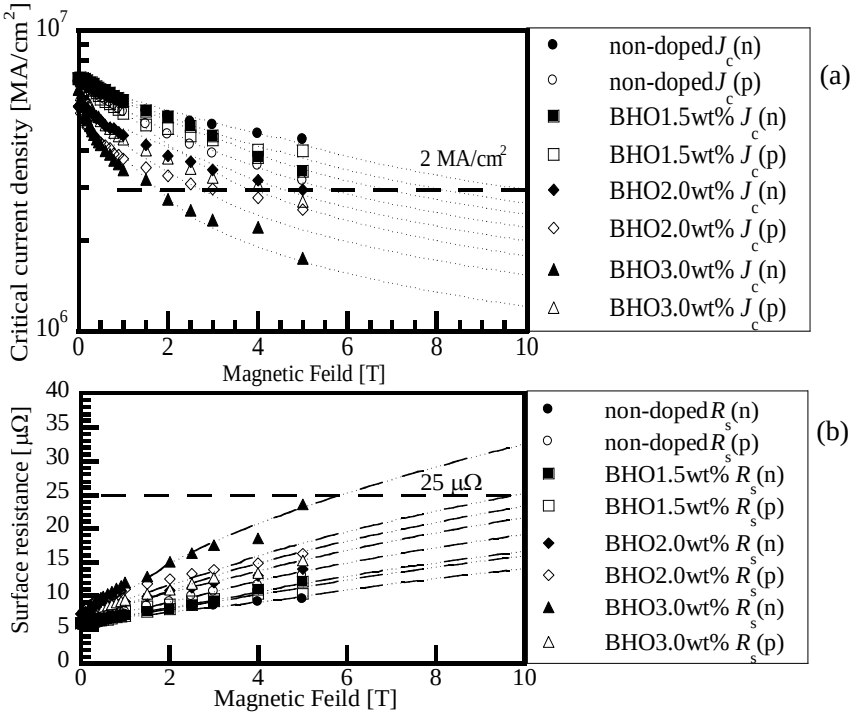

Figure 3. (a): J_c of the YBCO films as a function of magnetic field applied parallel and vartical to the c-axis of the films. (b): Magnetic field dependence of the R_s calculated from the magnetic field dependence of J_c.

4. Conclusion

We measured the J_c of 0, 1.5, 2.0 and 3.0 wt% BHO doped YBCO thin films. In addition, we have calculated the correlating equation of J_c. The J_c(p) of BHO1.5 was 7.4 % bigger than non-doped in a zero magnetic field. The J_c(p) of BHO1.5 in 5 T was 41 % bigger than non-doped. J_c(n) was also comparable to the non-doped. The R_s(n) and R_s(p) of BHO1.5 of 10 T were smaller by approximately two orders of magnitude than the R_s of copper. Therefore, we clarified the possibility that the most effective way to fabricate thin film for NMR coil application is by doping YBCO with BHO 1.5 wt%.

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