$BaHfO_3$ 완충층을 사용한 IBAD MgO 기판 위에 제조된 고임계전류밀도의 $GdBa_2Cu_3O_y$ 박막

High-J_c $GdBa_2Cu_3O_y$ films on $BaHfO_3$-buffered IBAD MgO template

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Abstract: The $BaHfO_3$ (BHO) buffer layer on the IBAD MgO template was turned to be effective for a successful fabrication of $GdBa_2Cu_3O_{7-\delta}$ (GdBCO) films with high critical current density (J_c). Both the BHO buffer layers and GdBCO films were prepared by pulsed laser deposition (PLD). The effects of the PLD conditions, including substrate temperature (T_s), oxygen partial pressure (PO_2), and deposition time on the *in-plane* texture, surface roughness, and microstructures of the BHO buffer layers on the IBAD MgO template were systematically studied for processing optimization. The *c*-axis oriented growth of BHO layers was insensitive to the deposition temperature and the film thickness, while the *in-plane* texture and surface roughness of those were improved with increasing T_s from 700 to 800°C. On the optimally processed BHO buffer layer, the highest J_c value (77 K, self-field) of 3.68 MA/cm^2 could be obtained from GdBCO film deposited at 780°C, representing that BHO is a strong candidate for the buffer layer on the IBAD MgO template.

Key Words: $BaHfO_3$, GdBCO, buffer layer, superconductor, IBAD, critical current density.

1. Introduction

The PLD process has been extensively employed for the preparation of $YBa_2Cu_3O_{7-\delta}$ (YBCO) coated conductors (CCs). Up to now, two different buffer materials of $LaMnO_3$ (LMO) and CeO_2 have been successfully employed for the fabrication of long-length CCs. Using LMO buffer layer on the IBAD MgO template, Superpower, Inc. has scaled YBCO tapes (Metal-Organic Chemical Vapor Deposition) up to 790 m length with a minimum performance of 150,100 A/m [1]. On the other hand, on the buffer architecture of CeO_2/IBAD-GZO templates, Fujikura Ltd. reported GdBCO tape (PLD) with end to end I_c of 304.8 A for a 368 m long tape, corresponding to $I_c \times L$ values of 112,166 A/m [2]. Recently, we have successfully fabricated a highly-textured long-length IBAD MgO template using continuous reel-to-reel process [3]. We also reported the optimum PLD processing conditions for the LMO buffer layer on highly-textured IBAD MgO template and *in-plane* texture correlation between the LMO layer and IBAD MgO template [4]. On the LMO-buffered IBAD MgO template, 27m-long $SmBa_2Cu_3O_y$ CC with the minimum I_c of 305 A/cm-width could be successfully fabricated by a batch-type reactive co-evaporation method [5].

Recently, however, $GdBa_2Cu_3O_{7-\delta}$ (GdBCO) CCs have drawn a great attention since they exhibited J_c-B properties superior to YBCO CCs [6]-[9]. Takahashi *et al.*[8] fabricated both GdBCO and YBCO CCs with various thicknesses on the CeO_2-buffered IBAD-$Gd_2Zr_2O_7$ (GZO) template by the PLD method. They reported that all GdBCO CCs exhibited less degradation of J_c in magnetic fields than YBCO CCs. In 2006, Ibi *et al.* [9] reported the thickness dependence of critical current, I_c and the ratio of *a*-axis oriented grains to *c*-axis oriented grains for both PLD-processed GdBCO and YBCO CCs. They reported that the higher I_c values of GdBCO films compared with those of YBCO films are attributable to the smaller amount of *a*-axis oriented grains. They also reported that GdBCO exhibits higher material yield and production rate than YBCO. Recently, Fuji *et al.* of Fujikura Ltd. [10] have reported the fabrication of a long high I_c GdBCO CC on the CeO_2/IBAD-GZO template by the PLD method; 368 m-long GdBCO tape with end to end I_c of 304.8 A at 77K, corresponding to $I_c \times L$ values of 112,166 A/m. More recently, S. Hanyu *et al.* of Fujikura Ltd. reported the fabrication of 300 m-long GdBCO CC with I_c of 180 A at 77K on CeO_2/IBAD MgO template [11].

While LMO[1], [12], [13] and CeO_2 [14], [15] are the most widely used buffer layers on the IBAD MgO template, there have been many reports for various other buffer layers such as $SrTiO_3$ [16], $SrRuO_3$ [17], Ba_2YNbO_6 [18], $Sm_xZr_{1-x}O_y$ [19] on the

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IBAD MgO template. However, further study to develop a new effective buffer layer is still required for the IBAD MgO template. In this study, we have selected $BaHfO_3$ as a potential buffer layer on the IBAD MgO template since it shows an excellent chemical compatibility with YBCO and also a very small lattice-mismatch with MgO (a=0.421 nm) [20], [21]. We here report the effects of PLD processing parameters, including T_s, PO_2, and deposition time on the crystallinity, texture, and surface roughness of BHO buffer layer. In addition, we report a successful fabrication of high-J_c GdBCO films on the BHO-buffered IBAD MgO template.

2. Experimental

The 2-inch diameter BHO targets were prepared by the solid-state reaction employing precursors of $BaCO_3$ and HfO_2 (all 99.9% purity), respectively. All the substrates were cut from long-length IBAD MgO template fabricated by continuous reel-to-reel process [3]. On these IBAD MgO templates with *in-plane* texture of ~6.5° and RMS surface roughness of ~5 nm in 5×5 μm^2 area, BHO thin films were deposited by PLD using KrF excimer laser (Lambda Physik, λ= 248 nm). The T_s and deposition time for BHO layer growth were varied from 700 to 800°C and 5 to 20 min, respectively. On the other hand, the 2-inch diameter GdBCO targets for PLD were prepared by the conventional solid-state reaction employing precursors of Gd_2O_3, $BaCO_3$, and CuO (all 99.9% purity). For the PLD conditions of BHO buffer layers and GdBCO films, the energy density, laser frequency, and target-to-substrate distance were fixed at 2 J/cm^2, 10 Hz, and 6 cm, respectively. GdBCO films were deposited at various T_s from 770 to 800°C for 25 min. The ambient oxygen pressure for the BHO buffer layer and GdBCO film was kept at 100 mTorr and 400 mTorr, respectively. As-deposited GdBCO films on the BHO buffer layer were post-annealed at 500°C for 30 min in a pure oxygen atmosphere for a full oxygenation. After the oxygen-annealing, all GdBCO films were silver-coated by DC magnetron sputtering and then annealed at 450°C for 1 h with the O_2 flowing rate of 0.5 l/min for the measurements of superconducting properties.

To determine the crystalline phases and *in-plane* textures (Full-width at half maximum, $\Delta\phi$) of the deposited films, θ - 2θ and $\Delta\phi$ scans were routinely examined by X-ray diffraction (XRD, Bruker D8 Discover). The surface roughness of the BHO buffer layer was analyzed in 5×5 μm^2 area by atomic force microscopy (AFM, Nanostation II). The surface morphologies of BHO layers were analyzed by field-emission scanning electron microscopy (FE-SEM, JEOL, JSM-6330F). The atomic element distributions in the GdBCO film were analyzed by an electron probe micro analyzer (EPMA, JXA-8900R). The critical temperature, T_c and the critical current, I_c values of GdBCO films were measured by the standard four-probe method using 1 μV/cm criterion.

3. Result and Discussion

BHO layers were deposited on IBAD MgO template at PO_2 of 100 mTorr. As shown in Fig. 1, only (00*l*) reflections without other orientations are observed from BHO films deposited at 700-800°C. The lattice parameters of BHO layers determined from XRD measurements were 4.18 Å, which is similar to JCPDS value (a=4.171 Å). As shown in Fig. 2, the surface morphologies of BHO layers deposited at 700-800°C are almost identical. The FWHM values for ϕ-scans of BHO (110) exhibited 6.46, 5.89, and 5.85° as T_s was increased from 700 to 750, finally to 800°C, respectively. Though not represented, AFM image analyses for BHO films grown at 700, 750, and 800°C revealed relatively high surface roughness, corresponding to RMS values of 6.9, 6.4, and 5.7 nm, respectively. Thus, from these results, it can be noted that the effect of the substrate temperature on the crystallinity and surface morphology of BHO buffer layer is insignificant. However, the *in-plane* texture and surface roughness of BHO buffer layer are dependent on the substrate temperature.

At the fixed T_s of 800°C and PO_2 of 100 mTorr, the deposition time was altered from 5 to 20 min to identify their effects on the BHO films. As shown in Fig. 3, only (001) reflections for BHO films are observable from the deposition time of 5 min (~30 nm thickness) to that of 20 min (~130 nm thickness). AFM image analyses for BHO films deposited for 5, 10, 15, and 20 min revealed RMS values of 3.8, 5.5, 6.1, and 6.2 nm, respectively, implying that the degree of surface roughness is increased with the increasing film thickness. Also, ϕ-scans of BHO (110) exhibited 6.01, 5.85, 6.02, and 6.09° as the film thickness is increased from 30, 50, 95, and finally to 130 nm, respectively.

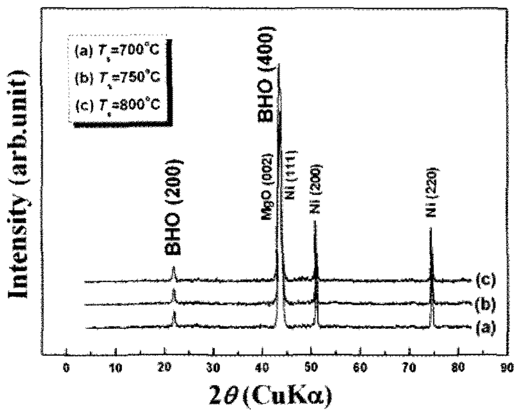

Fig. 1. X-ray diffraction patterns of BHO films deposited at (a) 700°C, (b) 750°C and (c) 800°C.

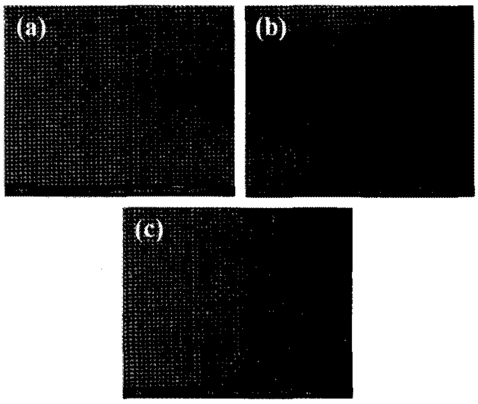

Fig. 2. The plan-view images for BHO layers deposited at (a) 700°C, (b) 750°C and (c) 800°C.

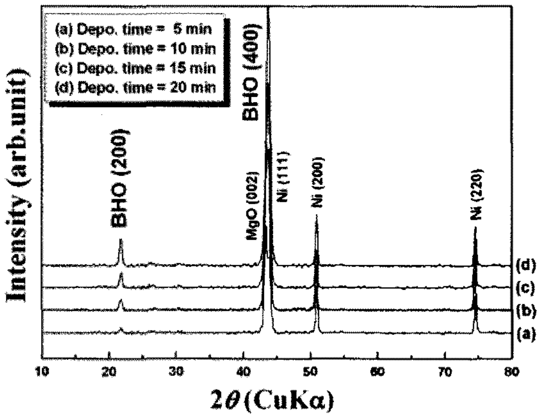

Fig. 3. X-ray diffraction patterns of BHO films deposited for 5, 10, 15 and 20 min.

To identify the effectiveness of BHO as a buffer layer, GdBCO films have been deposited on the BHO buffer layer which was deposited at the PO_2 of 100 mTorr and T_s of 800°C. Fig. 4 shows XRD θ-2θ scan of the GdBCO films on BHO-buffered IBAD MgO template deposited at T_s range of 770-800°C. The film thickness of GdBCO was ~280 nm. The major peaks in the pattern correspond to the (00l) reflections of GdBCO phase. Therefore, we can see that all GdBCO films are highly *c*-axis oriented. However, as shown in Fig. 4(a)-(c), GdBCO films deposited at T_s of 770-800°C show GdBCO (200) peak which indicates a small amount of *a*-axis oriented grains. As can be seen in Fig. 4(d), *a*-axis-free GdBCO film could be obtained at T_s of 800°C. Therefore, the substrate temperature of 800°C is required for obtaining only *c*-axis oriented GdBCO film on the BHO buffer layer. With increasing the deposition temperature from 770, 780 to 790°C, FWHM values of GdBCO (102) are slightly improved from 4.5, 4.37° to 4.2° and then degraded to 4.7° at 800°C. All GdBCO films showed four-fold symmetry (102) diffraction peaks without 45°-rotated peaks.

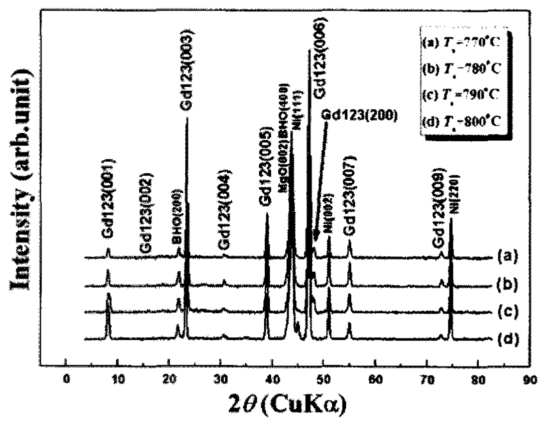

Fig. 4. X-ray diffraction patterns of GdBCO films deposited on BHO-buffered IBAD MgO template with deposition temperature of (a) 770, (b) 780, (c) 790 and (d) 800°C. The arrow indicates GdBCO (200) peak corresponding to the a-axis grains of GdBCO.

The surface morphologies of GdBCO films are shown in Fig. 5. In Fig. 5(a)-(c), *a/b*-axis oriented grains are undetectable even though small amounts of XRD peaks for *a*-axis oriented grains are observed. As shown in Fig. 5(a), GdBCO film deposited at 770°C represents that the grain connectivity of GdBCO is degraded because of grain boundaries between rectangular shaped grains. One can see that GdBCO film deposited at 780°C show a dense microstructure without a distinct grain boundary in the matrix (See Fig. 5(b)). In Fig. 5(c), a few out-grown particles and small sized pores are observed in GdBCO sample deposited at 790°C. As T_s increases up to 800°C, it is apparently observable that the number of outgrown particles is increased (See Fig. 5(d)). In order to identify this phase, we performed EPMA analysis for GdBCO sample deposited at T_s of 800°C. From the SEM image of Fig. 6(a), it is observable that white particles are slightly protruded from the surface of GdBCO film. The elemental mapping images for Hf, Gd, Ba, and Cu atoms are shown in Fig. 6(b)-(e). As can be seen in Fig. 6(b), the region of Hf element in mapping image coincides with that of the white particles shown in Fig. 6(a). In Fig. 6(d), the amount of Ba element is almost uniform on the

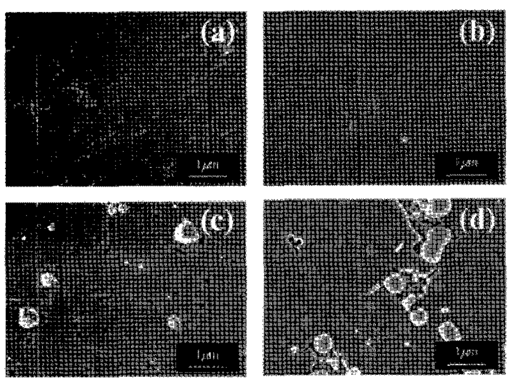

Fig. 5. Surface morphologies of GdBCO films deposited on BHO-buffered IBAD MgO template with deposition temperature of (a) 770, (b) 780, (c) 790 and (d) 800°C.

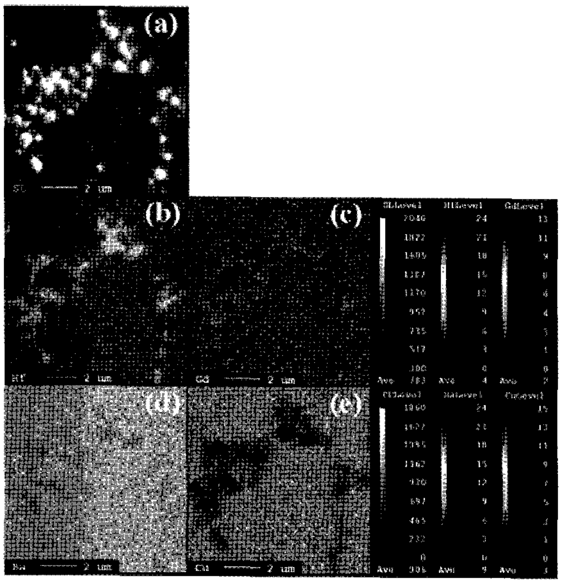

Fig. 6. Elemental mapping images ((a) SEM image, (b) Hf, (c) Gd, (d) Ba, and (e) Cu elements) of GdBCO film deposited on BHO-buffered IBAD MgO template with T_s of 800°C.

entire surface since Ba is a common element in GdBCO and BHO phases. However, in Fig. 6(c) and (e), one can see that Gd and Cu elements are deficient in the area corresponding to the white particles. Thus, BHO particles on the GdBCO film surface might be formed by ionic diffusion of Ba^{2+} and Hf^{4+} from the BHO buffer layer at T_s of 800°C.

The $T_{c,zero}$ values of GdBCO films grown at T_s of 770, 780, 790 and 800°C exhibited 90, 91.5, 91.6 and 88 K, respectively. I_c values for GdBCO films are shown in Fig. 7. As T_s increases from 770 to 780°C, the I_c values increase from 39 to 103 A/cm-width at 77K in self-field. However, the I_c value of the film grown at T_s of 790°C was abruptly reduced to 12 A/cm-width. From these I_c values of GdBCO films deposited at 770, 780 and 790°C, the estimated J_c values (77 K, self-field) were 1.4, 3.68, and 0.43 MA/cm^2, respectively. The lowest J_c value from GdBCO film deposited at 790°C can result from the existence of BHO particles diffused into GdBCO superconducting matrix. The I_c value of GdBCO film deposited at 800°C showed "0" which may be attributed to an increase in BHO particles diffused into GdBCO layer because they interrupt supercurrent-paths in GdBCO superconducting phase. Thus, the highest J_c value (77 K, self-field) of 3.68 MA/cm^2 could be obtained from GdBCO film deposited at 780°C. The significant difference of J_c values according to the deposition temperature indicates that the processing window for obtaining a high-quality GdBCO film using BHO buffer layer is quite narrow. Futher study should be performed to identify the origin for such an abrupt microstructural variation at 800°C. Nevertheless, the $T_{c,zero}$ and J_c values obtained from GdBCO films deposited on the BHO buffer layer demonstrate that BHO is very effective buffer layer on the IBAD MgO template.

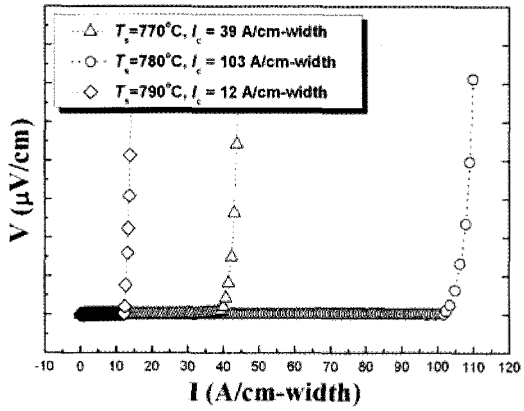

Fig. 7. I_c curves of GdBCO films deposited on BHO-buffered IBAD-MgO template with deposition temperature of 770, 780 and 790°C.

4. Summary

High-J_c GdBCO films on the $BaHfO_3$-buffered IBAD MgO template were successfully fabricated by optimizing PLD processing parameters, including oxygen partial pressure, target-to-substrate distance, and substrate temperature. This achievement was, in turn, enabled by optimizing the PLD processing condition of the $BaHfO_3$ buffer layers on the IBAD MgO template. The crystallinity of BHO layers was insensitive to the deposition temperature and the film thickness. However, the *in-plane* texture and surface roughness were improved with increasing T_s from 700 to 800°C. On optimally-processed BHO buffer layers, the highest J_c value (77 K, self-field) of 3.68 MA/cm^2 could be obtained from GdBCO film deposited at 780°C. Although BHO particles diffused into the GdBCO matrix at the deposition temperature over 790°C and thus the processing window for high-J_c GdBCO film on the BHO buffer layer was quite narrow, BHO must be a promising buffer layer on the IBAD-MgO template.

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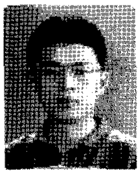
저 자 소 개



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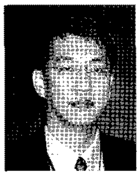

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