Enhancement of Critical Current Density of Yttrium Barium Copper Oxide Thin Films by Introducing Nano dimensional Cerium Oxide Defects

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Abstract

The critical current density, J_c has been the most important parameter used in the design and engineering of effective devices which is one of the implementation of high temperature superconductors (HTSC). In this work, an effort has been made to further improve the critical current density of YBa₂Cu₃O_{7-x} (YBCO) thin films by preventing the magnetic flux line lattice against the Lorentz force by pinning it in place with the aid of nano-dimensional defects. These defects were generated by distributing nano sized CeO₂ islands after YBCO layer was created on LaAlO₃ substrates perpendicular to the film using pulsed laser deposition (PLD) technique. Three samples with buffer layers of CeO₂ were prepared. CeO₂ with 50 pulses, 100 pulses and 150 pulses, after each 1000 pulses of YBCO were prepared five layers for each of the samples. The structural characterization of YBCO/CeO₂ and YBCO pristine films were carried out using x-ray diffraction (XRD) and scanning electron microscopy (SEM). Superconducting proprieties were measured using a vibrating sample magnetometer (VSM). J_c for the pure YBCO and the YBCO/CeO₂ films were calculated from magnetization (M) versus Field (H) loops using Bean's model. J_c for the 50 pulses of YBCO/CeO₂ films was found to be increased slightly by an order of magnitude of about 40% with respect to those of YBCO films without the nano dimensional defects.

Keywords: high temperature superconductivity, thin films, YBCO, CeO₂, critical current density.

1. Introduction.

YBa₂Cu₃O₇₋x (YBCO) film has attracted a lot of attention in electrical power applications due to its high critical transition temperature T_c (>90 K) and critical current density J_c (>1 MA cm⁻²) (Lei, Zhao, Xu, Wu, & Chen, 2011; Haugan, Barnes, Brunke, Manrtense & Murphy, 2003). The growth of YBa₂Cu₃O_{7-x} (YBCO) thin films is of great interest for superconducting applications because of its power transmission magnetic shielding, low phase noise oscillators, magnetic resonance imaging receiver coils (Chen et al., 2016; Matsuunotoet al., 2004; Rejith, Vidya &Thomas, 2015; Zhao, Ito & Goto, 2014). Fine precipitates, dislocations, grain boundaries and vacancies are many kinds of crystalline defects, being considered as pinning centers (Zhao, Ito & Goto, 2014; Kujur, Sahoo, Panda, Asokan & Behera, 2013). The pinning centers depend on size, shape and concentration to achieve effective defects. The critical current density is highly influenced by flux lattice motion due to thermal fluctuations and Lorentz force due to applied magnetic fields. To maintain the necessary levels of high J_c in high applied magnetic fields we need high spatial densities or naturally occurring growth defect to suppress the thermal fluctuations to stop the vortex mobility by pining them. Although nanodots of CeO₂ have been shown to induce additional flux pinning, this has always been due to strain in the YBCO lattice because of these inclusions. Such strain however, could have detrimental effect on the YBCO. Aside from the thickness dependence of J_c, there is also a limitation placed on the thickness at which such films could be grown (Uzun & Avci, 2014; Zhao, Iton & Goto, 2014; Sueyoshi, Kotaki, Fujiyoshi, Mitsugi, Ikegami & Ishikawa, 2013).

(Huang, Li, Wang, Qi, Sebastain, Haugan, &Wang, 2017), reported the enhanced flux pinning properties of YBCO thin films with various pinning landscapes a magnetic nanocomposite of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO))x (CeO₂)_{1-x} was incorporated into YBCO as either a cap layer or a buffer layer but the defect pinning and magnetic pinning are introduced in the systems giving the J_c at 77K to be around 4.66MA/cm² (Xu et al., 2012), reported the influence

of CeO₂-cap layer on the texture and critical current density of YBCO film, here the found that the texture and J_c of YBCO film were largely dependent on the texture of CeO₂-cap layers under optimized deposition conditions, with the increase of the degree of in-plane and out-plane texture of CeO₂-cap layers, there was a decrease in the Jc of the YBCO thin films from 4.23MA/cm² to 0.47MA/cm². (Lee, Kim, Park & Park, 1996), agreed that addition of CeO₂ into YBCO will increase the critical current density to an extent at 77K, they reported $2x 10^4$ A/cm² an increased by lowering the measuring temperature. (Solovyov, Bagarinao, Li, Si, Win, Zhou, Wiesmann & Qing, 2010), used active (001) Ceria (CeO₂) buffer to modify the structure of the epitaxial high temperature superconductor YBCO which resulted with 0.8μ m thick film exhibiting strong enhancement of the critical current density of 4.2MA/cm². (Feng et al., 2015), used CeO₂ cap layers for high temperature superconducting, were YBCO films were epitaxially deposited on the as-growth CeO₂/ YSZ (001) stack using water- free metal organic deposition (MOD) method to obtain 1.92MA/cm² as J_c at 77K at 0.5Pa deposition pressure.

Pulsed laser deposition (PLD) which is one of the physical deposition methods has been a promising technique for thin film growth and for fabricating nano-dimensional/nano-dots within epitaxial, textured or polycrystalline thin film matrices in recent years (Bhaumik et al., 2017; Haque, Pant and Narayan, 2018; Haque and Narayan, 2018; Karnati, Haque, Taufique and Ghosh, 2018). In the present study, we have generated the nanostructures on LaAlO₃ substrates by depositing YBCO and YBCO/CeO₂ multilayer thin films. To investigate the effects of introducing nano dimensional CeO₂ defects on (current density) Jc and the micro structure of YBCO.

2. Experimental Procedure

Several alternating layers of YBCO/CeO₂ thin films were grown on single crystal LaAlO₃ substrates using pulsed laser deposition technique. YBCO target of 99.9% with density of 5g/cm³ and 99.9% of CeO₂ target, both were ordered from MTI corporation were used for this experiment. The substrate which is LaAlO₃ with orientation (100) and size of (10×10) and (5×5) mm were used for the deposition, cleaned with acetone, methanol, and ethanol for 10 mins each, under ultrasonic to remove dirt and oil, after that the substrate was dropped inside hydrogen fluoride to also remove oil particle if there is any left and then dry. The deposition chamber was cleaned with acetone and methanol to remove dirt particle inside it. We then mounted the cleaned substrate on the holder with silver paste. Ablation was performed using KrF laser source of a wavelength 248nm. The ablation was done at (300) mJ of laser energy with a repetition of 5Hz. The aperture size was 12 x 4 mm² and laser spot size was 1 x 4 mm^2 . The target-substrate distance was maintained at 4cm. A based pressure of 10^{-7} Torr was maintained before the deposition; the deposited films were cooled down naturally in 600 mTorr of oxygen gas. To determine the optimum oxygen pressure for YBCO depositions, experiments were carried out at oxygen pressures of 400 mTorr. For this study three samples were chosen based on their yield and structural quality. They are identified as 50 Pulses, 100 Pulses and 150 Pulses samples based on the number of laser pulses used for a layer of CeO₂. The films were deposited on LaAlO₃ single crystal substrates with (001) direction. Starting with YBCO followed by CeO₂, 10 layers of films were deposited for each sample tabulated below.

LAYER	Sample I: 50 Pulses	Sample 2: 100 Pulses	Sample 3: 150 Pulses
YBCO	1000 Pulses	1000 Pulses	1000 Pulses
CeO ₂	50 Pulses	100 Pulses	150 Pulses
YBCO	1000 Pulses	1000 Pulses	1000 Pulses
CeO ₂	50 Pulses	100 Pulses	150 Pulses
YBCO	1000 Pulses	1000 Pulses	1000 Pulses
CeO ₂	50 Pulses	100 Pulses	150 Pulses
YBCO	1000 Pulses	1000 Pulses	1000 Pulses
CeO ₂	50 Pulses	100 Pulses	150 Pulses
YBCO	1000 Pulses	1000 Pulses	1000 Pulses
CeO ₂	50 Pulses	100 Pulses	150 Pulses

Table 1. The multilayer deposition showing all the samples

2.1 Multilayer Deposition of YBCO/CeO₂

The 50 Pulses sample contains five layers of YBCO and 5 layers of CeO_2 . The difference between the three samples is differences in the thickness of the CeO_2 layers. Single and multilayer samples were prepared using pulsed laser deposition. Also, a fourth sample of pure YBCO with 5000 pulses was made to be used for comparison of vital data such as the superconducting transition temperature and critical current density. The

experimental conditions for deposition of pure YBCO films are: Deposition temperature of 800° C, 825° C, 850° C, after several depositions we settled at 850° C for our optimum deposition temperature, an oxygen pressure of 400 mTorr, laser energy of 300 mJ, and laser pulse repetition rate of 5Hz. The pure YBCO films were cooled under an oxygen pressure of 600mTorr. The deposition parameters for CeO₂ nano dimensional were same for the pure YBCO in terms of the deposition temperature.

X-ray diffraction (XRD) and Scanning Electron Microscopy (SEM) were carried out to investigate the film microstructures and to calculate/measure oxygen deficiency in the samples. The superconducting properties of all the samples were measured using physical property measurement system(PPMS), it has an option called vibrating sample magnetometer (VSM) where the sample was attached to and then inserted inside the PPMS. Magnetization (M) Vs Temperature (T) in low field of 5 Oe were carried out to determine the critical temperature for each of the samples. (Haywoods, Oh, Kebede, Pai, Sankar, Christen, Pennyccok & Kumar, 2008). The magnetic field dependence of the critical current density J_c (H) was calculated using bean model from the magnetization versus field of the hysteresis loop (M-H) for several fixed temperatures.



Figure 1. Schematic diagram of multilayer deposition of YBCO/CeO₂. 1000 pulses of YBCO was first deposited on the first layer of the substrate followed by CeO₂ which was varied (50,100,150) Pulses. These layers were done five times ending it with CeO₂

3. Results and Discussion

X-ray diffractometer was performed on all the films to determine film crystallinity and specially to analyze the non-superconducting impurity diffraction peaks. A high quality epitaxial YBCO film has been observed.



Figure 2. XRD pattern of Pure YBCO and YBCO/CeO₂ where 0P is for the pure YBCO and 50P,100P and 150P are for the YBCO/CeO₂

The (001) peaks in the XRD-spectra indicate that YBCO thin films are highly c-axis oriented, which is commonly observed and corresponds to the natural growth of the material. The XRD pattern from the YBCO and YBCO/CeO₂ deposited on LaAlO₃ at 850°C and 300mTorr of oxygen pressure, is shown in figure 2. From the graph above we can observe that (001) peaks are (001) at 7.56°, (002) at 15.18°, (003) at 23.7°, (004) at 31.2°, (005) at 38.5°, (006) at 47°. The peaks of the substrate were also observed with corresponding degrees at 23.5° and 48.3°. All the samples of YBCO and YBCO/CeO₂ was highly c-axis oriented with good sharp and strong peaks. The CeO₂ appears in the graph below as an independent phase. The peak of CeO₂ are observed only at 32.8° (200) orientation from the graph we can observe that the intensity increases as the number of pulses are added. The XRD pattern for the 50P YBCO/CeO₂ was not resolvable, probably due to the small amount of the CeO₂ size and low density. The presence of (001) confirmed that the deposition parameters used for this work were good and suitable for the growth of YBCO on LAO substrates. No second phase such as Y_2Cu_2O and CuO were found in this case. Only YBCO and CeO₂ phase was formed with a high intensity. From the XRD the oxygen deficiency was calculated for the samples using equation 1 (Coll, Gazque, Huhne, Holzapfel, Morilla, Lopez, Pomar, Sandimenge, Puig & Obradors, 2009), and the results are shown on Table 2.

Oxygen deficiency calculated from c-axis

$$c = 12.736 - 0.1501(7 - x)$$

3

Or

$$x = \left(\frac{c - 11.6853}{0.1501}\right) \tag{1}$$

Table 2. Oxygen deficiency calculated from c-axis

Sample Name	c- axis pattern (°)	Oxygen deficiency(°)
Sample I (800°C)	11.7056	0.148
Sample II (825°C)	11.7053	0.133
Sample III (850°C)	11.7003	0.099
Sample IV (50Pulses)	11.6978	0.083
Sample V (100Pulses)	11.6958	0.063
Sample VI (150Pulses)	11.6901	0.031

The scanning electron microscopy (SEM) was performed on the samples, it revealed the surface morphologies of thin films. Figure 3 (a and b) shows the SEM image of pure YBCO and 50 pulses of YBCO/CeO₂. Figure 3 (c and d) shows the SEM of the YBCO/CeO₂ samples for (100 and 150) pulses respectively with nano dimensional defects, the pure YBCO reveals that they YBCO are well packed with long and extended grains randomly oriented in all direction with sizes vary from $1-5\mu$ m.

$$D = \frac{0.9\lambda}{\beta \cos\theta}$$
(2)

Where,

D = Grain size,

 λ = Wavelength of X-ray used,

 β =Full width at half maxima of the peak (FWHM) in radians,

 $\theta =$ Bragg's angle

Table3.	Calculated	Crystalline	size	of CeO ₂
				/

Sample name	Peak Width	2theta	Theta	Wavelength	Grain Size
50 pulses	NIL	NIL	NIL	1.5406nm	NIL
100 pulses	0.881°	33.29°	16.64°	1.5406nm	1.64nm
150 pulses	0.587°	33.12°	16.56°	1.5406nm	2.46nm



Figure 3. SEM images showing the microsctructrue of the samples, (A) Pure YBCO, (B) 50pulses, (C)100pulses, (D) 150pulses of YBCO/CeO₂

To measure and confirm the critical temperature (T_c) of HTSC samples, zero field cooled magnetization measurement was performed using the Vibrating Sample Magnetometer (VSM) option on the PPMS. For these measurements, the sizes of the samples both the pure YBCO and the multilayer YBCO and CeO_2 were reduced to (2 x 2) mm² by cutting it with a diamond cutter. the samples were mounted in such a way that the magnetic field was parallel to the c-axis, that is, the magnetic field was perpendicular to the film surface. After mounting the sample on the holder, the PPMS system was cooled down to 10K. As can be seen in figure 4 the magnetization versus temperature measurement was carried out for all the samples. The results of the measurements are summarized in Table 3 below. The transition to the superconducting phase was sharp for more samples, and the T_c in the range of 89 K to 92 K was obtained and this agrees with some of the literature (Kumar, Apte, Pinto, Sharon & Gupta, 1994; Haruta et al., 2013; Bobylev, Gerasimov & Zyuzeva, 2015; Pahike et al., 2016; Liu, Song, Wang & Zhang, 2016; Zhai et al., 2015). It was also observed the that deposition temperature of 800°C has given a larger volume fraction of superconductivity than the other two deposition temperatures and it is about twice the fraction than for the 850°C. In this case the factors influencing the volume fraction may include inconsistency in controlling the growth parameter. This experiment shows the effect of different pulses of CeO_2 on the transition temperature and the superconducting volume fraction. As can be seen the superconducting volume fraction is highest for the 150 Pulses compared with the 100 Pulses and 50 Pulses. On the other hand, the 50 Pulses sample gave the highest T_c. This is because the number of CeO₂ was small compared to others.



Figure 4. Magnetization Versus Transition Temperature (K) of all the sample, pure YBCO at different deposition temperature and (50, 100.150) pulses of YBCO/CeO₂ at the same deposition temperature

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Sample Name	Deposition Temperature °C	Oxygen pressure (m Torr)	Laser Energy(mJ)	T _c (K)
Sample I (YBCO)	800	400	300	87.2
Sample II (YBCO)	825	400	300	89.0
Sample III (YBCO)	850	400	300	91.1
Sample IV YBCO/CeO ₂ (50Pulses)	850	400	300	92.0
Sample V YBCO/CeO ₂ (100Pulses)	850	400	300	91.0
Sample VI YBCO/CeO ₂ (150Pulses)	850	400	300	87

Figure 5 shows T_c as a function of number of pulses for the onset transition temperature. From the plot, we can deduce that the T_c was high for Pure YBCO (0 pulses) and after the introduction of 50 pulses of CeO₂ the T_c increased slightly. Similar changes were observed at 100 pulses and 150 pulses the T_c decreases as the number of pulses were added. This is a clear indication that the CeO₂ defects were randomly introduced into superconducting matrix. Figure 6 shows that the obtained magnetic moment is in the emu units, which was converted to magnetization (emu/cm³) by dividing the magnetic moment by the volume of the sample. The area of the thin film sample was (2 x 2) mm². The thickness of the film was maintained at 100nm for pure YBCO.

The magnetic moment was obtained for the field swept from desired positive field to zero field and zero field to desired positive field. The value of the current density depends on several parameters including deposition temperature (T_D), the number of pulses (P), external magnetic field (H) and the temperature at which it is measured (T). i.e $J_c = (T_D, P, H, T)$. (T_D and P) are sample parameters that can be controlled during the sample growth. Keeping these two parameters fixed, J_c depends on H and T. The most determining sample condition for the value of J_c are the flux pinning centers introduced by the CeO₂ nano dots and randomly distributed in the superconducting matrix, the pinned flux experiences forces because thermal fluctuations and the Lorentz force that lead to flux lattice melting. Using Bean's formula, critical current density (J_c), was calculated for pure YBCO and multilayer CeO₂ nano dimensional defect. Bean's formula is given as (Matsuunoto, Horide, Osamura, Mukaida, Yoshida, Ichinose & Horii, 2004; Rejith, Vidya & Thomas, 2015; Zhao, Ito & Goto, 2014)

$$J_{c} = \frac{30 \,\Delta M}{a} \tag{3}$$

where ΔM is the difference of magnetization at a field, in emu/cm³, a is the area of the inscribed circle, in cm and J_c is critical density in A/cm².



Figure 5. Transition Temperature Versus Number of pulses of all the samples, Pure YBCO and (50,100,150) Pulses of YBCO/CeO₂



Figure 6. Magnetization Vs Magnetic field (hysteresis loop at 77K of 50 pulses)



Figure 7. Current Density Versus Magnetic Field H(T) for Pure YBCO and (50,100,150) Pulses of YBCO/CeO2

Figure 8 shows the plots of the current density J_c against the field in Tesla, at 77K it explains that the 50 pulses of the YBCO/CeO₂ in the multilayer gave highest J_c which was about 4.1 MA/cm² this agrees with other report (Huang, Li, Wang, Qi, Sebastain, Haugan &Wang, 2017; Xu, Liu, Wang, Zhu, Zhu & Li, 2012; Lee, Kim, Park & Park, 1996; Feng, Zhang, Qu, Huang, Xiao, Zhu, Lu, Shi, &. Han, 2015) against the pure YBCO and we observed that the more CeO₂ was added in the multilayer the less the J_c , and T_c we got, and this did not affect its structural properties. Table 5 below shows some current density of other literature which agree with our result. (Zhu, Wei, Yan, Tei, Jing, Liang, Bin, He &Wei, 2016). Figure 8 shows critical current density J_c as a function of measured temperature of all the samples.



Figure 8. Current density Versus Measured Temperature (K) of all the samples at 10K, 50 K and 77K

Table 5. Comparing the current density with other literatures

Literatures	Current Density at 77K
Huang et al. (2017)	4.66MA/cm ²
Xu et al. (2012)	4.23MA/cm ²
Lee et al. (1996)	$2x10^4$ A/cm ²
Solovyov et al. (2010)	4.2MA/cm ²

4. Conclusion

In conclusion, we conclude that enhancing flux pinning in superconductors is central to improving their current carrying capability (J_c) and their ability to be used in magnetic fields but in this study only on 50 pulses that the J_c increases slightly. Unlike other superconducting parameters, such as critical Temperature (T_c) and critical magnetic field. J_c is not intrinsically limited to the material of system. In this work, the micro structure and critical current density o YBa₂Cu₃O_{7- δ} films have been found to improve very slightly with the introduction of nano dimensional multilayer CeO₂. Evidence from magnetic properties and XRD, shows that no second phase such as Y₂Cu₂O and CuO were found. Only YBCO and CeO₂ phase were formed with a high intensity, while SEM shows that pure YBCO has long and extended grain randomly oriented in all direction with size vary from 1-5 μ m with the grain very closely packed to each other. TEM analysis and X-ray diffraction texture analysis of CeO₂ layers will be incorporated in the future work.

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