

**MICROSTRUCTURE AND CRITICAL CURRENT
DENSITY OF $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ + BaSnO_3 THICK
FILMS GROWN WITH PRE-MIXED PULSED
LASER ABLATION TARGET**

J. Burke^{1,3}, C.V. Varanasi^{1,3}, L. Brunke^{1,3}, H. Wang², J.H. Lee²,
P.N. Barnes³

¹ University of Dayton Research Institute Dayton
Dayton, OH, 45469-0170, USA

² Texas A&M
College Station, TX, 77843-3128, USA

³ Air Force Research Laboratory
Wright-Patterson AFB, OH, 45433, USA

ABSTRACT

$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) + BaSnO_3 (BSO) thin films with BSO nanocolumns have been shown to have improved critical current density (J_C) in applied magnetic fields. Previously, a sector target was used to grow thick ($> 2.5 \mu\text{m}$) YBCO + BSO films. In the present study, a premixed YBCO + BSO (20 mol %) target was used to grow thick films ($> 3 \mu\text{m}$) to determine if similar high quality thick films can be obtained as with the sector target approach. In the case of the premixed target, BSO material is continuously supplied as opposed to the sector target method. YBCO + BSO thick film samples processed using a premixed target were also found to have high J_C at high fields with $J_C > 10^4 \text{ A/cm}^2$ at 8 T at 77 K, whereas typical YBCO films carry only 10^2 A/cm^2 . Transmission electron microscopy (TEM) on these films indicated that BSO nanocolumns with a diameter of $\sim 8\text{-}11 \text{ nm}$ extend through the thickness of the films. The critical transition temperature (T_C) for the films was found to be $\sim 87 \text{ K}$, regardless of thickness.

KEYWORDS: high-temperature superconductors, BaSnO_3 , critical current density, superconducting transition temperature, $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

INTRODUCTION

YBa₂Cu₃O_{7-x} (YBCO) coated conductors have excellent self-field critical current density (J_C) but need improvement in applied magnetic fields like in motors, generators, and other electrical power applications [1,2]. To minimize this drop, flux pinning centers are introduced to the YBCO. Pinning in YBCO films can be provided via crystalline defects or other non-superconducting materials in nanodimensional additions. Examples of such material inclusions are Y₂BaCuO₅, BaZrO₃ (BZO), Y₂O₃, etc when dispersed in a controlled manner in a sufficiently high density, to provide this enhancement [3-6]. In the case of BaSnO₃ (BSO), it has been shown to create nanocolumns in YBCO when proper processing conditions were used [7], similar to BZO. These BSO nanocolumns particularly contribute to pinning in the c-axis orientation, with J_C increasing by more than 2 orders of magnitude in high fields.

In the literature, undoped YBCO also shows rapid degradation in J_C as the thickness of the film is increased (>1 μm) [8,9]. In a previous study, thick YBCO + BSO films were grown with a sectored target approach to address this issue [10]. By only varying the time of the deposition, thick (3 μm) YBCO + BSO films were grown and found to maintain J_C in applied magnetic fields due to the presence of the nanocolumns. These nanocolumns were uniformly straight and extended throughout the thickness of the films, for thicknesses ranging from 300 nm to 3 μm . The study presented here was performed to investigate if thick YBCO + BSO films with a pre-mixed target in place of a sectored target can achieve similar results. This is not clear since the supply of BSO is fairly continuous during film growth in the pre-mixed target case as opposed to an intermittent supply of BSO in the case of a YBCO/BSO sectored target.

EXPERIMENTAL

All the films were made by pulsed laser deposition (PLD). The PLD target used in this experiment was made using NEXANS YBCO powder and Cerac BSO powder (-325 mesh, 99%). The powders were mixed and ground together in the appropriate ratio to yield a final composition of YBCO + 20 mol% BSO. This ratio was used for its similarity to the BSO content of the previous experiment that used a YBCO + BSO sectored target. The mixed powder was pressed and then sintered for 72 hours at 850 °C and 168 hours at 920 °C to an approximate density of ~90%.

The film depositions were carried out in a Neocera chamber with a target to substrate working distance of ~6 cm. A Lambda Physik excimer laser ($\lambda=248\text{nm}$) was used to ablate the target. The laser was operated at 4 Hz, 625 mJ with an energy density on the target of ~2-4 J/cm². The films were deposited on single crystal (100) LaAlO₃ substrates in a 300 mTorr O₂ atmosphere at 780 °C. Samples were oxygenated during cool down at 500 °C, 600 torr O₂ for ½ hour, and then cooled to room temperature. The thickness of the films was varied by changing only the deposition time. Final film thicknesses ranging from 250 nm to 3 μm were obtained by varying the deposition times from 20 minutes up to 4 hours.

Magnetization hysteresis loops were taken in a Quantum Design PPMS Vibrating Sample Magnetometer (VSM). The Bean model was used for calculation of magnetization J_C from the data taken from these loops. The magnetic dipole moment of the sample was also measured as temperature was varied in the VSM to obtain the critical transition temperature (T_C) of the samples. The film thickness was measured with a KLA Tencor profiler. X-ray diffraction data was obtained on the samples using a Rigaku DMAX B. A FEI Sirion high resolution scanning electron microscope (SEM) was used to obtain electron micrographs of the samples' microstructure. A FEI Tecnai F20 analytical electron microscope with a point-to-point

resolution of 0.21 nm (TEM) was used after standard sample preparation to observe the nanocolumns of BSO in the YBCO matrix.

RESULTS AND DISCUSSION

The T_C data of YBCO + BSO samples of different thickness are shown in FIGURE 1. All the films for this study showed a T_C of ~ 87 K. It can be seen that the T_C does not degrade as the film thickness is increased, even up to 3 μm . This offers initial confirmation that 30 min of oxygenation is sufficient for even the thick films. A slight reduction of T_C to 87 K is consistent with the thin films of YBCO + BSO as reported earlier [11].

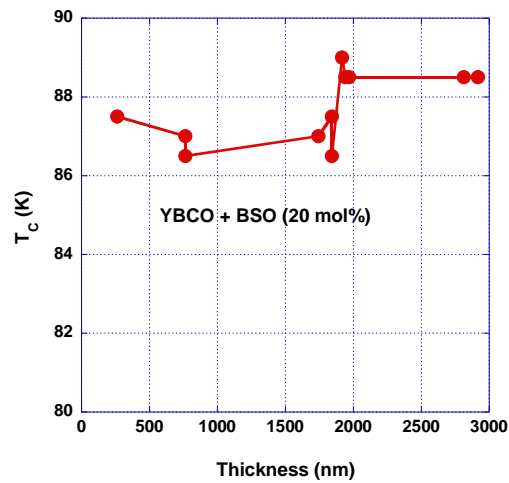


FIGURE 1. T_C in YBCO + 20 mol% BSO films with varying thickness.

A plot of the J_C of different thickness samples is shown as a function of applied magnetic fields in FIGURE 2. The results show that the samples continue to maintain their J_C very well even when the thickness is increased, especially at higher fields. At 8 Tesla of

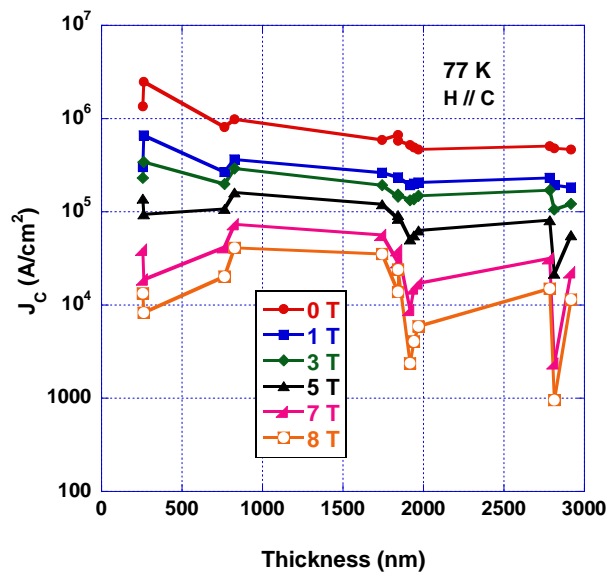
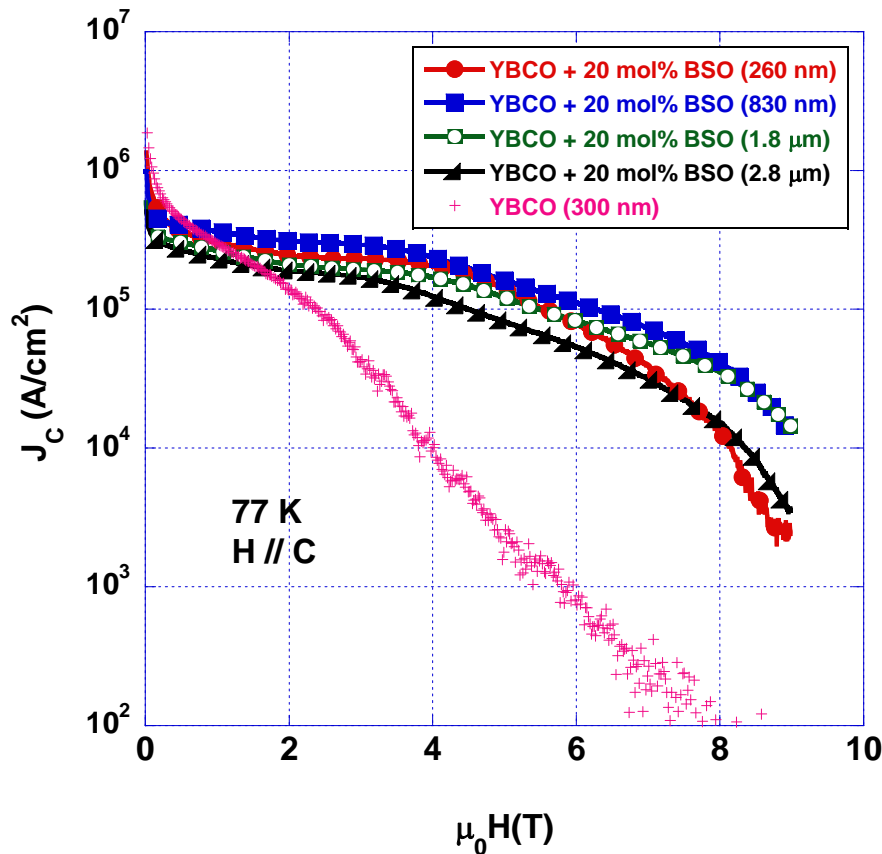


FIGURE 2. J_C values for varying thickness YBCO + 20 mol% BSO films at 77 K and $H // C$.

applied magnetic field, the J_C of a YBCO + BSO sample of 2.8 μm thickness was $1.5 \times 10^4 \text{ A/cm}^2$, which is similar to the J_C of a 260 nm thick YBCO + BSO sample. Although there is some reduction in J_C evident at very low fields, there appears to be a slight improvement at the higher fields as the thickness approaches the 1 μm range.

FIGURE 3 displays several magnetization J_C curves of thick YBCO + BSO samples. These are compared with a YBCO 300 nm film standard. After 1.8 Tesla, all YBCO + BSO films show improvement from YBCO. At 8 T it can be seen that the YBCO + BSO films are at least 2 orders of magnitude better than the undoped YBCO.

Theta - two theta x-ray diffraction data taken from different samples is shown in FIGURE 4. The area of interest has been expanded and compared for 4 representative thickness samples. All the samples show good texture for (001) type YBCO peaks. However, an additional peak appears in the thicker samples which corresponds to YBCO (103) peak. This peak becomes more intense in the 2.8 μm thickness sample. These misoriented grains are expected as the temperature at the growing surface could be lower in thicker films than thin films as the films are heated from the back of the substrate. These misoriented grains could be the primary cause for the slight drop in self-field J_C as observed in films with thickness $> 2 \mu\text{m}$.

**FIGURE 3.** VSM J_C data for selected YBCO + 20 mol% BSO film thicknesses compared with 300 nm YBCO at 77 K and $H // c$.

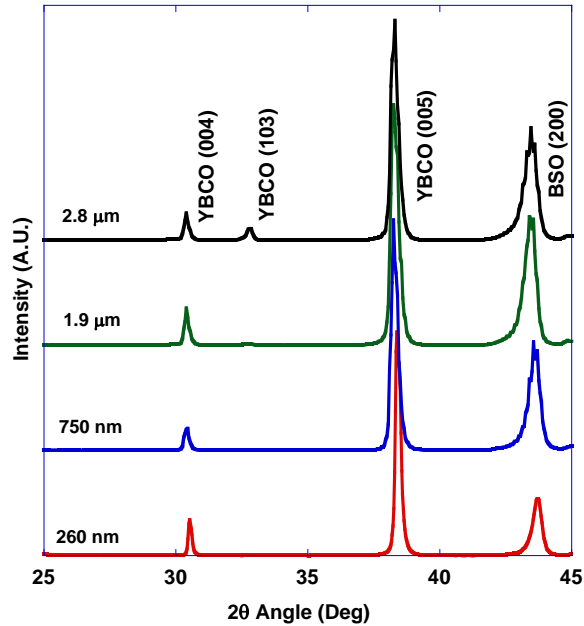


FIGURE 4. θ - 2θ x-ray scans of YBCO + 20 mol% BSO of varying thickness.

FIGURE 5 shows the low magnification (1.5k x) scanning electron surface micrographs of a) 260 nm and b) 2.8 μm thick YBCO+BSO samples. The 260 nm shows standard surface features for YBCO + BSO with small particles (note that these particles are not the planar view of the nanocolumns) on the surface that can be observed at low magnification. The 2.8 μm sample, however, shows misoriented grains such as a-axis grain growth occurring in the sample. Despite these misoriented grain growth (possibly created by the lower surface temperature as discussed before) in thick films, the YBCO + BSO microstructure still shows nanocolumn formation of BSO continuing into the upperparts of the films. FIGURE 6 shows at higher magnification (150k x) the a) 260 nm sample and b) 2.8 μm sample. Both display a uniform distribution of BSO nanoparticles at the surface. These are the cross-sections (a-b planar view) of the BSO nanocolumns as observed in cross-sectional transmission electron micrographs shown later.

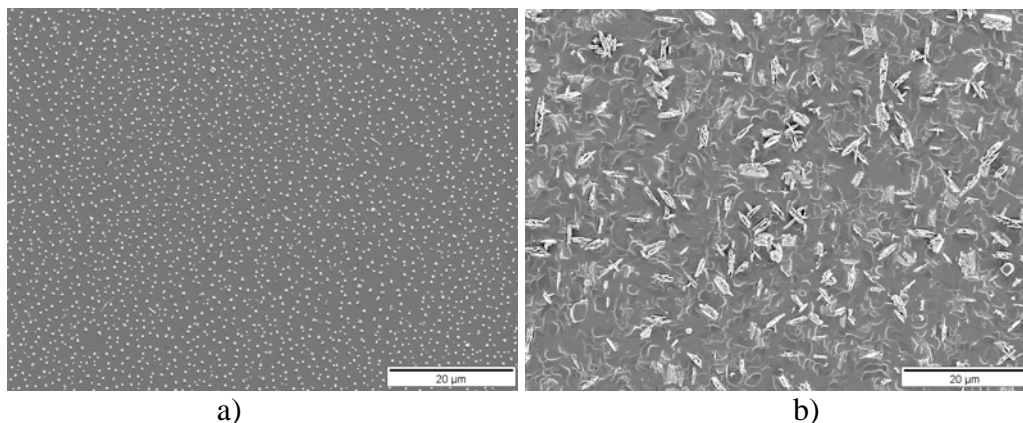


FIGURE 5. SEM photomicrograph showing surface microstructure of a) 260 nm sample and a b) 2.8 μm sample

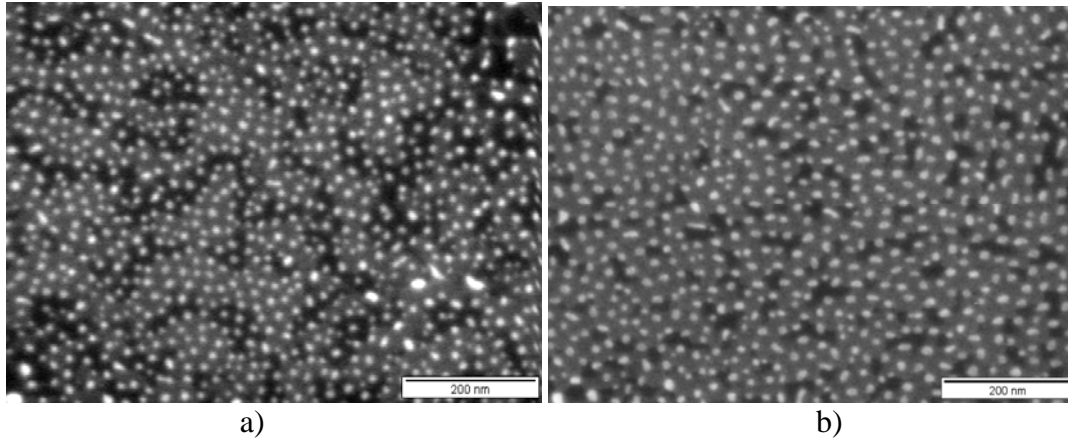


FIGURE 6. SEM photomicrograph showing uniform BSO distribution in YBCO on a) 260 nm sample and a b) 2.8 μm sample

Cross-sectional TEM was done on the samples to investigate the extent of the BSO nanocolumnar formation in the samples as well. FIGURE 7 shows a YBCO + BSO film of a) 750 nm thickness and b) 2.8 μm thickness. Both samples show nanocolumns with a diameter of ~ 8-11 nm. Both also show continuous nanocolumns that extend through the thickness of the film. They appear to be relatively unaffected by the appearance of a-axis growth. The nanocolumns were similar to the nanocolumns shown in the YBCO + BSO thick film made with a sectored target.

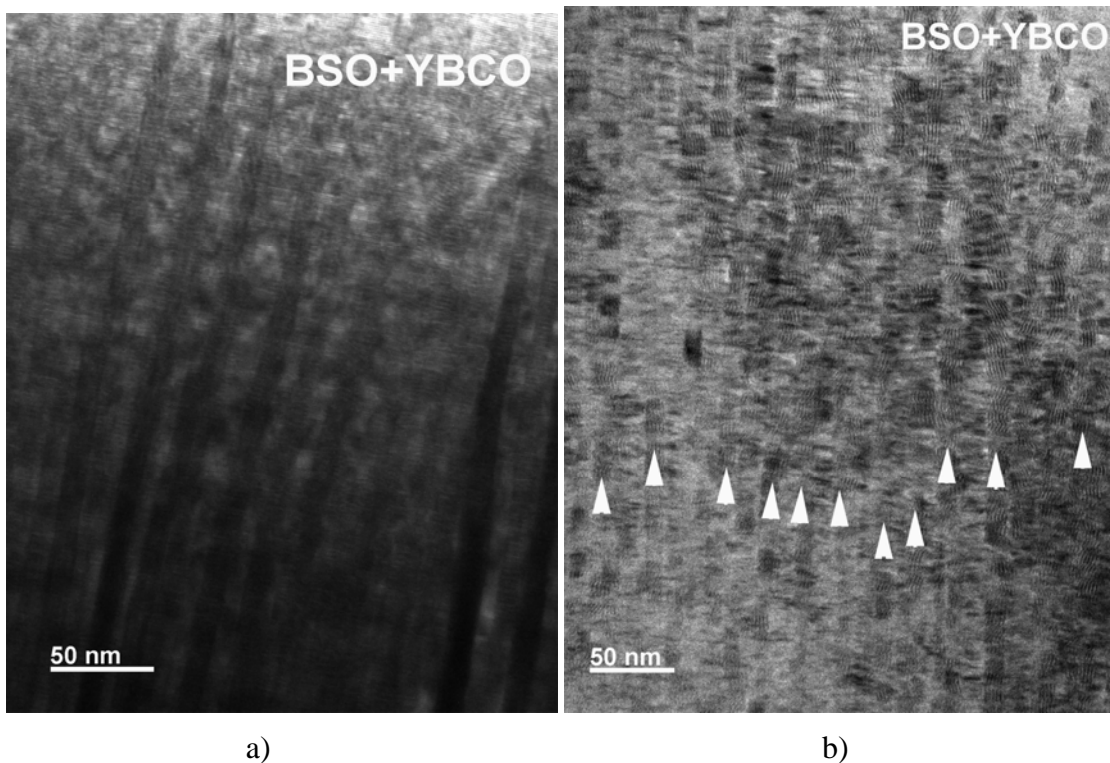


FIGURE 7. Cross sectional TEM image showing through-thickness, uniform BSO nanocolumns in a) 780nm film and a b) 2.8 μm film

CONCLUSIONS

It has been shown that quality, thick YBCO + 20 mol% BSO films up to 3 μm can be grown by just varying the deposition time. This quality growth occurs whether BSO is supplied from a continuous source like a pre-mixed target or intermittent source as the sectored target. The YBCO + BSO thick films show a T_C of $\sim 87\text{K}$ regardless of the thickness. The samples show little degradation of J_C as thickness is increased, with samples ranging from 260 nm to 2.8 μm showing $>10^4 \text{ A/cm}^2$ at 8 T, H//C. The samples also all show continuous 8-11 nm nanocolumns of BSO extending throughout the sample despite the presence of a-axis growth in the thicker samples.

ACKNOWLEDGEMENTS

Funding Support was provided by the Air Force Office of Scientific Research (AFOSR) and the Propulsion Directorate of the Air Force Research Laboratory (AFRL). The authors would like to thank John Murphy for technical assistance.

REFERENCES

1. Larbalestier, D., Gurevich A., Feldman, D.M., and Polyanski, A., *Nature* **414**, pp 368-377 (2001).
2. Barnes, P.N., Sumption, M.D., and Rhoads, G.L., *Cryogenics* **45**, pp 670-686 (2005).
3. Haugan, T.J., Barnes, P.N., Wheeler, R., Meisenkothen, F., and Sumption M., *Nature* **430**, pp 867-870 (2004).
4. MacManus-Driscoll, J.L., Foltyn, S.R., Jia, Q.X., Wang, H., Serquis, A., Civale, L., Maiorov, B., Hawley, M.E., Maley, M.P., and Peterson, D.E., *Nat. Mater.* **3**, pp 439-443 (2004).
5. Kang, S., Goyal, A., Li, J., Gapud, A.A., Martin, P.M., Heatherly, L., Thomson, J.R., Christen, D.K., List, F.A., Paranthaman, M., and Lee, D.F., *Science* **311**, pp 1911-1914 (2006)
6. Campbell, T.A., Haugan, T.J., Maartense, I., Murphy, J., Brunke, L., and Barnes, P., *Physica C* **423**, pp 1-8 (2005).
7. Varanasi, C., Barnes, P.N., Burke, J., Carpenter, J., and Haugan, T.J., *Appl. Phys. Lett.* **87**, pp 262510-262510-3 (2005).
8. Foltyn, S.R., Tiwari, P., Dye, R.C., Le, M.Q., and Wu, X.D., *Appl. Phys. Lett.* **63**, pp 1848-1850 (1993).
9. Foltyn, S.R., Civale, L., Macmanus-Driscoll, J.L., Jia, Q.X., Mairov, B., Wang, H., and Maley, M., *Nat. Mater.* **6**, pp 631-642 (2007).
10. Varanasi, C.V., Burke, J., Wang, H., Lee, J.H., and Barnes, P.N., *Appl. Phys. Lett.* **93**, pp 092501-092501-3 (2008)
11. Varanasi, C.V., Burke, J., Brunke, L., Wang, H., Lee, J.H., Barnes, P.N., *J. Mater. Res.* **23**, pp 3363-3369 (2008)