Afyon Kocatepe Üniversitesi Fen ve Mühendislik Bilimleri Dergisi

Afyon Kocatepe University Journal of Science and Engineering

AKÜ FEMÜBİD **14** (2014) OZ5740 (255-260) AKU J. Sci. Eng. **14** (2014) OZ5740 (255-260)

Improvement of Flux Pinning Properties of YBCO Superconducting Films with $BaMeO_3$ (Me:Zr, Mn, Hf and Ir) Perovskite Nanoparticles by TFA-MOD Technique

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Geliş Tarihi:28.10.2012; Kabul Tarihi: 11.11.2013

Key words
High temperature superconductors; YBCO; $BaMeO_3$; Flux pinning; STO; TFA-MOD

Abstract

Superconducting materials increase performances of machines by intensively using in MRI in medicine, thermal bolometer, energy stroge systems in transformer, magnetic separators, levitation, NMR, generators, engines, cables, superconducting wires and tapes, accelerators, electromagnets, and electronic transistors. In these applications, the high temperature superconductors (HTS) possess very high crtitical current density (J_c) values at low temperature even in high magnetic fields. In these kinds of studies, many kinds of crystalline defects, such as fine precipitates of non-superconducting phases, dislocations, vacancies, grain boundaries, twin boundaries and so on, are considered to act as pinning centers. However, the J_c values rapidly decrease with increasing temperature in magnetic field. The main reason of the J_c depression is the lack of effective pinning in the structure. Depending on these reason, a novel technology has been developed by means of a nanostructure engineering to create artificial pinning centers in HTC materials. With this regard, increasing critical current density and improvement of flux pinning properties of $YBa_2Cu_3O_{6.57}$ (YBCO) superconducting films with $BaMeO_3$ (Me:Zr, Mn, Hf and Ir) perovskite nanodots, nanorods or nanoparticles, as pinning centers, on $SrTiO_3$ substrate were aimed.

TFA-MOD Tekniğiyle $BaMeO_3$ (Me:Zr, Mn, Hf ve Ir) Perovskit Nanopartiküllü YBCO Süperiletken Filmlerin Akı İğnelenmesi Özelliklerin Geliştirilmesi

Anahtar kelimeler
Yüksek sıcaklık süperiletkenler; YBCO; $BaMeO_3$; Akı iğnelenmesi; STO; TFA-MOD

Özet

Süperiletken malzemeler tıpta MRI cihazlarında, enerji depolama sistemlerinde, termal kameralarda, manyetik ayrıştırıcılarda, analiz cihazlarında (NMR), jeneratörlerde, motorlarda, kablolarda, süperiletken tel ve şeritlerde, hızlandırıcı mıknatıslarda ve transistörlerde, cihazların performanslarını arttırmak amacı ile kullanılmaktadır. Bu uygulamalarda, yüksek sıcaklık süperiletken malzemeleri (HTS) düşük sıcaklık ve yüksek manyetik alanlarda, çok yüksek kritik akım yoğunluğu (J_c) değerlerine sahiptir. Bu çeşit çalışmalarda, süperiletken olmayan fazların nano boyutta çökeltileri; dislokasyonlar, boşluklar, tane sınırları, ikiz sınırları gibi çoğu kristal hatalar, akı iğnelemesi merkezleri olarak düşünülmektedir. Ancak J_c değerleri manyetik alanda artan sıcaklıkla hızlı bir şekilde düşmektedir. J_c'nin düşmesinin ana sebebi etkili iğnelenme merkezlerinin olmamasıdır. Bu nedenlere bağlı olarak yeni teknoloji HTS malzemelerde yapısal iğnelenme merkezleri nanoyapılı mühendislik ile geliştirilmektedir. Bunu sağlamak amacıyla TFA-MOD metodu kullanılarak, yapısal iğnelenme merkezleri olarak $BaMeO_3$ (Me: Zr, Hf, Ir, vs.) perovskit yapılı nanonoktalar, nanoçubuklar veya nanopartiküller şeklinde yapıların $YBa_2Cu_3O_{6.57}$ (YBCO) süperiletken filmlerin içine ilave edilerek kritik akım yoğunluğunun artırılması ve akı iğnelenmesi özelliklerin geliştirilmesi hedeflenmiştir.

1. Introduction

Production of long-length and low cost YBCO films has been an essential aim in the field of superconducting processing. Besides, YBCO films need to possess a high critical current density (J_c) under high magnetic fields for practical applications as power and high field applications (Gutierrez *et al.*, 2007; Puig *et al.*, 2005). Improvement of their performance in magnetic fields has become one of the interesting topics on

high temperature superconductors (HTS). However, the ability for HTS to carry currents is significantly reduced in the presence of magnetic fields. In order to counteract this effect, various methods to increase the HTS current carrying abilities in magnetic fields by flux pinning have been developed through the pinning of the quantized flux lines by nanoscale crystalline defects and impurities (Strickland *et al.*, 2008; Engel et al., 2007).

Vacuum deposition techniques like pulsed-laser deposition (PLD) and metal organic chemical vapour deposition (MOCVD) have been used to produce these nanocomposite YBCO films. However high cost and small exposure area of these deposition techniques have hindered the large scale production (Liu et al. 2008). Because of this reason, the more practical and cost effective chemical solution deposition method (CSD) is preferred recently.

The way for flux pinning in the structure can be the use of nanoparticles-modified substrate surfaces, mixing rare earth doping in to the structure and doping with $BaMeO_3$ nanoparticles. Among these approaches, Ba(Zr, Mn, Hf and Ir)O_3 nanoparticles in YBCO thin films are one of the most popular ones which can prevent the vortex motion at high fields. The amount of dopant in the structure needs to be high enough to generate the density of defects, which is needed to enhance flux pinning in magnetic fields. However, excess amounts of this nonsuperconducting content can suppress the self-field and in-field J_c values significantly (Chen et al., 2009).

In the present work, undoped and $BaMeO_3$ doped YBCO thin films were grown with a non-vacuum, cost effective chemical solution deposition method. Their microstructures and dependence of the critical current on applied magnetic fields were investigated as a function of dopant concentration.

2. Material and Method

The precursor solutions of undoped and doped YBCO were prepared by dissolving acetates of Y, Ba and Cu with different amounts of Me(IV)-2,4-pentanedionates in propionic acid with a quantitiy of TFA. It is followed by a solution purification process. The concentration of the solution was fixed to 0.25 M with addition of acetone and propionic acid with a certain ratio. Three different doped solutions were prepared with 6, 12 and 18 mol% $BaMeO_3$. Film deposition on 10 mm x 10 mm STO (100) single crystal substrate was performed by spin coating technique at a rotation speed of 6000 rpm and acceleration speed of 6000 rpm s^{-1}.

Coated samples were heat treated according to the profile shown in Figure 1. During the heat treatment process, dry gas treatment up to 60^{o}C prevents the gel film from absorbing humidity that would deteriorate film integrity. Above 60^{o}C, humidified O_2 is introduced to suppress the sublimation of Cu trifluoroacetate. Metal trifluoroacetates decompose and harmful gaseous residues are removed during the pyrolysis to give a precursor film. After that, the fluoride containing precursor film is fired at a maximum temperature of 780^{o}C under humidified N_2 mixed with 100 ppm O_2 and then oxygenated at 450^{o}C to obtain the YBCO superconducting film.

X-Ray Diffraction (XRD) was carried out using a Philips diffractometer with Co Kα radiation to ascertain the phase purity of the undoped and doped films. Scanning Electron Microscopy (SEM, Philips XL20) was used to characterize the surface morphology of the final films. The critical transition temperature (T_c) and critical current density (J_c) of the films were measured by an inductive method. Transport measurements up to 6 T at 77 K on bridges of 0.8 mm length and 50 µm widths were carried out with a physical properties measurement system (PPMS)

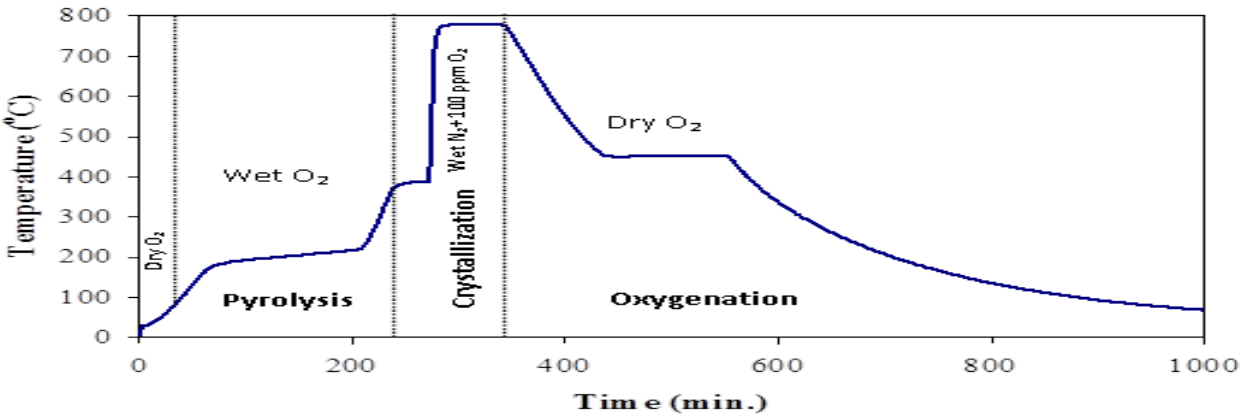


Figure 1. Heat treatment profile for the production of undoped and $BaMeO_3$ doped YBCO films. All films are crystallized at 780°C and oxygenated at 450°C.

3. Results and Discussion

Figures 2, 3, 4 and 5 show the θ-2θ diffraction patterns of undoped and $BaMeO_3$ doped YBCO films. In these patterns, the major peaks correspond to the (001) reflections of the YBCO phase and STO substrate which indicates that the YBCO film has a strong c-axis texture. Depending on the amount of dopant concentration, there is a slight increase at the $BaZrO_3$ (200) peak intensity. The structural integrity of the YBCO film can be easily affected by the doping process. In all doped samples (103) orientation of the YBCO phase becomes observable which represents that these small amounts of dopant deteriorate the textured structure of the film. Also, reduction in the (008) YBCO peak intensity with increasing BZO amount is evidence for this structural deterioration. A similar effect was found by J Hänisch et al., (2006) with nanoscale precipitates of Hf dopant in YBCO films.

Figures 6, 7, 8 and 9 depict surface morphologies of undoped and doped samples. These images show that all films have a smooth, crack free surface and all of them are generally formed by c-axis oriented grains. BMeO doped YBCO films present a denser surface structure with decreasing porosity compared with the undoped YBCO films. On the other hand, 18 mol% BZO doped sample surface has bigger sized grains in comparison to the fine grains of 6 and 12 mol% BZO doped sample surfaces.

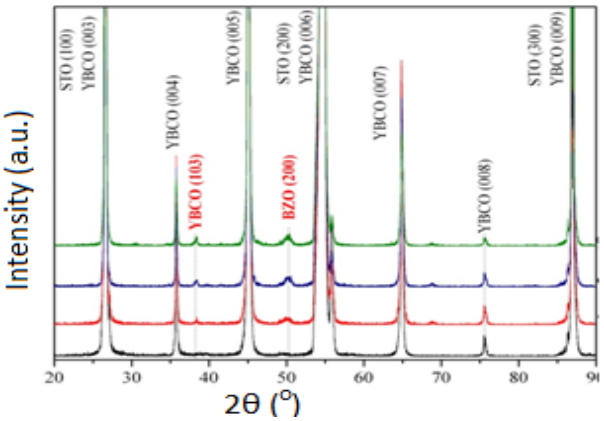


Figure 2. X-ray θ-2θ scans of undoped and BZO doped YBCO films with a Co Kα radiation.

The dependence of inductively measured critical transition temperature (T_c) on the amount of BMeO in the structure is shown in Figures 10, 11,

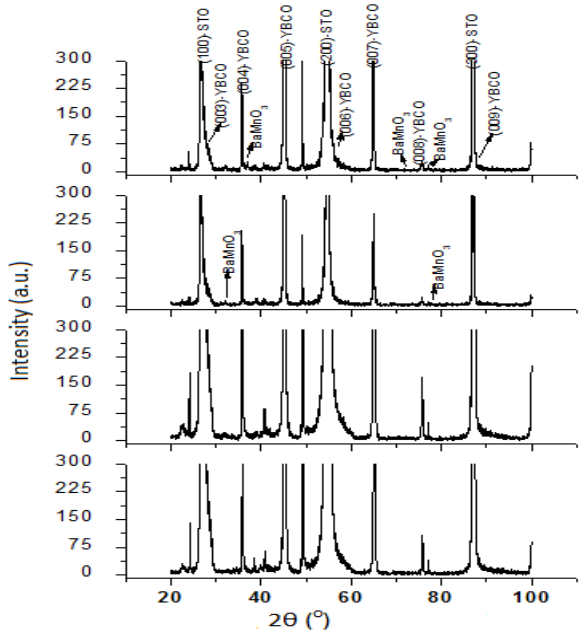


Figure 3. X-ray θ-2θ scans of undoped and BMnO doped YBCO films with a Co Kα radiation.

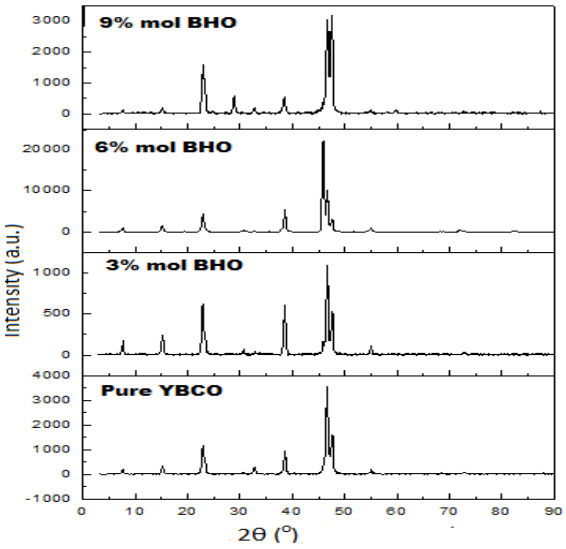


Figure 4. X-ray θ-2θ scans of undoped and BHfO doped YBCO films with a Cu Kα radiation.

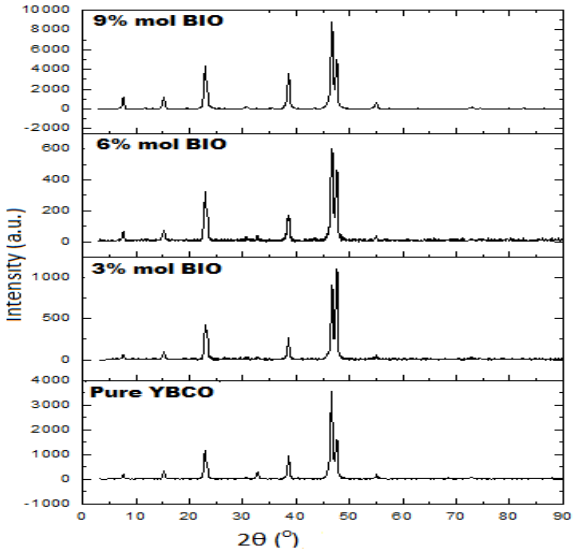


Figure 5. X-ray θ-2θ scans of undoped and BIrO doped YBCO films with a Cu Kα radiation.

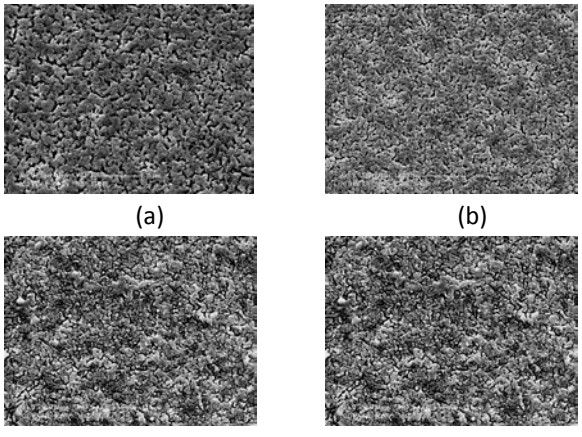


Figure 6. Surface morphologies of (a) pure YBCO film and YBCO films with (b) 6 mol%, (c) 12 mol% and (d) 18 mol% BZO addition.

12 and 13. The T_c value of 90.1 K for an undoped YBCO film decreases gradually to 88.2 K with the increasing amount of dopant inside for $BaZrO_3$doped films. Zr diffusion into YBCO and substitution on yttrium (Y) sites might be the reason for lowered T_c. Besides, the transition width of T_c (ΔT_c) stays below 1.0 K for undoped, 6 mol% and 12 mol% doped samples but is 2.0 K for 18 mol% doped sample which indicates the higher inhomogeneity of this sample.

As reported by Campbell & Evetts, (1972) the most accurate quantitative measurements of flux pinning are those of J_c as a function of field and temperature. Therefore, transport measurements were performed up to 6 T at 77 K in order to determine the magnetic field dependence of the critical current density as shown in Figure 14.

It can be seen that the 6 mol% BZO doped YBCO sample has the highest J_c value for all magnetic fields. At fields lower than 4 T, the undoped sample has higher Jc value than 12 mol% doped sample. However, the drop rate of 12 mol% doped sample is slower than the pure YBCO and J_c value of this sample exceeds the value of undoped YBCO sample at fields higher than 4 T. Within all the samples, the 18 mol% doped YBCO film has the lowest J_c value even at high fields which means that the superconducting structure of YBCO film is destroyed; self-field and in-field properties are suppressed with the excess amount of dopants concentration.

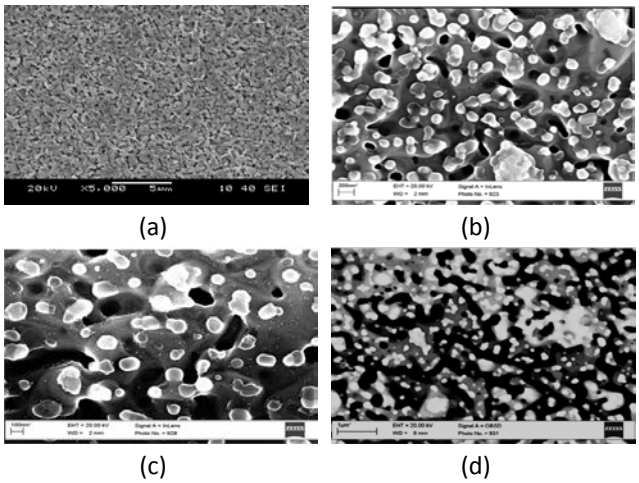
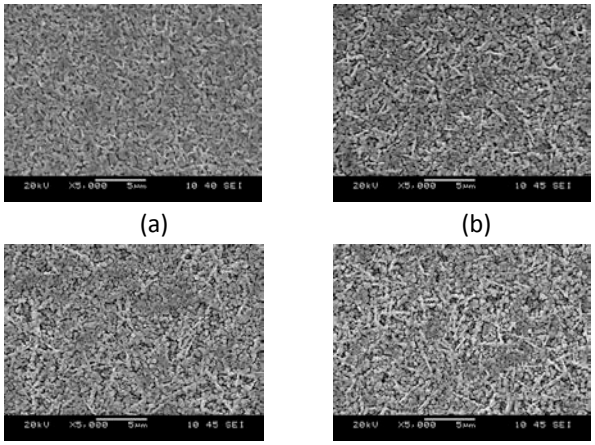
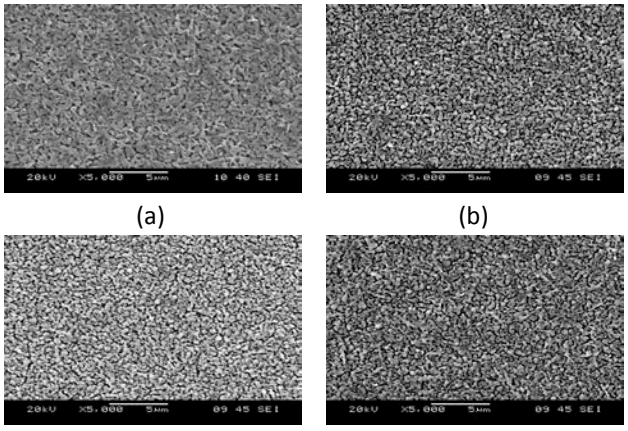


Figure 7. Surface morphologies of (a) pure YBCO film and YBCO films with (b) 6 mol%, (c) 12 mol% and (d) 18 mol% BMnO addition.



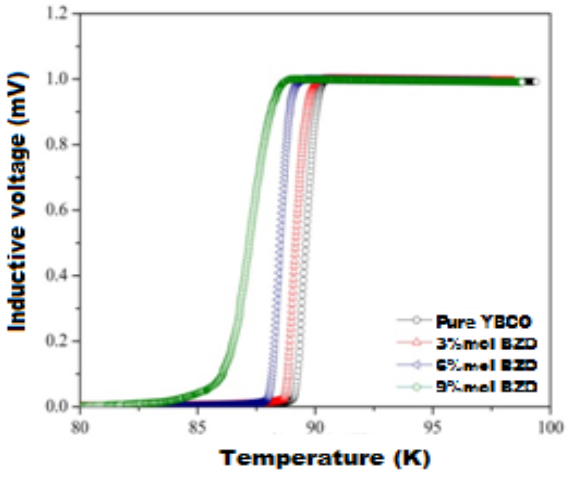
(a) (b) (c) (d)

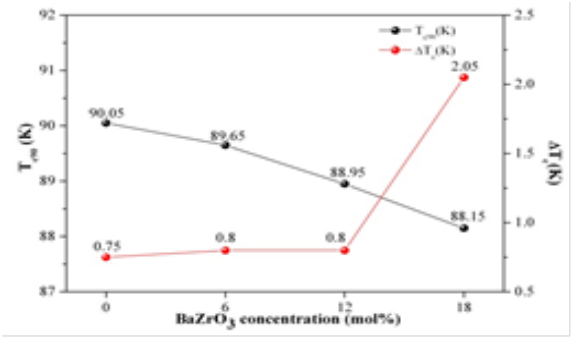
Figure 8. Surface morphologies of (a) pure YBCO film and YBCO films with (b) 6 mol%, (c) 12 mol% and (d) 18 mol% BHfO addition.



(a) (b) (c) (d)

Figure 9. Surface morphologies of (a) pure YBCO film and YBCO films with (b) 6 mol%, (c) 12 mol% and (d) 18 mol% BIrO addition.





$BaZrO_3$ concentration (mol %)

Figure 10. Dependence of critical temperature T_c and transition width ΔT_c on the amount of BZO in YBCO. T_c value is decreasing with increasing amount of BZO.

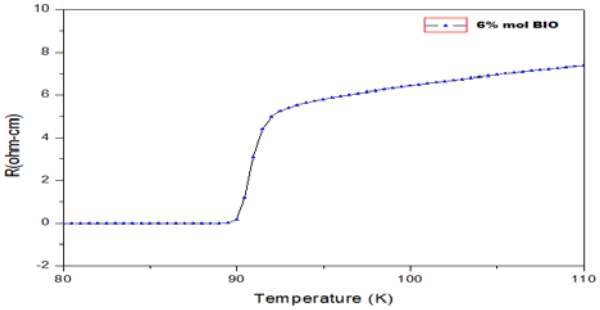


Figure 11. Critical temperature T_c of 6% mol BIO doped YBCO film.

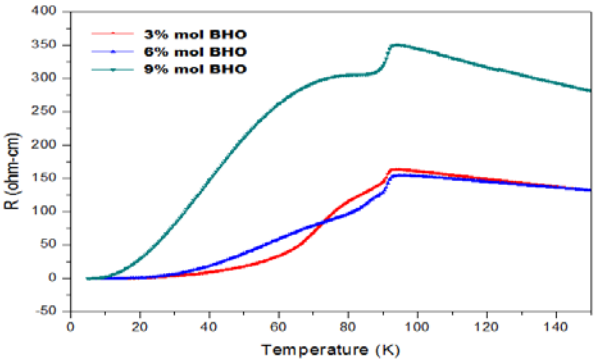
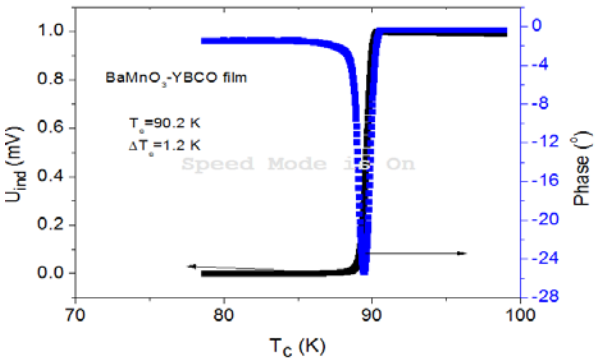


Figure 12. Dependence of critical temperature T_c on the amount of BHO in YBCO.



(a)

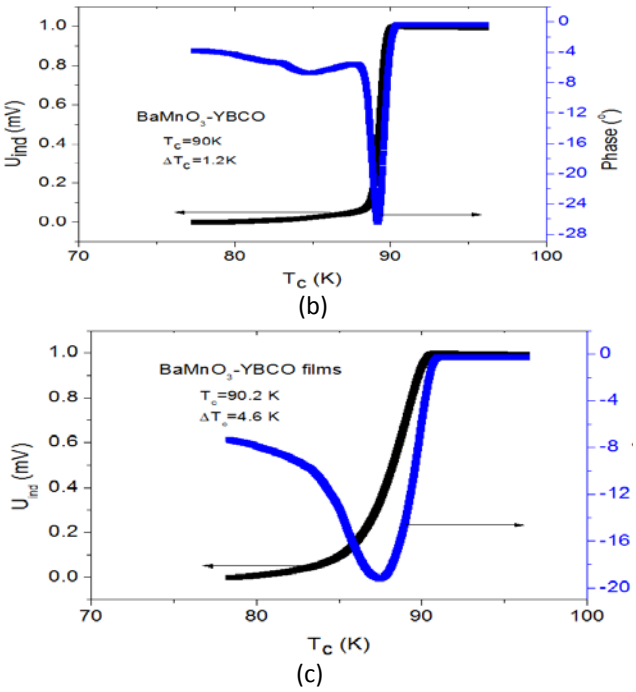


Figure 13. Dependence of critical temperature T_c and transition width ΔT_c on the amount of BaMnO in YBCO. T_c value is decreasing with increasing amount of BaMnO.

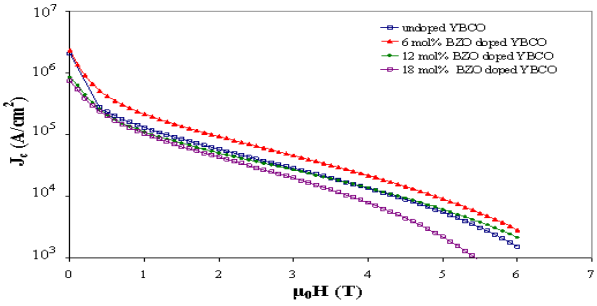


Figure 14. Field dependence of the critical current density for all samples on a single crystal STO substrate.

4. Conclusion

Barium zirconate (BZO) nanoparticles were introduced into the YBCO structure by the chemical solution deposition (CSD) method. The textured structure of undoped YBCO thin film was deteriorated with increasing dopant concentration. Both critical transition temperature (T_c) and critical current density (J_c) decreased with increasing dopant concentration. Jc measurements performed at 77 K to 6 T and highest Jc value has been reported for 6 mol% BZO doped sample. Further efforts will focus on improving the J_c value of films under high magnetic fields as well as the application of CSD method with other dopants and REBCO systems.

Acknowledgement

This study was derived from the PhD thesis of Graduate School of Natural and Applied Sciences of Dokuz Eylül University. Project was funded by The Scientific and Technological Research Council of Turkey (TUBITAK), under project number 109M054 and EU-FP6 Research Project "NanoEngineered Superconductors for Power Applications" NESPA no. MRTN-CT-2006-035619.

References

Campbell, A M., & Evetts, J. E, 1972. Flux vortices and transport currents in type II superconductors. *Advances in Physics*, *21*, 199-428.

Chen, Y., Selvamanickam, V., Zhang, Y., Zuev, Y., Cantoni, C., Specht, E., et al., 2009. Enhanced flux pinning by $BaZrO_3$ and $(Gd,Y)_2O_3$ nanostructures in metal organic chemical vapor deposited GdYBCO high temperature superconductor tapes. *Applied Physics Letters,* **94** (062513), 1-3.

Engel, S., Thersleff, T., Hühne, R., Schultz, L., & Holzapfel, B., 2007. Enhanced flux pining in $YBa_2Cu_3O_7$ layers by the formation of nanosized $BaHfO_3$ precipitates using the chemical deposition method. *Applied Physics Letters,* **90 (**102505) 1-3.

Gutierrez, J., Llordes, A., Gazquez, J., Gibert, M., Roma N., Richart, S., et al., 2007. Strong isotropic flux pinning in solution-derived $YBa_2Cu_3O_{7-x}$ nanocomposite superconductor films. *Nature Materials,* **6,** 367-373.

Hänisch, J., Cai, C., Stehr, V., Hühne, R., Lyubina, J., Nenkov, K., et al., 2006. Formation and pinning properties of growth-controlled nanoscale precipitates in $YBa_2Cu_3O_{7-\delta}$/transition metal quasi-multilayers. *Superconductor Science and Technology*, **19**, 534-540.

Liu, M., Suo, H., Ye, S., Shi, D., Zhao, Y., & Tang, X., 2008. Preparation and properties of YSZ-doped YBCO films grown by the TFA-MOD method. *Superconductor Science and Technology,* 21 (115012).

Puig, T., Gonzalez, J. C., Pomar, A., Mestres, N., Castano, O., Coll, M., et al., 2005. Influence of growth conditions on the microstructure and critical currents of TFA-MOD $YBa_2Cu_3O_7$ films. *Superconductor Science and Technology*, **18,** 1141-1150.

Strickland, N. M., Long, N. J., Talantsev, E. F., Hoefakker, P., Xia, J., Rupich, M. W., et al., 2008. Enhanced flux pinning by $BaZrO_3$ nanoparticles in metal-organic deposited YBCO second-generation HTS wire.

Physica C, **468**, 183-189.