



Investigation of physical and structural properties of cs doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superconductors

Öznur BAĞ^{1,*} , Saffet NEZİR² 

¹ Gaziosmanpaşa University, Department of Physics, Faculty of Arts and Science, Tokat/TURKEY

² Kırıkkale University, Department of Physics, Faculty of Arts and Science, Kırıkkale/TURKEY

Abstract

In this work, $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %) samples were prepared by using solid state reaction method. Some electrical, physical and structural properties of these compounds were examined by using SEM (scanning electron microscopy), XRD (X-ray diffraction), electrical resistivity, critical current density and AC susceptibility measurements, respectively. On the basis of the SEM measurements, it would seem that increasing the amount of Cs doping, the porous structures decrease and the grain size increases up to approximately $50 \mu\text{m}$. Unit cell parameters were calculated by employing XRD measurements. On the basis of the data obtained from X-ray diffraction, Cs atoms displaced Ba atoms in the crystal structure. From the measurements of electrical resistivity at 80 K - 120 K temperature, it was determined that the highest transition temperature was 91.5 K after addition of 0.05 wt. % Cs. The critical transition temperature was decreased by increasing the amount of Cs doping. Critical current density measurements on the same samples showed that as the amount of Cs doping increases, the values of J_c decrease. AC magnetic susceptibility measurements showed a sharper transition to the superconducting state in $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt.%) samples with the increase in the additive amount.

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1. Introduction

The discovery of high-temperature superconductor Y–Ba–Cu–O in 1987 has stimulated the intensity of scientists in development of the superconducting application theoretically [1]. $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) superconducting oxides are expected to be used in many technological applications such as magnetic levitation, high-field magnets, magnetic shields, motors and generators, because of their high critical current density (J_c) at 77 K and a high magnetic field [2]. There has been a considerable interest [3-4] in the preparation of good quality Y-Ba-Cu-O superconductors [5]. By adding different elements, such as Nb [6], Pr [7], Au [8], to the Y-Ba-Cu-O superconductor, the material is tried to be made more suitable for use in technological fields [9-10-11-12].

The synthesis of new ceramic materials with improved superconducting properties is one of the major concerns of today's researchers. Since the discovery of high temperature superconductors, many have tried to obtain compounds with better physical and morphological properties using simpler methods. The most commonly used chemical synthesis methods are solid state [13], sol-gel [14], automatic combustion [15], metal organic

chemical vapor deposition (MOCVD) [16] and microwave-based methods [17]. The most commonly used methods can be considered as a solid state reaction method, melting-casting method and sol-gel (nitrate), thin-film method. A solid-state method is the most widely preferred by superconductivity research groups due to the ease of use and cheapness [10].

The aim of this study was to synthesize and characterize the YBCO high temperature superconductor doped with different Cs element percentages and to investigate the effects of Cs doping on the properties of these superconducting compounds. A solid state reaction method was used to prepare $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %) superconducting compounds. We have studied the physical, structural and electrical properties of produced samples with XRD (X-ray powder diffraction), SEM (scanning electrical microscope), electrical resistivity, critical current density and AC susceptibility measurements.

2. Materials and Methods

The starting composition of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %) samples were prepared by using high purity nitrate compounds ($\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$,

*Corresponding author. e-mail address: oznurbag@gmail.com

$\text{Ba}(\text{NO}_3)_2$, $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ and $\text{Cs}(\text{NO}_3)$). The samples were subjected to calcination process for 4 h at 650 °C. After cooling to room temperature, the samples were ground for 2 h and pressed under a pressure of 440 MPa into pellets. The samples were put into a furnace at room temperature. The samples were sintered at 935 °C and attended under flowing oxygen atmosphere for 40 h, and then cooled to room temperature. The heating and cooling temperature rates were chosen to be 5 °C/min⁻¹ and 1 °C/min⁻¹, respectively. A schematic representation of sintering process is shown in Figure 1.

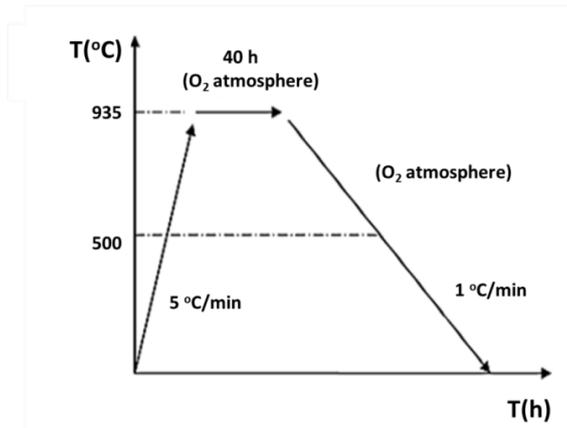


Figure 1. Schematic drawing of sintering process for the samples preparation.

The morphology of all superconducting samples are examined by using scanning electron microscope (Model Jeol JSM 5600). X-ray diffraction data were recorded using a Rigaku diffractometer with $\text{Cu-K}\alpha$ radiation over the range $2\theta = 10^\circ - 60^\circ$. The lattice parameters of superconducting samples were estimated by utilizing X-ray diffraction measurements. The superconducting transition temperature (T_c) and critical current density of samples was determined by a standart four-probe method [18]. Magnetic measurements were performed using a 7130 AC susceptometer of Lake Shore at temperature range of 40 K-100 K with a fixed magnetic field (80 m/A) and fixed frequency (111 Hz).

3. Results and Discussion

SEM images of the samples are shown in Figure 2. In the light of SEM analysis, it has been detected that the average particle size boosts as the amount of the addition grows [19, 20, 21]. The particles in the sample with 0.05 Cs addition are by and large circular and have 10 – 15 μm while the particles in the sample with 0.1 Cs addition are in the shape of a rectangular prism and their particle height can extend up to 30 μm . As illustrated in the Fig. 2, the particle height can reach up

to 50 μm in the samples which assume 0.2 and 0.3 addition rate and their width is about 10 μm on average. Furthermore, it can be inferred that the gaps among the particles have vanished to a great extent unlike first two samples, which leads to better contact among the particles [22]. As a result, it is contemplated that it will end in higher values of critical current density compared those of others. It is believed that to be increased the formation of liquid phase in the YBaCuO with Cs addition and it has also lead to remarkable growth in the grains.

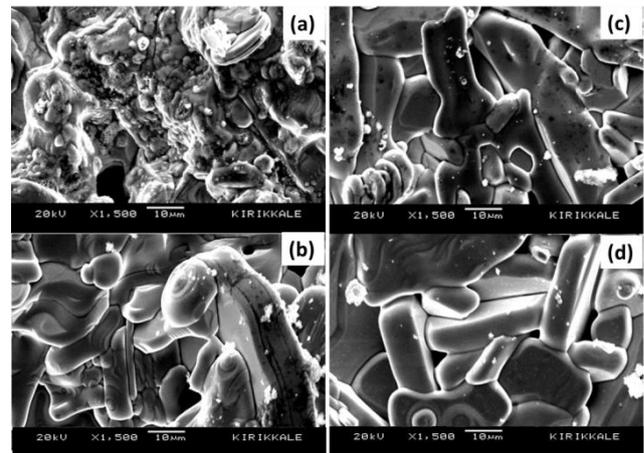


Figure 2. SEM micrographs of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %) a) 0.05 Cs b) 0.1 Cs c) 0.2 Cs d) 0.3 Cs.

The XRD patterns of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0, 0.05, 0.1, 0.2$ and 0.3 wt. %) are shown in Figure 3. All samples have the $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %) phase which is responsible of the superconducting state. As seen in Figure 3, it is understood that the samples completed the structural phase formation as a result of annealing and all the peaks obtained are compatible with the literature [23, 24, 25]. As can inferred from the figure, there appeared no superconductor phase and peaks belonging to tetragonal phase with low oxygen level. The (103), (113) and (123) peaks are seemed to decrease with increasing of Cs addition. Peaks such as (003), (004), (005) and (006) at the direction of c axis appear noticeably. That demonstrates that particles on the samples acquire acceleration throughout c axis as the amount of the addition increases. On the basis of ASTM data, no characteristic Cs peak has been observed as a result of comparison and contrast of characteristic peaks belonging to Cs with sample peaks. Such a portrait proves the fact that all Cs atoms replacing Ba atoms have largely occupied the spaces of Ba atoms bearing a crystal structure.

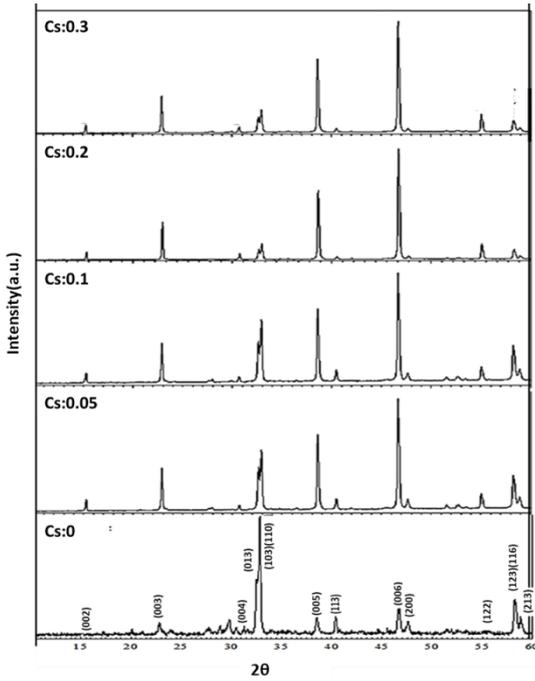


Figure 3. The X-ray diffraction patterns of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0, 0.05, 0.1, 0.2$ and 0.3 wt. %).

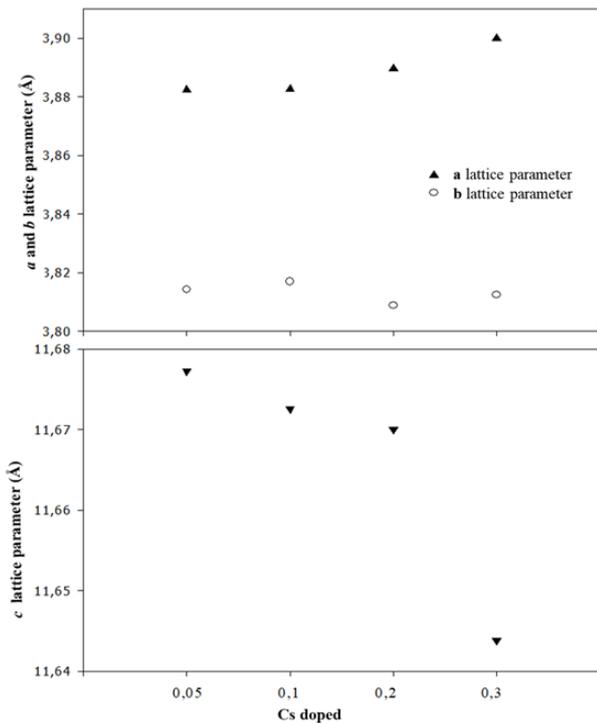


Figure 4. The lattice parameters of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %).

The lattice parameters obtained from X-ray diffraction of the samples and are plotted in Figure 4. The a parameter is found to increase continuously and the c parameter decrease with increasing of Cs concentration. It is estimated that this is caused by the fact that the ionic radius of Cs entering instead of Ba is larger, due to the O (5) gaps, the a axis expands a little

and thus the c axis is slightly contracted [26]. It is thought that there is not much change in the b parameter due to the O (1) ions located in the b axis.

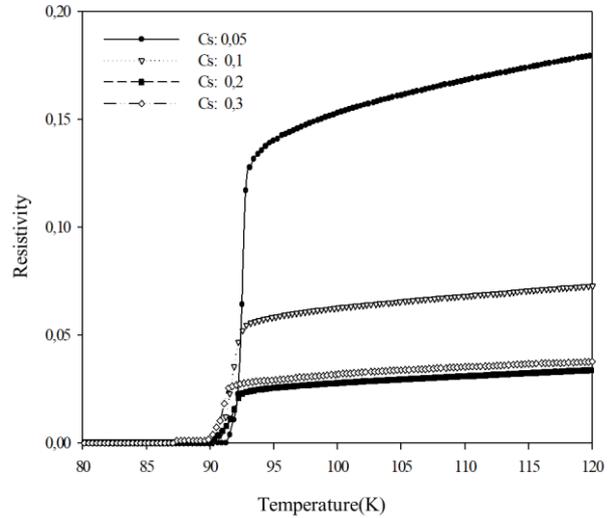


Figure 5. The electrical resistivity measurements of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %).

Figure 5 is shown that electrical resistivity measurement of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0, 0.05, 0.1, 0.2$ and 0.3 wt. %). As can be deduced from the figure, there happens a slight decrease in the resistance values in the normal zone the amount of the addition increases. It has been observed that superconductor transition (ΔT_c) is too sharp (app. 1.5 K). On the other hand, in the sample with 0.05 Cs addition its 91.5 K value decreases to 90 K in the sample in which the addition amount grows up to 0.3 in terms of weight. That effect may be traced from the graphic of the change of normalized impedance by heat illustrated in the Fig. 5. The study by M. Ausloos and his colleagues in which they replaced Ba with 0.05 Cs reported the critical transition heat as 80 K [27]. Whereas, 91.5 K heat was measured in the sample which we manufactured using the same addition.

Critical current density measurements were conducted applying 5 μA current through standard four-probe method. Figure 6 shows the measurement results. On the basis of measurements, it has been perceived that J_c values increase as the amount of the addition increases. While J_c is about 40-55 A/cm^2 in the samples with low addition, it moves to its maximum value, that is 160 A/cm^2 , in the sample with 0.3 Cs addition. It is estimated that the increase in the contact among the particles due to the decrease in the porosity and the increase in the particle size lead to such a change as can be inferred upon the analysis of SEM images.

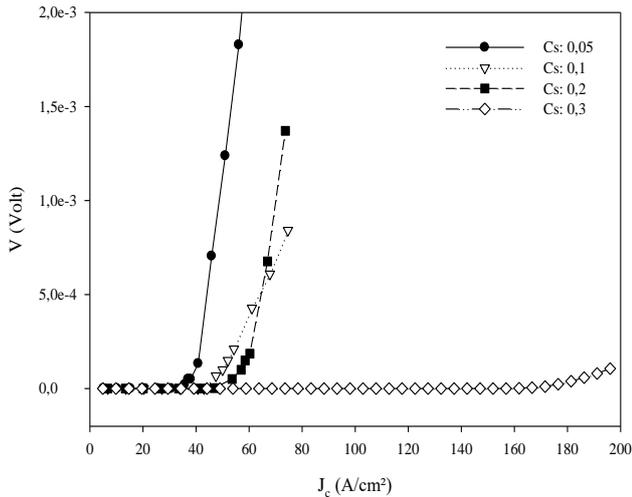


Figure 6. The critical current density J_c of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %).

Figure 7 shows the AC magnetic measurements at the temperature range of 40-100 K. As mentioned in the literature [28, 29, 30], coupling diamagnetic shielding seems clearly in relatively more granular the samples. In the samples where the Cs additive ratio is 0.2 and 0.3, the grain effect decreases considerably and the material passes into superconductivity more sharply.

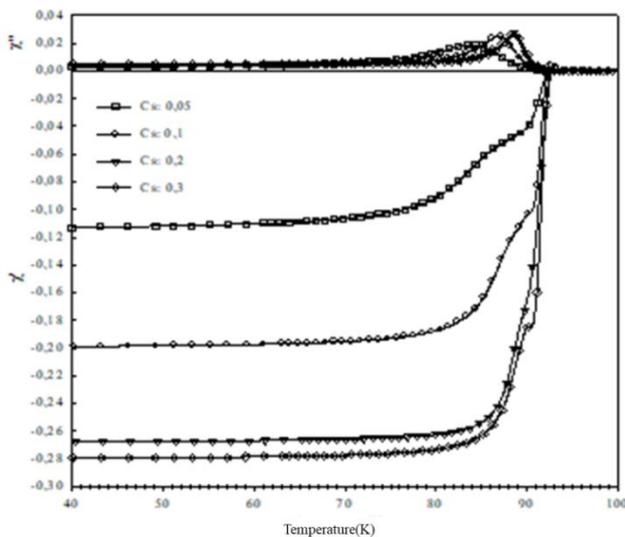


Figure 7. AC magnetic susceptibility of $\text{YBa}_{2-x}\text{Cs}_x\text{Cu}_3\text{O}_{7-\delta}$ ($x = 0.05, 0.1, 0.2$ and 0.3 wt. %).

4. Conclusions

In summary, Cs-added YBCO compounds were produced using the compounds of nitrated unlike conventional samples and we have identified some electrical, physical and structural properties of Cs-added YBaCuO superconducting samples. Scanning electron microscopy (SEM) images were obtained for annealed (at 935°C for 40 h) samples. It can be seen

that the porous structure in the samples has decreased with the increasing of the amount of the additive.

According to XRD measurements, (001) peak in the grains of all superconducting samples was observed to be significantly with the increasing of the amount of the additive. There appeared no superconductor phase and peaks belonging to tetragonal phase with low oxygen level. So it is appeared that the samples were completed the structural phase formation. By calculating the lattice parameters, it is observed a significant decrease in the lattice parameter c . This is estimated to cause the Cs atoms settle in the place of Ba atoms. The electrical resistivity measurement shows the resistivity of the samples and it is observed to occur in a slight decrease with increasing the amount of contribution. The study by M. Ausloos and his colleagues in which they replaced Ba with 0.05 Cs reported the critical transition heat as 80 K [26]. Whereas, 91.5 K heat was measured in the sample which we manufactured using the same addition. According to the results of the AC magnetic susceptibility measurements, as the amount of additive increased, it was seen that there was a visible decrease towards the ideal value of -1 in the real part of magnetic susceptibility. Cs addition enhances the formation of liquid phase in the YBCO superconducting compounds and as a consequence, it is believed that the coarse-grained structure emerged.

Conflicts of interest

The authors state that did not have conflict of interests.

References

- [1] Wu M.K., Ashbourn J., Torng C. J., Hor P.H., Meng R.L., Gao L., Huang Z.J., Wang Y.Q. and Chu C.W., Superconductivity at 93 K in a new mixed-phase Y-Ba-Cu-O compound system at ambient pressure, *Phys. Rev. Lett.*, 58 (1987) 908.
- [2] Zhang R.X., Yang H.X., Tian H.F., Chen G.F., Wu S.L., Wei L.L., Li J.Q., Superconductivity in the orthorhombic phase of thermoelectric $\text{CsPb}_x\text{Bi}_{4-x}\text{Te}_6$ with $0.3 \leq x \leq 1.0$, *Journal of Solid State Chemistry*, 232 (2015) 50–55.
- [3] Mumkani M., Melt processing of YBaCuO superconductors and critical currents, *Modern Physica Lett. B* 4(3) (1990) 163.
- [4] Yanmaz E., Drake A., Harris I.R. and Abell J.S., Melt processing of powdered arc-cast $\text{YBa}_2\text{Cu}_3\text{O}_y$ materials, *J. Alloys Compounds*, 195 (1993) 23.

- [5] Gencer A., Ateğ A., Aksu E., Nezir S., Çelebi S. And Yanmaz E., Microstructural and physical properties of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superconductors prepared by the flame-quench-melt-growth (FQMG) method, *Physica C*, 279 (1997) 165-172.
- [6] Naik S.P.K., Raju P.M.S., Seshubai V., Role of Sm and Nb on the preform optimized infiltration growth processed YBCO superconductors, *Materials Chemistry and Physics*, 182 (2016) 503-507.
- [7] Vovk R.V., Khadzhai G.Y., Dobrovolskiy O.V., Resistive measurements of the pseudogap in lightly Pr-doped $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ single crystals under high hydrostatic pressure, *Solid State Communications*, 204 (2015) 64–66.
- [8] Dadras S., Gharehgasloo Z., Effect of Au nanoparticles doping on polycrystalline YBCO high temperature superconductor, *Physica B*, 492 (2016) 45–49.
- [9] Guo N.N., Leu M.C., Additive manufacturing: technology, applications and research needs, *Front. Mech. Eng.*, 8 (2013) 215–243.
- [10] Wei X., Nagarajan R.S., Peng E., Xue J., Wang J., Ding J., Fabrication of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) superconductor bulk structures by extrusion freeforming, *Ceramics International*, 42 (2016) 15836–15842.
- [11] Volochova D., et al., YBCO bulk superconductors doped with gadolinium and samarium, *Physica C-Superconductivity and Its Applications*, (2013) 494.
- [12] Volochova D., et al., Contamination of YBCO bulk superconductors by samarium and ytterbium, *Physica C-Superconductivity and Its Applications*, (2014) 496.
- [13] Grigoryan S., et al., A new way of preparing the Y–Ba–Cu–O high-temperature superconductor using the sol–gel method, *Supercond. Sci. Technol.*, (2003), 16 (10) 1202.
- [14] Tang X., Zhao Y. and Grivel J.C., Influence of initial pH on the microstructure of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ superconducting thin films derived from DEA-aqueous sol-gel method, *Ceramics International*, (2013) 39.
- [15] Suan M.S.M. and Johan M.R., Synthesis of Al_2O_3 nanoparticles highly distributed in $\text{YBa}_2\text{Cu}_3\text{O}_7$ superconductor by citrate-nitrate auto-combustion reaction, *Physica C-Superconductivity and Its Applications*, 492 (2013) 49.
- [16] Li S.W., et al.. Morphology and superconducting properties of photo-assisted MOCVD processed YBCO film by variation of sublimation temperature of the Cu-based precursor, *Physica C-Superconductivity and Its Applications*, (2012) 478.
- [17] Baghurst D.R., Chippindale A.M., and Mingos D.M.P., Microwave syntheses for superconducting ceramics, *Nature*, (1988) 332.
- [18] Schildermans I., Van Bael M.K., Knaepen E., Yperman J., Mullens J. and Poucke L.V., *Physica C*, (1997) 4848278, 55.
- [19] Cheng C.W., Innes A.C.R., McN Alford N., Harmer M.A. and Birchall J., The effect of porosity on the superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_x$ ceramic, *Supercond. Sci. Technol.* 1, 113 (1988).
- [20] Murakami M., Supercond. Processing of bulk YBaCuO , *Sci. Technol.* 5, 185 (1992).
- [21] LaGraff J.R. and Payne D.A., Chemical diffusion of oxygen in single-crystal and polycrystalline $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ determined by electrical-resistance measurements, The American Physical Society, *Phys. Rev. B.* 47 (1993) 3380.
- [22] Dadras S., Dehghani S., Davoudiniya M., Falahati S., Improving superconducting properties of YBCO high temperature superconductor by Graphene Oxide doping, *Materials Chemistry and Physics.*, 193 (2017) 496-500.
- [23] Yvon K. and François M., Crystal structures of high-Tc oxides, *Z. Phys. B - Condensed Matter.*, 76(1989) 413.
- [24] Alecu G., Crystal Structures of Some High-Temperature Superconductors, *Romanian Reports Of Physics.*, 56 (2004) 404.
- [25] Cava R.J., Batlogg B., van Dover R.B., Murphy D.V., Sunshine S., Siegrist T., Remeika J.P., Rietman E.A., Zahurak S. and Espinosa G.P., Bulk superconductivity at 91 K in single-phase oxygen-deficient perovskite $\text{Ba}_2\text{YCu}_3\text{O}_{9-6}$, *Physical Review Letter.*, 59 (1987) 1676.
- [26] Kikuchi M., Syono Y., Tokiwa A., Oh-Ishi K., Arai H., Hiraga K., Kobayashi N., Sasaoka T. and Muto Y., *Japanese journal of Applied Physics.*, 26 (1987) L1066.
- [27] Ausloos M., Superconductivity in $\text{YBa}_{1.95}\text{Cs}_{0.05}\text{Cu}_3\text{O}_{7-y}$ granular ceramics, *Physical Review B.*, 39 (1989) 2729.
- [28] Goldford R.B., Lelental M. and Thompson C.A., *Alternating-Field Susceptometry and Magnetic Susceptibility of Superconductors.*, NISTIR. 3977 (1991) 1.

- [29] Nikola M. and Goldfarb R.B., Flux creep and activation energies at the grain boundaries of Y-Ba-Cu-O superconductors, *Physical Review B.*, 39 (1989) 6615.
- [30] Calzona V., Cimberle M.R., Ferdeghini C., Putti M., Sırı A.S., Vaccarone R., Supercurrent lengthscale in sintered YBCO and critical state model, *Physica C.*, 157 (1989) 425.