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The Effect of Ag on the Structure and Oxygen Content of YLEBCO Superconductor

I.B. Alit Paramarta, I.W. Supardi, N. Wendri, W.G. Suharta*

Department of Physics, Faculty of Mathematics and Natural Sciences, Udayana University

*Corresponding author's email: wgsuharta@gmail.com

Abstract. The effect of doping Ag on the oxygen content of $\text{Y}_{0.90}\text{La}_{0.05}\text{Eu}_{0.05}\text{Ba}_2\text{Cu}_3\text{O}_{7-d}$ (YLEBCO) superconductor has been carried out. YLEBCO superconductor was synthesized by the addition of Ag with the variations of 0.010, 0.025, and 0.050 of molar ratio. The synthesis process was made using a wet-mixing method with calcination at 600 °C for 3 hours and sintering at 900 °C for 5 hours. The samples were characterized by means of X-ray diffraction (XRD), Fourier Transform Infrared (FTIR), and Micro-Oxygen. The results of XRD analyses showed the domination of YLEBCO-123 phase with a volume fraction of between 85-92%, while the impurity phase Y-113 was also detected with a volume fraction between 8-15%. The increase in Ag (0.010-0.050) gave rise to enlarge the *a* and *b* axis. Meanwhile, the smaller change on *c* (3.891971-3.884080 Å) was observed with the increase in Ag content. Orthorhombic strain decreased (1.593%-1.126%) with the increase in doping Ag. The addition of Ag doping caused YLEBCO oxygen content decreased (6.938-6.674 Å).

Keywords: Superconductor, YLEBCO, wet-mixing, oxygen content, structure

1. Introduction

The substitution of Y ion in YBCO superconductors with a rare earth atom has been studied for one (Nd, Eu, Gd, Sm, Dy) ion [1,2] or a combination of several ions (Y-Nd, Y-Gd, Y-Eu, Nd-Eu-Gd) [3–6]. The substitution of atoms can increase the critical temperature of YBCO (90 K) to 94 K in REBCO superconductors. The latest may be indicated as an RE-123 phase. The substitution of the rare earth (RE) atom may give rise to change the lattice parameters, where the RE-123 lattice parameters are greater than Y-123 caused by the difference in ion radius of Y^{3+} instead of RE^{3+} . The similar phenomena also work for other superconducting families.

The application of superconductor demands the quality improvements for several parameters such as critical temperature, critical current density, as well as the critical magnetic field. In addition, the efforts to increase the value of J_c and H_c continue to be carried out, one of which is the substitution of Ag and Ce atoms [7–9]. The substitution Ag and Ce will fill the porosity, reduce the grain boundaries, resistivity to normal conditions and influence the weak-link. The various methods in the synthesis process of Y-123 and RE-123 superconductors have also been carried out to increase the value of T_c , J_c and H_c such as the solid reaction method, top-seeded melt growth (TSMG) method [10], melt-textured method [11] and oxygen-controlled melt growth method (OCMG) [12].



The properties of YBCO are closely similar to REBCO which is strongly influenced by the oxygen content in the compound. The oxygen content is determined by the treatment in the synthesis process. Usually, the determination of the oxygen content is done using the iodometric titration method. So far, many studies have been performed for the ion substitution for Y (RE), or Ba, or Cu sites on Y-123 as well as RE-123 phases. It is rare to find the effect of silver-doped Y-123 or RE-123 associated with its oxygen content and their structures. We prepared a series of Ag-doped RE-123 and calculated the oxygen content of each sample using micro-oxygen and their structures.

2. Methods

YLEBCO superconductor was synthesized using Y_2O_3 (99.9%), La_2O_3 (99.9%), Eu_2O_3 (99.9%), BaCO_3 (99.9%), CuO (99.9%), and AgNO_3 as the raw materials. The raw materials were weighed according to the composition of $\text{Y}_{0.90}\text{La}_{0.05}\text{Eu}_{0.05}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$. The Ag varied as 0.010, 0.025, and 0.050 in molar composition. The synthesis was conducted using the wet-mixing route prior to sintering. The raw materials were dissolved using HNO_3 then stirred to obtain a homogeneous solution. The homogeneous solution was heated at 100 °C to obtain a crust phase. The calcination process was carried out at 600 °C for 3 hours and followed by sintering at 900 °C for 5 hours. All the samples have been characterized by X-ray diffractometer (XRD), and Fourier Transform Infrared (FTIR). Meanwhile, the oxygen content was analyzed by micro-oxygen.

3. Results and Discussion

$\text{Y}_{0.90}\text{La}_{0.05}\text{Eu}_{0.05}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (YLEBCO) superconductor with doping Ag was successfully synthesized by calcinating at 600 °C for 3 hours and sintering at 900 °C for 5 hours prepared by means of the wet-mixing route. The XRD spectra of YLEBCO with various Ag (0.010, 0.025, and 0.050) are shown in Figure. 1. In general, the XRD spectra of all samples showed similar patterns with different intensities.

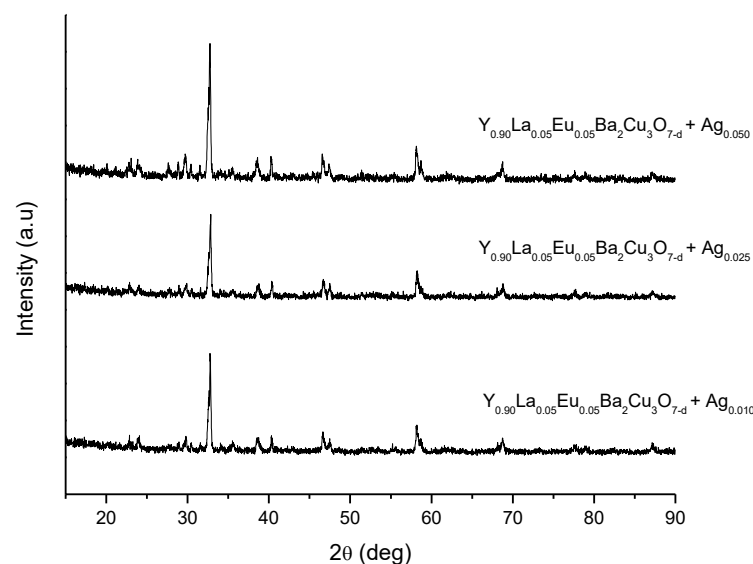


Figure 1. XRD spectra of $\text{Y}_{0.90}\text{La}_{0.05}\text{Eu}_{0.05}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ with doping Ag (0.010, 0.025, 0.050)

The phase analysis showed a dominance of the appearance of YLEBCO superconductors with a volume fraction between 85 and 92%. These results referred to $\text{YBa}_2\text{Cu}_3\text{O}_{6.8}$ (Y-123), $\text{EuBa}_2\text{Cu}_3\text{O}_7$ (Eu-123), and $\text{La}_{1.1}\text{Ba}_{1.9}\text{Cu}_3\text{O}_7$ (La-123); each with an entry of 00-041-0172 [13], 00-039-0486 [14], and 00-039-1414 [15]. All samples in the reference above had an orthorhombic crystal structure with a Pmmm (47) space group. The impurity phase in the form of Y-113 was also detected with a volume fraction between 8-15%. Y-113 referred to $\text{Ba}(\text{Sm}_{0.2}\text{Y}_{0.8})\text{Cu}_3\text{O}_{6+x}$ with the entry of 00-046-0143 [16]. The silver-doped YLEBCO histograms

in Figure 1 was typical for RE-123 phase. The peak profiles were certainly different from another common superconducting phase of La-214 [17,18] or Bi-2223 [19].

Rietveld analysis was carried out using Rietica software [20] with Newton-Raphson refinement strategy, normal calculation method, and Voigt peak profile. Rietveld analysis results for YLEBCO for Ag = 0.01 are shown in Fig. 2. The lattice parameters of YLEBCO superconductors with various Ag (0.010, 0.025, 0.050) are shown in Table 1. It is readily seen that the increase in doping Ag caused the lattice parameter towards a and c increased. Meanwhile, b decreased with the increase in the silver content. The increase in Ag also caused their orthorhombic strain decreased. The decrease in orthorhombic strain could be used to indicate a change in the crystal structure of orthorhombic to tetragonal or vice versa. The reliability and goodness-of-fit values are shown in Table 2. The value of goodness-of-fit of the samples ranged between 1.58-1.75. These values indicated the compatibility between the observed data and the reference was acceptable.

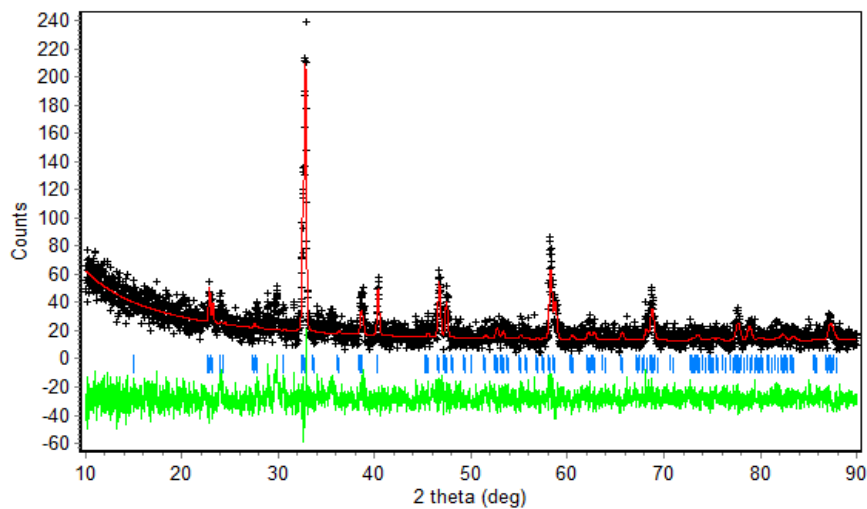


Figure 2. The refinement results of YLEBCO with doping Ag (0.010)

Table 1. The values of lattice parameter and orthorhombic strain of YLEBCO with various Ag contents of 0.010, 0.025, and 0.050

Sample	a (Å)	b (Å)	c (Å)	Orthorhombic strain
YLEBCO + Ag _{0.010}	3.830462	3.891971	11.656456	1.593
YLEBCO + Ag _{0.025}	3.837283	3.890462	11.678265	1.376
YLEBCO + Ag _{0.050}	3.840599	3.884080	11.702741	1.126

The spectra of FTIR for Ag-doped YLEBCO showed a curve with a similar pattern as shown in Fig. 3. The curve shows absorption peaks appearing at the wavenumbers of 3739.97, 3533.59, 2987.74, 2873.94, 2281.79, 1797.66, 1743.66, 1705.07, 1514.12, 1238.30, 1082.07, 952.84, 542.00, 505.35, and 443.63 cm^{-1} . These wavenumbers were related to the absorption of CO_3^{2-} bending vibration, CH_3 bending vibration, M-O vibration, and C-H stretching respectively.

Table 2. The values of reliability and goodness-of-fit of YLEBCO with doping Ag (0.01, 0.025, 0.05)

Sample	R_p	R_{wp}	R_{exp}	GoF
YLEBCO + Ag _{0.010}	23.60	18.67	18.75	1.58
YLEBCO + Ag _{0.025}	25.45	19.80	19.25	1.75
YLEBCO + Ag _{0.050}	23.69	18.36	18.69	1.61

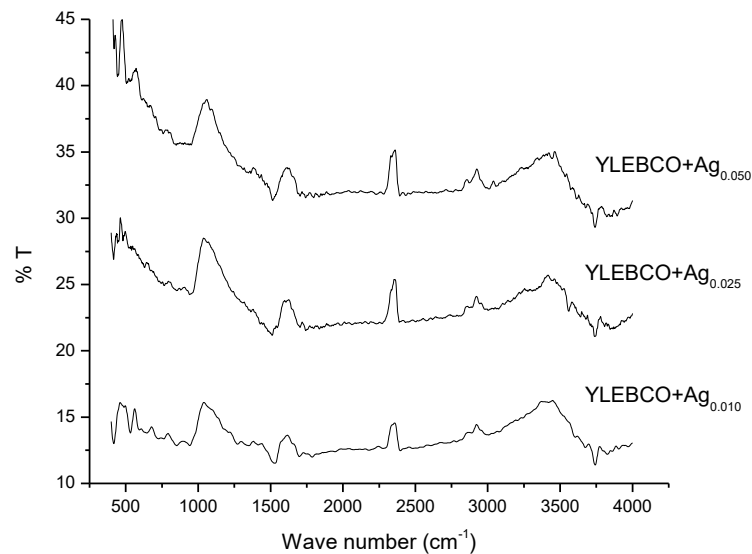


Figure 3. FTIR spectra of YLEBCO with doping Ag (0.010)

The oxygen content was calculated using micro-oxygen with the CHN628 series. The sample was placed on the micro-oxygen holder and heated at 1300 °C. The micro-oxygen characterization results of Ag-doped YLEBCO are shown in Fig. 4. The molar of oxygen content was calculated by dividing atomic weight (BA) of oxygen with the molecular weight (BM) of compounds following the equation below.

$$16.52\% = \frac{BM_{Oksigen}}{BM_{Senyawa}} = \frac{XBA(O)}{0.9BA(Y) + 2BA(Ba) + 3BA(Cu) + 0.05BA(Eu) + 0.05BA(La) + 0.01BA(Ag) + XBA(O)}$$

$$X = 6.93797$$

The results of the calculation of all samples are displayed in Table 3. It shows that the addition of doping Ag resulted in a decrease in the oxygen content of YLEBCO.

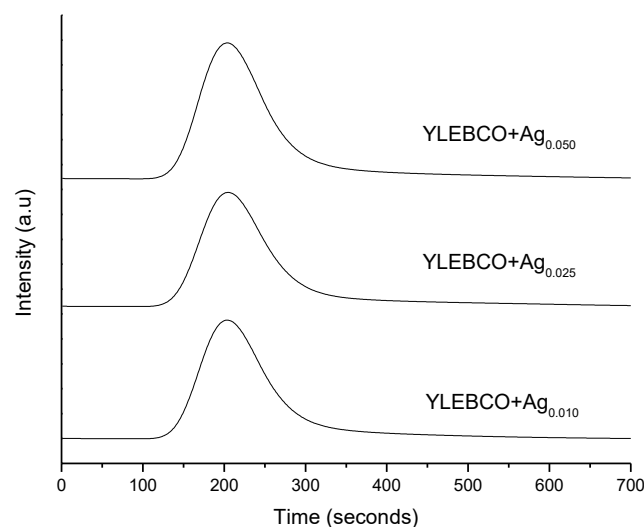


Figure 4. The micro-oxygen characterization results of YLEBCO with doping Ag (0.010, 0.025, 0.050)

The obtained micro-oxygen characterization is displayed in percent to its total compound. By employing the equation, the actual molar ratio of chemical formula on its compound is shown in Table 3. The reduction of oxygen content by the increase in silver in the YLEBCO is reasonably acceptable. The valence of the stable existing and the oxygen content was lower than RE.

Table 3. The micro-oxygen characterization and content oxygen

Sample	%content	molar ratio
YLEBCO + Ag _{0.010}	16.52%	6.938
YLEBCO + Ag _{0.025}	16.24%	6.817
YLEBCO + Ag _{0.050}	15.89%	6.674

4. Conclusion

From data analysis, we may conclude that the increase in Ag content of 0.010 - 0.050 gave rise to increase the lattice parameters of a (3.830462-3.840599 Å) and c (11.656456-11.702741 Å). On the other hand, the increase in silver content reduced b -lattice from 3.891971 to 3.884080 Å. It was further obtained that the orthorhombic strain decreased from 1.593 to 1.126% with the increase in Ag content. The addition of Ag doping caused the oxygen content of YLEBCO decreased from 6.938 to 6.674.

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