The Microstructure of Y-359 Superconducting Phase with Various of Y-211 Compound

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Abstract. The synthesis of Y-359 superconducting phase with a certain amount of Y-211 green phase compound has been carried out. The samples were synthesized by means of sintering method which were prepared by a wet-mixing route using Y$_2$O$_3$ (99.99%), BaCO$_3$ (99.99%), and CuO (99.99%) as raw materials. The purpose of this study was to find the microscopic properties of the Y-359 superconductor in terms of the changes on lattice parameters and morphology. This study consists of three steps. The first step was the formation of the Y-211 compound; the second is the formation of the Y-359 superconductor, the third is the formation of the Y-359 superconductor doped Y-211 compound. The wet-mixing method consists of mixing raw materials with HNO$_3$ and stirred using a magnetic stirrer. The processes were followed by calcination at 600 °C for 3 hours and subsequently followed by sintering at 900 °C for 5 hours.

The characterizations of the samples were performed using X-ray diffractometer (XRD), scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS). The addition of Y-211 causes the intensity, the volume fraction, orthorhombic strain and density of Y-359 superconductor increase. Also, the addition of Y-211 causes the lattice parameters of $a$ and $b$ is decreasing from 3.833434 to 3.830971 Å and from 3.890728 to 3.888263 Å respectively. On the other hand, the lattice parameter of $c$ increased from 31.048117 to 31.052530 Å. Furthermore, we found that the particle size of the samples ranged between 500 to 750 nm.

Keywords: Wet-mixing route, Y-359 superconductor, Y-211 compound, lattice parameter.

1. Introduction

The Y-358 superconductor which is a development of the Y-123 family has never been separated from the attention of researchers, considering its critical temperature (97 K) is higher than the critical temperature of the Y-123 superconductor (90 K). Various experiments have been carried out related to this material. Regarding the synthesis method, generally, researchers used the solid-state reaction method. The Y-358 superconductor synthesis reported by Landinez et al., it was carried out by a solid-state reaction method followed by calcination at 960 °C for 12 hours and sintering at 920 °C for 24 hours [1]. Kruaheong also reported the Y-358 superconductor synthesis, it also made by a solid-state reaction method followed by calcination at 950 °C for 24 hours, sintering at 950 °C for 24 hours and annealing at 500 °C for 24 hours [2]. Meanwhile, a previous work performed it with calcination at 840 °C for 24 hours and sintering at 900 °C for 24-48 hours [3].
In addition, to obtain superconductor materials with high homogeneity, the wet-mixing route used instead of the direct solid-state reaction method, as Sumadiyasa reported in growing YLBCO superconductors [4]. Several works reported an effort to obtain superconducting phases which were employing the wet-mixing method to grow the Bi-2212 phase [5], and BSCNGCO superconductor nanoparticle [6].

The Y2BaCuO5 (Y-211) compound is a green phase, which is the initial phase formed before becoming Y-123 phase. The presence of green phase Y-211 is useful for increasing critical current density (Jc) [7]. In this study, the addition of various Y-211 phase was carried out to obtain the growth of Y-359 phase. In this work, the wet-mixing method using HNO3 as digest agent was employed prior to sintering of both phases of Y-211, Y-359, and the mixture of them.

2. Method
The experimental procedure of this study consisted of three steps. The first step was to prepare the Y-211 compound by dissolving raw materials Y2O3, BaCO3 and CuO with HNO3. The mixture stirred with a magnetic stirrer to get a homogeneous solution. The homogeneous solution calcined at 600 °C for 3 hours and sintered at 900 °C for 5 hours. The second step was to prepare the Y-359 with the same synthesis process as Y-211 preparation, only in this process, sintering was carried out at 850 °C for 5 hours. The third step was to add Y-211 compounds into Y-359 superconductor with a different fraction of 5, 10, and 15 wt%. Each mixture composition was ground homogenously using a mortar. The last process was sintering all mixtures at 850 °C for 5 hours. All samples were characterized by means of X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS).

3. Results and Discussion
The XRD pattern of the Y-211 compound showed in Figure 1.

![XRD pattern of the Y-211 compound](image)

**Figure 1.** XRD pattern of the Y-211 compound. symbol o for Y-211, and symbol x for Y2O3

Phase identification was conducted using the Match software. The XRD spectrum is prominently dominated by Y-211 phase, and it was followed by the appearance of the Y2O3 as an impurity phase. The phase identification of Y-211 compound refers to Wong [8] with ICDD 00-038-1434, while Y2O3 phase refers to Grier [9] with ICDD 00-043-1036. The calculation results showed that the volume fraction value of Y-211 phase is 89%, while the volume fraction of the Y2O3 phase is 11%. This indicated that the synthesis process with wet-mixing method followed by the calcination at 600 °C for 3 hours and it sintering at 900 °C for 5 hours was an effective way to form Y-211 compounds.
We performed a Rietveld analysis using Rietica software for the Y-211 compound. The reliability of $\chi^2$, $R_p$, $R_{wp}$, and $R_e$ are 2.19, 20.9, 26.9, and 18.2, respectively. It was obtained that the lattice parameter Y-211 samples were 7.129663, 12.176950, and 5.658031 Å for $a$, $b$, and $c$ respectively. Figure 2 is the XRD pattern of Y-359 superconductor with the addition of different Y-211 compound. In this case, Y-211 is the impurity phase. As seen in the figure, Y-359 superconductor phase is shown by an empty circle (o), while Y-211 impurity phase is represented by a solid circle (●). The histograms of Y-359 as depicted in Figure 2, is ready can be distinguished from other superconducting phases of La-214 [10, 11] or Bi-2223 [12] families. Figure 3 is a selected Bragg’s diffraction angle of 26-35° to clarify the increase of intensity of the highest peak.

![XRD pattern of Y-359 with doping of the Y-211. Symbol: ● (Y-359), o (Y-211)](image)

It is found that the addition of Y-211 phase caused the intensity of Y-359 superconducting phase to increase. The volume fraction of Y-359 compound increased from 89 to 94% with the rise in the Y-211 compound from 5 to 15 wt %. The lattice parameters of the Y-359 superconductor with the addition doping of the Y-211 were obtained from Rietveld analysis using the Rietica software [13]. Rietveld analysis was performed using a model obtained from the result Landinez [1]. The lattice parameters of each sample obtained in this analysis are shown in Table 1. It can be seen that the addition of Y-211 compound from 5 to 15 wt % causes the lattice parameter of $a$ and $b$ is decreasing, while the lattice parameter $c$ increase. The lattice parameter $a$ decreases from 3.833434 to 3.830971 Å. The $b$ lattice also decreases from 3.890728 to 3.888263 Å. In contrary, the $c$ lattice increases from 31.048117 to 31.052530 Å.

From the lattice parameters, we obtained a measure of orthorhombic strain. The orthorhombic strain increased with the addition Y-211 as shown in Table 1. This indicates that the addition of Y-211 resulted in the orthorhombic structure of the crystal significantly visible.

| Sample | $a$ (Å)       | $b$ (Å)       | $c$ (Å)       | Orthorhombic strain (%) |
|--------|---------------|---------------|---------------|-------------------------|
| Y-359 + 5 wt % Y-211 | 3.833434(2)   | 3.890728(9)   | 31.048117(7)   | 1.483501                |
| Y-359 + 10 wt % Y-211 | 3.831881(4)   | 3.889173(0)   | 31.051232(3)   | 1.483971                |
| Y-359 + 15 wt % Y-211 | 3.830971(8)   | 3.888263(8)   | 31.052530(2)   | 1.484020                |
Figure 3. Selected area of Y-359 histograms with various Y-211.

Landinez et al. reported the lattice parameter of Y-358 superconductor with \(a\), \(b\), and \(c\) are 3.9211, 3.8514, and 31.0170 Å, respectively [1]. In general, the lattice parameter of Y-359 superconducting phase obtained in this work differs from the Y-358 phase. The lattice parameter of \(a\) and \(b\) for Y-359 is greater than Y-358 phases, while the \(c\) lattice parameter Y-359 shows smaller than Y-358. From the refinement, we also found the samples’ density. The reliability, Chi-square and density values of Y-359 superconductor with variations in addition of Y-211 compounds of 5, 10, and 15 wt% is shown in Table 2. It is seen that the sample density increased with the addition of the Y-211 compound.

| Sample                | \(Rp\) | \(Rwp\) | \(Rexp\) | \(\chi^2\) | density g/cm³ |
|-----------------------|--------|---------|----------|-----------|--------------|
| Y-359 + 5 wt % Y-211 | 20.66  | 26.67   | 17.24    | 2.39      | 6.272        |
| Y-359 + 10 wt % Y-211| 20.41  | 27.18   | 16.32    | 2.61      | 6.275        |
| Y-359 + 15 wt % Y-211| 20.68  | 27.51   | 16.92    | 2.42      | 6.278        |

The SEM images of Y-359 with a various fraction of a Y-211 phase is shown in Fig. 4 and 5, respectively. As illustrated in Figures 4 and 5, the addition doping of the Y-211 compound indicates that the crystal size is more distributed. This is consistent with the results of the study reported by M.R. Koblischka [14], where the Y-211 phase does not only function as a flux pinning center and increases the critical current, but also can produce a more homogeneous grain size due to the interaction between Y-211 and Y-359 phases. The particle size of the sample ranged from 500 to 750 nm which were calculated using ImageJ software.

Table 3. EDS analysis of the Y-359 + 5 wt % Y-211.

| Atom | keV  | Mass% | Atomic% |
|------|------|-------|---------|
| O    | 0.525| 23.66 | 64.56   |
| Cu   | 8.040| 18.67 | 12.83   |
| Y    | 1.922| 24.79 | 12.16   |
| Ba   | 4.464| 32.88 | 10.45   |
Figure 4. The SEM image of Y-359 + 5 wt% Y-211.

Figure 5. The SEM image of Y-359 + 15 wt% Y-211

Figure 6. EDS spectrum of the Y-359 + 5 wt % of Y-211

Figure 6 is the EDS spectrum of Y-359 superconductors with 5 wt % of Y-211 compound (inset: area of inspection). It is seen that all of the starting materials for the Y-359 and Y-211 compounds were contained in the sample. Further analyses inform us that the percentage of each element was not suitable with the initial composition of the sample, as shown in Table 3. This is understandable because the characterization of EDS takes several observation points, so it does not represent the entire sample.

4. Conclusion
The wet-mixing method followed by calcination process at 600 °C for 3 hours and sintering at 900 °C for 5 hours is an effective way to form the Y-211 compound. The addition of Y-211 green phase affects the intensity of Y-359 superconductor increase. The addition of doping Y-211 from 5 to 15 wt% causes the increasing of the volume fraction of Y-359 superconductor from 89 to 94%. We also found that the orthorhombic strain of Y-359 phase increase from 1.483501 to 1.484020 which also increased of density from 6.272 to 6.278 g/cm³. The addition of the Y-211 compound from 5 to 15 wt % decreases the lattice parameter if the Y-359 phase for a and b, but an increase of c. The lattice a and b decrease from 3.833434 to 3.830971 Å and from 3.890728 to 3.888263 Å respectively. While the c lattice parameter increased from 31.048117 to 31.052530 Å. The average particle size of Y-359 is in the range of 500-750 nm.
References

[1] Landénez DA, T´ellez M, Cabrera B´aez and Roa-Rojas J 2012 Modern Physics Letters B 26 11.
[2] Kruaehong T 2014 International Journal of Physical Sciences 9 16 360-367.
[3] Roaa F Al-Masoodi, Emad K Ai-Shakarchi, International Journal of Advanced Research in Physical Science 2 7 33-39.
[4] Sumadiyasa M, Adnyana IGAP, Widagda IGA, Suharta WG 2016 Journal of Physics: Conference Series 725 1 012001.
[5] Suharta WG, Widagda IGA, Putra K, Suyanto H 2017 Journal of Physics: Conference Series 820 1 012006.
[6] Sudarmini IAM, Suharta WG, Suarbawa IKN, Sukarasa K, Suryatika IBM International Journal of Geomat 15 50 155-160.
[7] Sun JZ, Webb DJ, Naito N, Char K, Hahn MR, Hsu JWP, Kent AD, Mitzi DB, Oh B, Beasley MR, Beballe TH, Hammond RH, Kapitulnik A 1987 Phys. Rev. Lett. 58 1574-1576.
[8] Wong-Ng W, McMurdie H, Paretzkin B, Hubbard C, Drago A 1987 ICDD Grant-in-Aid, NBS (USA).
[9] Grier D, McCarthy G, North 1991 ICDD Grant-in-Aid Dakota State University, Fargo, North Dakota, USA.
[10] Sutjahja, I.M, Diantoro,M, Darminto, D, Nugroho, AA, Tjia, M.O., Menovsky, AA, Franse, J. J.MF. Fish-tail effect and superconducting phase diagram of LaNdSrCuO4 single crystal, Physica C 378-381(2002) 541-545.
[11] Sutjahja, I.M, Aarts, J., Nugroho, A.A. Diantoro, M, Tjia, M.O., Menovsky, A.A. and Franse, J.J.M., Doping and field effects on the lowest Kramers doublet splitting in La1.6-xNd0.4SrxCuO4 single crystal, Physica C 392-396 (2003) 207-212.
[12] Kovác, P, Husek, I., Pachla, W., Diantoro, M., Bonfait, G., Maria, J, Fröhlich, K, Kopera, L., Diduszko, R, and Presz A, Material for resistive barriers in Bi-2223/Ag tapes, Supercond. Sci. and Technol., 14 (2001) 11.
[13] Hunter BA, 1997 Rietica for Windows version 1.7.7.
[14] Koblishka, Koblishchka-Veneva A, Reddy ES, Schmitz GJ, Ogasawara K, Murakami M 2003 Physica C 392-396 589-595.