Nanostructural Characterization of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ Superconductor Tapes Fabricated by TFA-MOD Process Using Starting Solutions with Different Cation Ratios

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Trifluoroacetate-metal organic deposition (TFA-MOD) method is one of low cost process for fabrication of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) superconductor tapes. It is reported that the YBCO films fabricated by the TFA-MOD process using starting solutions with cation ratios of Ba-deficient (Ba/Y < 2.0) have higher critical current ($I_c$) values than that of stoichiometric composition (Ba/Y = 2.0). We fabricated the TFA-MOD YBCO films using starting solutions with the Ba-deficient ($\text{Y}:\text{Ba}:\text{Cu} = 1.0:1.5:3.0$) and the stoichiometric composition ($\text{Y}:\text{Ba}:\text{Cu} = 1.0:2.0:3.0$) on Hastelloy$^\text{TR}$ metal substrates with a CeO$_2$/LaMnO$_3$/MgO/Gd-Zr-O buffer layer [1,2]. Fig. 1 shows the relationship between the $I_c$ values of the holding time in the crystallization process. The highest $I_c$ values for the respective films were achieved with a 100 min crystallization process. After further long time crystallization, the $I_c$ values of the stoichiometric composition films decreased, while those of the Ba-deficient films stayed the same [3]. In this study, we observed the nanostructures of these films using scanning electron microscopy (SEM), a focused ion beam (FIB)-SEM dual beam system, and transmission electron microscopy (TEM) to compare the nanostructural changes due to the influence of the starting solution composition during the crystallization process.

Figure 2 shows cross-sectional TEM images of (a) Ba-deficient and (b) stoichiometric composition films after 100 min crystallization process. Fig. 3 (a) - (d) shows the results of EDS elemental maps of Y, Ba, Cu, and F corresponding to the region surrounded by the red-dotted line in Fig. 2 (a), and Fig. 3 (e) – (h) shows Y, Ba, Cu, and F maps corresponding to the region by the red-dotted line in Fig. 2 (b). Both Ba-deficient and stoichiometric composition films, after the 100 min crystallization process, are mainly composed of $c$-axis oriented YBCO grains. In addition, CuO, $\text{Y}_2\text{Cu}_2\text{O}_5$ grains, and $\text{BaF}_2$ regions with an average size of 200 nm are distributed in the films. However, the quantity of $\text{BaF}_2$ grains of the stoichiometric composition film are quite larger than that of the Ba-deficient film. Fig. 4 (a) and (b) shows a cross-sectional TEM image of the Ba-deficient and the stoichiometric composition film after 180 min crystallization process, respectively. Fig. 5 (a) - (d) shows the results of EDS elemental maps of Y, Ba, Cu, and F corresponding to the region surrounded by the red-dotted line in Fig. 4 (a), and Fig. 5 (e) – (h) shows Y, Ba, Cu, and F maps corresponding to the region by the red-dotted line in Fig. 4 (b). After 180 min of crystallization process, some of $\text{BaF}_2$ grains convert to $\text{BaCuO}_2$ grains in the stoichiometric composition film. Fig. 6 shows a 3D reconstructed image of cracks formed in the stoichiometric film after 180 min crystallization process using the FIB-SEM dual beam system. These cracks have the radial shapes and seemed to be occurred around the larger sized secondary phases.
(BaCuO$_2$, CuO, Y$_2$Cu$_2$O$_5$). Moreover, YBCO c-axis oriented grains are broken around the cracks. These nanostructural changes can be observed only in the stoichiometric composition films after 180 min crystallization process with its $I_c$ reduction. Therefore, the formation of the larger size secondary phase and the cracks in the stoichiometric composition YBCO film are one of the reasons for its $I_c$ degradation.

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FIG. 4. Cross-sectional TEM images of 180 min crystallization process. (a) Ba-deficient, (b) stoichiometric composition. Moreover, YBCO c-axis oriented grains are broken around the cracks. These nanostructural changes can be observed only in the stoichiometric composition films after 180 min crystallization process with its $I_c$ reduction. Therefore, the formation of the larger size secondary phase and the cracks in the stoichiometric composition YBCO film are one of the reasons for its $I_c$ degradation.

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References

FIG. 1. Relationship between $I_c$ values of the holding time in crystallization process.

FIG. 2. Cross-sectional TEM images of 100 min crystallization process. (a) Ba-deficient, (b) stoichiometric composition.

FIG. 3. Results of EDS elemental maps. (a) Y, (b) Ba, (c) Cu, (d) F map corresponds to red-dotted line region in Fig. 2 (a). (e) Y, (f) Ba, (g) Cu, (h) F map to red-dotted line in Fig. 2 (b).

FIG. 4. Cross-sectional TEM images of 180 min crystallization process. (a) Ba-deficient, (b) stoichiometric composition.

FIG. 5. Results of EDS elemental maps. (a) Y, (b) Ba, (c) Cu, (d) F map corresponds to red-dotted line region in Fig. 4 (a). (e) Y, (f) Ba, (g) Cu, (h) F map to red-dotted line in Fig. 4 (b).

FIG. 6. 3D reconstructed image of cracks formed in the stoichiometric film after 180 min crystallization process using the FIB-SEM dual beam system.