Study on the formation of YBCO phase and texture transformation of Ag substrate during the heat treatment of TFA-MOD process

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Abstract. The YBCO films were directly deposited on cold-rolled Ag substrates by MOD (metal-organic deposition) method. The formation of YBCO phase and change of Ag textures were studied during firing precursor films. It was found that YBCO phase starts to be formed at 800°C when Ag {110}<110> texture (most suitable texture for YBCO epitaxial growth) appears. With the increase of firing temperature, pure YBCO phase is obtained and Ag {110}<110> texture gradually becomes stronger. The textures of both {110}<110> texture in Ag substrate and biaxial orientation in YBCO film were optimized after holding the film at 900°C for 30 minutes. Finally, the YBCO film deposited directly on cold rolled Ag substrate has a good biaxial texture, a smooth and dense surface as well as high critical current density of 1.5×10^4 A/cm^2. The results show that cold-rolled Ag can be used as a substrate material to directly deposit YBCO film without any buffer layers by MOD method.

1. Introduction

YBCO superconductor is the most potential candidate material for coated conductor applications due to its high irreversibility field and low AC losses [1]. However, high Jc strongly depends on the biaxial textures of the YBCO films [2]. The most essential techniques for obtaining good textures are to epitaxially grow YBCO films on textured substrates with various buffer layers. Although the Jc values exceeding 1MA/cm² are obtained [3], these techniques are not appropriate for industrial applications owing to complicated process and high cost. The {110}<110> textured silver is one of the promising substrate for the coated conductors in view of its chemical compatibility with high-Tc materials [4]. Therefore, it would be economically viable and industrially practical to deposit YBCO films directly on textured Ag tapes without any buffer layers, which has been proved in some reports [5]. MOD has many unique advantages, such as precise controllability of composition and a low cost non-vacuum approach for depositing YBCO films. Besides, the doped additives can be added to the precursor solution in order to improve the YBCO film properties at high field [6]. Long lengths of YBCO tapes with high Jc values have been reported on buffered Ni tapes by this method [7]. In this paper, the YBCO films were directly deposited on cold-rolled Ag substrates by MOD method. To determine whether cold-rolled Ag can be used as a substrate material for the direct
deposition of the textured YBCO film, the formation of YBCO phase and change of Ag textures were studied systematically during firing of the precursor films.

2. Experiment

TFA precursor solution was prepared by dissolving three acetates of Y, Ba and Cu in a cation ratio of 1:2:3 in distilled water with stoichiometric quantity of trifluoroacetic acid (TFA), and the water and acetic acid were removed by an evaporator to yield a blue glassy residue. The coating solution with total metallic concentration of 1.5mol/l was made by dissolving the residue into methanol. The gel solution was coated onto cold-rolled Ag substrates to obtain the gel films by spin-coating method at a speed of 4000 rpm for 2 minute.

The post annealing of the coated films was carried out in two stages. In the first calcination stage, the gel film was decomposed to an amorphous precursor film by slowly heating up to 400°C in a humid oxygen atmosphere. In the second firing stage, the amorphous precursor film was heated up to 900°C in humid argon atmosphere and held for 30 minutes in dry argon. Humid atmosphere used in the heat-treatment was produced by bubbling the inlet gas through an attached reservoir of de-ionized water. After two steps annealing, the samples were slowly cooled down to 500°C. Post-oxygenation was performed at 500°C for 90 minutes, followed by naturally cooling to room temperature.

The YBCO phase purity and textures of both YBCO films and Ag substrates were analyzed by X-ray diffraction, ODF and pole figures. SEM was used to evaluate the surface morphology of films. The critical current density ($J_c$) of the YBCO films was measured using a standard four-probe method at 77K and zero field.

3. Results and discussion

3.1 The formation of YBCO phases during firing precursor films.

Fig. 1 shows X-ray diffraction patterns of films fired at different temperatures. The results show that the films grown at temperature range between 400°C-600°C contain mainly oxides, fluoride and oxy-fluorid that is produced from the decomposition of the trifluoroacetate salts such as BaF$_2$. Their diffraction peaks are wide and weak, indicating that these phases are amorphous or weakly crystallized after the heat-treatment at low temperatures. With the increase of the annealing temperature to 700°C, the diffraction peaks of Y$_2$Cu$_2$O$_5$ phase are found, which illustrates that oxide, fluoride and oxy-fluorid may have been reacted each other to form other intermediate phases. When the annealing temperature is increased to 800°C, YBCO phase starts to be formed, meanwhile there are still some impurities such as BaF$_2$ to be maintained in the films. When the firing temperature reaches 900°C, BaF$_2$ phase completely disappears, and the pure YBCO film with both (00L) peaks (the YBCO grains with c-axis orientation) and (103) peak (the YBCO grains with a-axis orientation) is obtained. Pure c-axis aligned YBCO film without a-axis orientation can be obtained after holding the sample at 900°C for a longer time of 30 minutes (see Figure 1).

Figure 1. XRD results of YBCO films fired at different temperatures

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3.2 The transformation of Ag textures during firing precursor films

In order to study texture changes of cold-rolled Ag substrates, the ODFs of Ag substrates at different temperatures of firing stage were calculated through the data of three measured pole figures, and the result is shown in Fig.2. It was found that the textures of Ag substrates are strongly random distributed when annealing temperatures are lower than 800°C. These orientations consist of \{023\}<011> (\(\phi_1=90^\circ\), \(\varphi=35^\circ\), \(55^\circ\), \(\varphi_2=0^\circ\)), \{013\}<893> (\(\phi_1=50^\circ\), \(\varphi=18^\circ\), \(72^\circ\), \(\varphi_2=0^\circ\)) and \{011\}<811> (\(\phi_1=10^\circ\), \(\varphi=45^\circ\), \(\varphi_2=0^\circ\)). When the temperature is higher than 800°C, the strongest intensity is concentrated on the \{023\}<110> orientation (\(\phi_1=90^\circ\), \(\varphi=35^\circ\), \(55^\circ\), \(\varphi_2=0^\circ\)), which is found to be gradually transformed into \{110\}<110> orientation (a most suitable texture for YBCO epitaxial growth) when the firing temperature is further increased. The single \{110\}<110> orientation is formed when the firing temperature reaches 900°C. The maximum ODF value of Ag substrates reaches 20 after holding YBCO films at 900°C for 30 minutes. The observed result by ODF is consistent with XRD result shown in Fig.1. The diffraction peaks of [111]_Ag, [200]_Ag and [220]_Ag can be detected when the firing temperature is lower than 800°C, indicating no preferred orientations are formed at lower temperatures. The (111) Ag and (200) Ag diffraction peaks become rapidly disappeared after annealing above 800°C, meanwhile (220) Ag diffraction peak, which is a typical peak of Ag \{110\} texture, is dominant. The intensity of Ag \{110\} texture reaches the maximum value when holding YBCO films at 900°C for 30 minutes.

![Figure 2. ODF results of Ag substrates annealed at different temperatures (\(\varphi_2=0^\circ\))](image)

3.3 The properties of YBCO films

In order to determine whether the cold-rolled Ag can be used as a substrate for the direct deposition of the YBCO film, the quality of the texture of YBCO film are analyzed. Fig. 3 (a) and (b) show the (006) and (113) pole figures of a YBCO film on cold-rolled Ag substrate, respectively. One dense area in the (006) pole figure and four dense areas in the (113) pole figure show a good out-plane and in-plane texture of the YBCO film, respectively. The YBCO film is biaxially textured and growing epitaxially on the Ag substrate with \{110\}<110> texture. The epitaxial relationship between the YBCO film and the Ag substrate is \(\langle 100 \rangle_{\text{YBCO}} \parallel \langle 110 \rangle_{\text{Ag}}\) direction, and \{001\}_\text{YBCO} \parallel \{110\}_\text{Ag} plane. Fig. 3 (c) presents the SEM image of the YBCO film on the cold-rolled Ag substrate. The film has a dense and flat surface, as well as a good connectivity. The thin disc-shape particles are observed without rectangular shape of a-axis crystals [8][9], which also indicates a better c-axis alignment of...
YBCO film. The as obtained film has a $J_c$ value of $1.5 \times 10^4 \text{A/cm}^2$ at 77K and zero magnetic field, which is much lower than that of films produced on either LaAlO3 or SrTiO3 single crystals by same method. However, this value compares favourably with the $J_c$ values obtained for YBCO films on polycrystalline Ag substrates prepared by other chemical methods [10]. The YBCO films with better superconductivity could be expected by further optimization of the deposition conditions.

![Diagram](image)

Figure 3. (006) pole figure (a); (113) pole figure (b) and SEM image (c) of YBCO film deposited on cold-rolled Ag substrate.

4. Conclusion
The YBCO films were directly deposited on cold-rolled Ag substrates by MOD method. It was found that both YBCO phase and Ag $\{110\}<110>$ texture tended to be formed at the same temperature of 800°C. With the increase of the firing temperature, the impure phases in precursor films disappeared and Ag $\{110\}<110>$ texture gradually became stronger. The textures of both $\{110\}<110>$ texture in Ag substrate and biaxial orientation in YBCO film were optimized after holding the film at 900°C for 30 minutes. Finally, YBCO film with a strong biaxial texture was obtained, which has a smooth and dense surface, a transport $J_c$ value being $1.5 \times 10^4 \text{A/cm}^2$. The results showed that cold-rolled Ag could be used as a substrate material to directly deposit YBCO film without any buffer layers by MOD method.

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