The Improvement of Utilization Ratio of Metal Organic Sources for the Low Cost Preparation of MOCVD-synthesized YBCO Films based on a Self-heating Technology

Ruipeng Zhao¹, Qing Liu¹, Yudong Xia², Bowan Tao¹ and Yanrong Li¹

¹State Key Lab of Electronic Thin Films and Integrated Devices, University of Electronic Science and Technology of China, Chengdu 610054, PR China
²School of Physical Science and Technology, Southwest Jiaotong University, Chengdu 610031, PR China
Email: zhaoruipeng1990@sina.com

Abstract: We have successfully applied metal organic chemical vapor deposition (MOCVD) to synthesize biaxially textured YBa₂Cu₃O₇₋δ (YBCO) superconducting films on the templates of LaMnO₃/epitaxial MgO/IBAD-MgO/solution deposition planarization (SDP) Y₂O₃/Hastelloy tape. The YBCO films have obtained dense and smooth surface with good structure and performance. A new self-heating method, which replaced the conventional heating-wire radiation heating method, has been used to heat the Hastelloy metal tapes by us. Compared with the heating-wire radiation heating method, the self-heating method shows higher energy efficiency and lower power consumption, which has good advantage to simplify the structure of the MOCVD system. Meanwhile, the utilization ratio of metal organic sources can be increased from 6% to 20% through adopting the new self-heating method. Then the preparation cost of the YBCO films can be also greatly reduced.

1. Introduction
In the past few years, REBa₂Cu₃O₇₋δ ((REBCO), RE= rare earth elements) high temperature superconducting tapes have been amply researched by many research teams for its application in electric power field [1-3]. Because of its high current carrying capacity and large irreversible field, REBCO high temperature superconducting tapes have great potential for application in the aspects of superconducting cable, strong magnet, superconducting motor, generator, superconducting current limiter and so on [4-6]. Metal organic chemical vapor deposition (MOCVD) has been proved as one of the most promising techniques to massively prepare biaxially textured REBCO superconducting thin films on the surface of biaxially textured substrates. However, the low utilization ratio of metal organic sources makes it difficult to reduce the fabrication cost of REBCO films. Therefore, a new self-heating method was used to prepare the MOCVD-synthesized YBCO superconducting films to improve the utilization ratio of metal organic sources and reduce the cost. For the new self-heating method, the current is introduced to the metal tapes through the brush [7], as is shown in figure 1.

Nowadays, the research and preparation of the buffer template, which is used to prepare the YBCO superconducting films, is mainly based on the ion beam assisted deposition (IBAD) [8] and rolling assisted biaxially textured substrates (RABiTS) [9] technology. Compared to RABiTS, the IBAD process has the better advantage in the cost and product performance. Therefore, the IBAD technology is widely used in the world. In our experiments, the templates of LaMnO₃/epitaxial MgO/IBAD-MgO/solution deposition planarization (SDP) Y₂O₃/Hastelloy tape are adopted to prepare YBCO films,
as is shown in figure 2. In this paper, we focus on the development of the heating technology to improve the utilization ratio of metal organic sources. What’s more, the structure, morphology and performance of the prepared YBCO films based on the novel heating method have been also investigated.

**Figure 1.** The schematic diagram of the self-heating method for the preparation of YBCO films

**Figure 2.** The schematic diagram of the buffer layers on the Hastelloy tape

2. **Experimental**

In the experiments, the precursors of metal organic sources were prepared by dissolving Y(tmhd)₃, Ba(tmhd)₂·(1,10-phenanthroline)₂ and Cu(tmhd)₂ (tmhd: 2, 2, 6, 6-tetramethyl-3, 5-heptanedionate) in tetrahydrofuran (THF). The tapes of LaMnO₃/epitaxial MgO/IBAD-MgO/solution deposition planarization (SDP) Y₂O₃/Hastelloy tape, with flat surface and good structure, were used as the templates (LMO template). The YBCO films were quickly prepared on the LMO template by metal organic chemical vapor deposition (MOCVD). Firstly, the prepared precursors were quickly atomized by a home-made nozzle and evaporated rapidly by the heating evaporator. Then, the vapors of metal organic sources, which were mixed with the argon, oxygen and nitrous oxide, were transported into the reaction chamber and reacted to grow YBCO films on the surface of the heating tapes. At last, the prepared YBCO superconducting tapes were annealed in flowing oxygen at 500 °C and furnace cooled to room temperature.

X-ray diffraction (XRD, Bede D1 system) patterns were measured using Cu Kα radiation (λ=1.5406 Å) in the mode of θ-2θ step-scan, ω-scan, φ-scan and Chi-scan for the prepared samples. The RMS roughness of LMO template layer was characterized by atomic force microscope (AFM, Seiko SPA300HV). The surface morphology was characterized by a scanning electron microscope (SEM, JEOL7500F). And the composition of thin films was characterized by an energy dispersive...
spectrometer (EDS, Oxford INCA). The thickness was measured by a step profiler (Veeco Dektak 150). Moreover, the critical current density ($J_c$) and critical current ($I_c$) at 77 K and 0 T were obtained by the Leipzig $J_c$-scan system and four-probe-method measurement system.

3. Results and Discussion

The used LMO template is characterized by the AFM and SEM system. Figure 3a shows the AFM image of LaMnO$_3$ template for the preparation of YBCO superconducting film. The root-mean-square (RMS) roughness of $5 \times 5 \mu m^2$ is 2nm, which meets the demand of the preparation of biaxially textured YBCO films. Figure 3b is the SEM image of LaMnO$_3$ film morphology, which shows that its surface is very dense and smooth. Therefore, the used LMO templates are very suitable for the growth of the YBCO superconducting thin film.

As we adopt the new self-heating method, it avoids the problem that the temperature of the shower is too high due to using the radiation heating methods. So the shower head can be very close to the surface of template tapes. Thus, the YBCO films prepared by the heating-wire radiation heating method and the self-heating method respectively are characterized by the XRD $\theta$-2$\theta$ step-scan, as is shown in figure 4. The diffraction peaks of YBCO (00l) can be clearly observed in the curves of $\theta$-2$\theta$ patterns, which indicate that the grains of YBCO films are purely $c$-axis-oriented growth. However, the intensity of YBCO (00l) diffraction peaks of YBCO film based on the self-heating method is obviously stronger than that of the radiation heating method. It indicates that the YBCO films prepared by the self-heating method crystallize better. Moreover, the XRD $\chi$-scan of YBCO (102) has also been performed, which is used to characterize the $c$-axis-oriented grains and the $a$-axis-oriented grains. For the two $\chi$-scan curves, there is only one peak around 56.6°, representing the diffraction of (102) plane of $c$-axis oriented YBCO grains, which indicate that all the prepared YBCO grains aligned their $c$-axis perpendicular to the surface of the LMO templates.
Figure 4. (a) The XRD $\theta$-2$\theta$ scanning patterns of YBCO films based on the different heating methods; (b) The XRD $\chi$-scan curves of YBCO (102) based on the different heating methods.

What’s more, the XRD $\omega$-scan of YBCO (005) and the XRD $\varphi$-scan of YBCO (103) have also been performed, which are used to characterize out-of-plane and in-plane textures of YBCO films. Based the radiation heating method, the rocking curve of YBCO film obtained through YBCO (005) ($\Delta\omega$) yields the FWHM value of 1.35° and the $\varphi$-scan curve obtained through YBCO (103) ($\Delta\varphi$) yields the FWHM value of 3.18° in figure 5a. Similarly, for the self-heating method, the rocking curve of YBCO film obtained through YBCO (005) ($\Delta\omega$) yields the FWHM value of 1.27° and the $\varphi$-scan curve obtained through YBCO (103) ($\Delta\varphi$) yields the FWHM value of 2.8° in figure 5b. It can be seen that the FWHM values of YBCO films prepared based on the novel self-heating method is even smaller than that of YBCO films prepared based on the radiation heating method, which also indicates that the biaxial texture can be better improved through adopting the self-heating technology.

Figure 5. (a) The XRD $\omega$-scan and $\varphi$-scan curves YBCO films based on the radiation heating method. (b) The XRD $\omega$-scan and $\varphi$-scan curves YBCO films based on the self-heating method.

The surface morphologies of the superconducting films are observed by the SEM system. Figure 6 shows the morphology images of these prepared samples based on the different heating method. There are some larger outgrowths formed on the surface of YBCO film prepared by the radiation heating method. In comparison, the surface of YBCO film prepared by the self-heating method is flat and there are not basically large outgrowths. For these samples prepared by the different heating
method, the critical current density ($J_c$) at 77K and 0T are measured by the induction and four-probe methods and the corresponding measured values are shown in Table 1. Owing to the narrow distribution of grain alignments, the $J_c$ of the sample prepared based on the self-heating method is 5 MA/cm$^2$ (0 T, 77 K), which is higher than the $J_c$ of 3.6 MA/cm$^2$ based on the radiation heating method. Meanwhile, depending on the amount of metal organic sources and the thickness of the corresponding YBCO films, the utilization ratio of metal organic sources can be increased from 6% to 20% through adopting the new self-heating method. And the deposition rate of YBCO films can be greatly increased.

![Figure 6.](image)

(a) The SEM image of YBCO film based on the radiation heating method. (b) The SEM image of YBCO film based on the self-heating method

| Heating method    | Deposition rate (nm/min) | Utilization ratio | Critical current density (MA.cm$^2$) |
|-------------------|--------------------------|-------------------|-------------------------------------|
| Radiation heating | 300                      | ~6%               | 3.6                                 |
| Self-heating      | >600                     | ~20%              | 5.0                                 |

4. Conclusion
In this study, a novel self-heating method is used to prepare YBCO films by us. Different from the conventional radiation heating method, the shower head can be very close to the metal tapes without excessive temperature. Therefore, the utilization ratio of metal organic sources can be greatly improved. It is concluded that the YBCO film with high $J_c$ at a high metal organic sources utilization ratio of 20% could been successfully prepared.

5. References
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