Optimization of YbBa$_2$Cu$_3$O$_y$ Thick Film Formation on MgO Substrates using Yb$_2$BaO$_5$ and liquid phase

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Abstract. The production of best thick YbBa$_2$Cu$_3$O$_y$ (Yb123) films on MgO substrates are crucial for the utilization of this technology in silver sheated wire as similar to Bi-2223. In this work, in order to optimize YbBa$_2$Cu$_3$O$_y$ films on MgO substrates, a mixture of alpha-terpineol and 2-ethyl acetate was utilized as a solvent to which 100 nm sized stoichiometric powder mixture of Yb$_2$BaO$_5$, BaCuO$_2$ and CuO was added and grinded for several hours to form a highly dense paste. Thick film precursors were prepared by spreading the paste over MgO substrates with a screen printing technique and various sintered and quench studies are done. Growth temperature is systematically varied between 800 and 950 °C for 30 minutes in air and quenched in liquid nitrogen. Similarly, sintering durations are varied systematically from 15 minutes to 8 hours. XRD reveals conformed that large Yb123 grains are formed when the thick films are sintered at 825 °C for 1 hour on MgO substrate. SEM micrographs are also in congruence with the XRD results depicting the giant Yb123 crystals. The present results clearly demonstrate that optimization of the growth temperature and time is crucial to obtain the large thick YbBa$_2$Cu$_3$O$_y$ films on MgO substrates.

1. Introduction

In the current day trend, most of the research carried on High $T_c$ superconductor technology is focused on developing high performance, good quality cheaper superconducting tapes and power cables operating at 77 K [1]. Since the first generation Bi-based silver coated cables and tapes, the REBa$_2$Cu$_3$O$_y$ (RE – rare earth element) type coated conductors represent the second generation superconductors [2,3]. Few challenges in thin films along sides production cost is the alignment of grains and grain connectivity issues. But these can be addressed with high temperature biaxial texturing which gives alignment and improves grain connectivity. But, practical applications also include thick films, where the impact of biaxial texturing decreases with increase in thickness. Our aim is to process YbBa$_2$Cu$_3$O$_y$ coated conductors cheaply, for commercial applications while countering the difficulties. In past few years, we have fabricated good quality RE-123 (RE – Gd, Er) thick films on MgO and Ag substrates using RE-210, BaCuO$_2$ and CuO mixture [4,5]. The Ba-Cu-O precursor in the mixture is a low melting compound (900 °C), which improves the density as well as quality of the thick film and quench studies pointed out that the corresponding 123 phase can be synthesized below silver’s melting point [4]. Also, since the melting point is below Ag, it is easy to use for applications alongside with Ag. Because of this low
melting nature, it is worth studying the properties of Yb-123, which is a low melting RE-123 material as this promotes the application based attributes.

In our present work, we processed Yb-123 thick film on MgO substrate and characterized it after quenching. We have optimized the temperature, dwelling time and other parameter for good growth of Yb-123 thick film. XRD, DTA and SEM analysis have been done to explain the trend and other properties of thick films synthesized.

2. Experimental details
Homemade Yb$_2$BaO$_5$ powders were made by using the solid state reaction technique and then mixed with high purity commercial powders of BaCuO$_2$ and CuO in stoichiometric propositions to obtain of YbBa$_2$Cu$_3$O$_y$. Later this mixture is ball milled for 2 hours to promote the number of nanoscale particles (around 100 nm) [4]. These powders were air dried for 24 hours, then vacuum dried at 200 °C for 6 hours, and finally cooled to room temperature. A mixture of alpha-terpinol and 2-ethyl acetate was used as a solvent to which the ball milled powder mixture was added and grinded for several hours to form a highly dense paste. Thick films (about 0.5 mm thick) were then fabricated on commercial MgO single crystal substrates using screen printing technique, where the dense paste is spread on the substrate manually using printing head through a 165-mesh screen.

Two types of quench studies were done to optimize the temperature as well as dwelling time. In the first experiment, to optimize the temperature profile, the films were heated to and maintained at particular growth temperatures, which are varied from 800 to 950 °C in a box furnace. After attaining the required temperatures, the samples were held at that temperature for 30 minutes and then rapidly quenched in liquid nitrogen (77 K). Subsequently, the samples were cleaned in an ultrasonic cleaner and dried in a drying box, to be ready for characterization. In the second experiment, similar quenching studies are done to optimize the dwelling time required for maximum Yb123 growth. The structure of the samples was determined by means of a high-resolution automated X-ray powder diffractometer RINT2200 with a step size of 0.01° from 5° to 90°, using Cu-Kα radiation generated at 40 kV and 30 mA. The microstructure of these films was studied with a scanning electron microscope (SEM).

3. Results and discussion
3.1) Quench studies for temperature optimization to obtain good quality Yb-123 thick films on MgO substrate:

After depositing the mixture paste and preparing the thick film samples using screen-printing, they are subjected to the specific heat cycle as mentioned in experimental section. The growth temperatures are systematically varied such as 800, 825, 850, 875, 900, 925 and 950 °C (see figure 1-(a)).
The thick films are characterized using XRD for identifying the phases formed. From the XRD results, it can be seen that 825 °C is the best temperature condition for optimum Yb123 growth. At high temperatures, the dominant phase is Yb211 as well as liquid phase as we can observe the intensity of Yb123 dropping as well as disappearing (at 950 °C), while the peak intensity of Yb211 and liquid phase starts increasing (see figure 2), maximum at 950 °C.

Microstructural studies from FESEM showed that large crystals of Yb123 are formed at 825 °C which is in accordance with our XRD results. When the temperature is 800 °C small grains of Yb123 are formed (see figure 3-left) and large crystals are formed at the temperature 825 °C (see figure 3-middle). While at high temperatures such as 950 °C Yb211 particles can be seen minor amount of Yb123 (see figure 3-right) which can also be observed from XRD. Hence, when the growth temperature is around 825 °C the growth of crystalline Yb123 is optimum in Yb-123 thick films grown on MgO substrates.

3.2) Quench studies for dwell time optimization to obtain good quality Yb-123 thick films on MgO substrate:
It is also equally important to optimize the dwelling time or soaking time for optimum for large crystal growth. In order to optimize the dwelling time, we fabricated Yb-123 thick films using 825 °C as growth temperature and vary the sintering duration or dwell time. In this study, we systematically varied the dwell time such as 0.25, 0.5, 1, 2, 4 and 8 hours (see figure 1-(b)). XRD analysis reveals that, highest Yb123 is formed when the dwelling time is 1 hour as the peak intensity corresponding to the Yb123 is
highest of all the other samples (see figure 4). It can be seen that the Yb123 peak intensity reduces with increase in sintering duration after 1 hour, while the Yb211 peak intensity increases with sintering duration.

![XRD of thick film sample sintered at various durations](image)

**Figure 4.** XRD of thick film sample sintered at various durations.

![Low magnification SEM images of thick film samples sintered at various sintering durations](image)

**Figure 5.** Low magnification SEM images of thick film samples sintered at various sintering durations; left-15 minutes, middle- 1 hour and right-8 hours.

Microstructural studies revealed that large and long crystal of Yb123 are observed in thick film, which was subjected to 1-hour sintering duration (see figure 5). Hence we can get obtain huge Yb123 crystals in thin films when the optimum sintering conditions such as 825 °C and 1 hour dwelling time are followed.

**4. Conclusions**

In this study, we have successfully optimized the sintering conditions required to fabricate the Yb123 large crystal thick films on MgO substrate. From XRD and SEM microstructural studies, we found that when the films are sintered at 825 °C for 1-hour optimum content of large Yb123 crystals are formed. The present results indicate that stoichiometric powder mixture of Yb$_2$BaO$_5$, BaCuO$_2$ and CuO can help to prepare the large–area thicker Yb-123 films in a short processing time.
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References

[1] Dusastre V, Larbalestier D, Gurevich A, Feldmann D M and Polyanskii A 2001 High-Tc superconducting materials for electric power applications Nature 414 368–77
[2] Haugan T, Barnes P N, Wheeler R, Meisenkothen F and Sumption M 2004 Addition of nanoparticle dispersions to enhance flux pinning of the YBa2Cu3O7-x superconductor Nature 430 867–70
[3] Chen Z, Kametani F, Chen Y, Xie Y, Selvamanickam V and Larbalestier D C 2009 A high critical current density MOCVD coated conductor with strong vortex pinning centers suitable for very high field use Supercond. Sci. Technol. 22
[4] Muralidhar M, Sakai N, Jirsa M and Tanaka S 2009 Solid phase (RE 2BaO 4)-Liquid phase (BaCuO 2) reaction: The way to highly oriented ErBa 2Cu 3O y superconducting thick films on commercial silver substrates Cryst. Growth Des. 9 2404–8
[5] Muralidhar M, Sakai N, Murakami M, Hirabayashi I and Tanaka S 2006 RE-Ba2Cu3Oy formation mechanism via (RE)2BaO4 Phys. C Supercond. its Appl. 445–448 282–5