# Texture and self-biased property of an oriented M-type barium ferrite thick film by tape casting*

Chen Da-Ming(陈大明), Liu Ying-Li(刘颖力)†, Li Yuan-Xun(李元勋), Yang Kai(杨 楷), and Zhang Huai-Wu(张怀武)

*State Key Laboratory of Electronic Films and Integrated Devices, University of Electronic Science and Technology of China, Chengdu* 610054, *China*

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In this paper, the oriented M-type barium ferrite (BaM) thick films with different thicknesses are prepared by tape casting. It is found that the crystallographic alignment degree ($f$), the pore and the squareness ratio ($M_r/M_s$) are not affected by the thickness of the film. XRD and SEM results show that the thick film has hexagonal morphology with a crystal texture of $c$-axis grains perpendicular to film plane. The hysteresis curve indicates that the BaM thick film exhibits a self-biased property with a remanent magnetization of 3.30 T, a squareness ratio ($M_r/M_s$) of 0.81, and a coercivity of 0.40 T. The results show that the BaM thick film has potential for use in self-biasing microwave devices, and also proves that the tape casting technique is capable of fabricating high-quality barium ferrite films, thus providing a unique opportunity to realize the large area production of thick film.

**Keywords:** barium ferrite, magnetic material, thick films

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## 1. Introduction

With the rapid development of communication systems, such as mobiles and satellites, the monolithic microwave integrated circuits (MMIC) require that the microwave magnetic devices (circulator and phase shifter) will be planar and self-biased in future. M-type barium ferrite (BaM) with a crystal structure of the so-called magnetoplumbite structure that can be described as RSR*S*,[1] is widely utilized in microwave devices due to its large saturation magnetization, low conductivity and high uniaxial magnetic anisotropy field.[2,3] In the past, BaM magnet bulk was used to provide an external biasing magnetic field to control microwave propagation in microwave magnetic devices,[4,5] so it is disadvantageous in the sense of planarizartion and miniaturization of the microwave devices. The tendency today is to replace the magnet bulk with thick and thin films integrated into substrate to reduce the size and weight.[6,7] Hence, a high-quality BaM film with self-biasing (high remanence, $M_r$) is needed to be developed so that it can eliminate the requirement for an external biasing magnetic field provided by magnet bulk.

In recent years, many deposition methods, for example, pulsed laser deposition (PLD)[8] and liquid phase epitaxy (LPE)[9] have been used to try to grow BaM films on semiconductor substrates with thicknesses ranging from 1 μm to 100 μm. Although thin films with a high crystallographic alignment degree and low microwave loss can be deposited on substrates successfully by PLD and LPE, some problems still exist in processing. For example, the thickness of the PLD film is too thin for practical applications, while the large-area LPE films are difficult and expensive to produce.[10]

Tape casting is a well-established technique widely used in large-scale production of ferroelectric thick films and substrates.[11,12] However, this technology is not used to fabricate oriented ferrite thick films. In this work, we present detailed procedures to fabricate the textured BaM thick film with a high crystallographic alignment degree.

## 2. Experiment

The M-type barium ferrite ($BaFe_{12}O_{19}$) powder was prepared using the traditional solid-state method.

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†Corresponding author. E-mail: lyl@uestc.edu.cn

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The raw materials, $BaCO_3$ and $Fe_2O_3$ were mixed by the molar ratio and ball-milled for 12 h, then dried and sintered at 1200 °C in air for 2 h. For preparing BaM thick film, powder, solvent, dispersing agent and binder were mixed together in certain proportions, then ball milled for 24 h to form slurry. Subsequently the slurry was casted by the tape casting facility. The thickness of film was controlled by a doctor blade. Before sintering, the film was subjected to a magnetic field of 1 T to align. Then the film was dried at 60 °C for 15 min, and removed off the PET film. Several films were under isostatic pressing to obtain thick films with different thicknesses. Finally, the thick film was sintered in air at different temperatures for 4 h.

Crystal structures of the sintered BaM thick films were measured by an X-ray diffractometer (XRD, Bede TM 2000) with a Cu K$\alpha$ radiation). Morphologies and chemical analyses of the sintered ferrite thick films were investigated by SEM–EDAX (scanning electron microscopy–energy dispersive X-ray spectra, Quanta 200, Philips, Netherland). The $M$–$H$ loops of the sintered ferrite films were measured by a vibration sample magnetometer (VSM), with the applied field varied in a range from –15 T to +1.5 T.

## 3. Results and discussion

A typical XRD patten of BaM is shown in Fig. 1. It is clear that single BaM phases are visible, no other phases are detected (Compared with JCPDS card NO.07-0276 and the data in Ref. [13]). The diffraction peaks of (008), (0014), and (006) are stronger than others, indicating that the sample has a crystal texture of $c$-axis grains perpendicular to the film plane, which is further proved by the SEM results as shown in Fig. 2. Since the crystalline orientation of specimens has an evident effect on the squareness ratio and microwave loss (ferromagnetic resonance linewides),[7,14] it is necessary to calculate the degree of the crystallographic alignment in the sintered BaM. According to the Lotgering way,[15] the crystallographic alignment degree ($f$) can be quantitatively expressed as

$$f = (P - P_0)/(1 - P_0), \tag{1}$$

where $P$ denotes the ratio between the total intensities of the ($00l$) and ($hkl$) reflections of the oriented sample, while the $P_0$ represents that of the non-oriented sample whose diffraction intensities were obtained from the randomly oriented powder. Based on Eq. (1), the crystallographic alignment degree of the ($00l$) plane in our oriented specimen can be calculated to be 0.75, illustrating that most of the BaM crystallites were aligned up by the magnetic field. Though this crystalline orientation is below the results of the quasi-single crystal bulk sample,[16] it is stronger than those of the samples obtained by the polymer network-assisted alignment method.[17] In addition, it has been proved that the microwave loss is directly proportional to the degree of orientation,[14] hence our sample must have low microwave loss due to the high degree of orientation. In addition, it was also observed that the crystallographic alignment degree was not affected by the thickness of film, as shown in Fig. 3.

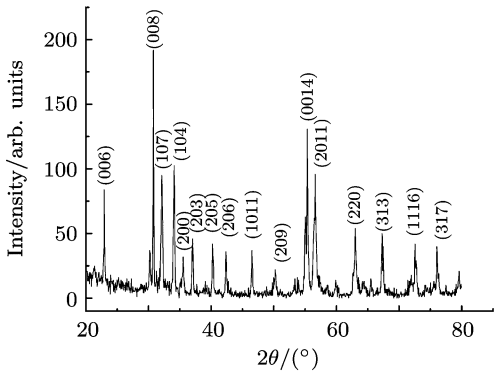



**Fig. 1.** XRD patten of the BaM thick film.

Figure 2 shows the SEM images of the BaM thick film of surface (a), cross sections (b) at 1000 °C, (c) 1200 °C with 50 μm in thickness and (d) with 400 μm in thickness, respectively. The grain size is believed to be a key factor in crystalline orientation, and the size is smaller than or close to single domain size (0.5 μm–1 μm), good for alignment and in turn for high remanence.[16] In our specimen, the average grain size is about 1 μm, implying that it is good for our sample to obtain a high crystalline orientation and high remanence, which are confirmed by the results plotted in Figs. 1 and 4. Furthermore, it can be seen that the crystal texture of $c$-axis grains are normal, to thick film planes with a hexagonal shape as confirmed by Figs. 1 and 2. Unfortunately, some pores were observed in the sample, and it has been proved that the pores give rise to an enhancement of microwave loss,[7] so it is important to reduce the pore number. However, the crystalline grains become bigger and the number and size of pores is greatly reduced with the increase in sintered temperature. On the other hand, it is worth noting that the crystalline grain and pore are not improved by the thickening of the film, compared Fig. 2(c) with Fig. 2(d).

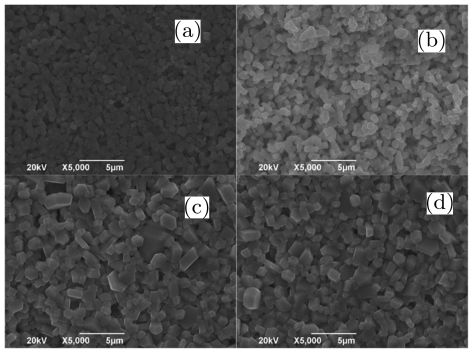



**Fig. 2.** SEM images of the BaM thick film of surface (a), cross section (b) in 1000 °C and (c) 1200 °C with 50 μm, and (d) with 400 μm, respectively.

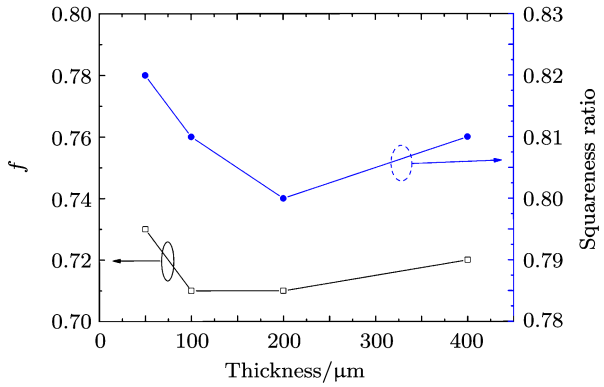



**Fig. 3.** Crystallographic alignment degree ($f$) and squareness ratio versus thickness of film.

The variation of squareness ratio with the thickness of film is shown in Fig. 3. It can be seen that the variation is similar to that of crystallographic alignment degree, so it is reasonable that the crystallographic alignment degree is sensitive to the squareness ratio. In order to further learn the effect of texture on thick film, hysteresis curves of the BaM thick film are given in Fig. 4, measured with the external magnetic field perpendicular to plane (PTP) and in plane (IP) respectively. It is interesting to note that in the cases of PTP and IP, the hysteresis loops are quite different. The sample is easily saturated in the former case, whereas it is not even saturated at the external field of 1.5 T in the later one. Hence, it is reasonable to deduce that the specimen exhibits the magnetocrystalline anisotropy property, which is attributed to the fact that the thick film has a crystal texture of $c$-axis grains perpendicular to the film plane. From the hysteresis loops, it can be seen that the thick film has a remanent magnetization ($M_r$) of 3.30 T, a squareness ratio ($M_r/M_s$) of 0.8 and a coercivity of 0.40 T in the case of PTP. This result is of great significance, and it illustrates that 80% of the saturation magnetization remains when the external magnetic field is removed, which indicates that this thick film exhibits self-biased properties. All of these results enable this thick film to be used in self-biased microwave devices.

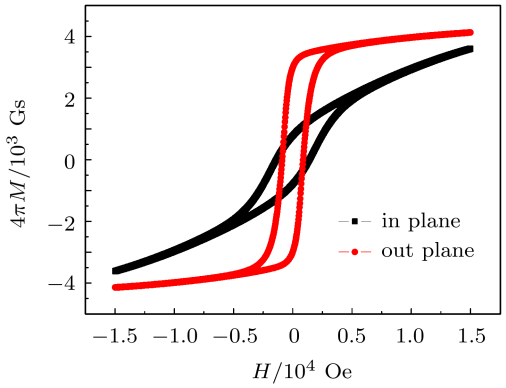



**Fig. 4.** Hysteresis curves of the BaM thick film, 1 Oe = 79.5775 A/m, 1 Gs = $10^{-4}$ T.

## 4. Conclusions

Oriented BaM thick films with a high crystallographic alignment degree are prepared by tape casting. The specimen exhibits self-biased properties with a remanent magnetization ($M_r$) of 3.30 T, a squareness ratio of 0.8, and a coercive field of 0.40 T. Though microwave loss cannot be directly evaluated, it is reasonable to deduce that the thick film possesses a low microwave loss due to the high crystallographic alignment degree and less pores in the sample. All of these results enable the thick film to have potential for use in self-biasing microwave devices. Anyway, in this paper it is proved that the tape casting technique provides an ability to fabricate oriented BaM thick film with self-biased property.

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