SYNTHESIS AND CHARACTERIZATION OF $\text{BaFe}_{12}\text{O}_{19}$ THIN FILMS USING SUSPENSION OF NANO POWDERS

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BaM thin films have been synthesized by dispersing the dried gel nano powders prepared by Sol-Gel method. The solution was made by dissolving iron nitrate $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, barium nitrate $\text{Ba}(\text{NO}_3)_2$ and citric acid in deionized water and methanol. This sol was slowly evaporated until a dried gel was formed. This dried gel was then added to ethylene glycol. The final solution was vigorously shaken and mixed in ultrasonic cleaner for 30 min to disperse particles sufficiently. Then the prepared solution spin coated on Si(110) substrate. The obtained thin films were dried at 120 °C and then calcined at 900 °C for 1 h. The films were characterized using X-ray diffraction (XRD) and vibrating sample magnetometer (VSM).

Keywords: barium hexaferrite, thin films, nanoparticles.

1. Introduction

Barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$, BaM in short) is a hexagonal ferrimagnetic ceramic with hard magnetic properties. Thin films of barium ferrite have been recognized as one of the appealing candidates in the fabrication of low-noise high-density recording media due to their high coercivity H_c , high saturation magnetization M_s , high degree of crystalline anisotropy and excellent chemical stability.¹⁻⁴ In magnetic recording media, decreasing the crystallite size has always been a path towards increasing the recording density.²⁻⁴ Furthermore to develop the thin films for high density magnetic recording media, the medium must have coercivity above 3.0 kOe in order to record bits with narrow transition length and a moderate remanent magnetization M_r to achieve relatively high signal to noise ratio.^{5,6} The higher coercivity can also be

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attributed to the reduced grain size in thin film structure. Accordingly, the major objective of this research is to prepare nanostructure barium hexaferrite thin films. In this work, we present a procedure to prepare good quality and easily reproducible nanostructure barium ferrite thin films. This route uses a suspension of the dried gel nano powders synthesized by sol-gel method.

2. Experimental Procedure

A suspension of the dried gel powders was prepared to synthesis BaM thin films. This powders were made by Sol-Gel method. The sol was prepared by dissolving iron nitrate (Fe(NO₃)₃·9H₂O), barium nitrate (Ba(NO₃)₂) and citric acid (C₆H₈O₇) in distilled water while stirring at 80°C. In this solution Fe/Ba ratio was 10 and the pH was also adjusted to 2 using ammonia (25% solution) NH₃·H₂O. Sol was slowly evaporated until an amorphous powder (Gel) was formed. An amount of powders were calcined at 900 °C and the remained powders were then added to ethylene glycol. The final solution was vigorously shaken and mixed in ultrasonic cleaner for 30 min to disperse particles sufficiently.

The prepared suspension was spin-coated on the Si (110) single crystalline substrates which had been cleaned by distilled water and acetone. The spin speed was 2000 rpm and the spin period was 20 s. The obtained samples were dried at 120 °C for 24 h and then calcined at 900 °C for 1 h in air.

The structure of the calcined barium hexaferrite thin films was characterized by X-ray diffraction (XRD) with Cu K α radiation. XRD measurements were also used for determination of the average crystallite size of the ferrites. The crystallite size was determined using Scherrer equation by applying the full-width at half-maximum intensity of the main diffraction peaks. Magnetic properties were also measured using a vibrating sample magnetometer (VSM) at a maximum applied field of 10 kOe.

3. Results and Discussions

Fig. 1 shows the XRD patterns of the dried gel and the calcined powders at 900 °C. As can be seen the dried gel is amorphous but after calcination at 900 °C the BaFe₁₂O₁₉ phase was crystallized. However, some characteristic reflections corresponding to the BaFe₂O₄ and α -Fe₂O₃ phases can also be observed in the calcined powders at 900 °C. It shows that although the ferrite phase was formed at 900 °C predominantly but it seems that this temperature is not still sufficient to make the single phase hexaferrite. The crystallite size of the Ba-hexaferrite phase is about 20 nm.

The XRD pattern of the thin film obtained from a suspension of the dried gel and then calcined at 900 °C is also presented in Fig. 2. Compare to Fig. 1, in this figure only a small traces of α -Fe₂O₃ and BaFe₂O₄ could be observed which shows more complete formation of Ba-hexaferrite. It seems that making a second gel from suspension of the first gel dried powder prepared the better condition for more efficient calcination. The

crystallite size of BaM phase in this sample is also about 25 nm (Table. 1), which satisfy the requirements for high-density magnetic recording media.⁶ This size is smaller than the size obtained for the sample with Fe/Ba=10 in the presence of ammonia calcined at 900 °C which has been reported in previous works.³

Fig. 1. XRD patterns of Ba-hexaferrite powder calcined at 900 °C (▼: BaFe₁₂O₁₉, ●: α-Fe₂O₃, ■: BaFe₂O₄).

Fig. 2. XRD patterns of Ba-hexaferrite thin film calcined at 900 °C (▼: BaFe₁₂O₁₉, ●: α-Fe₂O₃, ■: BaFe₂O₄).

Table 1. Comparison of the crystallite size between the calcined dried gel and the thin films synthesized by suspension of the dried gel.

Sample	Crystallite Size (nm)
Ba Hexaferrite powder synthesized by calcination of the dried gel	20
Ba Hexaferrite powder synthesized from calcination of the dried gel nano powder	25

The in-plane and out-plane hysteresis loop of BaM thin film are observed in Figs. 3 and 4, respectively. This thin film exhibits high magnetic coercivity ($H_{c\parallel} = 4.9$ kOe, $H_{c\perp} = 4.8$ kOe), relatively high remanent magnetization ($M_{r\parallel} = 123$ emu/cm³, $M_{r\perp} = 118$ emu/cm³), and rather large saturation magnetization ($M_{s\parallel} = 196$ emu/cm³, $M_{s\perp} = 195$ emu/cm³).

The high coersivities of in-plane and out-plane confirm the formation of nanostructure in the film and suitability of that for high recording densities. The amounts of Mr as already stated can also provide a relatively high signal to noise ratio. Compare to the results of the single phase Ba-hexaferrite film obtained from the spin coating of the first gel and calcination at lower temperature reported before in which only in plane loop could be detected,¹ these findings show that the novel process of using the second gel from suspension of the first gel dried powder in the process of spin coating resulted in perpendicular orientation which makes the film suitable for perpendicular recording applications as well.

Fig. 3. In-plane hysteresis loop of BaM thin film

Fig. 4. In-plane hysteresis loop of BaM thin film

4. Conclusions

Highly reproducible, barium ferrite thin film with thickness of 150-160 nm was successfully prepared by spin-coating of the suspension of the dried gel nano powder on the Si (110) single crystalline substrate. It was found that the 900 °C is a suitable calcination temperature to get good magnetic properties.

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