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PREPARATION OF BARIUM HEXAFERRITE BY SOL GEL SELF COMBUSTION USING POLYVINYL ALCOHOL

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ABSTRACT

PREPARATION OF BARIUM HEXAFERRITE BY SOL GEL SELF COMBUSTION USING POLYVINYL ALCOHOL. Barium hexaferrite powders were produced by sol-gel auto-combustion. Ferric and barium nitrate in the molar ratio of 1:2, and Poly Vinyl Alcohol (PVA) are used as starting materials. After gelation with ammonium hydroxide, dehydration and subsequent mechanical milling, auto combustion and calcination at temperature from 300 °C to 1250 °C were performed to produce barium ferrite powder. XRD pattern showed barium ferrite phase started to form at temperature of 500 °C and grew to single phase at 1000 °C, with hexagonal structure. At this stage, the powder has saturation and remanence magnetization of 50.9 emu/g and 33.6 emu/g, respectively, and coercivity of 2.94 kOe.

Key words : Barium hexaferrite, Sol gel auto combustion, PVA

ABSTRAK

PREPARASI BARIUM HEKSAFERIT MELALUI SOL GEL *AUTO COMBUSTION* MENGGUNAKAN POLIVINIL ALKOHOL. Telah dilakukan penelitian pembuatan barium heksaferit melalui proses *sol gel auto combustion*. Bahan baku awal adalah ferit dan barium nitrat dalam perbandingan molar 1:2, dan menggunakan *Polyvinyl Alcohol (PVA)*. Serbuk barium ferit terbentuk setelah melalui proses pembentukan gel setelah penambahan amonium hidroksida, pengeringan, penggilingan, *auto combustion* dan kalsinasi pada suhu 300 °C hingga 1250 °C. Hasil *XRD* menunjukkan fasa ferit mulai terbentuk pada suhu 500 °C dan fasa tunggal mulai terbentuk pada suhu 1000 °C dengan struktur heksagonal. Karakteristik serbuk pada penelitian ini adalah magnetisasi jenuh 50,9 emu/g, magnetisasi sisa 33,6 emu/g dan koersivitas 2,94 kOe.

Kata kunci : Barium heksaferit, Sol gel *auto combustion*, *PVA*

INTRODUCTION

Barium hexagonal ferrite has been produced and extensively studied in the many recent years. It has attracted attention due to potential applications in many technological device applications, such as conventional permanent materials [1], and magnetic materials for recording media [2]. Barium ferrite posses excellent magneto crystalline anisotropy, high Curie temperature and chemical stability against corrosion [3,4]. The requirement of specific material properties for device applications has driven extensive study of magnetic materials.

There were numerous methods for producing barium ferrite materials, e.g. conventional method of powder metallurgy [5], micro emulsion [6], glass crystallization [7], hydrothermal reaction [8], aerosol pyrolysis [9], and sol-gel technique [10,11]. Recently, sol gel auto combustion method was efficiently produced barium ferrite at relatively low temperature. The method introduced fuel precursors containing carbonize solution as starting materials beside Ba-Fe nitrat solution, such as citric acid [12-13].

In this study, polyvinyl alcohol is used as fuel precursor in the sol gel auto combustion of barium hexaferrite. The crystalline phase and magnetic properties are studied as function of calcination temperatures, subsequent mechanical milling of the as burnt barium ferrite precursors.

EXPERIMENTAL METHOD

The starting materials used in this study is analytical reagent of ferric nitrate hydrous $Fe(NO_3)_3.9H_2O$, barium nitrate unhydrous $Ba(NO_3)_2$, and polyvinyl alcohol $(CH_2\text{-}CHOH\text{-}CH_2\text{-}CHOH\text{-}CH_2)n$

(PVA). The ferric and barium nitrates in the molar ratio of 12 were dissolved in de ionized water. PVA of 10 wt% was added to the metal nitrate solution. Under continuous stirring proper amount ammonium hydroxide NH_4OH solution was poured into the prepared mixed solution until the final solution reach pH of 8, and viscous brown gels was formed.

All the mixing process were carried out at room temperature. The gels was then heated at 100 °C for 24 hours in the oven to ensure water evaporated, and form dark-brown loose agglomerate. The agglomerate was mechanically milled for about 6 hours to obtain fine particles of barium ferrite precursors (the as burnt powder), and calcined at temperature of 300 °C, 500 °C, 800 °C, 900 °C, 1000 °C and 1250°C for 2 hours.

Powder X-ray Diffractometer (XRD) (type D/max 2550 V) using Cu K_γ radiation was performed to examine crystallite phase of the as burnt powder and powder calcined at temperature of 300 °C, 500 °C, 800 °C, 900 °C, 1000 °C and 1250 °C. Hitachi S-2500 Scanning Electron Microscope (SEM) was used to reveal the surface morphology of powder calcined at 800 °C, 900 °C and 1000 °C. Permagraph C vibrating sample magnetometer (VSM) was employed to measure the magnetic properties of the ring sample shaped of powder calcined at 1000 °C.

RESULTS AND DISCUSSION

Figure 1 shows the XRD pattern of the as burnt and powder barium ferrite calcined at 300 °C, 500 °C, 800 °C, 900 °C, 1000 °C and 1250 °C.

The as burnt XRD pattern of barium ferrite indicates several peaks attributed to Fe_3O_4 and $Ba(NO_3)_2$ (Figure 1a). At this stage no indication of barium ferrite is formed; but physically this powder is attracted by magnet bar. As calcined temperatures are increased to 300 °C and 500 °C, γ Fe_2O_3 and α Fe_2O_3 started to grow (Figure 1b and Figure 1c), and Fe_3O_4 disapear. In fact, seed of barium ferrite ($BaFe_{12}O_{19}$) phase appears at calcined temperature of 500°C ($2\theta = 35.61^o$) (Figure 1d). As the calcined temperature of 800 °C is achieved there grow multi crystallites of barium ferrites and α Fe_2O_3. At higher temperature only $BaFe_{12}O_{19}$ and α Fe_2O_3 peaks are observed. The later peak disapears at calcined temperature of 1250 °C. Evidently, the phenomena is found similar to the work of Bahadur [14], without carboneous removal at 450 °C.

The use of PVA provide sufficient fuel for self-combustion, which enhance the nucleation but inhibit the growth rate of particles. Using Scherrer formula, the particle size extracted from line broadening of X-ray highest peak intensity of powder calcined at 1000 °C is 100 nm1to 50 nm. This size is not in agreement with the grain size depicted in SEM photograph, due to the fact that the photograph is revealing a surface morphology of grains that are randomly oriented while

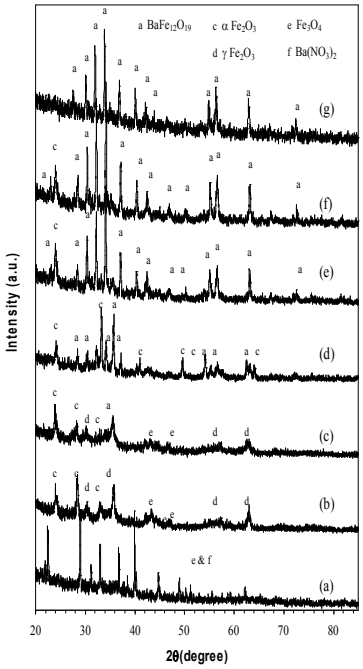


Figure 1. XRD pattern of (a) as burnt powder, and powder calcined at (b) 300 °C, (c) 500 °C, (d) 800 °C, (e) 900 °C, (f) 1000 °C, (g) 1250 °C.

XRD measurement averages the crystallinity in all directions of crystallites [15].

Figure 2 shows the surface morphology of the barium ferrite powder calcined at 800 °C, 900 °C, and 1000 °C. It is clearly noted that the microstructure are strongly affected by calcined temperatures. The grain size of powder calcined at 800 °C exhibit rode-like grain with narrower grain size distribution Figure 2a.

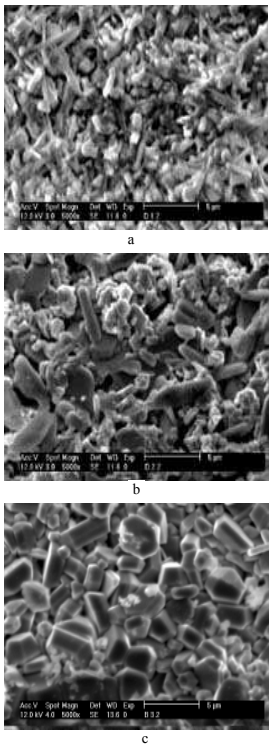


Figure 2. SEM photographs of $BaFe_{12}O_{19}$ powder calcined at (a) 800 °C, (b) 900 °C, and (c) 1000 °C

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As temperature is increased several grains grow, with a small amount of hexagonal shape Figure 2b. The grain shape is then transformed to mostly hexagonal grain with various different size at calcination temperature of 1000 °C Figure 2c, as observed by Huang et al. [16].

The magnetic properties were measured using Permagraph with an applied field of 20 kOe at room temperature. Figure 3 shows magnetization hysterisis loop of the powder calcined at 1000 °C.

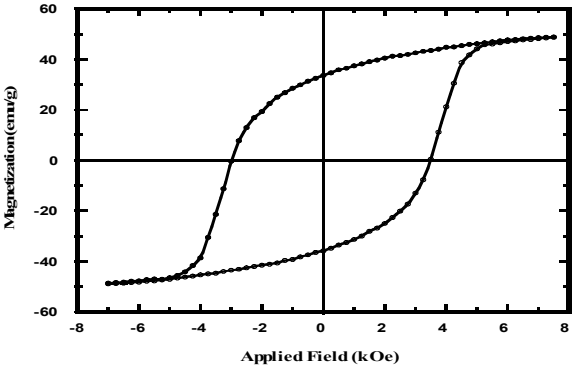


Figure 3. M-H hysterisis loop of barium ferrite powder calcined at 1000 °C

Table 1 depict the magnetic properties of the powder calcined at 800 °C, 900 °C and 1000 °C. The values of saturation (M_s) and remanence (M_r) magnetization, as well as coercive force H_c are extracted from magnetization hysterisis loops.

Table 1. Magnetic properties of calcined powder.

Sample Calcined at	M_s (10 kOe) (emu/g)	M_r (emu/g)	H_c (kOe)
800°C	41.7	22.0	1.67
900°C	42.1	28.1	2.83
1000°C	50.9	33.6	2.94

The three samples have similar magnetization saturation at 12.6 kOe. With increasing calcined temperature from 800 °C to 1000°C, M_s, M_r, and H_c grew to significant values of 50.9, 33.6 emu/g, and 2.94 kOe, which indicated that barium hexaferrite single phase was produced. However, the value M_s is slightly lower than that of theoretical calculation of 72 emu/g performed on a single crystal of $BaFe_{12}O_{19}$ [17]. The maximum BH product of the later sample is 0.59 MGOe.

CONCLUSION

Sol gel auto combustion using PVA precursors with metal-nitrates produce barium ferrite phase started at calcination temperature of 500 °C and grew to single phase at 1000 °C, with hexagonal structure. At calcined temperature of 1000 °C has yielded powder of barium hexaferrite with saturation and remanence magnetization of 50.9 emu/g and 33.6 emu/g, respectively, and the coercive force of 2.94 kOe.

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