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Remanence properties of barium hexaferrite

P.E. García.Casillas, A.M. Beesley, D. Bueno, J. A. Matutes-Aquino, C.A. Martínez

Abstract

Barium hexaferrites have been widely used as permanent magnets, however one of the most important parameters that characterizes these ferrites is the switching field distribution (SFD). This work calculates the SFD on a small particle barium ferrite ($BaFe_{12}O_{19}$) obtained by coprecipitation from chlorides in an alkaline medium using the irreversible component of the magnetization. Forward and reverse switching field distribution curves were obtained by differentiation of isothermal remanent magnetization (IRM) and dc demagnetisation (DCD) curves. It was found that both values differ by a factor of 3.5, quite away from the value of non-interacting systems. The Henkel plot was built from thee data sets, indicating a predominant region with demagnetising interaction between particles, and a small region in which the particles interact constructively to the magnetization, according to the Preisach model framework.

Introduction

Barium ferrite ($BaFe_{12}O_{19}$) has been commonly used as permanent magnet since its development in the beginning of the 1950s by Phillips researches. In fact, this material presents high saturation magnetization, high Curie temperature, high intrinsic coercivity and rather large crystal magnetic field anisotropy [1,2]. Due to these properties, many methods of synthesis have been developed to obtain a low production cost of fine particles of barium ferrite with a good chemical homogeneity, in this sense coprecipitation seems to be the most suitable at the moment [2–5]. However it is of great importance the level of stacking of Ba ferrite particles which is believed to determine the magnetic interparticle interactions, affecting essential properties such as thermal duplication, media noise and resolution in longitudinal recording media [1,6]. The deviation from the Stoner–Wohlfarth model is a technique to estimate interparticle interactions but it is restricted to uniaxial single domain particles. Barium ferrite obtained by coprecipitation consists of hexagonal plates and can be considered as a uniaxial system, however the size of these particles determines whether or not they are single- or multi-domains. Therefore a more general theory is needed to describe these interactions. The Preisach hysteresis model provides the framework to make the Wohlfarth relation valid for multi-domain systems. This model considers the magnetic material as a distribution of small hysteresis loops or "hysterons", which are determined by (h_c, h_u) units in which h_c represents its coercive field and h_u is the displacement of the hysteron from origin (0, 0) in the magnetization versus field plane [7]. These hysterons can be considered as Stoner–Wohlfarth particles only if they represent the reversible component of the magnetization. The values h_c and h_u of each hysteron determine the Preisach plane (h_c versus h_u). The three remanent states, magnetizing (M_r), demagnetising (M_d) and saturation (M_∞) are associated with three partitions of the Preisach plane [7,8]. Considering that the Preisach distribution is concentrated along the h_c-axis ($h_u = 0$), the upper limit of the Henkel plot region is determined by the relation $M_d = M_\infty - 2M_r$, as the Wohlfarth model. In that case if $M_d \geq (<) M_\infty - 2M_r$, the particles present a magnetizing (demagnetizing) nature. On the other hand if the Preisach distribution is focused around a local coercivity h_0 in which all the units present the same coercivity value $h_c = h_0$, it leads to the lower limit of the Henkel plot region as $M_d = -M_r$ [7,8]. According to this, the known expression that determines the Henkel plot regions, are deduced from the following inequalities $-M_r \leq M_d \leq M_\infty - 2M_r$ [7,8]. The interparticle interaction process consists mainly on two contributions, the mean field effects and the local disorder of the particles, which compete with each other giving rise to magnetizing and demagnetizing effects. In this work the type of interaction that governs our system is deduced using the Henkel plot and the δm curves. Isothermal remanent magnetization (IRM) and dc demagnetization (DCD) data were taken and analysed in this framework. The first differential of these curves gives a direct measurement of irreversible changes in the magnetization states, from which the switching field distribution was calculated.

Experimental

Barium ferrite powders were produced by coprecipitation of Fe(III) and Ba(II) nitrate solutions in an alkaline aqueous medium. The ratio Fe:Ba was kept at 11:1. The alkaline solution was a mixture of NaOH (50 g) and Na_2CO_3 (12.5 g), with pH 13. The precipitate appeared after adding both nitrate solutions into the alkaline medium under vigorous stirring, after which it was washed and dried at 40 ◦C for 24 h. Further calcination at different temperatures (600, 700, 750, 800, 925, 950, 1000 and 1100 ◦C) for 2 h was carried out in order to study the influence of the calcination temperature on the magnetic properties. In each case the mean particle size was measured using an isothermal adsorption technique.

The magnetic properties of the samples were measured at room temperature using a Vibrational Sample Magnetometer LDJ Electronics 9600, with a maximum applied field of 1.5 T. The morphology of the samples were examined by transmission electron microscopy using a Phillips CM-200 microscope. Associated to this, the mean particle size was measured using the Brunauer–Emmett–Teller method (BET).

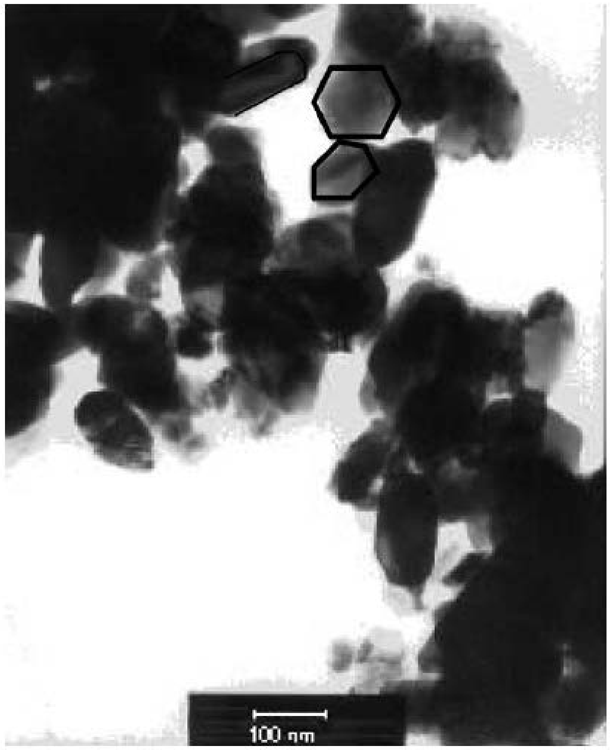

Fig. 2. Transmission electron micrograph of barium ferrite powder, calcinated at 925 °C.

The IRM curve was obtained after ac demagnetization followed by a positive applied field from where the remanent magnetization was then recorded. This procedure was repeated while gradually increasing the field strength until positive saturation remanence was obtained. The DCD curve was obtained in a similar manner. Initially the sample was saturated with a positive field of 1.5 T. A negative field was applied and the remanent magnetization was recorded. This procedure was repeated while gradually increasing the field strength until negative saturation remanence was obtained. The δm values were obtained from the relation $M_d - [M_\infty - 2M_r]$ and the switching field distribution from the irreversible contribution was calculated as $(M_\infty/H_c)/\chi_r$ $|H_c$.

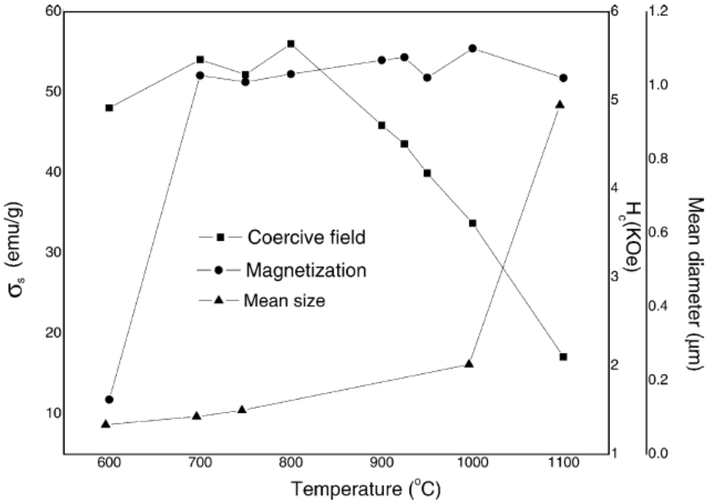

Fig. 1. Magnetic properties of barium ferrite powder calcinated at different temperatures.

Results and discussion

Fig. 1 shows the specific magnetization, intrinsic coercive field and mean particle size value as function of the calcination temperature. The particle size obtained at different temperatures, determines the magnetic properties of the barium ferrite, as expected [9]. The intrinsic coercivity increases with the reduction of particle size up to 700 °C after which tends to decrease. On the other hand the specific magnetization is dependant of the amount of crystalline barium ferrite obtained at different calcination temperatures. X-ray diffraction measurements indicate that the crystalline phase appears when the precipitate was calcinated at 700 °C [10], and in fact the particle size shown in Fig. 1 starts to increaseat this temperature. At lower temperatures (<600 °C) thesamples present a mean specific magnetization of 12 emu/g, rising to 53 emu/g between 700 and 1100 °C. These results are comparable with those found in the literature [9,11].

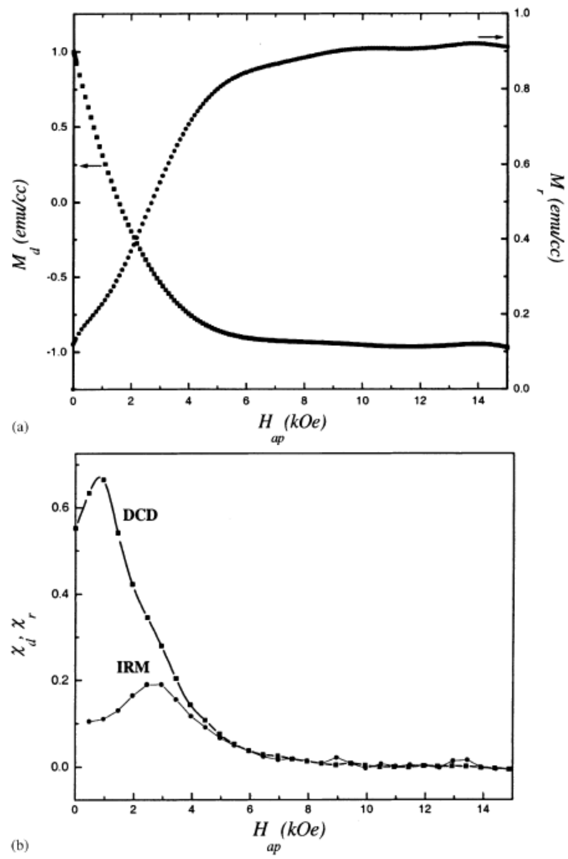

Fig. 3. (a) Remanent magnetization curves IRM and DCD; (b) IRM and DCD magnetic susceptibilities as function of the applied field, for a Ba ferrite calcinated at 925 °C.

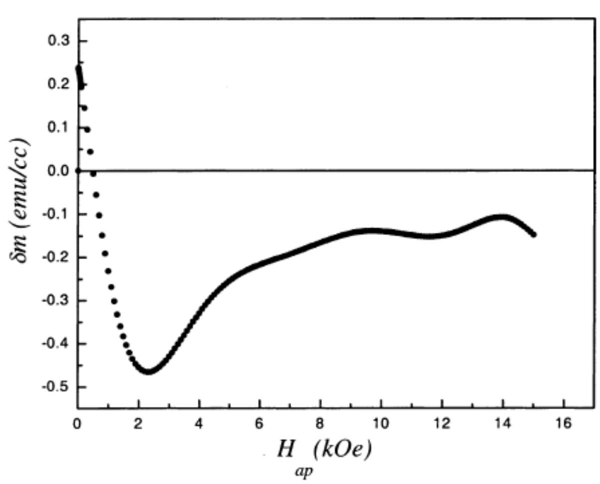

Fig. 4. Behaviour of the δ_m curve with the magnetic applied field, for a Ba ferrite calcinated at 925 °C.

The morphology of the sample treated at 925 °C is shown in Fig. 2. The hexagonal plates observed are typical of well crystalline barium ferrite. The particle size is in agreement with those found by BET. The thickness of the particles, varies around of 80–300 nm. According to the relation obtained by Pfranger et al. [11], the dependence of the domain wall width D on the thickness L of barium ferrite crystals along the magnetically preferred direction is given by $D = 1.53L^{0.32}$ for L < 500 μm. Using this relation the domain wall width in our particles lie around 6.2–9.5 nm. In this sense it is possible to study the interparticle interactions using the Wohlfarth relation if the system is treated under the Preisach framework.

The IRM and DCD remanence curves are shown in Fig. 3a. The remanence at saturation M_∞, corresponds to the M_r value at 15 kOe, equal to 0.92 emu/cm^3. The first differential of these curves gives a direct measure of irreversible changes in the magnetization states χ_r and χ_d, shown in Fig. 3b. By differentiation of the Wohlfarth relation, one obtains the remanence susceptibility ratio as $\chi_d/\chi_r = 2$, for non-interactive systems. In this case the amplitude of χ_d and χ_r peaks, differ by a factor of 3.5 with no coincidence of widths or maximum positions.

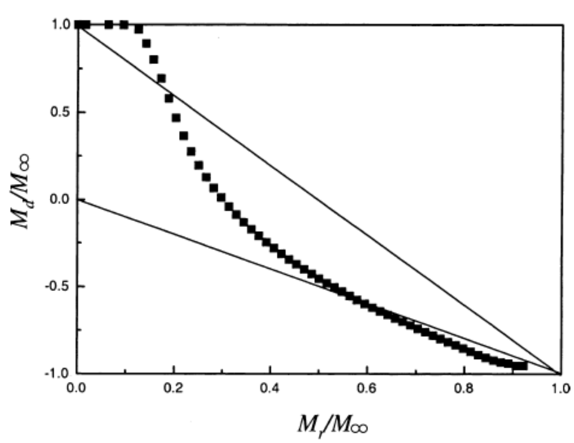

Fig. 5. Henkel plot for a Ba ferrite calcinated at 925 °C, showing the upper and lower limits in dashed lines

This indicates a strong interparticle interaction that could be due to local disorder of the system or to the mean magnetic field distribution associated to each particle, producing magnetizing or demagnetizing-like effects, at certain values of magnetic field. The switching field distribution was calculated considering only irreversible events, i.e. $(M_\infty/H_c) / \chi_e|H_c$, leading to a value of 0.61 [12].

Fig. 4 shows the δm plot as a function of the applied field. For a non-interactive system, this plot would show a horizontal line through the origin. In this case this plot presents a positive region between zero applied field and 500 Oe, associated to interparticle interactions that contribute constructively to the magnetization following the applied field (magnetizing-like effect). The opposite occurs at higher applied fields; from 500 Oe to saturation the particles interact against the field producing a demagnetizing-like effect. The values of δm in the positive region are comparable to those found in hard disks and tapes [6,12]. On the other hand the values of δm in the negative region are comparable to those found in floppy disks [6,12].

The Henkel plot built from the IRM and DCD data I shown in Fig. 5, where the upper and lower limits are als represented. The positive region of the Henkel plot and it shape is associated according to the literature to a system in which the particles are interacting with each other governed by the mean field distribution of the system up to a value of field around 500 Oe, at that point the competition starts between the mean field and the local disorder of the particles and the latter finally governs the system as the curve approaches to the lower limit of the Henkel region.

Conclusion

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It has been studied the remanence properties of barium ferrite using the Wohlfarth relation in the Preisach framework. The system shows a magnetizing interparticle interaction where the mean field effects are dominating between zero applied field and 500 Oe. Above this field the system shows a demagnetizing-like interparticle interaction with government of local disorder. A further investigation will clarify the influence of the particle size on the remanent properties.

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