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Magnetic interactions and Preisach distributions of nanostructured barium hexaferrite

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Abstract

Barium hexaferrite (phase M) samples with different nanostructures were studied. Sample M1 is composed of nanocrystals of $BaFe_{12}O_{19}$ produced after milling the elemental oxides (Fe_2O_3 and $BaCO_3$) and heating in air atmosphere. Two more samples were produced by milling the same oxides and a 20% excess of α-Fe. The resulting powder (composed of phase M and a ~20% hematite) was heat-treated in different conditions, resulting in samples MF1 (with a partially sintered structure) and MF8 (with almost completely sintered structure).

In order to have an insight into the interactions in each sample, Preisach distributions were obtained using first-order reversion curves (FORCs) measurements. The Preisach distribution corresponding to M1 is a Gaussian-shaped one, with a maximum around 4.1 kOe. The distribution of MF1 has a narrow and high peak at 5.3 kOe, a number of overlapping small peaks down to 2.6 kOe and a distinct and low-intensity peak at 2 kOe. MF8 has a Preisach distribution with a succession of equally spaced distinct peaks from 4.2 to 1.5 kOe.

The found Preisach distributions suggest that the interactions occur among nanocrystals inside conglomerates with different number of particles.

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It is generally accepted that the coercive field H_c of hard magnetic materials is well described by the modified Brown's equation [1]:

$$H_c = 2\frac{K_1}{M_S}\alpha_\phi\alpha_K\alpha_{ex} - N_{eff}M_S, \tag{1}$$

where K_1 is the anisotropy constant, M_S is the saturation magnetization, N_{eff} is the effective demagnetization factor, and the coefficients α_ϕ, α_K and α_{ex} take into account, respectively, the grains' orientation distribution in the magnet, the reduction in anisotropy in regions near internal surfaces, and the effect of exchange coupling between neighboring grains. These coefficients are $\leqslant 1$, thus resulting in a diminution of the coercive field of the magnet.

It is evident then, that the coercive field depends on the entire system. The Preisach Model [2] is a powerful tool to describe the magnetic behavior of a system of particles that considers the coercivity of each particle (h_c) as well as the internal field that acts on them (h_u). The Preisach distribution function $f(h_c, h_u)$ depends on both the coercive and interaction fields and reflects how the particles

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of the system are distributed in the plane (h_c, h_u). By means of the identification $h_c \leftrightarrow H_c$ it is possible to study the magnetic interactions in different systems.

Our interest is to enlighten the current knowledge about the interaction mechanisms in barium hexaferrite using the Preisach Model. In order to do so we study three samples with different nanostructures.

Sample M1 was obtained after milling the elemental oxides (Fe_2O_3 and $BaCO_3$) in stoichiometric proportions in a planetary ball mill and then heating at 1000°C in air atmosphere for 1 h. Samples MF1 and MF8 were obtained by first milling the same oxides and a 20% excess of α-Fe in the same conditions than M1. Sample MF1 was then heat-treated for 1 h and compacted into platelets 1–2 mm thick, while sample MF8 was compacted into platelets before it was heat-treated for 8 h in the same conditions than MF1.

Sample M1 is composed of highly crystalline barium hexaferrite (phase M) with very small traces of hematite, and mean particle size $D = 50$ nm. Samples MF1 and MF8 are composed of phase M and approximately 20% hematite. As the heat treatment on MF1 was performed on the powders before compacting, there is a partial and short-range sintering between particles. On the other hand, BMF8 was first compacted and then heated, so there is a larger degree of sintering due to the intimate contact amongst the particles. In this case, a high density was obtained, approximately 70% of the theoretical value for the mixture. The value of D for MF1 and MF8 is 50 and 60 nm, respectively.

The Preisach distributions of the samples were calculated with the aid of first-order reversal curves (FORCs), using that

$$f(\alpha,\beta) = -\frac{\partial^2 M(\alpha,\beta)}{\partial\alpha\partial\beta},$$

where $\alpha = (h_c + h_u)/2$, $\beta = (h_c - h_u)/2$ and $M(\alpha,\beta)$ is the magnetization of the sample at a point (α,β) of a FORC [2,3].

In order to check the validity of these Preisach distributions, we compared the measured values of the irreversible susceptibility $\chi^{\mathrm{i}}_{\mathrm{irr}}$ with the calculated $\chi^{\mathrm{i-calc}}_{\mathrm{irr}}$ using

$$\chi^{\mathrm{i-calc}}_{\mathrm{irr}} = \chi^{\mathrm{i}}_{\mathrm{tot}} - \chi^{\mathrm{i}}_{\mathrm{rev}} = \frac{\partial M}{\partial H_{\mathrm{i}}} - \chi^{\mathrm{i}}_{\mathrm{rev}} = \int_0^\infty f(h_c, h_c - H_{\mathrm{i}})\,\mathrm{d}h_c - \chi^{\mathrm{i}}_{\mathrm{rev}},$$

where superscript i indicates derivatives with respect to the internal field H_{i}. The reversible susceptibility $\chi^{\mathrm{i}}_{\mathrm{rev}}$ was determined by calculating the slope of a small loop performed at different fixed fields, after saturating the sample. The agreement between $\chi^{\mathrm{i}}_{\mathrm{irr}}$ and $\chi^{\mathrm{i-calc}}_{\mathrm{irr}}$ is remarkable, indicating that the Preisach distributions found describe the three systems correctly.

The Preisach distribution obtained for M1 is a Gaussian-shaped one, with a maximum centered in $h_c \approx 4.1$ kOe and standard deviations $\sigma_c \approx 1$ kOe (on the variable h_c) and $\sigma_u \approx 0.2$ kOe (on the variable h_u). The peak value is very close to the field for which the irreversible susceptibility of this sample has a maximum.

The Preisach distributions obtained for MF1 and MF8 (Figs. 1 and 2, respectively) are more complex and nothing like this has yet been reported for systems composed of a single magnetic phase. The distribution of MF1 has a narrow and high peak at 5.3 kOe, a number of overlapping small peaks down to 2.6 kOe and a distinct and low-intensity peak at 2 kOe. MF8 has a Preisach distribution with a succession of equally spaced distinct peaks of roughly the same intensity, centered at different h_c from 4.2 to 1.5 kOe.

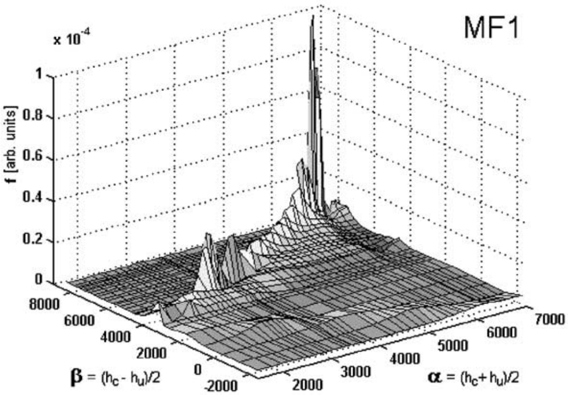

Fig. 1. Preisach distribution for MF1 as a function of the variables $\alpha = (H_c + h_u)/2$, $\beta = (H_c - h_u)/2$.

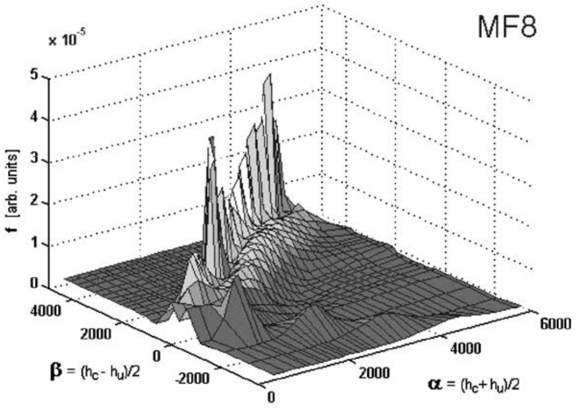

Fig. 2. Preisach distribution for MF8 as a function of the variables $\alpha = (H_c + h_u)/2$, $\beta = (H_c - h_u)/2$.

The already mentioned Gaussian distribution of M1 could be attributed to the fact that this sample is composed of phase M alone (practically) and there is no reason to believe that the heat treatment would produce other kind of particles distribution on the plane (h_c, h_u).

However, a completely different behavior is noticed in the other studied samples. The multiple peaks observed in the Preisach distributions of MF1 and MF8 suggest us that they represent different groups of conglomerates that invert their magnetization at the fields h_c of the maxima. The sintering process has produced a distribution of conglomerate sizes, as sintering occurs among different number of particles. It is highly probable that each conglomerate will have a well defined h_c value, the larger the number of particles per conglomerate the lower the corresponding h_c. The origin of this kind of distribution lies in the fact that both MF1 and MF8 are composed not only of barium hexaferrite but also of 20% hematite. This phase surrounds conglomerates of phase M with different number of particles preventing them from welding with each other during the sintering process. Also the pores in the samples play a role in the formation of the conglomerates. In MF1 there are more pores because this sample was prepared by first heat-treating the powder and then compacting it. More conglomerates with few particles are expected in this case. This is reflected in the Preisach distribution of this sample because, as it can be seen in Fig. 1, the highest maximum is attained at high fields. On the other hand, BMF8 was first compacted and then sintered, so in this case there was a smaller proportion of pores and a wider distribution of conglomerates that underwent the sintering without losing certain 'individuality'.

We identify the maxima h_c of the Perisach distribution with the coercive fields $H_c^{\exp}$ that correspond to different conglomerates. So, using Eq. (1) each conglomerate will have an effective anisotropy given by

$$K_1^{\mathrm{eff}} = K_1 \alpha_K \alpha_{\mathrm{ex}} = \frac{H_c^{\exp} M_S + N_{\mathrm{eff}} M_S^2}{2\alpha_\phi},$$

with $\alpha_\phi = \frac{1}{2}$ for a randomly oriented structure.

As the exchange length in these systems ($l_{\mathrm{ex}} = 8$ nm) is lesser than D, we deduce that there is a partial coupling between the particles of phase M inside each conglomerate and there are regions of lower coercivity produced by the exchange coupling. If we accept that there is a distribution of conglomerates with different number of particles that invert their magnetization at different coercive fields, we can infer that the exchange interaction in these systems depends on the number of particles per conglomerate. Further work is being done in order to find a physical model that explains this behavior.

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