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Improvement of the remanence properties and the weakening of interparticle interactions in $BaFe_{12}O_{19}$ particles by B_2O_3 addition

Ugur Topal *

TUBITAK-UME, P.K. 54, 41470 Gebze-Kocaeli, Turkey

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ABSTRACT

In the present study, the effects of B_2O_3 addition on the remanence properties of barium ferrite magnets are examined. The relationship between isothermal magnetization remanence $M_R(H)$ and demagnetization remanence $M_D(H)$ for non-interacting single domain particles, $M_D(H)=M_R(H_{max})-2M_R(H)$, was used in order to investigate the interactions between particles. We have found that remanence magnetization M_R increased by 40% in magnitude with B_2O_3 addition in addition to the weakened couplings between particles. The B_2O_3 addition seems to supply the required conditions for usage of these materials in the magnetic recording media.

1. Introduction

Finding the ways of increasing the capacity of magnetic recording media has been the subject of many works in the past decades. Barium ferrite, as a good candidate, has received much attention due to its potential for such an application [1–4]. In order to improve the properties of recording media, the particle size and the interaction between particles must be optimized. With this purpose, many different techniques have been proposed, such as co-precipitation [5,6], hydrothermal synthesis [7,8], sol–gel [9], organometallic precursor [10], microemulsion [11,12] and ammonium nitrate melt [13], for the synthesis of barium ferrite particles having optimal properties not only for the recording media but also for other applications. Recently, we synthesized high-quality barium ferrite particles ($BaFe_{12}O_{19}$) with addition of small amounts of B_2O_3 and with calcination at relatively much lower temperatures (850 °C) and durations (2 h) [14,15]. Resultant products are of single phase and have particles in the single domain limit. Both the magnetization remanence M_R and the saturation magnetization M_S have also been increased by almost double in magnitude compared to the undoped one. Such high magnetization values (M_R ~31 emu/g and M_S ~56 emu/g) together with the convenient coercivity (between 2 kOe and 3 kOe) governed by B_2O_3 addition may facilitate the usage of these materials for high-density magnetic recording media.

To assess the suitability of a material for use in magnetic recording it is preferred to investigate the magnetic particle interactions using the well known Wohlfarth [16] relation.

$M_D(H)=M_R(H_{max})-2M_R(H)$ for an array of non-interacting particles, where $M_D(H)$ is demagnetization remanence and $M_R(H)$ is isothermal magnetization remanence. Deviation from this equality reveals the existence of interaction between particles. The equation above was re-formulated later by Kelly et al. [17] by adding δm, $\delta m(H)=m_D(H)-(1-2m_R(H))$,which gives the sign and relative strength of the interactions. Here $m_R(H)$ and $m_D(H)$ are the remanences normalized to the saturated magnetization remanence value $M_R(H_{max})$. In order to optimize properties of the recording media, such as high output and low noise, the minimal δm values are desired (i.e. the particle interactions must be minimized). However, it must be noted that interaction between the particles may be observed even in ferromagnetic/non-magnetic neighborhoods as stated in Ref. [18].

In the present study, we examine the evolution of the particle interactions with the B_2O_3 addition in the $BaFe_{12}O_{19}$ using the modified Wohlfarth equation given above and thus, we will find the optimal B_2O_3 doping level which makes these materials convenient for uses in the recording media.

2. Experimental

A series of B_2O_3-doped M-type barium hexaferrite was synthesized using the conventional ceramic preparation method [14]. Structural analysis was done by a Shimadzu XRD-6000 diffractometer

* Tel.: +90 262 6795000; fax: +90 262 6795001.
E-mail address: ugur.topal@ume.tubitak.gov.tr

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(CuKα) and by a scanning electron microscope (JEOL 6335F Field Emission Gun). Isothermal magnetization remanence, $M_R(H)$, was measured on thermally demagnetized samples. A small field is applied, then removed and the remanence magnetization is measured. The remanence was measured as a function of increasing field until saturation remanence had been reached. Demagnetization remanence, $M_D(H)$, is measured as follows: the sample is initially saturated in one direction and then a small reverse field is applied, removed and the remanence is measured. This process is repeated until negative saturation remanence is observed. In order to obtain true remanence values, the time dependent magnetization and demagnetization effects were taken into account by application of the applied field for at least 30 s and the relaxation at zero fields for the same time period prior to the remanence measurements [19].

3. Results and discussions

It was previously reported that the presence of impurity phases and the grain dimensions influence the remanence properties and the strength of interparticle interactions in barium ferrite samples [20]. Therefore, it will be supplementary to analyze the structural properties of the samples under investigation prior to the discussions over the magnetization data. The X-Ray analysis has shown that single phase $BaFe_{12}O_{19}$ can be obtained even with additions of quite small amounts of B_2O_3 although the undoped samples contain large amounts of $BaFe_2O_4$ and α-Fe_2O_3 secondary phases [14,15]. For instance, the undoped sample calcined at 900 °C is identified to be a mixture of the M-type hexagonal barium ferrite ($BaFe_{12}O_{19}$) and the impurity phases α-$BaFe_2O_4$ and α-Fe_2O_3 (Fig. 1a). Here, we must note that only the main characteristic peaks of the present phases were marked. Amounts of these impurities were determined to be ~31% and ~17% for α-$BaFe_2O_4$ and α-Fe_2O_3, respectively. On the other hand, the impurity phases disappear with addition of 0.5 wt% of B_2O_3 (Fig. 1b). Scanning electron microscopy (SEM) images of the samples calcined at different temperatures and containing different amounts of B_2O_3 are shown in Fig. 2. These pictures reveal that grain sizes are more or less 1 μm (single domain limit) in spite of the existence of some larger grains for the samples having B_2O_3 content up to 2.0 wt% (Fig. 2a, b and d, e, see also Ref. [14]). Further increase of B_2O_3 content, on the other hand, causes the grains growing up to 5–10 μm (Fig. 2c and f). It is also apparent from SEM images that particles get closer and stick to each other with the increase of boron concentration. Many studies have shown that the use of finer barium ferrite particles with improved particle size dispersion is essential for the increase of the density of information stored on magnetic media and the minimization of medium noise [21,22]. In these studies, it was shown that noise power is proportional to the square of the particle volume. Therefore, the dispersion of particle sizes on the samples having low concentrations of B_2O_3 seems more suitable than those having high concentrations of B_2O_3 for their usage in magnetic recording media.

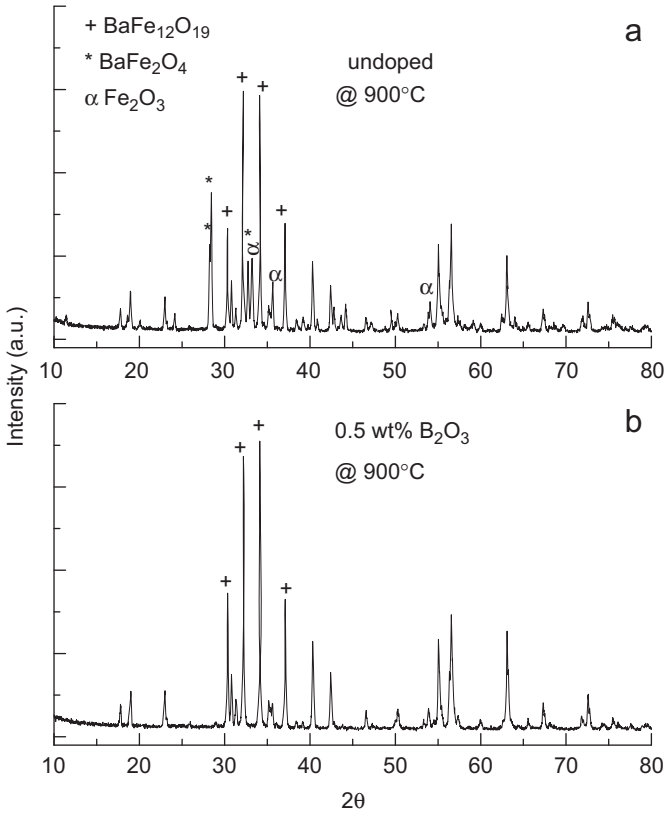

Fig. 1. XRD patterns of (a) undoped and (b) 0.5 wt% B_2O_3-doped samples calcined at 900 °C. Here, the main characteristic peaks of the relevant phases were marked.

The saturated magnetization remanence $M_R(H_{max})$ and the saturation magnetization M_S of some selected samples were determined from the M–H loops and reported in Ref. [14]. It was observed that squareness ratio $M_R(H_{max})/M_S$ of most of the samples is quite close to that of randomly oriented 3-D single domain particles [17,23]. Together with the SEM analysis, it will be correct to ignore the possible contribution of grain sizes to the δm values, at least for low B_2O_3 concentrations [20].

The $M_R(H)$ and $M_D(H)$ curves are shown in Fig. 3. It is seen that both remanences reach saturation at certain magnetic fields and their values increase significantly with B_2O_3 addition. For instance, the $M_R(H_{max})$ values of the undoped and the 0.2 wt% B_2O_3-doped samples, which are calcined at 1000 °C, are 22.3 emu/g and 31.0 emu/g, respectively. The general trend of the remanence parameters seen is initial increase, reaching maximum and then decreasing with the increase of B_2O_3 concentration. It was also recognized that the value of B_2O_3 concentration, on which the highest magnetization is achieved, decreases as the calcination temperature increases. For instance, the highest $M_R(H_{max})$ is seen on the 1 wt% B_2O_3-doped sample in the case where the calcination temperature is 850 °C. On the other hand, it will be seen on the 0.2 wt% B_2O_3-doped one if the calcination process is carried out at 1000 °C.

Fig. 4 shows the δm plots as a function of the applied field for the samples containing different amounts of B_2O_3 and calcined at different temperatures. For non-interactive particles these plots would show a horizontal line through the origin (see Fig. 4). As seen, the δm takes both positive and negative values for all studied samples. Initially observed positive δm values are followed by negative values above certain magnetic fields. Positive regions are seen below 5000 Oe (except 5.0 wt% B_2O_3) and width of the region decreases as the calcination temperature increases. It is also interesting that the fields at which positive maxima are seen mostly coincide with the coercive field. As mentioned before, deviations from the non-interaction line indicate that there is an interaction between the grains of the samples examined. The strength of these interactions seems to change as a function of boron concentration and the calcination temperature. With the increase of calcination temperature, the magnitude of positive peak decreases as the magnitude of negative peak increases. It can be interpreted as a decrease in resistance to the magnetization reversal in increasing reversing fields with the increase of calcination temperature. Taking also the granular natures of the samples into account (calcination at low temperatures results in