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Magnetic properties of B_2O_3-SiO_2-BaO-Fe_2O_3 glass-ceramics

D. Yousefi, M. Tavoosi *, A. Ghasemi

Department of Materials Engineering, Malek-Ashtar University of Technology (MUT), Shahin-Shahr, Isfahan, Iran

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ABSTRACT

The magnetic characterization of B_2O_3-SiO_2-BaO-Fe_2O_3 glass-ceramics during annealing process was the goal of this study. In this regard, different mixtures of BaO, Fe_2O_3, SiO_2 and B_2O_3 were melted and then quenched between two cold steel sheets. The annealing process was done in as-quenched samples in temperature range of 550 to 850 °C for different periods of time. The prepared samples were characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), differential scanning calorimetry (DSC) and vibrating scanning magnetometer (VSM). According to achieved results, fully amorphous structure can only be formed in $20B_2O_3$-$10SiO_2$-45BaO-$25Fe_2O_3$ system and the microstructures of as-quenched samples in other systems were combination of Fe_3O_4, $BaFe_{12}O_{19}$ and amorphous phases. The coercivity and saturation of magnetization of as-quenched samples were estimated about 85–120 Oe and 12–17 emu/g, respectively. By annealing the produced samples, the coercivity value was increased to about 3100 Oe as a result of precipitation of $BaFe_{12}O_{19}$ in nonmagnetic $Ba_2FeSi_2O_7$ matrix. The highest value of coercivity was achieved in annealed sample at 700 °C for 5 h.

1. Introduction

Magnetic properties of amorphous materials containing transition metal ions have been extensively studied in the past for both practical and fundamental reasons [1–3]. There has been considerable interest in iron-bearing oxide glasses because of their interesting magnetic, structural, and optical properties [4–6]. The controlled crystallization of these glasses is commonly employed to produce glass-ceramics with optimum magnetic properties starting from a mixture of Fe_2O_3, BaO, SrO, B_2O_3, SiO_2 and P_2O_5 precursors [7,8].

This process widely used to precipitation of hard-magnetic M-type hexaferrites, such as $SrFe_{12}O_{19}$ and $BaFe_{12}O_{19}$ with a definite crystallite size in a nonmagnetic matrix. For instance, Zaitsev et al. [9] prepared a $SrFe_{12}O_{19}/Sr_2B_2O_5$ composite microstructure with optimum magnetic properties by crystallization of SrO-Fe_2O_3-B_2O_3 glass-ceramics. Mekki [10] also presented one microstructure with best magnetic properties in SiO_2-Na_2O-Fe_2O_3 compound by means of annealing process. These composites are ideal choice for development of advanced recording media [11].

Although, there are a lot of investigations about the formation and magnetic characterization of Fe_2O_3-BaO-B_2O_3-SiO_2 glass ceramics [7–10], the exact effect of annealing condition on structural and magnetic properties of these glass-ceramics have not been properly investigated. Therefore, optimization of the annealing process in B_2O_3-SiO_2-BaO-Fe_2O_3 glass-ceramics in order to achieve the best magnetic properties was the goal of this study.

2. Materials and methods

H_3BO_3 (Merck, 99.8% purity), SiO_2 (Merck, 99.8% purity), $BaCO_3$ (Merck, 99.8% purity) and Fe_2O_3 (Merck, 99.8% purity) were used as raw materials. The initial powders with the composition of $20B_2O_3$-$10SiO_2$-(45 − x)BaO-(25 + x)Fe_2O_3 (x = 0.10.20) (mol.%) (based on Table 1) were melted in an alumina crucible at temperature range of 1300–1460 °C for 1 h and then were quenched between two cold steel sheets. For instance, the macrograph of $20B_2O_3$-$10SiO_2$-45BaO-$25Fe_2O_3$ as-quenched sample is presented in Fig. 1. Annealing procedure had been done at temperatures range of 550 to 850 °C for different periods of time until 10 h at ambient atmosphere.

Philips PW3710 XRD machine using a diffractometer with Cu K_α radiation (λ = 0.15406 nm; 40 kV) was used to follow the structural variation of the specimens (2θ range: 10–80°, step size: 0.05°; time per step: 1 s). The crystallite sizes of different phases estimated form XRD patterns using Scherrer methods [12]. Structural and morphological characterizations of samples were carried out by field emission scanning electron microscopy (VEGA-TESCAN-XMU) at an accelerating voltage of 20 kV. Before SEM investigations, the specimens were polished and etched in 0.5% HF for 75 s. Differential scanning calorimetry was also conducted to study the thermal stability of produced samples using the L81/1750 DTA differential thermal analyzer. The samples were placed in Al_2O_3 pans and heated in dynamic argon atmosphere

* Corresponding author.
E-mail address: ma.tavoosi@gmail.com (M. Tavoosi).

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Table 1
The chemical composition of different B_2O_3-SiO_2-BaO-Fe_2O_3 specimens which were investigated in this work.

S.N.	BaO/Fe_2O_3 ratio	Chemical Composition (mol.%)			
		B_2O_3	SiO_2	BaO	Fe_2O_3
1	45/25 = 1.8	20	10	45	25
2	35/35 = 1	20	10	35	35
3	25/45 ≈ 0.56	20	10	25	45

Fig. 1. The macrograph of $20B_2O_3$-$10SiO_2$-45BaO-$25Fe_2O_3$ sample after quenching between two cold steel sheets.

up to 900 °C at a heating rate of 20 °C/min. Magnetic properties of produced samples were measured using a vibrating scanning magnetometer (VSM) under an applied field up to 15 kOe.

3. Results and discussion

3.1. Glass preparation

There are several factors such as composition, superheat and solidification rate that influence the structure and properties of as-quenched magnetic glass-ceramics [7–10]. A slight variation in these parameters can often cause large variations in the microstructure and properties of the produced samples. In the present study, all of these parameters were kept nominally constant and the microstructure and magnetic properties of the BaO-Fe_2O_3-SiO_2-B_2O_3 glass-ceramics with different composition (different BaO/Fe_2O_3 ratio) were investigated. It is important to note that, the percentages of SiO_2 and B_2O_3 glass formers were chosen about 10 and 20 mol% based on Marghussan [13] report. In fact, this composition has the best glass forming ability and chemical durability in BaO-Fe_2O_3-SiO_2-B_2O_3 system.

3.1.1. Structural characterization

In order to understand the effects of initial composition on the microstructure of B_2O_3-SiO_2-BaO-Fe_2O_3 glass-ceramics, three different combinations of initial precursors, based on Table 1, were melted and quenched at same condition. It is important to note that, during fusing process, the alumina crucible contaminates the melt. The maximum amount of Al contamination was estimated about 5.2 at.%. Although, the aluminum oxide is a glass former, its effect on final magnetic properties is not negligible [14,15].

The XRD patterns of as-quenched samples at different BaO/Fe_2O_3 ratio are presented in Fig. 2. According to this figure, the structure of as-quenched $20B_2O_3$-$10SiO_2$-25BaO-$45Fe_2O_3$ (BaO/Fe_2O_3 = 0.56) sample consists of Fe_3O_4, $BaFe_{12}O_{19}$ and amorphous phases. The FE-SEM micrographs of this specimen which are shown in Fig. 3 also confirm this finding. As seen, these microstructures consist of blade and racemose shape of precipitates which are dispersed in glassy matrix. According to the EDS results, the blade and racemose precipitates can be related to Fe_3O_4 and $BaFe_{12}O_{19}$ phases, respectively.

By increasing the BaO/Fe_2O_3 ratio, the intensity of Fe_3O_4 and $BaFe_{12}O_{19}$ peaks decrease and these peaks vanish completely in sample with BaO/Fe_2O_3 = 1.8. By attention to these results, the fully amorphous phase only can be formed in sample with lower percentage of Fe_2O_3. The FE-SEM micrographs of this sample (Fig. 4) are in conferment with this result. This result is in agreement with Francis [16], Shirk and Buessem [17] reports for B_2O_3-BaO-Fe_2O_3 system.

3.1.2. Magnetic characterization

The hysteresis loops and magnetic properties of as-quenched BaO-Fe_2O_3-SiO_2-B_2O_3 with different composition are presented in Table 2. According to this table, several points can be concluded as:

1. There is no significant difference between magnetic properties of as-quenched samples with different BaO/Fe_2O_3 ratio.

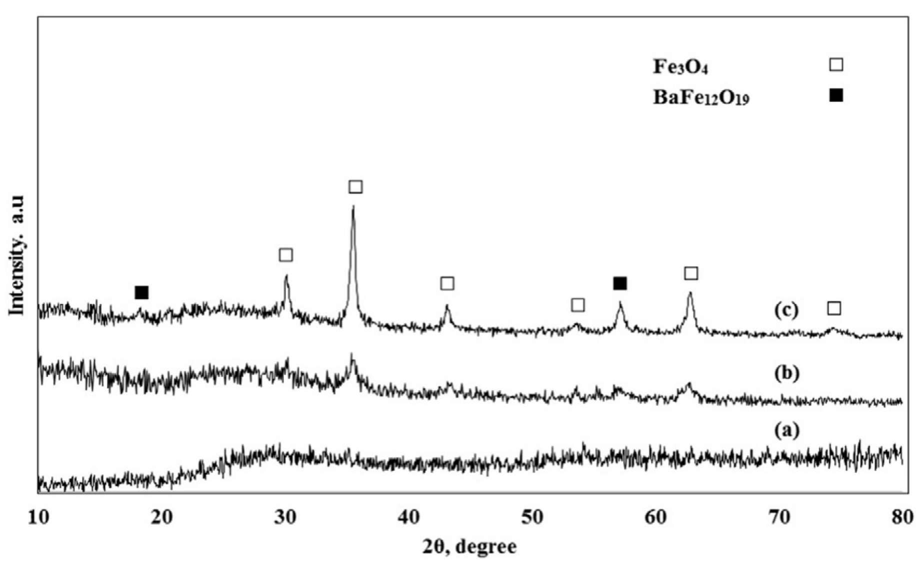

Fig. 2. The XRD patterns of as-quenched $20B_2O_3$-$10SiO_2$-(45 − x)BaO-(25 + x)Fe_2O_3 samples; a) x = 0 (BaO/Fe_2O_3 = 1.8), b) x = 10 (BaO/Fe_2O_3 = 1) and c) x = 20 (BaO/Fe_2O_3 = 0.56).