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Preparation and Magnetic Properties of Exchange Coupled Nanocomposite of $SrFe_{12}O_{19}$ and $Mn_{0.5}Zn_{0.5}Fe_2O_4$

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Abstract: Hard $SrFe_{12}O_{19}$ and soft $Mn_{0.5}Zn_{0.5}Fe_2O_4$ were prepared by solution combustion method and the composite was formed by mechanical mixing. XRD pattern of composite contains a small additional peak of α-Fe_2O_3 phase due to excess heat treatment. Hysteresis loop of composite indicates the presence of exchange coupling between the two phases. A small step in the demagnetization curve shows that the exchange coupling is weak. Coercivity of composite was obtained slightly less than that of hard material as predicted by theory. But remanence decreased due to the low value of saturation magnetization of soft ferrite when compared to hard ferrite.

Keywords: Nanocomposite; Exchange spring magnet; Exchange coupling; Magnetic Energy Product.

Introduction and Experimental:

Exchange spring magnets are on the cutting edge in the field of permanent magnetism. Kneller and Hawig put forward the concept of exchange hardening through mixing of nano-sized soft and hard magnetic materials [1]. It was proposed to improve the magnetic energy product $(BH)_{max}$ which is the figure of merit of permanent magnets. Although alloy composites possess high magnetic energy product, they have certain limitations such as poor corrosion resistance and high cost of raw materials. Extensive studies have been carried out on alloy composites to reduce the amount of rare earths to make the production more cost effective. Ferrite composites with enhanced magnetic energy product can overcome the above limitations up to an extent. The aim of present work is to investigate the effect of exchange coupling on magnetic properties of hard/soft $SrFe_{12}O_{19}/Mn_{0.5}Zn_{0.5}Fe_2O_4$ nanocomposite. To the best of our knowledge, the above system has not been studied so far.

The reagents used for synthesis were $Mn(NO_3)_2.4H_2O$, $Zn(NO_3)_2.4H_2O$, $Fe(NO_3)_3.9H_2O$ and $Sr(NO_3)_2$. $SrFe_{12}O_{19}$ and $Mn_{0.5}Zn_{0.5}Fe_2O_4$ ferrites were prepared separately by citrate nitrate auto-combustion method [2]. For the preparation of hard ferrite, $Fe(NO_3)_3.9H_2O$ and $Sr(NO_3)_2$ were dissolved in de-ionized water which was added to equimolar amount of citric acid solution under constant stirring. The pH of solution was adjusted to 6.5 by liquor ammonia. The aqueous mixture was heated to 90 ^oC and stirred until it turned to viscous gel. On further heating, the gel dried out and underwent spontaneous combustion which subsequently pre-sintered at 400 ^oC for 3 hr and later annealed at 1200 ^oC for 2 hr. Same procedure was adopted for the preparation of

$Mn_{0.5}Zn_{0.5}Fe_2O_4$[3]. Composite was formed by mechanical mixing of pure $SrFe_{12}O_{19}$ and $Mn_{0.5}Zn_{0.5}Fe_2O_4$ samples in 9:1 ratio followed by heating to 1200 ^oC.

All the samples were characterized by XRD to confirm the phase formation and the crystallite sizes were calculated using Scherrer formula, $D=K\lambda/(\beta Cos\theta)$, where variables have usual meaning. Room temperature magnetic properties of single phases and composite were investigated by Vibrating Sample Magnetometer (VSM).

Results and Discussion:

XRD pattern of hard ferrite, soft ferrite and composite are shown in Fig. 1. It has been confirmed that pure phases of both material were formed at 1200 ^oC. For samples sintered below 1200 ^oC, the spectra contain α-Fe_2O_3 peaks due to incomplete phase formation. Average crystallite sizes for $SrFe_{12}O_{19}$ and $Mn_{0.5}Zn_{0.5}Fe_2O_4$ are 39 nm and 38 nm respectively. The composite spectrum contains peaks corresponding to soft and hard ferrites which reveal the coexistence of both phases. However an additional peak of α-Fe_2O_3 shows presence of small percentage of additional phase in the composite due to excess heat treatment. The crystallite sizes of soft and hard phases increased further due to sintering and were obtained as 63 nm and 52 nm respectively.

Hysteresis loop of all the samples traced at a maximum applied field of 12.5 KOe are shown in Fig 2. Magnetic properties such as saturation magnetization (M_s), remanence magnetization (M_r), coercivity (H_c) and squareness ratio (α) are tabulated in Table 1. For composite, both saturation magnetization (47emu/g) and remanence magnetization (28 emu/g) are between that of soft and hard ferrite. One of the unique features of exchange spring magnet is remanence enhancement which was not observed here because of less saturation magnetization of soft ferrite when compared to hard ferrite and weak exchange coupling between the phases. There are several factors which influence the magnetic properties of a composite. In addition to the intrinsic properties, grain size, particle shape and distribution of both soft and hard phases substantially affect the exchange interaction between the phases. For strong exchange coupling, soft grain size should be about twice the domain wall width (δ_w) of magnetic hard phase [4]. The typical widths of domain wall for hard and soft ferrites are 9 nm and 25 nm respectively. Hence for optimum exchange coupling, soft and hard grain sizes should be in the range of 10-20 nm.

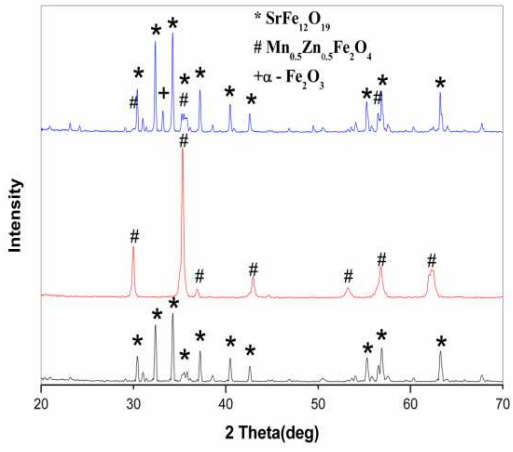

Fig. 1: X-ray diffraction spectra of $SrFe_{12}O_{19}$ ferrite, $Mn_{0.5}Zn_{0.5}Fe_2O_4$ ferrite and composite

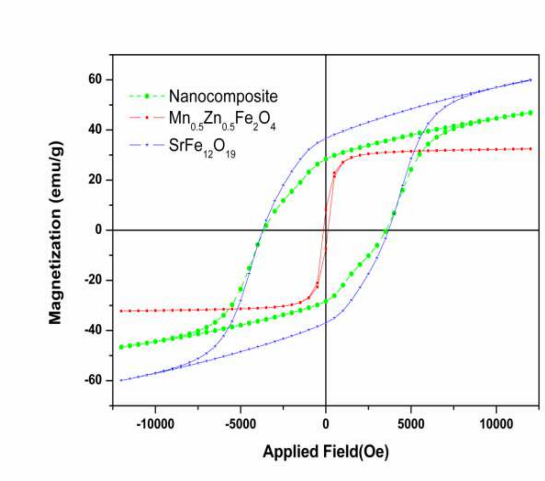

Fig. 2: Magnetic hysteresis loops of $SrFe_{12}O_{19}$ ferrite, $Mn_{0.5}Zn_{0.5}Fe_2O_4$ ferrite and composite

Table 1: Magnetic properties of individual ferrites and composite

Sample	M_s (emu/g)	M_r (emu/g)	$\alpha = M_r/M_s$	H_c (Oe)
$SrFe_{12}O_{19}$	60	37	0.62	3686
$Mn_{0.5}Zn_{0.5}Fe_2O_4$	32	8	0.25	134
$SrFe_{12}O_{19}$/ $Mn_{0.5}Zn_{0.5}Fe_2O_4$	47	28	0.60	3578

For overcritical dimensions, coupling will be weak and demagnetization curve will be of concave shape. The central region of soft phase will be decoupled from hard phase and when reverse magnetic field becomes equal to the reversal field of soft phase, a large reduction in saturation magnetization occurs which leads to a step in demagnetization curve as in Fig. 2 [5]. Coercivity of composite (3578 Oe) was reduced slightly due to additional demagnetizing field on the hard phase because of exchange interaction. This suggests that there exist coupling between phases. The squareness ratio for composite was obtained as 0.60 and is less than that of hard ferrite.

In conclusion, $SrFe_{12}O_{19}/Mn_{0.5}Zn_{0.5}Fe_2O_4$ composite of appreciable magnetic properties can be constructed by proper selection of preparation methods to control the microstructure. Synthesis methods which do not require high temperature sintering are preferred for the fabrication of ferrite composites to avoid grain growth and hence weakening of exchange coupling.

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