Pramāṇa, Vol. 19, No. 1, July 1982, pp. 103–106. © Printed in India.

Magnetic domain behaviour of $MgFe_2O_4$–$CoFe_2O_4$ system

VENKATESH RAO and H V KEER
Department of Chemistry, Indian Institute of Technology,
Bombay 400 076, India

MS received 15 March 1982

Abstract. Ferrites with the composition $Mg_{1-x}Co_xFe_2O_4$ $(0 < x \leqslant 0{\cdot}33)$ were synthesized by solid state reaction between MgO, α-Fe_2O_3 and $CoFe_2O_4$ at 1300°C. The compounds were analyzed from the results of x-ray diffraction, magnetic hysteresis and initial susceptibility (χ) studies. All the compounds of the series were found to be monophasic with no appreciable change in their lattice parameter. The saturation magnetization and coercive field (H_c) values increased with increasing cobalt ferrite content. Similarly, the shapes of the $\chi - T$ curves and temperature variations of H_c values indicated that the magnetic behaviour changes from multi-domain to single domain with increasing cobalt content. The results are similar to those observed for the $(MgFe_2O_4)_{1-x}$ (Co_3O_4) system.

Keywords. Mixed ferrites; magnetic domain behaviour; magnetic hysteresis; initial susceptibility.

1. Introduction

Ferrites have been the subject of extensive study because of their wide range of applications and of their importance in understanding the theories of magnetism. In this paper, the results of temperature variation of magnetic hysteresis parameters and initial susceptibility of $MgFe_2O_4$–$CoFe_2O_4$ system are reported and discussed. Both $MgFe_2O_4$ and $CoFe_2O_4$ are cubic-spinel ferrimagnets and their magnetic interaction can be explained on the basis of the Néel model (Néel 1948). However, $MgFe_2O_4$ has a negative crystalline anisotropy while $CoFe_2O_4$ has positive anisotropy. Therefore, an interesting variation in coercive field values may be expected for the mixed system. Recently it has been pointed out (Radhakrishnamurty and Nanadikar 1979; Nagarajan and Radhakrishnamurty 1981) that hysteresis and initial susceptibility studies at various temperatures can help to distinguish between multi-domain, single domain and super-paramagnetic behaviour. Therefore, an attempt has been made to use such studies and to determine the domain behaviour change, if any, as the concentration of cobalt ferrite is varied.

2. Experimental

Appropriate amounts of MgO and α-Fe_2O_3 were mixed with those of $CoFe_2O_4$ (up to 0·33 mole) prepared by the coprecipitation method (Hasegawa and Sato 1967), prefired at 700°C for 6 hr pelletized under $5\text{-}6 \times 10^6$kg/sq.m inch and finally sintered at 1300°C for 2-4 hr and then furnace cooled.

X-ray diffractograms of the starting materials as well as of the products after final sintering were recorded on a Phillips x-ray diffractometer (PW 1010) using MoK_α radiation ($\lambda = 0{\cdot}709$ Å) filtered through zirconium. The magnetic parameters such as saturation magnetization (σ_s) and coercive field (H_c) were calculated from the photographs of hysteresis loops taken at 80 and 300 K using an alternating current electromagnet-type loop tracer designed by Likhite *et al* (1965). Initial susceptibility studies were made from room temperature to 600°C using an instrument similar to the one reported by Radhakrishnamurty and Likhite (1970) and the Curie temperatures were determined.

3. Results and discussion

The relevant magnetic properties along with the lattice parameters and Curie temperatures for six samples have been listed in table 1; the hysteresis loops of some of them have been depicted in figure 1 and the χ-T curves in figure 2. The x-ray analysis indicated all the compositions to be monophasic cubic spinels. The unit cell parameter did not change appreciably from those of the parent compounds, $MgFe_2O_4$ ($a = 8{\cdot}38$ Å) and $CoFe_2O_4$ ($a = 8{\cdot}39$ Å). This may be attributed to the closeness of the ionic radii of Mg^{2+} (0·65 Å) and Co^{2+} (0·70 Å).

The results of the magnetic properties may be understood after recapitulating some of the pertinent observations made by Radhakrishnamurty and coworkers (Radhakrishnamurty and Nanadikar 1979; Nagarajan and Radhakrishnamurty 1981).

"Theoretically (Bean 1955) samples containing multi-domain (MD) or superparamagnetic (SP) particles should not show any hysteresis; practically, however some little hysteresis may arise due to defects and stresses in MD samples and interaction effects in SP ones. At low temperatures, there is a marginal increase in σ_r/σ_s for MD grains, while for SP samples, σ_r/σ_s increases considerably (Berkowitz and Schuele 1959). Similarly for MD samples, the relative increase in χ over the room temperature value is small and it sharply falls at the Curie temperature; a peak in the χ-T curve could be obtained only if the sample contains a substantial proportion of

Table 1. Structural and magnetic parameters for the $Mg_{1-x}Co_xFe_2O_4$ system.

No.	Nominal composition	Lattice parameter (Å)	Magnetisation σ_s (emu/g) 300 K	Remanence ratio at 300 K	Remanence ratio at 80 K	Coercive force H_c(Oe) 300 K	Coercive force H_c(Oe) 80 K	Curie Temp. (°C)
A_1	$Mg_{0.99}Co_{0.01}Fe_2O_4$	8·36	26·6	—	—	*	*	425
A_2	$Mg_{0.97}Co_{0.03}Fe_2O_4$	8·37	26·5	—	—	*	*	425
A_3	$Mg_{0.95}Co_{0.05}Fe_2O_4$	8·38	26·9	0·15	0·53	36	141	430
A_4	$Mg_{0.89}Co_{0.11}Fe_2O_4$	8·38	27·8	0·20	0·75	59	400	430
A_5	$Mg_{0.78}Co_{0.22}Fe_2O_4$	8·39	30·8	0·25	0·75	118	750	450
A_6	$Mg_{0.67}Co_{0.33}Fe_2O_4$	8·39	34·5	0·25	0·75	120	750	460

*Not decipherable with accuracy from hysteresis loop photographs but estimated to be < 10.

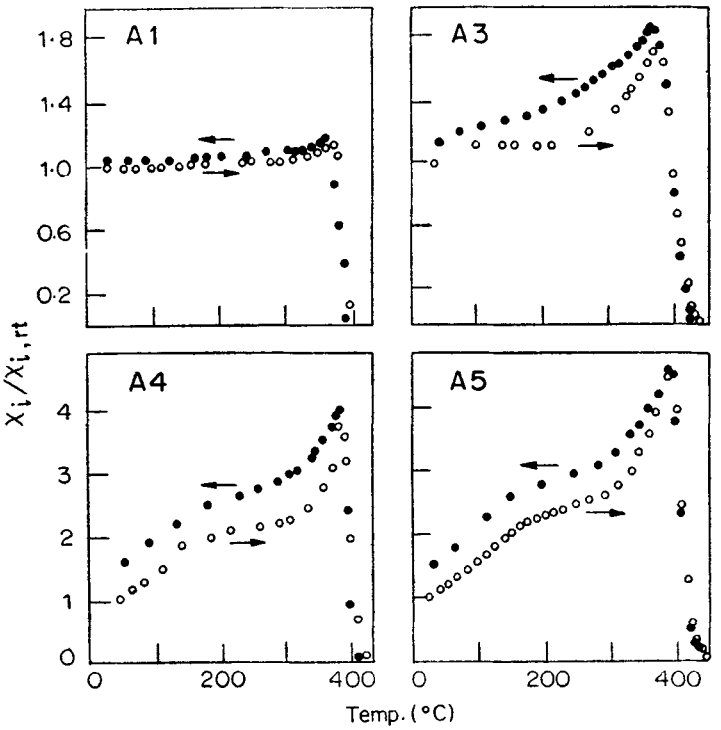

Figure 1. Magnetic hysteresis behaviour of $Mg_{1-x}Co_xFe_2O_4$ system.

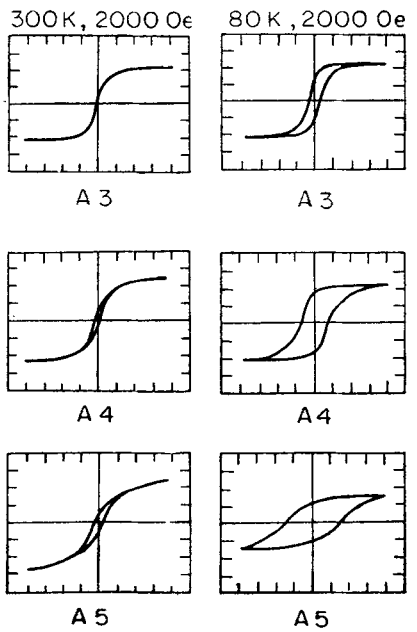

Figure 2. Temperature variation of normalized initial susceptibility of $Mg_{1-x}Co_x$ Fe_2O_4 system.

SD particles and it occurs at the blocking temperature (T_b) above which superparamagnetism sets in."

From the inspections of H_c values (table 1) and hysteresis loops at 80 and 300 K, the following points emerge:

(i) H_c increases with increase in $CoFe_2O_4$ content, which is not contrary to expectation, because the presence of Co^{2+} ions gives rise to large induced anisotropy due to relatively high (unquenched) orbital contribution to the magnetic moment.
(ii) For the samples containing up to 3 mole % $CoFe_2O_4$, MD behaviour prevails.
(iii) For compositions between 5 and 33 mole % $CoFe_2O_4$ a large increase in H_c on cooling to liquid nitrogen temperature is observed; this is indicative of SD behaviour.

The shapes of the χ-T curves generally corroborate the above observations; however, pure SD behaviour is not seen. Therefore, it may be concluded that there is at least a tendency for increase in the SD content with increasing $Co^{2+}/CoFe_2O_4$ concentration. Similar results were obtained by Venkatesh Rao (1975) and Radhakrishnamurty and Nanadikar (1979) for the $MgFe_2O_4$–Co_3O_4 system; which indicate that Co^{2+} ions, when incorporated, tends to introduce SD behaviour in the otherwise MD grains of a ferrite.

Acknowledgements

The authors are grateful to the late Prof. A B Biswas for constant encouragement and guidance and to Dr C Radhakrishnamurty, Tata Institute of Fundamental Research, Bombay, for critical comments. Further, one of them (VR) thanks the Council of Scientific and Industrial Research for the award of a fellowship.

References

Bean C P 1955 *J. Appl. Phys.* **26** 1381
Berkowitz A E and Schuele W J 1959 *J. Appl. Phys.* **30** 1345
Hasegawa K and Sato T 1967 *J. Appl. Phys.* **38** 4707
Likhite S D, Radhakrishnamurty C and Sahasrabudhe P W 1965 *Rev. Sci. Instrum.* **36** 1558
Nagarajan R and Radhakrishnamurty C 1981 *Bull. Mater. Sci.* **3** 217
Néel L 1948 *Ann. Phys.* **3** 137
Radhakrishnamurty C and Likhite S D 1970 *Earth Planet. Sci. Lett.* **7** 389
Radhakrishnamurty C and Nanadikar N G 1979 *Pramana* **13** 413
Venkatesh Rao 1975 *Synthesis and properties of some magnetic materials* Ph.D. Thesis, I I T., Bombay