CHAPTER - IV

HYSTERESIS AND MAGNETIZATION

CHAPTER - IV

Hysteresis and Magnetization Studies of Samarium doped Mg-Cd Ferrites

Section-A

4.1 Introduction

Ferrites are ceramic oxides that play an important role in electronics, mainly in the field of high frequency telecommunications. Material parameters have a greater say in deciding a particular application. The important physical properties of ferrites such as permeability, coercive force, hysteresis loss, depend to a greater extent on the grain size, porosity, inclusions etc. ie. on the microstructure of the product. A combined effort of various scientific and technical disciplines is under way for real improvements in the ferrite properties.

The magnetic parameters like saturation magnetization (σ_s), coercive field (H_c). remanance ratio (σ_r/σ_s) and permeability (μ) are related with hysteresis behaviour. Hysteresis study of ferrites, therefore, provides a valuable information on these parameters. On the basis of coercive field (H_c) ferrites have been classified into two catagories. Ferrites with low H_c are called as 'soft ferrites' and find use in the manufacturer of transformers, motors, generators and high frequency inductors in which high permeability, low hysteresis losses are essential requirements'. Ferrites with high H_c are called as 'hard ferrites' and are used as permanent magnets for various kinds of electric motors, loudspeakers, telephones and televisions for which high remanance is desirable.[2] The hysteresis properties are highly sensitive to crystal structure, heat treatment, chemical constituion, porosity and grain size.[3]

4.2 Magnetization

Spontaneous magnetization (M_s) is one of the most striking properties of a magnetic material. For many applications a high value of M_s is advantageous. In some microwave applications a material with low value of M_s is preferred.

Since the macroscopic moment is the sum of the moments of the atoms or ions in the solid, the following factors determine the value of M_s.

1) The magnitude of the atomic magnetic moments and the number of these ions per unit volume.

2) The magnetic ordering temperature (Curie temperature)

3) The pattern in which the magnetic moment are ordered i.e., all parallel or partly anti-parallel.

The magnetization in ferrites results from the distribution and alignment of magnetic ions on the octahedral and tetrahedral sites. The cations sitting on a certain sublattice are so oriented that their magnetic moments are in the same direction. For two successive sublattices moments are aligned anti-parallel to the moments of cations on the other sublattice as against ferromagnetism where they all are parallel. The parallel alignment of moments on each sub-lattice is a consequence of super exchange interaction[4] between the magnetic ions on one sublattice and those on the other. The parallel alignment of moments on each sub-lattice, which is a characteristics of ferrimagnetism, requires the sufficient concentration of the magnetic species on any one sub-lattice to maintain parallel alignment on the other neighbouring sub-lattice.

In 1948, Neel[5] explained ferrimagnetism in detail with the help of two sub-lattice model. The magnetic ions N per unit volume may be situated on A and B sites. The material possesses net magnetic moment for $\lambda \neq \mu$, where λ is a function of magnetization on A-site and $\mu = 1-\lambda$, is a function of magnetization on B-site. The unequal distribution of magnetic ions occurs either due to unequal number of sites on the two sub-lattices or due to the site preference of ions on one site over those on the other. the site preference depends on

i) ion size, site size,

ii) electron configuration of the ion, and

iii) symmetry and strength of the crystalline field at a site. For $\lambda = \mu$, material may exhibit ferrimagnetism if magnetic moments of ions on A and B sites are unequal. The unequality arises due to

i) same element in different ionic states, Fe^{2+}, Fe^{3+}.

ii) different elements in the same or different ionic states, e.g. Fe^{3+} or Co^{2+} .

iii) different crystalline fields acting at the two sites.

Also, non antiparallel arrangement may be possible which contributes to net magnetic moment.

4.3 Anisotropy

Anisotropy energy is by definition that part of the crystal energy that depends on the direction of magnetization. If this direction is given by its direction cosines with respect to the crystal axes, for cubic material the anisotropy energy contains terms such as

$$E_A = K_1 \left[\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2\right] + K_2 \, (\alpha_1^2\alpha_2^2\alpha_3^2) \qquad \ldots (4.1)$$

The anisotropy constants K_1, K_2 etc vary from one material to another and in addition are temperature dependent. The constants are determined by measuring the mechanical torque on a single crystal as a function of the direction of M. Anisotropy plays an important role in several magnetic properties such as susceptibility, resonance and magnetostriction etc.

4.4 Domain Structure

When sufficiently large magnetic field is applied to a ferro- or ferrimagnetic substance, the magnetization changes from zero to saturation value. Weiss explained this on the basis of his domain theory. The building blocks in this theory are the domains and the domain walls (or Bloch walls)[6,7]. The other independent variables describing the magnetic state are the direction of magnetization within the domains and the positions of the domain walls. The magnetization is constrained to be homogenous inside the domains and its directions to vary only within the domain walls.

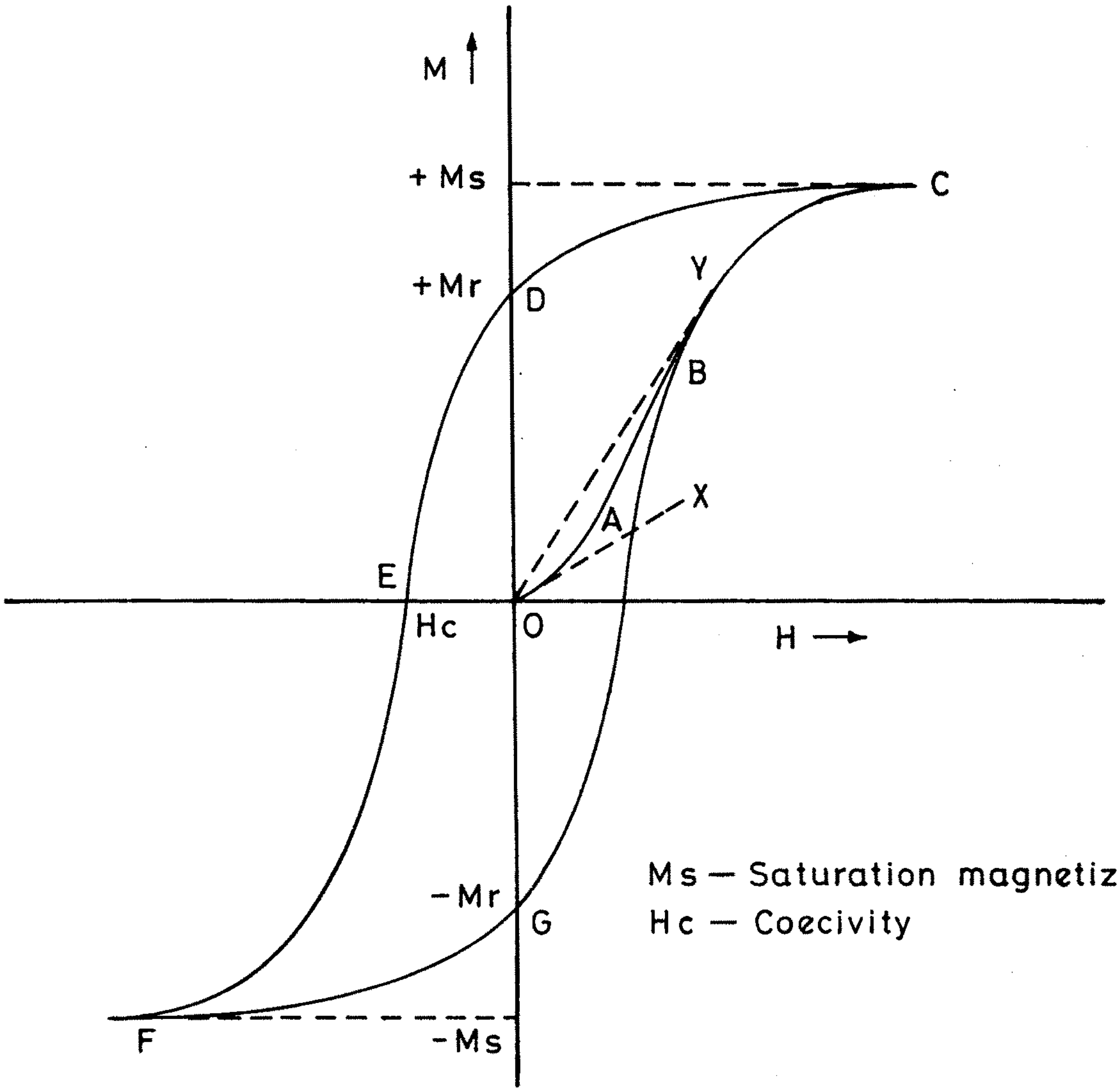
The origin of domain structure is based on the principle that a physical system tends to assume the lowest energy state when left to itself i.e. the free energy (F = E-TS) of a solid tries to reach a minimum value. In order to reduce the demagnetizing energy, arising due to the free poles at the surface, the crystal may be divided into a large number of domains or it may be reduced by the formation of 'closure domains'.

4.5 Hysteresis

When an external magnetic field is applied to a ferromagnetic substance its magnetization increases, with increasing applied field till it reaches the saturation value for a certain critical field. Now if the external field is decreased demagnetization is not recovered completely. Thus magnetization lags behind the applied field. This is known as the phenomenon of hysteresis. Weiss explained the hysteresdis phenomenon on the basis of the concept of magnetic domains.

The variation of magnetization with applied field for a virgin specimen is shown in Fig.4.1 Magnetization goes on increasing continuously with the increasing applied field and reaches a saturation value at a certain critical field. This behaviour is represented by the curve OABC and is called the magnetization curve. The variation of magnetization of the substance per cycle is indicated by the curve CDE FGC and is called as the 'hysteresis loop'. Area of this loop represents the loss of energy per cycle due to hysteresis.

Initial susceptibility (μ) is given by the slope of the tangent (OX) to the magnetization curve at the origin. Maximum



Ms — Saturation magnetization
Hc — Coecivity

Fig. 4·1 Magnetization curve(OABC) and Hysteresis loop (CDEFGC).

susceptibility occurs near the knee of the magnetization curve (slope - OY).

Retentivity or remanance (Mr) is the magnetude of magnetization of the specimen when field is reduced to zero (H=0). The value of reverse field for which magnetization is zero (M=0) is called coercive force (Hc). Saturation magnetization (Ms) is the maximum magnetization the specimen can permit. The coercive force and retentivity are the important properties in selecting the materials for permanent magnet applications.

Both irreversibility and hysteresis in ferromagnetic materials are attributed to impediments to the motion of domain walls offered by defects like inclusions, heterogeneity due to other phases and dislocations[8,9] The magnetization proceeds by reversible wall motion at small fields and above threshold field by irreversible wall motion, and at very high fields by irreversible rotations. Barkhausen jumps in magnetization are observed above the threshold field, when wall energy is maximum. This leads to irreversible increase in the magnetostatic and magnetoelastic energy of the material under the action of magnetic field. The thermal energy required for randomization and wall motion is not available. In addition, during magnetization reversal nucleation has to proceed before the sequence of reversible and irreversible wall motion and spin rotation can take place. Therefore for an unisolated specimen, the changes result in hysteresis during magnetization cycle signifying the energy loss. In soft magnetic materials magnetization reversal is produced by the nucleation and subsequent growth of domains of reversal magnetization due to wall motion.

4.6 Experimental

Experimental technique that are generally employed for the measurement of magnetic properties of ferrites are[10]

i) Ballistic method
ii) Vibrating coil magnetometer method
iii) Vibrating sample magnetometer method
iv) High field loop tracer method.

In the present work a high field hysteresis loop tracer was used for measuring the parameters viz. saturation magnetization (Ms), remance (Mr), etc. of the examples.

The high field hysteresis loop tracer (supplied by Arun electronics, Bombay) was used in the present case. It consists of an electromagnet operating on 50 Hz mains supply with the help of which a sinusoidal magnetic field of maximum peak value 3500 oersteds is produced in an air gap of about 1 cm. A special balancing coil gives a signal which after integration is proportional to the magnetic moment of the specimen and is fed to the horizontal plates of the oscilloscope. Thus CRO displays ~~magnetic~~ the hysteresis loop of the sample. The vertical deflection can be calibrated in terms of magnetic moment in emu and horizental scale in oersteds per division.

Measurement of M_s for each sample was carried out directly from CRO screen.Before introducing the sample in the balancing coil the stray fields, stray signals and phase mismatch were carefully got rid off with the help of controls provided. Even after introducing the sample the set up was once agains tested for above mentioned interferences keeping the current in the electromagnet at zero.The current was gradually increased and the hysteresis loop was obtained on the CRO screen. The vertical scale of the CRO (i.e. magnetization axis) was calibrated using standard nickel block having the saturation magnetisation of 53.54 emu/gm. Standard nickel block in the form of a pellet was loaded in between the pole pieces and the corresponding vertical scale displacement of hysteresis loop on CRO was noted. The procedure is repeated for another nickel block and calibration factor was calculated.

When the sample in the pellet form is introduced into the gap between pole pieces and R is the vertical scale reading on CRO in mV, the magnetization of the sample is given by

$$\sigma_s = R \times C.F. \qquad \ldots \quad (4.2)$$

$$\text{or} \quad \sigma_s = \sigma_s / m \quad \text{emu/gm}$$

The magnetic moment per molecule formula unit is given by the expression

$$\mu_B = \frac{\text{mol. wt.} \times \sigma'_s}{5585} \qquad \ldots \quad (4.3)$$

and magnetization M_s is given by the relation

$$M_s = (1 - p)\ dx\ .\ \sigma'_s \qquad \ldots \quad (4.4)$$

where p - porosity

dx - X-ray density

M - magnetization in emu/gm of the sample

To measure σ_s for different samples ($Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$ with x = 0.5 and y = 0, .1, .2, .3, .4, .5; sintered at 1000^{o}C for 40 hrs) it was introduced in the coil. The sample with x=0 is used as starting sample since it has maximum magnetization[11] and there arises minimum alterations in the setting of the apparatus for subsequent feeds of the samples.The current is slowly increased to get saturation magnetization. The same maximum current is used while taking the reading for other samples. Heights of saturation peak readings in mV for samples with y = 0, .1, .2, .3, .4, .5 were noted.

4.7 Results and Discussion

The representative hysteresis loops of our samples are shown in figures 4.2. The magnetization data for the present samples is given in tables 4.1 and 4.2. It can be observed from our results that the saturation magnetization and magneton numbers of the samples initially increase and then decrease with samarium content. This is due to the reduction of A-B interaction. According to Neel's model the Curie temperature for a series of ferrites is proportional to the product of the iron (Fe^{3+}) contents on the two sites and their inter sublatticeFe_A-O-Fe_B distances and angles. In our system substitution of Sm ions changes the iron ion concentration from 2 to 2-y thereby decreasing the number of ferrous ions on the octahedral sites.

The reduction of T_c can also be related to the reduction in Fe^{3+} ions on octahedral site on increasing of Sm concentration.

Table - 4.1

Hysteresis parameters and properties for ferrite samples

$Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$ (x = 0.5)

Y	Porosity	Saturation Magnetization $4\pi M_s$ gauss	Squareness ratio Mr/Ms
0	17.93	475.24	0.4
0.1	29.792	504.51	0.4
0.2	50.91	314.50	0.5
0.3	35.39	349.11	0.5
0.4	29.22	283.89	0.4
0.5	52.24	221.42	0.6

Table - 4.2

Data on molecular weight, magnetic moment at low temperature and at room temperature for samples $Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$ (x=0.5)

Y	Molecular weight	Magnetic moment n_B at low temp.	room temp.
0	244.05	-	1.6524
0.1	253.50	0.9039	1.8220
0.2	262.95	0.5055	1.1781
0.3	272.40	0.6814	1.3548
0.4	281.85	0.5952	1.1399
0.5	291.30	0.4083	0.9189

SECTION - B

PERMEABILITY

4.8 Introduction

In ferrites, permeability depends on microstructural conditions. The value of permeability with wide range makes the material suitable to various frequency range applications. When a magnetic material is subjected to an a.c. field it produces rapid movement of magnetic domain walls and hence the permeability is affected. The presence of any imperfections in the ceramic structure also reduces the permeability. The permeability is normally taken to be the real part of initial permeability. Schleomann[12,13] represented the formula for permeability in completely demagnetized state. The grain boundaries act as impediments to domain wall motion. This study was carried out by Bear and Schwartz[14], Perdujin and Peloschek[15] and Roses[16]. Most of the additives are simple oxides, having a very low melting point compared with the ferrite sintering temperature or being able to form low melting point phases in the spinel matrix. The increase in permeability is supposed to be originated while sintering by the presence of a liquid phase in the grain boundaries promotinf both sintering and grain growth[17,18,19]. Tasaki and Izushi[20] have studied the influence of sintering temperature on permeability.

The partially magnetized states are very important for the devices such as circulaters operating below ferromagnetic resonance, latching phase shifters and so on. The permeability spectra at microwave range is therefore essential. There are many microwave devices which operate in the region below saturation. Measurements of complex permeability of ferro and ferrimagnetic materials are often carried out at different points on the hysteresis loop with an audio frequency magnetic field.

4.9 Experimental

For the determination of permeability toroids were made with the help of a die having inner diameter of 1.0 cm and outer diameter of 1.50 cm. An insulated Cu wire was wound on toroide so as to form the core. By using a digital LCR meter (Aplab model 4910), the values of inductance and Q factor were noted with increasing temperature for the test frequency of 1 K Hz. The readings were noted at an interval of 25°C from room temperature to the Curie temperature of the sample. The temperature of the furnace was measured by chromel-alumel thermocouple. The permeability was calculated using the relation

$$L = 0.0046\, \mu\, N^2 h \log_{10} \left(\frac{OD}{ID} \right) \qquad \ldots \quad (4.5)$$

where L - is inductance, N is the number of turns on toroids, μ is the permeability of core, h is the thickness of the toroid and OD and ID are outer and inner diameters of the toroid respectively.

4.10 Results and Discussion

Polycrystalline ferrite is a complex system composed of crystallites, grain boundaries and pores. The magnetic properties of these ferrites are determined by chemical composition, porosity, grain size etc. Many studies on the composition related to magnetic properties have been made[21]. Several workers have attempted to clarify the effect of porosity and grain size on these properties[22,23].

The initial permeability has been observed to decrease with increasing concentration of Sm in the present system. According to Wijn et al[24], the initial permeability of polycrystalline spinels is given by

$$\frac{\mu_i - 1}{2\pi} = \frac{M_s^2}{k_1} \qquad \ldots (4.6)$$

where M_s is saturation magnetization and k_1 is the anisotropy constant. This equation is used to calculate k_1 from the observed values of M_s and μ_i. According to this equation the initial permeability is proportional to the square of saturation magnetization, hence the saturation magnetization mainly affects the initial permeability. Also, the soluble non-magnetic

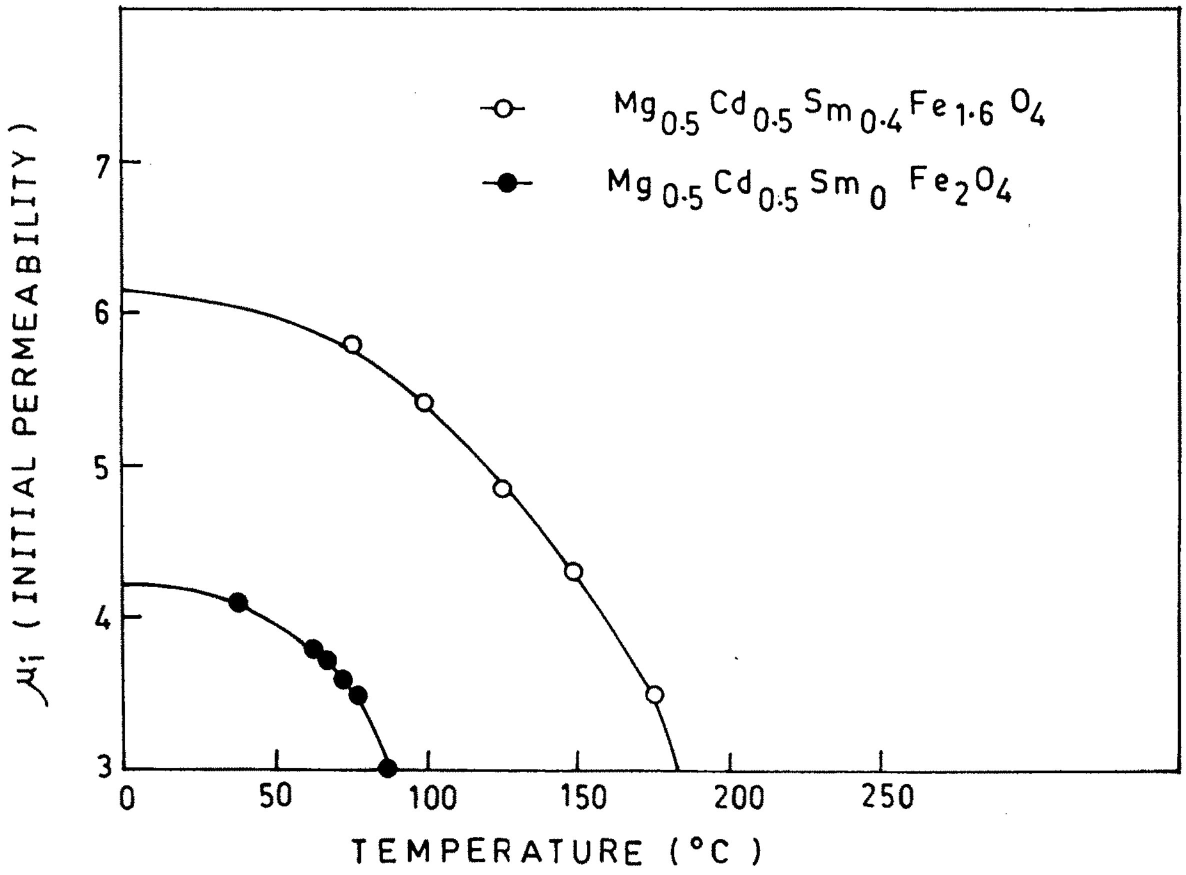


FIG. 4.3 — VARIATION OF INITIAL PERMEABILITY WITH TEMPERATURE OF SYSTEM $Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$.

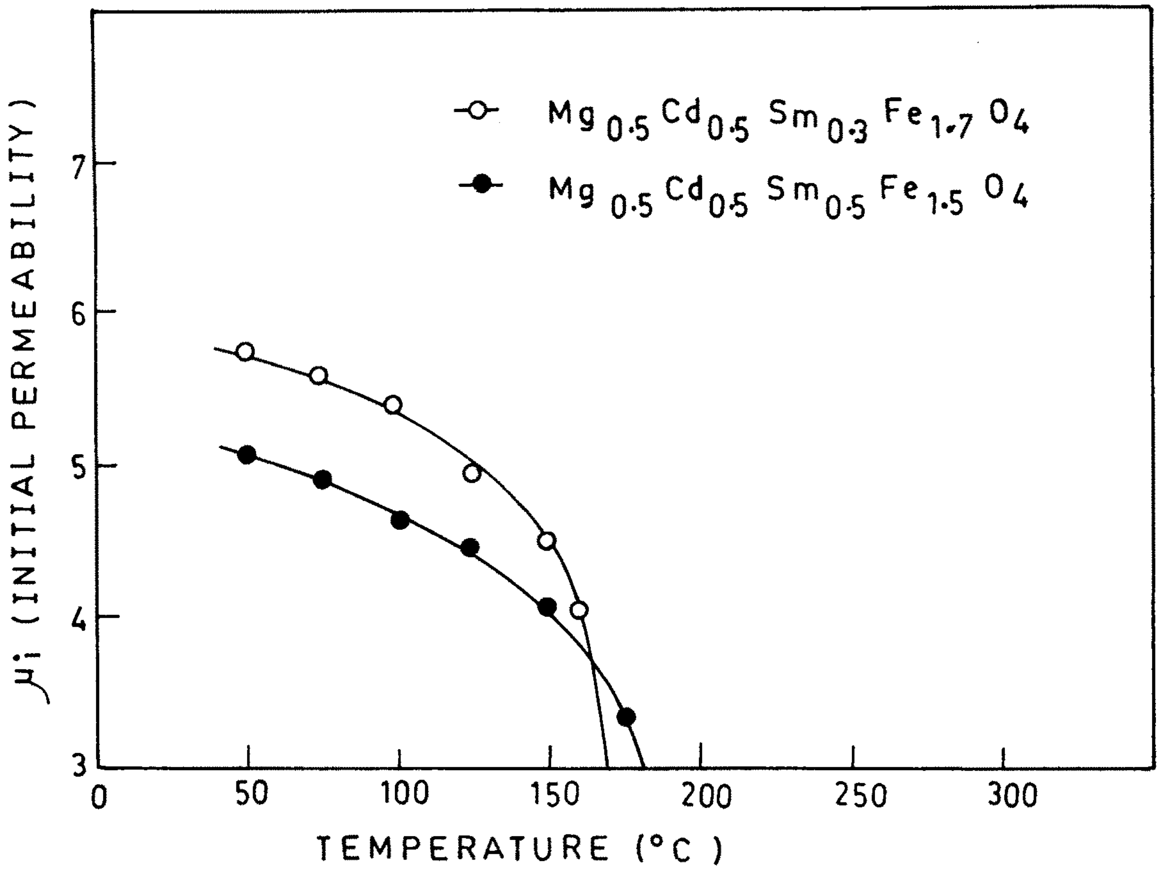


FIG. 4·4 – VARIATION OF INITIAL PERMEABILITY WITH TEMPERATURE OF SYSTEM $Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$.

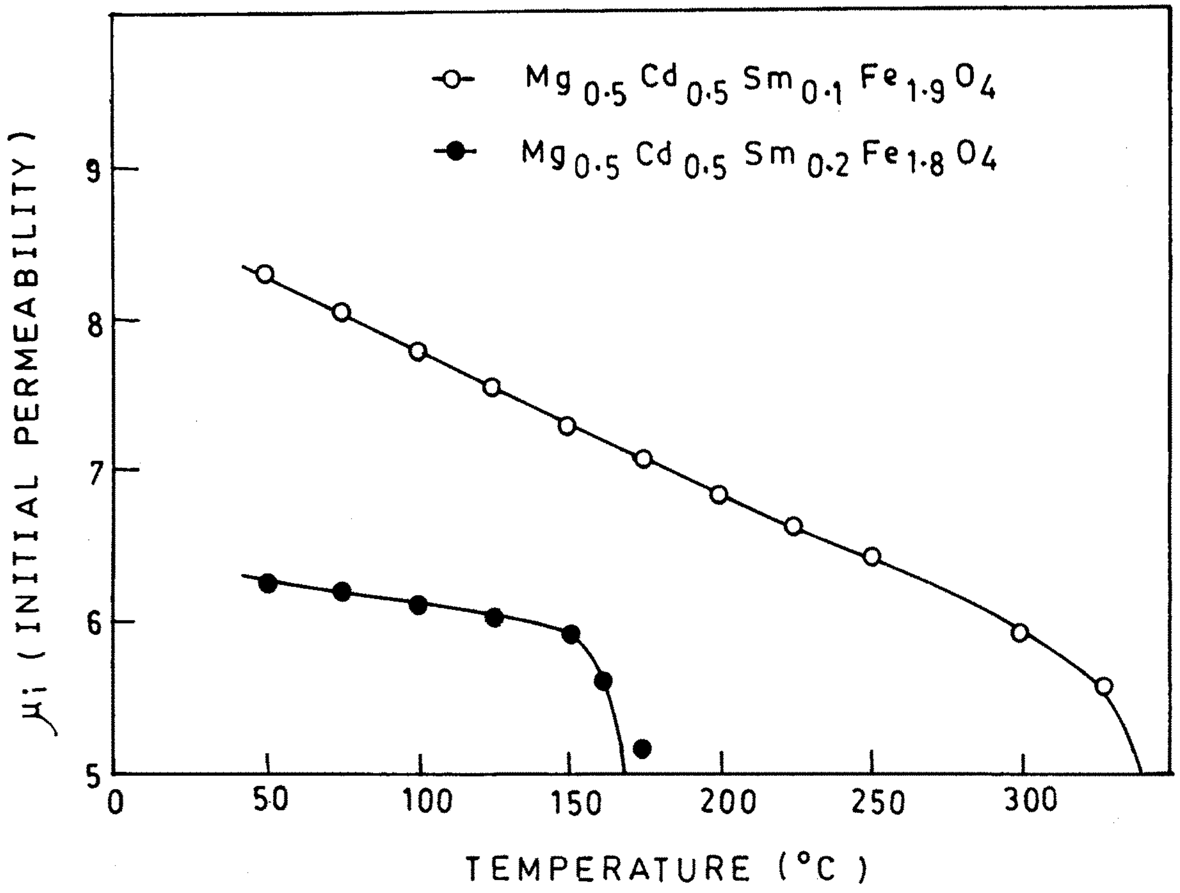


FIG. 4·5 — VARIATION INITIAL PERMEABILITY WITH TEMPERATURE FOR SYSTEM $Mg_xCd_{1-x}Sm_yFe_{2-y}O_4$.

impurities inside the grain hinder the reversible motion of domain walls and lower initial permeability.

The variation of initial permeability (μ_i) with temperature is shown in figures 4.3, 4.4 and 4.5.Initial permeability drops to zero at certain temperature.Some ferrite samples show slight tailing effect. This is attributed to minor impurity phases or weak interactions in the magnetization. The temperature at which μ_i drops to zero nearly coinsides with observed Curie temperature by other methods.

The permeability studies of magnesium-cadmium ferrites doped with samarium show that it decreases withthe addition of samarium since samarium replaces iron ions on octahedral site..

REFERENCES

1. Hoh S.R., Evaluation of high performance core materials, Tel. Tech. Vol.2 (1953) 86.
2. Richards E.K. and Lynch A.C., Soft magnetic materials for Telecommunications Per Jamon Press Ltd., (1953).
3. Lax B. and Butten K.J., Microwave ferrites and ferrimagnetics, Mc Graw Hill Book company, N.Y. (1962).
4. Sinha A.P.B. and Menon P.G. "Solid State Chemistry", Marcel Dekker N.Y., P. 384 (1974).
5. Neel L., Ann Phys., 3, 137 (1948).
6. Smit J. and Wijn H.P.J. "Ferrites", Centrex, Eindhoren (1961).
7. Chikazumi S., 'Physics of magnetism' J. Wiley and Sons N.Y. (1964).
8. Beaker R., Phys. Zeits 33, P. 906 (1932).
9. Kondorsky E., Physik 2. Sowjethnion 11, p.597 (1937).
10. Gorter E.W., Philips Res. Repts., (Netherlands) 9, p.321, 401 (1954).
11. Standly K.J., "Oxide Magnetic Materials" Oxford Univer. Press, London P. 64 (1962).
12. Schloemann E., J. Appl. Phys. 41 (1970) 204.

13. Schloemann E., J. Appl. Phys. 32 (1971) 443.

14. Beer A. and Schwartz T., IEEE. Trans. Magnet. 2 (1966).

15. Perduijn and Peloschek, Proc. Brit. Geram. Soc. 10 (1968) 283.

16. Roses Angew, Phys. 21 (1966) 391.

17. Jain G.C., Das B.K. and Kuman S. Proc. Int. Mag. Conf. 33-6 IEEE. Mag. Soc. (1978).

18. Takama E., Mishima S. and Kawahara S., J. Physique, 38, Suppl.-4, C-1 (1977) 349.

19. Craik D.J., 'Magnetic oxides' Wiley and Sons, London (1975).

20. Tasaki J. and Izushi T., J. Physique 38, suppl. 4, C-1 (1977) 175.

21. Smit J. and Wijn H.P.J., "Ferrites" Wiley and Sons, New York (1965).

22. Globus and Duplex P., IEEE, Trans. Mag. Maj. 2(3) (1960) 441.

23. Ernst Schloemann : IEEE Trans. Mag. Maj. 6 (1) (1970) 75.

24. Smit J. and Wijn H.P.J. : Advances in Electronics and Electron Physics (volume VI Physical properties of ferrites) Edited by L. Marton (1954) Academic Press Inc. Publishers, N.Y.