

## **DIELECTRIC AND ELECTRICAL PROPERTIES OF Cr SUBSTITUTED Mg FERRITES**

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### **ABSTRACT**

The spinel ferrites  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ( $0.0 \leq x \leq 1.0$ ) were prepared through the solid state reaction using conventional ceramic method at  $1300^\circ\text{C}$  in air. The homogeneous phase of the ferrite samples was observed from the X-ray diffraction study. Lattice parameter of the samples was found to decrease with increasing Cr concentration in the system obeying Vegard's law. The ac electrical resistivity, measured as a function of temperature, decreases with the increase of temperature indicating the semiconducting nature of all the samples. The activation energies were calculated and found to decrease with increasing Cr content. The lower activation energies are associated with higher electrical conductivity. With the increase of temperature, dielectric constant ( $\epsilon'$ ) and dielectric loss tangent are observed to be increased; while with the increase of frequency, dielectric constant ( $\epsilon'$ ) and dielectric loss tangent decrease for all the samples.

Key words: Ferrites, Dielectric properties, Resistivity, Activation energy

### **INTRODUCTION**

Spinel ferrites have gained much importance in the recent years because of their high resistance and magnetic characteristics. In the last two decades a good number of works have been carried out on spinel systems. Ferrites are one of the most studied magnetic systems and there have been a lot of experimental results and theoretical investigations on this material (Zakaria *et al.* 2003, 2004, 2005; Hilpert 1909, Lee 1999). Ferrites are especially convenient for high frequency uses because of their high resistivity. The electrical conductivity and dielectric behavior of ferrites are strongly depended on the sintering time and temperature, chemical composition, preparation conditions. A good number of studies on electrical conductivity of different ferrite samples with varying compositions have been reported by different workers (El Hiti 1990, Suryavanshi *et al.* 1991, Manjura Hoque *et al.* 2002).

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The study of dielectric properties provides valuable information on the behavior of localized electric charge carriers leading to greater understanding of the mechanism of dielectric polarization in ferrite system. The dielectric behavior of Nb doped Ni-Zn ferrite was studied earlier and observed that dielectric constant is of the order of  $>10^6$  by Rao *et al.* (2004). Normally in ferrites, the dielectric constant ( $\epsilon$ ) decreases with the increase of frequency. This decrease is rapid at the low frequency region and becomes slower at high frequencies. With the increase of temperature, dielectric constant ( $\epsilon$ ) was found to be increased. This behavior was observed in Ni-Zn, Co-Zn and Mg-Zn ferrites by Ahmed and Wasfy (2003) and Kumar *et al.* (2003). The local displacement of localized charge carriers includes dielectric polarization in ferrites.

The surface mounting devices have been rapidly developed for electronic applications, such as multilayer chip beads or inductors. They are the important components of technologically important products, such as cellular phones, video cameras, notebook computers and hard drives. Among available materials, polycrystalline ferrites have been widely used because of their high permeability in the RF region and environmental stability. There has been a growing interest in MgCr ferrites for the application in producing multilayer-type chips because these oxides can be sintered at relatively low temperature with a wide range of compositions.

The present work focuses on the study of structural, electric and dielectric properties of  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ferrites prepared in the solid state sintering technique.

## MATERIALS AND METHODS

Polycrystalline ferrite samples  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  with ( $x = 0.0, 0.2, 0.4, 0.6, 0.8$  and  $1.0$ ) were prepared by solid state sintering method at the Institute of Nuclear Science & Technology (INST), Atomic Energy Research Establishment, Dhaka, Bangladesh. The starting materials were high purity MgO,  $\text{Cr}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ . The materials were mixed thoroughly in correct stoichiometric proportions and wet blended with acetone in an agate mortar and pestle for about 2 hours and then ball milled for 6 hours in distilled water to produce a homogeneous mixture of the constituents. The finely mixed powders were dried and presintered at  $800^\circ\text{C}$  for 8 hours in a furnace. The presintered powders were mixed again in an agate mortar. The resulting powders were mixed with 1 wt.% polyvinyl alcohol (PVA) as a binder and pressed into small pellets discs. The compacts were then sintered in air at a temperature of  $1300^\circ\text{C}$  in a muffle furnace for 5 hours and finally cooled down to room temperature. X-ray diffraction measurements of all the samples were carried out in order to check their quality and phase purity using Phillips X'Pert PRO X-ray diffractometer with  $\text{CuK}_\alpha$  radiation ( $\lambda = 1.54178 \text{ \AA}$ ) at the Atomic

Energy Centre, Dhaka. The X-ray diffraction patterns, recorded in the angular range  $15^\circ \leq 2\theta \leq 65^\circ$  with a step size of  $0.02^\circ$ , showed sharp lines corresponding to single-phase cubic spinel structure for all the samples.

The bulk density of the pellet samples was determined after sintering the samples at  $1300^\circ\text{C}$ . The theoretical density of the samples has been calculated using the lattice parameter determined from X-ray diffraction data. The surfaces of  $\sim 1\text{mm}$  thick pellet samples were well polished and air-dried. Silver paste was applied on the flat surfaces to form electrodes for electrical measurements. The ac resistivity was measured by using a two-terminal method with electrodes connected to an electrometer (Keithley 6514, USA). The discs were placed between two electrodes inside the evacuated silica tube to avoid moisture absorption on the surface. The silica tube was supported within a furnace and a Ni-NiCr thermocouple to control and measure the temperature. The same specimen was used for measuring frequency and temperature dependent dielectric properties. The capacitance (C) of the samples was measured using a LCR meter (Hewlett Packard Impedance Analyzer, Model: 4192A) in the frequency range 1kHz to 1MHz. The dielectric constant ( $\epsilon'$ ) for all the samples was calculated. The resistivity of the pellet samples was measured by the same impedance analyzer from room temperature up to  $300^\circ\text{C}$ . From the temperature dependence of the resistivity data the activation energy of the samples were calculated.

## RESULTS AND DISCUSSION

X-ray diffraction (XRD) patterns of the  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ( $0.0 \leq x \leq 1.0$ ) ferrite samples at room temperature showed very sharp Bragg peaks indicating the formation of homogeneous and single phase cubic spinel structure corresponding to the space group  $\text{Fd}\bar{3}m$  for all the samples. One such representative XRD pattern for  $x = 0.6$  is shown in Fig. 1. The lattice constants were calculated from XRD data for all the samples. The lattice constant decreases with increasing  $\text{Cr}^{3+}$  ions concentration in the spinel oxides  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ . This was because  $\text{Cr}^{3+}$  ions have smaller ionic radii ( $0.53\text{\AA}$ ) than those of  $\text{Fe}^{3+}$  ions ( $0.645\text{\AA}$ ) in the octahedral sites. Therefore, a partial replacement of the  $\text{Fe}^{3+}$  ions by the  $\text{Cr}^{3+}$  ions caused shrinkage of unit cell dimensions, thereby decreasing the lattice parameter in the system (Shukla *et al.* 1999). This can be explained on the basis of the relative ionic radii of  $\text{Cr}^{3+}$  and  $\text{Fe}^{3+}$  ions present in the system. The lattice parameter decreased gradually with increasing  $x$  values satisfying the Vegard's law (Vegard 1921). The difference in the lattice parameter may also be explained by the cation migration since  $\text{Cr}^{3+}$  ions on the A-site exhibit a smaller ionic radius than on the B-site because of the covalence effects (Faizun Nesa *et al.* 2012).

The X-ray density ( $\rho_x$ ) of the samples has been calculated using the lattice parameter obtained from X-ray diffraction measurements. The bulk density ( $\rho_B$ ) was determined from the pellet samples sintered at 1300°C. The porosity of the samples has been determined from X-ray density and bulk density using the equation  $P = (1 - \frac{\rho_B}{\rho_x}) \times 100$ .

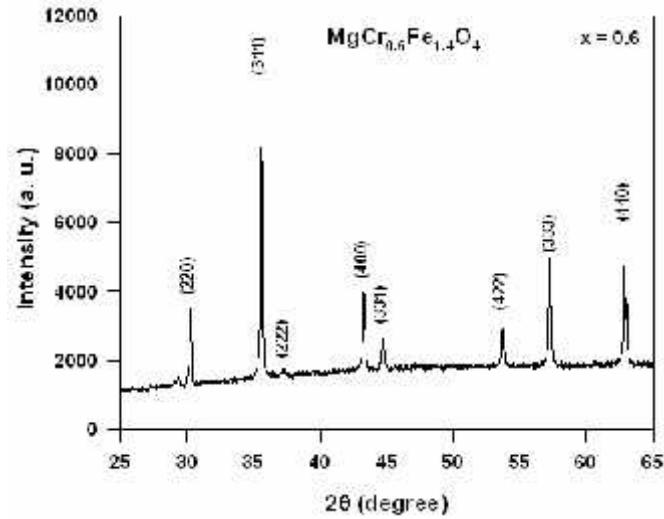


Fig. 1. X-ray diffraction pattern for the spinel oxide  $\text{MgCr}_{0.6}\text{Fe}_{1.4}\text{O}_4$  ( $x = 0.6$ ) showing single phase structure.

The lattice parameter, X-ray density, bulk density and porosity of  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ferrites sintered at 1300°C are presented in Table 1. The bulk density is found to be little lower than X-ray density. This is may be due to the existence of pores, which were created

**Table 1. Lattice parameter ( $a_0$ ), X-ray density ( $\rho_x$ ), bulk density ( $\rho_B$ ) and porosity (P) of  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ferrites sintered at 1300°C.**

Cr content (x)	Lattice parameter $a_0$ (Å)	X-ray density $\rho_x$ (gm/cc)	Bulk density $\rho_B$ (gm/cc)	Porosity P (%)
0.0	8.3740	4.55056	4.05323	10.9290
0.2	8.3690	4.54166	4.04524	10.9304
0.4	8.3670	4.52735	4.03508	10.8732
0.6	8.3660	4.51238	4.02339	10.8366
0.8	8.3610	4.50305	4.01327	10.8766
1.0	8.3590	4.48883	4.01066	10.6424

during the sample preparation or the sintering process. The X-ray density and bulk density decrease significantly with the increase of Cr concentration in the system. This is because of the greater atomic weight of Fe (55.845 gm/mol) than that of Cr (51.996 gm/mol). Porosity of the samples changes slightly with Cr content (x) in the system as shown in Table 1.

The variation of dielectric constant  $\epsilon'$  with frequency is shown in Fig. 2 for  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ferrites sintered at  $1300^\circ\text{C}$ . From the figure it is observed that the dielectric constant decreases with increasing frequency for all the samples exhibiting a normal dielectric behavior of ferrites. The dielectric dispersion is rapid at lower frequency region and it remains almost independent at high frequency side. The incorporation of Cr into Mg ferrites has no pronounced effect on the dielectric constant in high frequency, but it significantly decreases in the low frequency range. This is in agreement with earlier observations

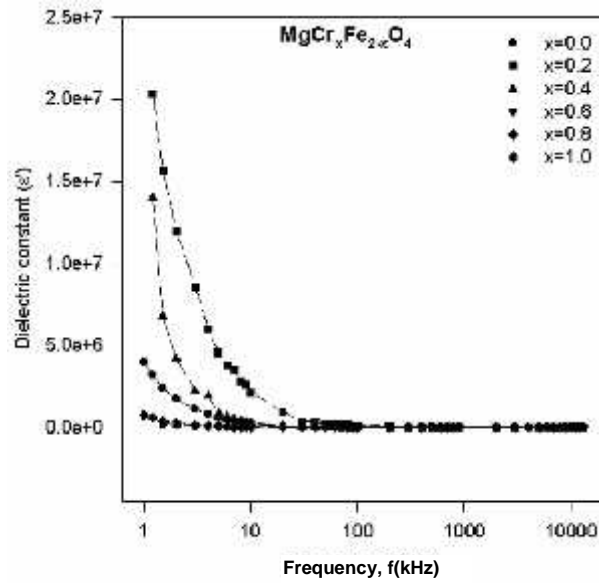
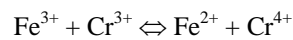


Fig. 2. Variation of dielectric constant ( $\epsilon'$ ) with frequency for the spinel oxides  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

This is in agreement with earlier observations of some other workers on Cr substituted spinel ferrites (Kuthari *et al.* 1990, Bhaskar *et al.* 2004). The dielectric behaviour of ferrites may be explained on the basis of the mechanism of dielectric polarization process and is similar to that of the conduction process. The electronic exchange  $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$  gives the local displacement of electrons in the direction of applied electric field, which induces the polarization in ferrites (Kuthari *et al.* 1990, Iwauchi 1971). In the present system, the following equilibrium may exist during sintering:



The magnitude of exchange depends on the concentration of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ion pairs present on B site for the present system. The samples with  $x = 0.2$  and  $0.4$  showed the maximum dispersion whereas for the samples with  $x = 0.6 - 1.0$  showed a least frequency dependence. The presence of  $\text{Fe}^{2+}$  ions in excess amount favors the polarization effects. Thus, the more dispersion observed in the sample with  $x = 0.2$  and  $0.4$  can be attributed to the presence of  $\text{Fe}^{2+}$  ions in excess amount which could be formed at elevated sintering

temperature. Similarly, the weak dependence of dielectric constant on frequency in  $x = 0.6-1.0$  samples can be due to the lack of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ions concentration. The observed decrease in dielectric constant with increase in frequency above certain frequencies the electronic exchange between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions is not dependent on the applied AC field.

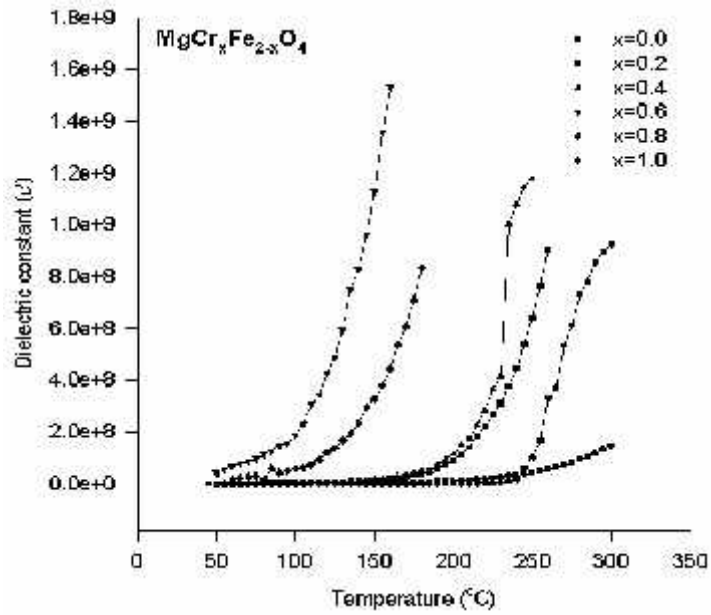


Fig. 3. Temperature dependence of the dielectric constant ( $\epsilon'$ ) for the spinel oxides  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

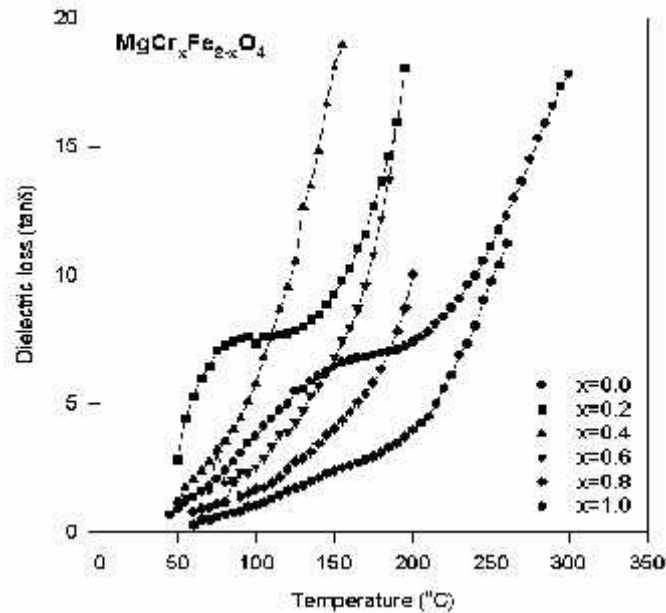


Fig. 4. Temperature dependence of the dielectric loss ( $\tan\delta$ ) for the spinel oxides  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

The temperature dependence of the dielectric constant  $\epsilon'$  and dielectric loss  $\tan \delta$  at 100 kHz is illustrated in Figs 3 and 4, respectively. As shown in Figs 3 and 4,  $\epsilon'$  and  $\tan \delta$  increase as the temperature increases, which is the normal dielectric behavior of magnetic semiconductor ferrites and the maximum value of temperature dependence dielectric constant is observed for the sample with  $x = 0.6$ . This is in accordance with the results observed earlier on some ferrites system (Ahmed *et al.* 1995, El Hiti *et al.* 1995, Ahmed *et al.* 1996). The hopping of electrons between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions (at adjacent B-sites) is thermally activated on increasing the temperature. The hopping of these strongly localized electrons in the d-shell causes local displacements in the direction of applied AC field. The local displacements of electronic charge carriers cause the dielectric polarization in ferrites. The dielectric behavior of ferrites may be explained on the basis of dielectric polarization process is similar to that of the conduction mechanism and are mainly by the hopping conduction mechanism (Bellad and Chougule 2000). As the temperature increases, the ac electrical conductivity increases due to the increase in the drift mobility of thermally activated electrons. As a result, the dielectric polarization increases causing an increase in  $\epsilon'$  and  $\tan \delta$ .

The increase in resistivity is observed with increasing Cr concentration for the spinel ferrites  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$  ( $0.0 \leq x \leq 1.0$ ) as shown in Table 2. In ferrites, the resistivity  $\rho$  at an absolute temperature T is given by the relation

$$\rho = \rho_0 e^{\frac{E_a}{K_B T}}$$

where,  $E_a$  is the activation energy and  $K_B$  is the Boltzmann constant.

In these polycrystalline materials, cations are surrounded by closed-packed anions and can well be treated as isolated from each other to a first approximation. The electrical conduction in ferrites can be explained by the Verwey model of electron hopping (Verwey and de Boer 1936) which involves exchange of electrons between ions of the same element present in different valence states, and distributed randomly over crystallographically equivalent lattice sites. In the present system, the conduction is taken to be due to the exchange of 3d electrons between ferrous  $\text{Fe}^{2+}$  and ferric  $\text{Fe}^{3+}$  ions in the octahedral site. The observed increase in resistivity with Cr concentration can be understood by considering cation distribution along with the hopping mechanism  $\text{Fe}^{2+} + \text{Fe}^{3+} \Leftrightarrow \text{Fe}^{3+} + \text{Fe}^{2+}$ . Since both  $\text{Fe}^{2+}$  and  $\text{Mg}^{3+}$  ions have a strong preference on B sites, they can cause conduction by the hopping mechanism. The substitution of  $\text{Cr}^{3+}$  for  $\text{Fe}^{3+}$  ions in  $\text{MgFe}_2\text{O}_4$  ferrites have a similar effect on resistivity, except that  $\text{Cr}^{3+}$  ions, due to their strong preference for B sites, can even displace small proportion of  $\text{Mg}^{2+}$  ions from B to A sites (Bradley 1971). This leads to the migration of an equal number of  $\text{Fe}^{3+}$  ions from A to B site which restrains the fast increase in resistivity. Thus, resistivity increases

slowly for  $x < 0.6$ . The rapid increase in resistivity for  $x > 0.6$  may be due to the dilution effect of  $\text{Cr}^{3+}$  ions wherein the increased concentration of  $\text{Cr}^{3+}$  ions inhibits the conduction process.

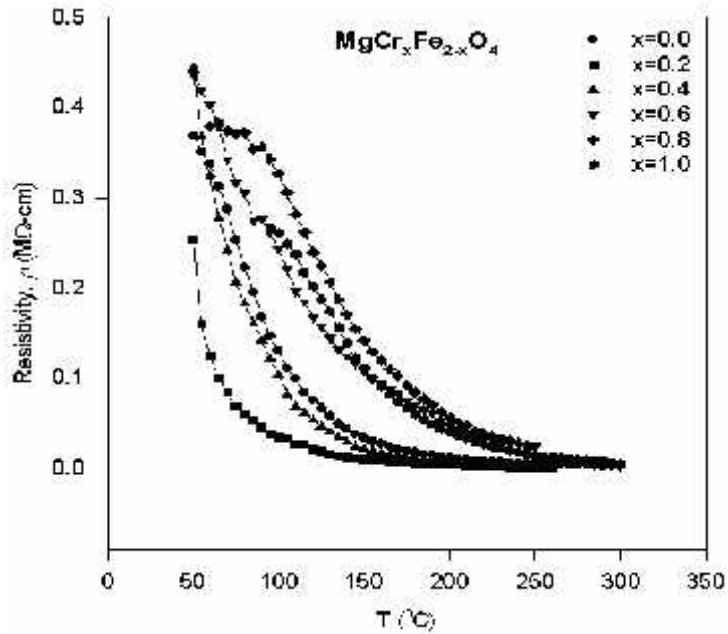


Fig. 5. Resistivity ( $\rho$ ) as a function of temperature for the spinel system  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

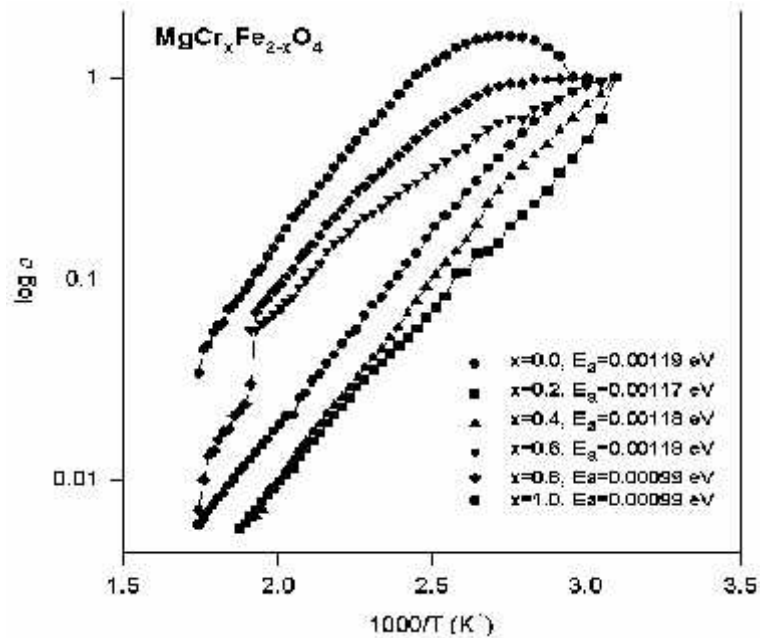


Fig. 6. Resistivity ( $\log \rho$ ) as a function of  $1000/T$  ( $\text{K}^{-1}$ ) for the spinel system  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .



The temperature dependence of resistivity for all the samples is shown in Fig. 5. The resistivity as a function of the reciprocal of absolute temperature ( $1/T$ ) is shown in Fig. 6. A remarkable change in the slope is observed for all the samples as has been observed by Josyulu and Subhanadri (1980) with similar type of systems. From the curves of this figure, a linear variation is observed at low temperatures. As temperature increases, a gradual change in slope is observed and the curves again become linear at higher temperature. The change in slope was originally anticipated to be due to the Curie temperature, but it is found that these temperatures do not correspond to the measured Curie temperature for all the samples. It may be due to the change in conduction mechanism. The observed change in the slope indicates that resistivity is affected by the superposition of a number of thermally activated contributions. Two parallel conduction mechanisms may exist with different values of activation energies. The curves of Fig. 6 are divided into two segments, segment 1 corresponds to the low temperature region and segment 2 to the high temperature region. The activation energies were calculated for the different slopes and are presented in Table 2. The activation energies are found to be in between  $1.19$  and  $10^{-3}$  eV and  $9.92$  and  $10^{-4}$  eV. The conduction mechanism in the low activation energy region (region I) is attributed to electron hopping, whereas in the high-temperature region (region II), Mg ion conduction may also be possible as indicated in the high value of activation energy. An increase in activation energy on passing from ferromagnetic to paramagnetic region is observed as in Table 2. This is due to the ordered states in the ferromagnetic region and disordered states in the paramagnetic region. The charge carriers in paramagnetic region need more energy to jump between the adjacent sites. This is in accordance with the results observed by other workers on some ferrites system (El Hiti 1994, Kunar and Srivastava 1994).

**Table 2. Resistivity,  $\rho$  ( $\Omega$ -cm) and activation energy,  $E_a$  (eV) for different concentration of  $MgCr_xFe_{2-x}O_4$  ferrites sintered at  $1300^\circ C$ .**

Concentration (x)	Resistivity $\rho$ ( $\Omega$ -cm)	Activation energy $E_a$ (eV)
0.0	0.9804	$1.19 \times 10^{-3}$
0.2	0.9960	$1.17 \times 10^{-3}$
0.4	1.0440	$1.18 \times 10^{-3}$
0.6	1.2520	$1.18 \times 10^{-3}$
0.8	1.3872	$9.9 \times 10^{-4}$
1.0	1.5680	$9.9 \times 10^{-4}$

The composition dependence of dielectric constant ( $\epsilon'$ ) and dielectric loss ( $\tan \delta$ ) are shown in the Fig. 7 at 100 kHz for the spinel  $MgCr_xFe_{2-x}O_4$  sintered at  $1300^\circ C$ . With the increase of Cr concentration dielectric constant ( $\epsilon'$ ) increases at first and then decreases

and dielectric loss ( $\tan \delta$ ) decreases. This can be explained in terms of electron hopping between ferrous and ferric ions on octahedral sites for n-semiconductor ferrites and in terms of hole hopping for p-type ferrites (Reddy *et al.* 1981). Cr ion has a strong preference for the tetrahedral (A) site (Bhise *et al.* 1991), while Mg and Fe ions occupy both A site and B site (Bijal *et al.* 1992). The increase of Cr concentration in the ferrite system replaces some Mg ions from the A site. As a result, other Mg and Fe ions will migrate from A site to B site. The number of ferrous  $\text{Fe}^{2+}$  and ferric  $\text{Fe}^{3+}$  ions between which the electron exchange interaction (hopping conduction mechanism) takes place at the B site increases, therefore, the electrical conductivity increases. Since the mechanism of dielectric polarization in ferrites is similar to that of electric conduction, the increase in the number of electrons exchange between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions results in local displacements in the direction of applied external electric field that determines the dielectric polarization. For  $x > 0.6$ , dielectric constant decreases with increasing Cr concentration in the ferrite system  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

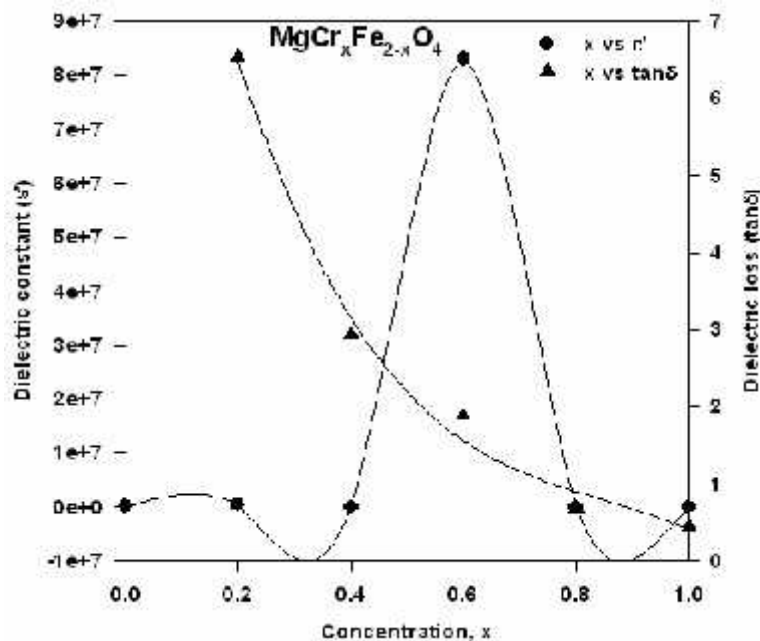


Fig. 7. Variation of dielectric constant,  $\epsilon'$  (left Y-axis) and dielectric loss,  $\tan\delta$  (right Y-axis) with Cr content in the spinel oxides  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ .

## CONCLUSION

The AC resistivity decreases with increasing temperature ensuring the semiconducting nature for all the samples of the spinel oxide  $\text{MgCr}_x\text{Fe}_{2-x}\text{O}_4$ . The activation energy decreases with increasing Cr concentration in the system. The lower activation energies are associated with higher electrical conductivity. The electrical

conduction in these ferrites is explained on the basis of the hopping mechanism. The increase in frequency enhances the hopping frequency of charge carriers resulting in an increase in the conduction process thereby decreasing the resistivity. The electrical conduction is related to the electron exchange interaction between the ferrous and ferric ions on the octahedral sites. The temperature dependence of the dielectric constant ( $\epsilon'$ ) and dielectric loss tangent increases while frequency dependence of the dielectric constant ( $\epsilon'$ ) and dielectric loss tangent decreases for all the samples. The resistivity changes monotonically with increasing Cr concentration in the system. The resistivity and dielectric constant ( $\epsilon'$ ) are found to decrease with increasing frequency exhibiting normal ferromagnetic behaviour of the samples. Dielectric constant ( $\epsilon'$ ) increases as the temperature increases, which is the normal dielectric behaviour of the magnetic semiconductor ferrite. The dielectric constant has abnormally high values and the dielectric loss exhibits a dielectric relaxation process or peaks at relatively higher temperatures.

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(Received revised manuscript on 18 November, 2014)