

**COMPOSITION, TEMPERATURE AND FREQUENCY DEPENDENT
MAGNETIC, DIELECTRIC AND ELECTRICAL PROPERTIES OF
MAGNESIUM-ZINC FERRITES**

M. MAHBUBUR RAHMAN^{*1}, MD. SHAHIN SHEAK¹, SOHELI PERVIN¹, NASIR UDDIN², FARID AHMED¹, MD. ABUL HOSSAIN¹, MASHUDUR RAHAMAN³, M. SHAHRIAR BASHAR³, TOFAZZAL HOSSAIN³ AND SHIREEN AKHTER⁴

School of Engineering and Energy, Murdoch University, Murdoch-6150, WA, Australia

ABSTRACT

Polycrystalline spinel $Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$, where $x = 0.10, 0.20, 0.25, 0.30$ and 0.35 ferrites (hereafter abbreviated as Mg-Zn) have been prepared by conventional double sintering technique. The samples were sintered at 1250°C in air for 6 hours. Measurements have been done at temperature and frequency ranges of $0 - 350^\circ\text{C}$ and $0 - 500$ kHz, respectively. In this work, some extrinsic magnetic properties such as Curie temperature, initial permeability, loss factor, quality factor, dielectric constant and resistivity of the samples have been studied. The Curie temperature and loss tangent of the samples decreased with increase in Cu-content whereas permeability, Q-factor, resistivity and dielectric constant have been noticed to be increased with the increase in Cu-content. The decrease in Curie temperature related to fact of weakening the strength of exchange A-B interaction. Maxwell-Wagner type of interfacial polarization might have found correlated with the normal dielectric behavior of the samples, however no relaxation peaks were observed in the dielectric dispersion curves of the Mg-Zn samples in virgin state or doped state.

Key words: Spinel ferrites, Curie temperature, Sintering temperature, Initial permeability, Dielectric constant

INTRODUCTION

Ferrites are ferrimagnetic materials composed of certain double oxide of iron and another metal which have two unequal sublattices and are ordered anti-parallel to each other. Each sublattice exhibits spontaneous magnetization at room temperature. Because of unequal magnitudes the magnetization is similar to ferromagnetics. Like ferromagnetics, ferrites have spontaneously magnetized domains and show the phenomena of magnetic saturation and hysteresis and have a critical temperature, T_c called the Curie temperature, above which they become paramagnetic. For these reasons, polycrystalline ferrites are highly demandable as very good dielectric materials and very useful materials

* Corresponding author: <mahbub235@yahoo.com>.

¹ Department of Physics, Jahangirnagar University, Savar, Dhaka-1342, Bangladesh.

² Department of Biomedical Engineering, University of Alberta, Edmonton, Canada.

³ Institute of Fuel Research and Development (IFRD), Bangladesh Council of Scientific & Industrial Research (BCSIR), Dhaka-1000, Bangladesh.

⁴ Materials Science Division, Atomic Energy Centre, Ramna-1000, Dhaka, Bangladesh.

for microwave devices as well (Rabinder and Latha 1999). Though all Mg, Zn and Cu are non-magnetic, they used to disproportionate the Fe^{3+} ions on the crystal lattice sites to provide or increase the magnetic moment. Pure Mg-ferrite has very high resistivity. Though substitution of Cu-ion decreases the resistivity, it substantially increases the permeability of the ferrite. Commonly it is categorized as non-microwave ferrite. The unique properties of non-microwave ferrites are high magnetic permeability and high electrical resistivity. So Mg-Zn-Cu ferrite is used in that case where minimization of eddy current loss is main point of interest. The resultant lower eddy current losses allow their use to higher frequencies than possible with metals. As soft magnetic materials Mg-Zn-Cu ferrites have been proved to be smart candidate of highest rank. The requisites for modern ferrite are so many in numbers that only extensive research in this field can meet the huge demand of technology. Mg-Zn-Cu ferrites with lower Cu content are widely used in high frequency and pulsed field applications for their unique combination of small coercivity and high electrical resistivity.

It is well established that properties of ferrites are very strongly dependent on composition, method of synthesis, temperature, frequency and sintering conditions (Rabinder and Latha 1999). Reslescy *et al.* (2006) showed that Mg-Zn ferrites have high resistivity, relatively higher T_c , cost effectiveness and have higher mechanical stability and hardness. Magnetic and electrical properties of Mg-Zn ferrites have been studied by different research groups around the globe (Murthy 2001, Yue *et al.* 2001a, b, Raju *et al.* 2006, Pujar *et al.* 1999) using additives. Addition of CuO in Mg-Zn ferrites have been found to work as sintering aid (Koh and Kin 1986, Park *et al.* 1997). Since CuO acts as sintering aid, it has been stated that easy and cost effective Mg-Zn ferrites of similar properties those that of Ni-Cu-Zn can be developed by ceramic technology at lower sintering temperature which also helps to avoid losing samples by evaporation. Rabinder and Latha (1999) reported normal dielectric behavior of undoped Mg-Zn ferrites. Abnormal dielectric behavior with dielectric relaxation peaks has been investigated by Haque *et al.* (2008a, b) as the effect of Cu-addition. However, in the present study normal dielectric and electrical behavior of Mg-Zn ferrites have been observed with the addition of Cu^{2+} ions. A few systematic study of T_c , loss tangent, Q-factor DC and AC resistivity of Mg-Zn ferrites with the substitution of Cu have been presented here.

METHODS AND MATERIALS

$\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$ (with $x = 0.10, 0.20, 0.25, 0.30,$ and 0.35) polycrystalline spinel ferrites have been prepared by conventional double sintering solid state reaction method. The laboratory facilities of Institute of Fuel Research and Development (IFRD), Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka-1000, Bangladesh have been used for this purpose. Appropriate proportions of raw materials

(Fe₂O₃, MgO, CuO and ZnO) were weighed according to their molecular weight. Mixing of these materials was performed using agate motor for about 5 hours. To obtain a homogeneous mixture of the materials the ball milling was carried out. The ball milling is the most common type of milling, which consists of a lined pot with hard spheres or rods inside the ball. To increase the degree of mixing, milling can be carried out in a wet medium such as ethyl alcohol. This method depends on the solid state inter-diffusion between the raw materials. Solid do not usually react at room temperature over normal time scale. Thus it is necessary to heat them at elevated temperature. The ground powders are then calcined in air at a temperature above 1000°C. For some time, this process is continued until the mixture is converted into the correct crystal phase. The calcined powders are again crushed into fine powders. The rod or plate shaped samples were prepared from this calcined powders by using die-punched assembly under a pressure of 1.70 and 1.15 ton/cm² to make the sample pellets and toroids shapes. Finally, the pellets and toroids were sintered at 1250°C in air for 6 hours and cooled inside the furnace. At the end stage, the final products were slowly heated in programmable muffle furnace to 650°C to avoid cracking in the samples. A heating rate of 2°C/min was maintained in the process.

Structural identification and phase analysis of Mg-Zn ferrites were carried out by XRD studies using Phillips X'Pert PRO X-ray (PW3040) diffractometer. XRD measurements were performed by Cu-K_α radiation ($\lambda = 1.54 \text{ \AA}$) in the range of $2\theta = 15$ to 75° in the steps of 0.02° . The samples were exposed to a primary beam power of 40 kV and 30 mA. Each data was collected in a time gap of 1.0 s. Spinel types of crystal structure without having any other intermediate phases and high purity levels of constituent powders were confirmed by XRD study. Results of these analyses are available elsewhere (Haque *et al.* 2008a,b, Sheak 2011). Permeability measurements with the toroidal samples were done with the Hewlett Packart (HP) impedance analyser (HP4192A). Pellet shapes sampled were prepared for electrical and dielectric measurements. Dielectric constant (ϵ') and resistivity of the samples was measured by HP impedance analyser. Dielectric constants as a function of frequency in the frequency range of 0 to 6 kHz at room temperature at different frequencies were carried out in conjunction with a laboratory made furnace. Dielectric constant was calculated from the relation:

$$\epsilon' = \frac{Cd}{\epsilon_0 A},$$

where C is capacitance of the pellet in Farad, d is the thickness of the pellet in meter, A is the area of cross section of the sample in meter² and ϵ_0 is the free space's permeability. The temperature dependence of the dc resistivity was done in the temperature range of 25 to 300°C while the ac resistivity was studied in frequency range of 0 to 5 kHz at room temperature.

RESULTS AND DISCUSSION

The bulk density, ρ of the samples was measured using the following relation:

$$\rho = \frac{m}{V} \quad (1)$$

where m is the mass of the bulk sample in g and V is volume in cc. The theoretical density, ρ_x was calculated from the following Eq. (2):

$$\rho_x = \frac{ZM}{Na_0^3} \quad (2)$$

where Z is the number of molecules in a unit cell of the spinel lattice which is 8 for the spinel structure, M represents the molecular weight of the ferrites, N is the Avogadro's number, a_0 the lattice constant of the samples and $V = a_0^3$.

Porosity of the samples in terms of percentage of each sample was calculated using the relation:

$$P(\%) = \left(1 - \frac{\rho}{\rho_x}\right) \times 100 \quad (3)$$

Bulk density, X-ray density and porosity of the samples are presented in Table 1. In the present study it has been observed that the density of Mg-Zn ferrites with and without the Cu-addition vary between 4.43 and 4.90 g/cc while the X-ray density varies between 4.72 and 5.10 g/cc. Density of polycrystalline ferrites play significant role in controlling the various properties. In both cases, the densities were found to increase linearly with the increase in Cu-content.

Table 1. Bulk density, theoretical density and porosity of Mg-Zn ferrites with Cu-additives.

Composition	x	Bulk density ρ (g/cc)	X-ray density ρ_x (g/cc)	Porosity P (%)
$Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$	0.10	4.90	5.10	4
	0.20	4.78	4.98	4.1
	0.25	4.65	4.88	4.8
	0.30	4.52	4.81	6
	0.35	4.43	4.72	6.2

It has been also observed that the bulk density of the samples is lower than that of the X-ray density. This reveals the existence and formation of pores in the samples during the synthesis and sintering processes. Another reason, might be due to the fact that the Mg^{2+} (0.65 Å) ions on the octahedral sites are being replaced by the massive Cu^{2+} (0.70 Å) ions. Unit cells expand to accumulate the larger ions. Addition of Cu^{2+} ions at the expense

of Mg^{2+} ions may cause the lattice parameters to enhance. Other possible reasons that might be associated with the densification of the polycrystalline samples are: formation of solid solution, almost all the copper ions entered into the spinel lattice during the heat treatment and thus activating the lattice diffusion, grain boundary diffusion (Haque *et al.* 2008). These types of possibilities have been also supported by few other groups (Gupta and Coble 1968, Coble and Gupta 1967). Present results have been found to be in good agreement with earlier reports [Yue *et al.* 2001, Haque *et al.* 2008a, b).

Table 2. Curie temperature of Mg-Zn ferrites with Cu-content.

Composition	x	Curie temperature, T_c (°C)
$\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$	0.10	175
	0.15	213
	0.20	245
	0.30	269

Curie temperature, T_c of Mg-Zn spinel ferrites have been measured from the temperature dependence of initial permeability with and without Cu-addition which is presented in Fig. 1. T_c has been taken at that temperature at which a sharp fall of permeability of the samples has been noticed (Rahman *et al.* 2012). At T_c , a magnetically ordered (ferromagnetic) substance converts to a magnetically disordered one (paramagnetic).

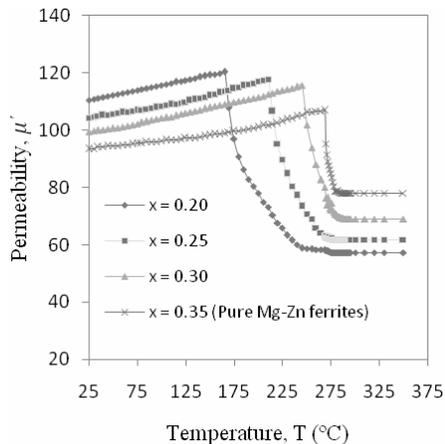


Fig. 1. Temperature dependence of permeability of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$ with Cu-content.

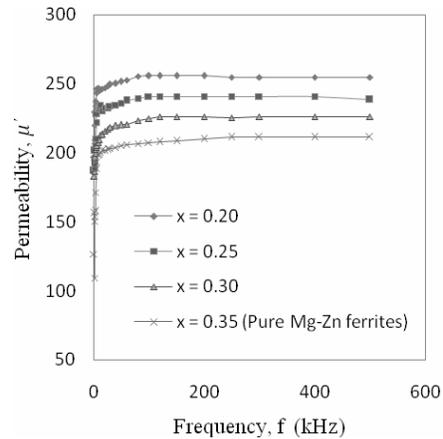


Fig. 2. Permeability of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$, as a function of frequency.

Curie temperature of the samples has been displayed in Table 2. According to Table 2, it is seen that T_c increases with the decrease of Cu-content. It is well established that T_c measures the strength of exchange interaction at which the thermal energy, $k_B T$ tending

to disorder the system winning over the exchange energy. Since the T_c of polycrystalline ferrites depend on the strength of A-B interaction (Shaikh *et al.* 2002), increase of non-magnetic Cu-content causes to a decrease in magnetic moment on tetrahedral A-site, which thereby weakens A-B interaction (Shaikh *et al.* 2002). This reflects the fact that due to the decrease in A-B interaction the T_c of Mg-Zn ferrites decrease with the increase in Cu-content. This type of results was supported in previous work of Cd-Cr-Cu ferrites (Greskovich and Lay 1972).

Permeability spectra of Mg-Zn ferrites with the addition of Cu-content have been presented in Fig. 2. Permeability data presented in Fig. 2 shows that the real part of the initial permeability of Mg-Zn ferrites increases with the addition of Cu to the pure samples. It also confirms that at low frequency permeability increase rapidly and at high frequency region the permeability becomes almost independent of the frequency. It might be due to the fact that the addition of Cu on Mg-Zn ferrites the density might have been increased which leads to an increase in the permeability. This is true because ferrites with higher density and large average grain size possess a higher initial permeability (Jiles 1998). Permeability of polycrystalline ferrites is related to the spin rotation and domain wall motion (Smit and Wijn 1959, Jun and Mi 2005). Spin rotation and domain wall motion are related as $\mu = 1 + \chi_w + \chi_{spin}$, where χ_w is the domain wall susceptibility, χ_{spin} is the intrinsic rotational susceptibility. The domain wall susceptibility and the intrinsic rotational susceptibility are given by the following Eqs.

$$\chi_w = \frac{3\pi M_s^2 D}{4\gamma} \quad (4)$$

$$\chi_{spin} = \frac{2\pi M_s^2}{K} \quad (5)$$

where M_s , K , D and γ are the saturation magnetization, total anisotropy, average grain diameter, and domain wall energy, respectively. Properties of soft ferrites are dependent on their compositions, additives, sintering conditions and microstructures. According to the Eqs. (4 & 5), it is clearly observed that the permeability is directly proportional to the square of M_s and inversely proportional to the anisotropy constant, K of the materials. As the temperature is increased both K and M_s are decreased. However, the rate of decrease of anisotropy constant is faster than that of the saturation magnetization. For this reason, permeability enhances very fast around T_c (the ferro/paramagnetic transition temperature of the sample Fig. 1) (Hoque *et al.* 2011, Nath *et al.* 2012). The initial permeability of ferrites depends on many factors such as reversible domain wall displacement, domain wall bulging as well as microstructural features *viz.*, intragranular porosity, chemical composition, average grain size and the presence of second phase etc. (Smit and Wijn 1959). At high sintering temperature and frequency, the initial permeability decreases

due to the decrease in average grain size of the samples. As a result, removal of the domain wall pinning sites at the grain boundary is also increased. Presence of Cu-content rich precipitation and porosity could also come into play to pin the domain wall and causing to reduce the initial permeability at higher sintering temperature and frequency. It is well known that the magnetic properties of ferrites are greatly influenced by the microstructures; the larger the grain sizes, the higher the saturation magnetization and larger initial permeability. Ferrites with lower initial permeability and saturation magnetization are suitable for microwave applications.

Fig. 3 shows the loss tangent of Mg-Zn ferrites as a function of frequency. This measurement was performed over the frequency range 1 to 500 kHz. According to the data presented in Fig. 3, it is noticed that the loss tangent of Mg-Zn ferrites decreases with the increasing of Cu-content. Similar effect has been observed for temperature dependence of loss tangent which is shown in Fig. 4. At low temperature, up to 230°C the loss tangent is unaffected but at high temperature (230 to 270°C) the loss tangent has been found to be high, afterwards it becomes almost invariant with the further rise in temperature.

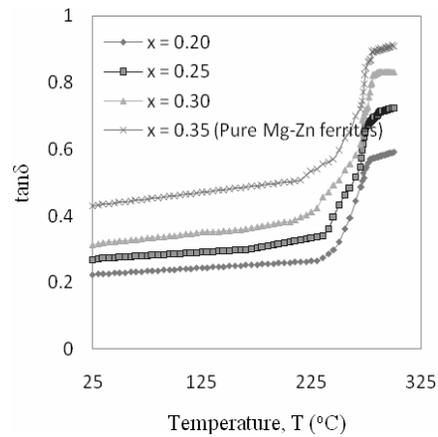
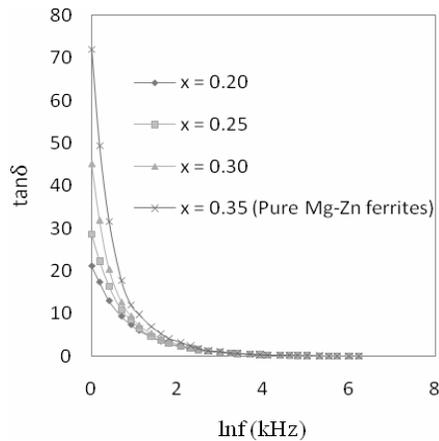


Fig. 3. Loss tangent of $Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$, as a function of frequency.

Fig. 4. Loss tangent of $Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$, as a function of temperature.

Loss tangent defines the amount of energy wasted on process other than magnetization that can prevent the AC applications of a given material. It also measures the inefficiency of a system. At high frequency and low temperature the loss tangent is very small which can be neglected. Generally, the loss of energy is related to various domain defects such as non-uniform and non-repetitive domain wall motion, domain wall bowing, localized variation of the flux density and nucleation and annihilation of domain walls (Overshott 1981). Moreover, in ferrites losses might also arise due to other factors: eddy current loss, hysteresis loss, and residual loss (Snelling 1989). Eddy current losses

can be minimized by increasing the resistivity of ferrites. Since the resistivity of ferrites depends on the composition, sintering environment, and cooling rate (Snelling 1989). Thus, a slow cooling rate is essential to reduce the trapping of pores within grains which in turn reduces the eddy current losses. Hysteresis loss mainly contributes by the irreversible wall displacements found to be increased by the increase of grain size and the impurity in the raw materials (Bellad and Chougule 2000) Residual losses have been found to arise due to the relaxation of domain walls (Bellad and Chougule 2000). All the phenomena of losses come into play when the permeability begins to drop which is well known as the ferrimagnetic resonance (Brockman *et al.* 1950). At resonance, maximum amount of energy is transformed from the applied ac magnetic field to the lattice and thus resulting in a quick decrease in relative Q-factor. At high frequencies, losses are to be lower if domain wall motion is inhibited and the magnetization is forced to change by rotation. Present observation has been found to be consistent with the Q-factor studies (Fig. 5), where it is observed that the Q-factor increases with the substitutions. This reflects the fact that with the addition of Cu the loss of energy has been reduced. The decreasing nature of the loss tangent attributes the relaxation phenomena of magnetization of domain wall (Jacobo *et al.* 1998). Addition of Cu^{2+} ions to the pure Mg-Zn samples might disrupts and weakens the Fe^{3+} -O- Fe^{3+} superexchange interaction and thereby accelerating the Fe^{2+} ions production. Thus, it can be concluded that due to combined effects of the above phenomena the loss of energy is decreased.

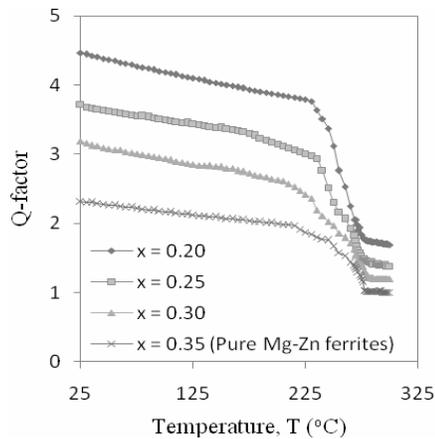


Fig. 5. Q-factor of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$, as a function of temperature.

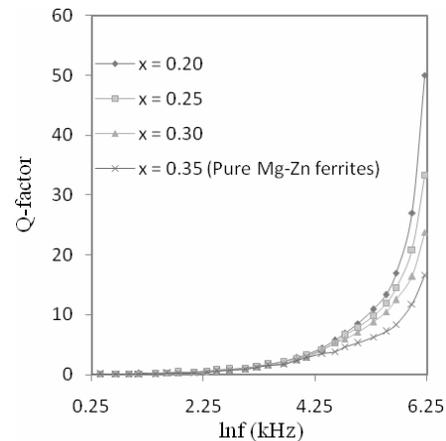


Fig. 6. Q-factor of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$, as a function of frequency.

Q-factor of Mg-Zn ferrites as a function of temperature with and without the addition of Cu-content has been displayed in Fig. 5. It is observed that the Q-factor of Mg-Zn ferrites increases as the Cu-content increased. In the low temperature region, Q-factor varies largely with the amount of Cu-doping. However, with the increase in

temperature Q-factor decreases slowly and at high temperature the Q-factor becomes almost invariant. Fig. 6 shows the Q-factor of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$ ferrites as function of frequency of sintered at 1250°C in air for 6 hours. It is observed that Q-factor increases with increasing frequency. According to the study of loss tangent and Q-factor, we observed that at low frequency region all the samples work better with low losses and high qualities. The variation in Q-factor with temperature and frequency may also be explained in the same way as that of the loss tangent, which is already been mentioned previously. From the frequency characteristic of Q-factor the perfect frequency band can be found at which these materials work well as soft magnetic material with low losses.

Fig. 7 shows the variation of dielectric constant in the frequency range from 1 to 500 kHz at room temperature for the samples $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$, with $x = 0.20, 0.25, 0.35$ sintered at 1250°C in air for 6 hours. The dielectric constant of the samples has been found to be decreased with the increase of Cu-content and frequency. This is a normal dielectric behavior observed in most ferromagnetic materials due to the interfacial polarization as suggested by Koops phenomenological theory of Maxwell and Wagner bi-layer model (Yue *et al.* 2001, Iwauchi 1971, Patil *et al.* 1991, Wagner 1913, Koops 1951). The dielectric constant decreases with the increasing frequency, which is very rapid at lower frequencies and slower at higher frequencies. At higher frequency, around 20 kHz and above dielectric constant become almost independent of frequencies. The dielectric behavior of ferrites can be explained on the basis of dielectric polarization, which is similar to that of the conduction mechanism that takes place mainly by the hopping of electrons reported by many research groups (Yue *et al.* 2001, Iwauchi 1971, Rabkin and Novikova 1960, Anderson 1964). Present results have been found to be in good agreements with earlier reports (Iwauchi 1971, Patil *et al.* 1991, Wagner 1913, Koops 1951, Dhanaraj and Rajesh 2011) without any signature of abnormal dielectric behavior as noticed by Haque *et al.* (2008b). Electronic conduction in ferrites is mainly due to hopping of electron between ions of the same element existing in more than one valence state and distributed randomly over crystallographically equivalent lattice sites. In this study, the frequency dependency dielectric constant of Mg-Zn ferrites has been determined by measuring capacitance. It is believed that in polycrystalline ferrites there are two layers: heterogeneous conducting layer and a very high resistive layer. The conducting layer is filled with grains while the resistive layer contains large number of grain boundaries (Koops 1951, Rabkin and Novikova 1960 Dhanaraj and Rajesh 2011). Dielectric properties of ferrites have been found to arise due to various types of polarization mechanisms (El Hiti 1999). The space charge polarization comes into play only at high temperature and low frequencies (El Hiti 1999). As the frequency increase the space polarization becomes weaker which in turn causes to a decrement of dielectric constant. On the other hand, various types of defects and dislocations in samples such as oxygen vacancies, defects in grain boundaries invite the interfacial polarization to be

active in the low frequency side (Mu *et al.* 2008, Haijun *et al.* 2003, Kuanr and Srivastava 1994). Due to this interfacial polarization the dielectric constant has higher magnitude at lower frequency region (Mu *et al.* 2008, Haijun *et al.* 2003, Kuanr and Srivastava 1994).

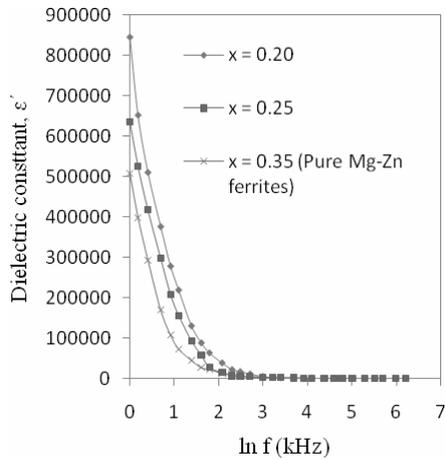


Fig. 7. Dielectric constant of $Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$, as a function of frequency.

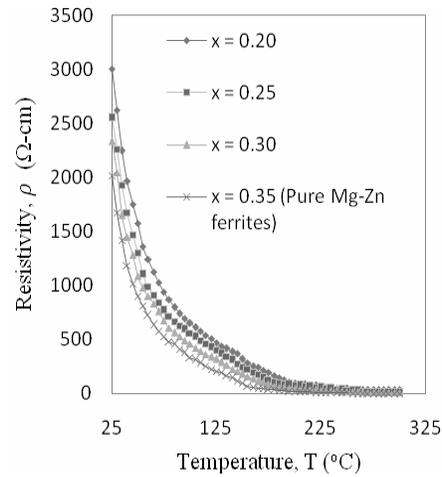


Fig. 8. DC resistivity of $Mg_xZn_{0.3}Cu_{0.7-2x}Fe_{2+x}O_4$, as a function of temperature.

Fig. 8 shows the temperature dependence of DC resistivity of Mg-Zn ferrites with the addition of Cu-content sintered at 1250°C in air for 6 hours. DC resistivity of the samples found to be increased due to the increase in Cu-content. It is also observed that the resistivity is maximum at room temperature and decreases with the increase in temperature. It was further noticed that the fall of resistivity is rapid from temperature 25 to 75°C, after that it decreases slowly until 175°C and then becomes nearly constant. The decrease of resistivity is related to the decrease of porosity, since pores are non-conductive, increase of which causes to increase the resistivity of the material (Iqbal *et al.* 2010). Moreover, the resistivity decreases with the decrease in porosity because the charge carriers on their way face less number of pores i.e., obstacles. Fig. 9 shows the AC resistivity of Mg-Zn ferrites sintered at 1250°C for 6 hours in air. Study of AC resistivity of the samples has been performed in the frequency range of 1 to 500 kHz. Cu-doped Mg-Zn ferrites have higher resistivity than undoped ones. All the curves show the significant dispersion with frequency, which is an important behavior of ferrimagnetics (Watawe *et al.* 2000). Conduction mechanism in ferrites is strongly related to the exchange of electrons between Fe^{2+} and Fe^{3+} ions. As the frequency increases it causes to increase the hopping of electrons. The increased electrons' hopping results in the increase of conductivity which eventually causes to a decrement of resistivity.

In the high frequency region, resistivity does not show remarkable variation with the frequency. This might be related to the fact that hopping frequency can no longer follow the frequency of the applied ac field (Watawe *et al.* 2000).

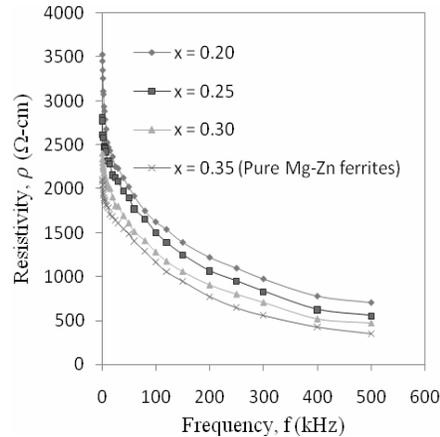


Fig. 9. AC resistivity of $\text{Mg}_x\text{Zn}_{0.3}\text{Cu}_{0.7-2x}\text{Fe}_{2+x}\text{O}_4$, as a function of frequency.

Resistivity becomes minimum when the frequencies of the hopping electrons become equal to that of applied field. This frequency is termed as the resonance frequency (Rahman *et al.* 2012) i.e., the jumping frequency of hopping charge carriers is almost equal to that of the applied field. Then again with the increase in frequency, the resistivity increases most linearly which may be due to the retardation of hopping charge carriers with increasing applied field frequency. In this investigation a rapid dielectric dispersion was observed at lower frequency region and it becomes almost independent in the higher frequency values. The dielectric behaviour of ferrites has been already explained on the basis of the mechanism of the dielectric polarization that has been found to be similar to that of conduction process in the discussion of dielectric properties.

Throughout the study, it was observed that addition of Cu played significant role in improving the magnetic, dielectric and electrical properties of Mg-Zn ferrites. The density of the samples increased with the increase in Cu-content. This happens because, the atomic mass of Cu is greater than that of the Mg and with the increase of Cu the Mg-content decreases in the present sample. T_c decreases as the Cu-content increases and at the same time the permeability has been found to be increased. It was also noticed that the initial permeability remains unaffected at high frequency with and without the addition of Cu-content. The loss tangent increases with the increase in temperature and decreases with increasing frequency Cu-content. The origin of the loss tangent can be attributed to various domain defects, which include non-uniform and non-repetitive domain wall motion, domain wall bowing, localized variation of the flux density and

nucleation and annihilation of domain walls. Studying the Q-factor of a sample the temperature could be identified at which the sample works as soft magnetic material with low losses. For inductors used in filter applications, the Q-factor is often used as a measure of performance. Q-factor decreases with the increase in temperature and increases with the increase in frequency and Cu-content. The observed decrease in dielectric constant with the increase in frequency might be due to the fact that above certain frequencies the electronic exchange between Fe^{2+} and Fe^{3+} ions does not follow the frequency of the applied AC field. The decrease of dielectric constant is faster at lower frequency side and slower at higher frequency region. This is the normal dielectric behavior observed in most of the ferrimagnetic materials. At room temperature resistivity has been found to be maximum and decreased with the increase in temperature and increased with the addition of Cu-content. AC resistivity decreases with the increase in frequency and increased with the increase in Cu-content. At 1 kHz, ac resistivity has been observed to be high while at 500 kHz it has been found to be low and become independent of frequency. Sintering temperature has a significant effect on the magnetic and other properties of ferrites. Low temperature sintering decreases the grain size, narrows the grain size distribution. Thus, it might be expected that the magnetic characteristics of ferrites could have been improved when sintered at low temperature. In addition, the optimization of the dynamic dielectric properties such as permeability, in the high frequency range requires a precise knowledge of magnetization mechanism. The magnetization mechanism contributing the permeability in polycrystalline ferrites have been a controversial subject for a long time and remain unresolved. Thus, still there are enormous scope of future work on this particular sample synthesizing at various lower and higher sintering temperature and changing the substituent amounts.

ACKNOWLEDGEMENTS

Authors are very grateful to Department of Physics, Jahangirnagar University, Dhaka-1342, Bangladesh; Bangladesh Council of Scientific and Industrial Research (BCSIR) and Materials Science Division, Atomic Energy Centre (AEC), Dhaka, Bangladesh.

REFERENCES

- Anderson, J.C. 1964. *Dielectrics*. Chapman and Hall, London.
- Bellad, S.S. and B.K. Chougule. 2000. Composition and frequency dependent dielectric properties of Li-Mg-Ti ferrites. *Mater. Chem. Phys.* **66**: 58.
- Brockman, F.G., P.H. Dowling and W.G. Steneck. 1950. Dimensional effects resulting from a high dielectric constant found in a ferromagnetic ferrite. *Phys. Rev.* **77**: 85.
- Coble, R.L. and T.K. Gupta, 1967. *Sintering and Related Phenomena*. G.C. Kuczynski, N.A. Hroton, C.F. Gibbon (Eds.). Gordon and Breach, New York, pp. 423.

- Dhanaraj, P.V. and N.P. Rajesh. 2011. Studies on the growth and characterization of tri (glycine) calcium (II) dichloride a nonlinear optical crystal. *Physica B: Condensed Matter* **406**: 12.
- El Hiti, M.A. 1999. Dielectric behaviour in Mg-Zn ferrites. *J. Magn. Magn. Mater* **192**: 305.
- Greskovich, C. and K.W. Lay. 1972. Grain Growth in very Porous Al₂O₃ Compacts. *J. Am. Ceram. Soc.* **55**: 142.
- Gupta, T.K. and R.L. Coble. 1968. Sintering of ZnO: I, Densification and Grain Growth. *J. Am. Ceram. Soc.* **51**: 521.
- Haijun, Z., L. Zhichao, M. Chenliang, Y. Xi, Z. Liangying and W. Mingzhong. 2003. Preparation and microwave properties of Co- and Ti-doped barium ferrite by citrate sol-gel process. *Mater. Chem. Phys.* **80**: 129.
- Haque, M. Manjurul, M. Huq and M.A. Hakim. 2008b. Influence of CuO and sintering temperature on the microstructure and magnetic properties of Mg-Cu-Zn ferrites. *J. Magn. Magn. Mater.* **20**: 2792.
- Haque, M. Manjurul M., Huq, M.A. Hakim. 2008. Densification, magnetic and dielectric behaviour of Cu-substituted Mg-Zn ferrites. *Materials Chem. and Phys.* **112**: 580.
- Hoque, S.M., A.K.M. Rezaul Haque, M.O. Rahman and N.H. Nghi. 2011. Ultra-soft magnetic properties and giant magneto-impedance of Co₆₈Fe_{4.5}Si_{12.5}B₁₅. *J. Noncrys. Sol.* **357**: 2109.
- Iqbal, M.J., M.N. Ashiq and I.H. Gul. 2010. Physical, electrical and dielectric properties of Ca-substituted strontium hexaferrite (SrFe₁₂O₁₉) nanoparticles synthesized by co-precipitation method. *J. Magn. Magn. Mater.* **322**: 1720.
- Iwachi, K. 1971. Dielectric Properties of Fine Particles of Fe₃O₄ and some Ferrites. *Jpn. J. Appl. Phys.* **10**: 1520.
- Jacobo, S.E., W.G. Fano, A.C. Razzitte, N.D. Digiovanni and V. Trainotti. 1998. *El. Insulation and Diel Phenomena Annual Report Conference* **1**: 273.
- Jiles, D. 1998. *Introduction to Magnetism and Magnetic Materials*. London: Chapman & Hall CRC.
- Jun, H. and Y. Mi. 2005. Preparation of high-permeability NiCuZn ferrite. *J. Zhejiang Univ. Sci.* **6B** (6): 580.
- Koh, J.G. and K.U. Kim. 1986. *New Phys.* (Korean Phys. Soc.) **26** (6): 540.
- Koops, C.G. 1951. On the dispersion of resistivity and dielectric constant of some semiconductors at audio frequencies. *Phys. Rev.* **83**: 121.
- Kuanr, B. K. and G. P. Srivastava. 1994. Dispersion observed in electrical properties of titanium-substituted lithium ferrites. *J. Appl. Phys.* **75**: 6115.
- Murthy, S.R. 2001. Low Temperature Sintering of MgCuZn Ferrite and its Electrical and Magnetic Properties, *Bull. Mater. Sci.* **24** (4): 379.
- Mu, C., N. Chen, X. Pan, X. Shen and X. Gu. 2008. Preparation and microwave absorption properties of barium ferrite nanorods. *Mater. Lett.* **62**: 840.
- Nath, S.K., K.H. Maria, S. Noor, S.S. Sikder, S.M. Hoque and M.A. Hakim. 2012. Magnetic ordering in Ni-Cd ferrite. *J. Magn. Magn. Mater.* **324**: 2116.
- Overshott, K.J. 1981. The causes of the anomalous loss in amorphous ribbon materials. *IEEE Trans. Magn.* **17**: 2698.
- Patil, R.S., S.V. Kakatkar, P.K. Maskar, S.A. Patil and S.R. Sawant. 1991. *Indian J. Pure Appl. Phys.* **29**: 589.

- Pujar, R.B., S.S. Bellad, S.C.Watawe and B.K. Chougule. 1999. Magnetic properties and microstructure of Zr^{4+} -substituted Mg-Zn ferrites. *Mater. Chem. Phys.* **57**: 264.
- Rabinder, D. and K. Latha. 1999. Dielectric behaviour of mixed Mg-Zn ferrites at low frequencies. *Mat. Lett.* **41**: 247.
- Raju, V.S.R., S.R. Murthy, F. Gao, Q. Lu and S. Komarneni. 2006. Microwave hydrothermal synthesis of nanosize PbO added Mg-Cu-Zn ferrites. *J. Mater. Sci.* **41**: 1475.
- Sheak, Md. Shahin. 2011. Investigation of the Effect Cu-Substitution on Magnetic and Electrical Properties of Mg-Zn Ferrites. MS Thesis, Jahangirnagar University, Dhaka-1342, Bangladesh.
- Rabkin, L.I. and Z.I. Novikova. 1960. Ferrites (Minks), USSR.
- Rahman, M. Mahbubur, M.K. Das, B. M. Sonia, F. Ahmed, M. Rahaman, M.S. Bashar, T. Hossain and S. Akhter. 2012. Study of Magnetic, Dielectric and Electrical Properties of Li-Mg-Ni Ferrites. Submitted to the *Indian J. Phys.*
- Shaikh, A.M., S.C. Watawe, S.A. Jadhav and B.K. Chougule. 2002. Preparation and characterization of Zn substituted Li-Mg ferrites. *Mater. Research Bull.* **37**: 2547.
- Smit, J. and Wijn H.P.J. 1959. *Ferrites*. Philips Tech. Library, Netherlands.
- Snelling, E.C. 1989. Proceedings of the Fifth International Conference on Ferrites (ICF-5), ICF-5, India 579.
- Yue, Z., Ji Zhou, Longtu Li, Xiaohui Wang and Zhilun Gui. 2001. Effect of copper on the electromagnetic properties of Mg-Zn-Cu ferrites prepared by sol-gel auto-combustion method, *Mater. Sci. Engg.* **86(1)**: 64.
- Yue, Z., Ji Zhou, Xiaohui Wang, Zhilun Gui and Longtu Li. 2001. Low temperature sintered Mg-Zn-Cu ferrite prepared by auto-combustion of nitrate-citrate gel. *J. Mater. Sci. Lett.* **20**: 1327.
- Wagner, K.W. 1913. Zur Theorie der unvollkommenen Dielektrika. *Anal. der. Physik.*, **345**: 817.
- Watawe, S. C., B. D. Sarwade, S. S. Bellad, B. D. Sutar and B. K. Chougule. 2000. Microstructure, frequency and temperature-dependent dielectric properties of cobalt-substituted lithium ferrites. *J. Magn. Mater.* **214**: 55.

(Received revised manuscript on 01 October, 2012)