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Structural and Magnetic Properties of Cr³⁺ Doped Mg Ferrites

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ABSTRACT

The polycrystalline $MgCr_xFe_{2-x}O_4$ ferrites ($0.0 \leq x \leq 1.0$) were prepared by conventional solid state ceramic sintering technique in air at 1300°C. X-ray diffraction experiments were carried out on all the samples in order to characterize the materials at room temperature. The X-ray diffraction patterns showed sharp peaks indicating the formation of single phased cubic spinel structure. The lattice parameters of the samples were determined from the X-ray diffraction data using Nelson-Riley extrapolation method. It was found that the lattice parameter decreased with increasing Cr concentration obeying Vegard's law. Magnetic properties of the samples were measured using an Impedance Analyzer. Real and imaginary parts of the complex permeability, loss factor and quality factor were measured as the function of frequency at three different sintering temperatures 1250°C, 1300°C and 1350°C for all the samples in the frequency range 1 kHz to 13 MHz. Frequency stability of the real part of permeability (μ') increases with increasing Cr concentration and also with sintering temperature. Imaginary part of permeability (μ'') decreases with increasing frequency and increased with increasing both of the Cr content and sintering temperature. Loss factor ($\tan\delta$) decreased with increasing frequency while the quality factor (Q) increased with increasing frequency for all the samples. The temperature dependence of initial permeability (μ_i) was measured for all the samples sintered at 1300°C. The Curie temperature (T_c) was determined from the μ'–T curves. The values of T_c were found to be 733 K, 657 K, 583 K, 468 K, 400 K and 317 K for x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0, respectively.

Keywords: Ferrites; Spinel Structure; Magnetic Properties; Permeability; Loss Factor; Curie Temperature

1. Introduction

Spinel ferrites have gained much importance in the recent years because of their interesting electrical and magnetic properties [1]. Usually ferrite materials have low conductivity and the order of magnitude of the conductivity greatly influences their dielectric and magnetic behavior [2]. Because of their high resistivity, spinel ferrites are the materials of great interest for high frequency inductive components. The magnetization mechanism contributing to the complex permeability in polycrystalline ferrites have been a controversial subject [3]. The optimization of their dynamic properties such as complex permeability in the high frequency range requires a precise knowledge of magnetization mechanism involved. Although a large number of spinel compounds have been studied, the existing theories and experimental results indicated that a complete understanding of the complex magnetic behavior arising from the change in magnetic ion concentration is yet to be explored.

Ferrites are one of the most studied magnetic systems and there have been a lot of reports on experimental and theoretical investigations on this material. Since a spinel ferrite is a multi-element oxide material having the formula AB_2O_4 (where A and B represents tetrahedral and octahedral lattice sites respectively), it can be fabricated by substituting various magnetic and non-magnetic ions for obtaining the specific properties. The substitution of elements and their compositions greatly affect the magnetic moments, lattice parameters and exchange interactions [4,5].

The system under investigation, $MgCr_xFe_{2-x}O_4$, contains three different cations Mg^{2+}, Cr^{3+} and Fe^{3+}. For such complex system, several new magnetic states are predicted depending on the strength of the anisotropy related

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to exchange interactions [6,7]. The advantage of the mixed spinel considered here is that, all interactions are well-defined near-neighbor antiferromagnetic with $|J_{AB}| \gg |J_{BB}| \gg |J_{AA}|$ and J_{AB} super an exchange interactions render the spinel ferrimagnetism. The parameter "x" may be varied from 0 to 2, which covers the entire range from Mg ferrite ($MgFe_2O_4$) to Mg chromate ($MgCr_2O_4$). According to the literature, $MgFe_2O_4$ is an inverse spinel taken to be collinear ferrimagnetic [8,9], whose degree of inversion depends on the cooling rate [8,10]. $MgCr_2O_4$ is a normal spinel with a canted ferromagnetic structure [11].

Ferrites are ceramic magnetic materials capable of combining the resistivity of an insulator with relatively high permeability, and have an enormous impact on the application as magnetic materials particularly at high frequencies. The term ferrite denotes a group of iron oxides, which have the general formula $MO \cdot Fe_2O_3$, where M is a divalent metal ion such as Mn^{2+}, Fe^{2+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, Mg^{2+}, or Cd^{2+}. The typical ferrite is magnetite, Fe_3O_4 (or $FeO \cdot Fe_2O_3$), which has been a well-known magnetic oxide since ancient time. The ferrites were developed into commercially important materials by Snoek [12] and his associates at the Philips Research Laboratories in Holland mainly during the years 1933-1945. In a classical paper published in 1948, Neel [13] provided the theoretical key to an understanding of ferrites. The subject has been covered at length in books by Smit and Wijn [14] and Standley [15] and in review papers by Smart [16] and Gorter [17].

In the present paper we have reported the structural and magnetic properties of $MgCr_xFe_{2-x}O_4$ ferrites prepared in the solid state sintering technique. The magnetic properties such as permeability, loss factor, quality factor etc. of the samples have been measured using an impedance analyzer (LCR Bridge) in the frequency range 1 kHz - 13 MHz sintered at different temperatures. The Curie temperatures (T_C) of the individual samples have also been reported.

2. Experimental

A series of ferrite sample $MgCr_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 & 1.0) were prepared in the solid-state reaction method using conventional ceramic technique at the Institute of Nuclear Science & Technology (INST), Atomic Energy Research Establishment, Savar, Dhaka, Bangladesh. For the preparation of samples, reagent of analytical grade MgO, Cr_2O_3 and Fe_2O_3 were used maintaining the exact stoichiometric ratios. The oxide powders were weighed separately and then thoroughly mixed with ethanol in an agate mortar and then ball milled for 6 hours in a stainless steel ball mill. For fine mixing of the ingredients a small amount of distilled water was used as a milling fluid. After milling, the materials were dried and then calcinated at 800°C in air for 6 hours in a muffle

furnace. After calcinations, the chunk of the samples was ground into very fine powder in an agate mortar. The powders were then pressed into pellets and toroidal rings for sintering. A small quantity of polyvinyl alcohol (PVA) was used as a binder. The compacts were then successively sintered in a muffle furnace in air at temperatures 1250°C, 1300°C and 1350°C for 5 hours and then cooled at room temperature.

X-ray diffraction study was carried out in order to check the quality and phase purity of the samples using an X-ray diffractometer with $Cu(K\alpha)$ radiation of wavelength 1.54178Å at Atomic Energy Center, Dhaka (AECD). The X-ray diffraction patterns recorded in the angular range $15° \leq 2\theta \leq 65°$ with a step size of 0.02 showed sharp lines corresponding to single-phase cubic spinel structure for all the samples. Measurements of magnetic permeability and magnetic loss tangents were carried out at different frequencies using an LCR Meter Bridge (Hewlett Packard Impedance Analyzer, Model: 4192ALF) at room temperature. Curie temperatures of all the samples were determined from the $\mu'-T$ curve as described by Soohoo [18].

3. Results and Discussion

X-ray diffraction (XRD) patterns of the spinel system $MgCr_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) at room temperature are shown in **Figure 1**.

The patterns showed well defined sharp Bragg peaks and no impurity phase has been identified in any of the samples sintered. The fundamental reflections from the crystal planes (220), (311), (222), (400), (331), (422), (333) and (440) characterizing the spinel ferrites are clearly identified. Reflections from the planes (222) and (422) have been observed with weak intensities. The analysis of XRD patterns indicates that samples have formed the homogeneous single phased cubic spinel belonging to the space group Fd3m.

The variation of the lattice parameter (a_o) with Cr content (x) is presented in **Figure 2**. The lattice parameter decreases with increased substitution of Cr^{3+} ions in the spinel oxides $MgCr_xFe_{2-x}O_4$. This can be explained on the basis of the relative ionic radii of Cr^{3+} and Fe^{3+} ions present in the system. Since Cr^{3+} ions have smaller ionic radii (0.615Å), than those of Fe^{3+} ions (0.645Å) in the octahedral sites, a partial replacement of the Fe^{3+} ions by the Cr^{3+} ions causes shrinkage of unit cell dimensions, thereby decreasing the lattice parameter [19]. The lattice parameter decreases gradually with increasing x values satisfying the Vegard's law [20]. The difference in the lattice parameter may also be explained by the cation migration since Cr^{3+} ions on the A-site exhibit a smaller ionic radius than on the B-sites because of the covalence effects.
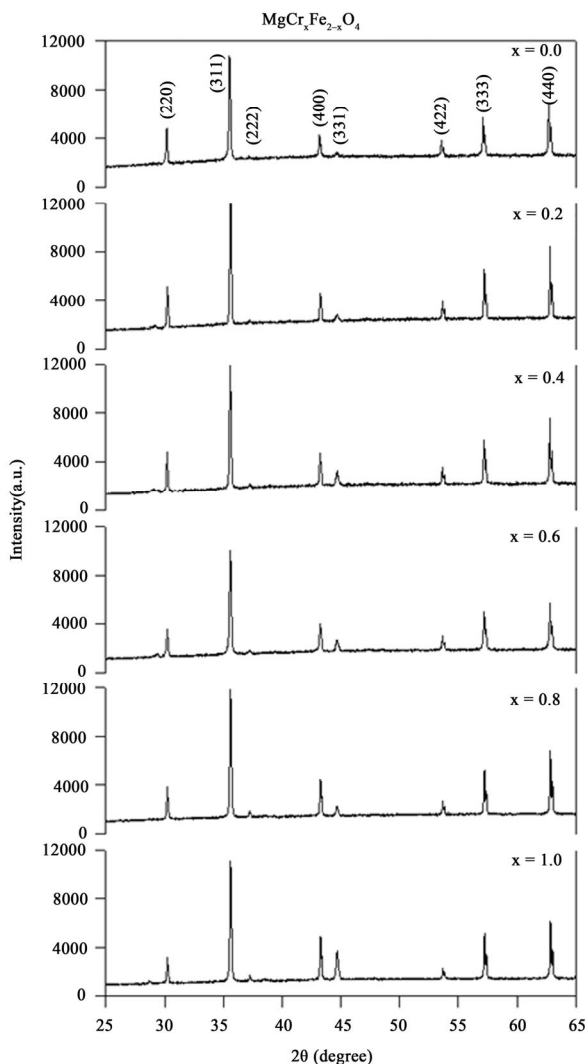
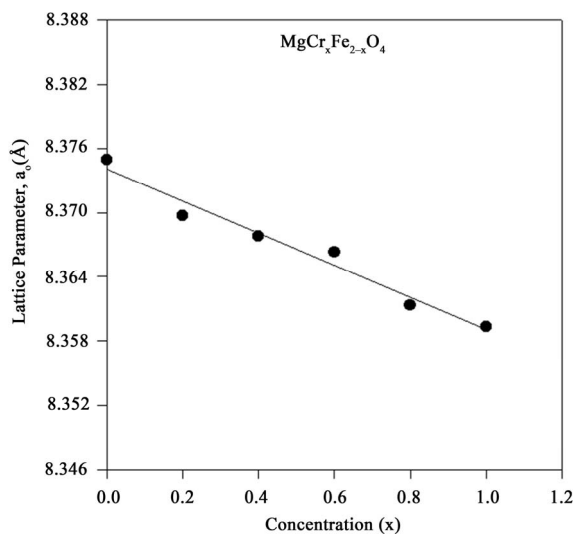**Figure 1. X-ray diffraction patterns showing single phase structure for the spinel oxides $MgCr_xFe_{2-x}O_4$.**

The bulk density ($\rho_B = m/v$) of the system under study were determined from the pellet samples after sintering at 1300°C. The X-ray density ($\rho_x = 8M/Na_o^3$) of the samples have been calculated using the lattice parameter (a_o) obtained from X-ray diffraction measurements. The variation of the X-ray density (ρ_x) with Cr content in the system is shown in **Figure 3**.

The porosity of the samples have been determined from X-ray density and bulk density using the equation $P = (1 - \rho_B/\rho_x) \times 100$. The lattice parameter, X-ray density, bulk density and porosity of $MgCr_xFe_{2-x}O_4$ 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 formed and developed during the sample preparation or the sintering process. The X-ray density and bulk density decrease with the increase of Cr content in the system. This is because of the greater atomic weight of Fe (55.845 gm/mol), while this value for Cr is (51.996 gm/mol) [21]. Porosity of the samples changes monotonically with Cr content (x) in the system as shown in **Table 1**.

Temperature dependence of the permeability for Mg $Cr_xFe_{2-x}O_4$ ferrites have been measured at a constant frequency 100kHz and are presented in **Figure 4**, from which Curie temperatures (T_C) have been determined. In the μ'-T curves, it can be seen that the initial permeability (μ') increases with increasing temperature attaining a maximum value just below the Curie temperature (T_C). The maximum value of μ' just below T_C is a manifestation of Hopkinson peak [22] attributed to the minimization of anisotropy energy with temperature. Beyond this peak value, initial permeability (μ') sharply falls to a very low value indicating the ferromagnetic to paramagnetic phase transition. T_C has been taken at the temperature
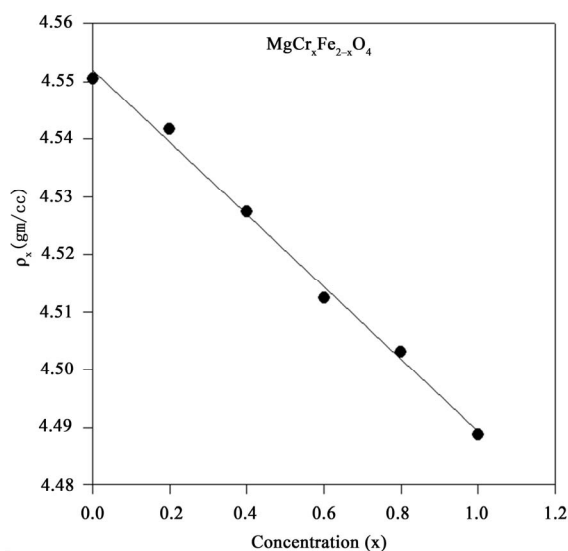

Figure 2. Variation of lattice parameter with Cr content.

Figure 3. Variation of the X-ray density (ρ_x) with Cr content in the spinel oxides $MgCr_xFe_{2-x}O_4$.
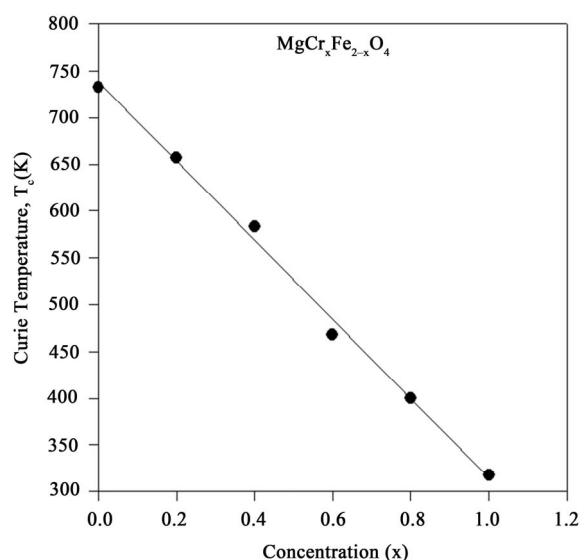
Table 1. Lattice parameter (a_o), cell volume (a_o^3), X-ray density (ρ_x), bulk density (ρ_B) and porosity (P) of $MgCr_xFe_{2-x}O_4$ ferrites sintered at 1300˙C.

Cr content (x)	Lattice parameter a_o (Å)	Volume a_o^3 (Å³)	X-ray density ρ_x (gm/cc)	Bulk density ρ_B (gm/cc)	Porosity P (%)
0.0	8.3740	587.2173	4.55056	4.05323	10.9290
0.2	8.3690	586.1661	4.54166	4.04524	10.9304
0.4	8.3670	585.7460	4.52735	4.03508	10.8732
0.6	8.3660	585.5360	4.51238	4.02339	10.8366
0.8	8.3610	584.4867	4.50305	4.01327	10.8766
1.0	8.3590	584.0674	4.48883	4.01066	10.6424
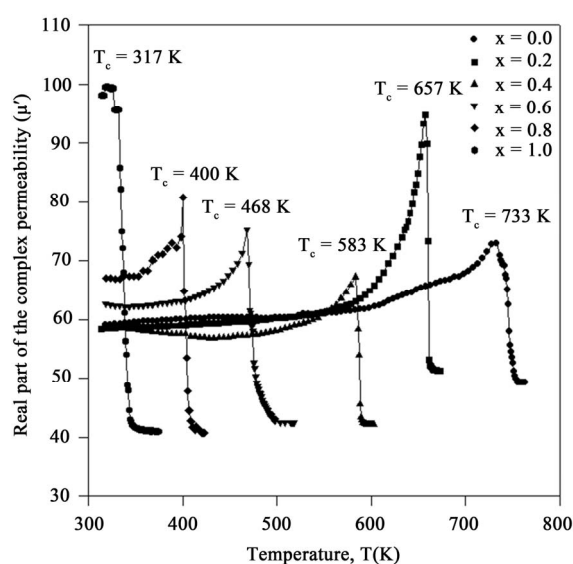
Figure 4. Permeability as a function of temperature for $MgCr_xFe_{2-x}O_4$ spinel ferrites.

Figure 5. Curie temperature (T_C) as a function of Cr concentration.

where a sharp fall of permeability is observed. The sharpness of the fall of permeability indicates the homogeneity and the phase purity of the studied samples [23, 24], which have also been confirmed by the XRD experiment mentioned above.

The values of Tc, as estimated from **Figure 4**, are found to be 733 K, 657 K, 583 K, 468 K, 400 K and 317 K for x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0, respectively in $MgCr_xFe_{2-x}O_4$ ferrites. The Curie temperature decreases linearly with increased substitution of Cr^{3+} ions as shown in **Figure 5** and can be explained on the basis of the number of magnetic ions present in the two sublattices and their mutual interactions. As Fe^{3+} ions are gradually replaced by Cr^{3+} ions, the number of strong magnetic ion begin to decrease at both the sites which also weakens the strength of A-B exchange interactions. Thus, the thermal energy required to offset the spin alignment decreases, thereby decreasing the Curie temperature in the system.

Mg-Cr ferrite is a mixed normal-inverse spinel type ferrite with a general formula $(Fe_{1-\partial}Mg_\partial)_A [Cr_{1-x}Fe_{x+\partial}Mg_{1-\partial}]_B O_4$, where the term within the square brackets indicates the octahedral (B) sites and the first term within the first bracket is tetrahedral (A) sites. The Cutie temperature in the ferrite samples is determined by the overall strength of the intersublattice AB interactions [25], but sometimes the intrasublattice AA and BB interactions may become important. The decrease in T_C with increasing concentration of Cr may be explained by a modification of A-B exchange interaction strength due to the change of Fe^{3+} distribution between A and B sites. The Curie temperature of magnetic material with an ideal crystal chemical composition has been found to be closely related to the number of Fe_A^{3+}-O-Fe_B^{3+} linkages per Fe^{3+} ion per formula unit and also on their distribution over A and B sites. The chains of such linkages are indefinitely long in a compound of ideal chemical composition. However, when iron ions are reduced in number due to the substi-

tutions, the number of active linkages decrease and thereby weakening the A-B exchange interaction. The internal energy required to offset the spin alignment will decrease and the Curie temperature is expected to fall. This could also be attributed to the increase in distance between the moments of A and B sites, which has been confirmed by the decrease in the lattice parameter with increasing Cr content in the system.

Anisotropy constants vary considerably with temperature. In most cases, anisotropy decreases faster than magnetization from a high value at low temperature as the temperature increases and finally becomes zero at T_C [22, 24]. At T_C there is no preferred crystallographic direction for the magnetization of a domain. It is observed that the initial permeability increases with temperature to a maximum value just below the T_C. This is due to the fact that the crystal anisotropy normally decreases with increasing temperature [26]. The initial permeability varies as $\mu_i \approx M_S/K^{1/2}$ [22,27]. Since anisotropy decreases faster than magnetization on heating the initial permeability increases with temperature, tends to infinity just below T_C and then drops sharply at T_C.

The variation of the real part of complex permeability (μ') in the frequency range 1 kHz to 13 MHz for the Mg Cr$_x$Fe$_{2-x}$O$_4$ ferrite series at three different sintering temperatures 1250°C, 1300°C and 1350°C, respectively are shown in **Figures 6-8**. The general characteristic of the μ'-f spectra is that, μ' remains constant up to a certain frequency range, while at higher frequencies, after a small rise, it drops rapidly to a very small value. The real part of initial permeability increases with increasing sintering temperature. The increase in permeability with increasing sintering temperature was observed by Nakamura [28] for NiZnCu ferrites.
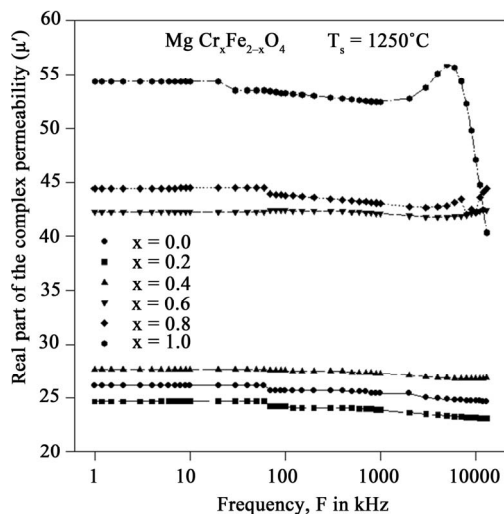
The initial permeability (μ_i) increases with increased substitution of Cr^{3+} ions as shown in **Figure 7**. The contributions to initial permeability arise due to domain wall displacement and domain rotation and are given by the expressions [29,30].
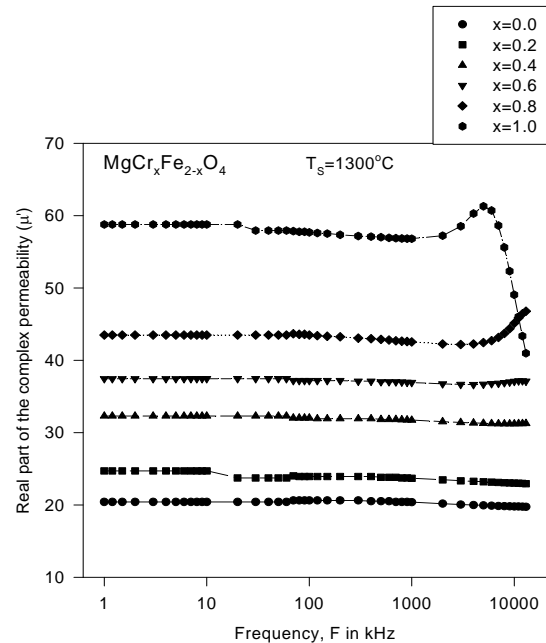

Figure 7. Variation of the real part of complex permeability (μ') in the frequency range 1 kHz to 13 MHz for the MgCr$_x$Fe$_{2-x}$O$_4$ series at 1300°C.

Figure 6. Variation of the real part of complex permeability (μ') in the frequency range 1 kHz to 13 MHz for the MgCr$_x$Fe$_{2-x}$O$_4$ series sintered at 1250°C.
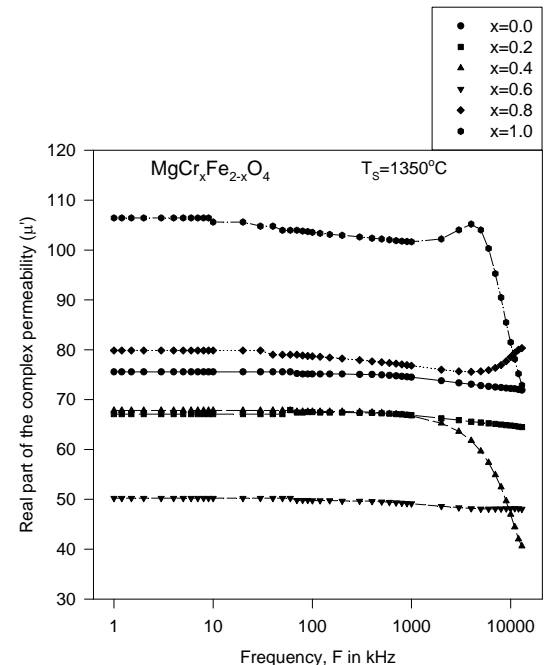

Figure 8. Variation of the real part of complex permeability (μ') in the frequency range 1 kHz to 13 MHz for the MgCr$_x$Fe$_{2-x}$O$_4$ series sintered 1350°C.

$$(\mu_{ic} - 1)_w = \frac{3\pi M_s^2 D_m}{4\gamma}$$

and

$$(\mu_{ic} - 1)_{rot} = \frac{2\pi M_s^2}{K_1}$$

where μ_{ic} represents the initial permeability corrected for density, D_m is the mean grain diameter, K_1 is the magnetocrystalline anisotropy constant, and γ is the wall energy per unit area. The contributions of magnetoelastic anisotropy and dipolar energy have not been considered as they remain practically constant and do not change with composition unless the substituted ions produce lattice distortions. Also, since D_m does not change significantly, the variations of μ_i can be attributed primarily to the variations of M_s and K_1. The change in K_1 with increased substitution of Cr^{3+} ions can be estimated quailtatively by applying the single ion anisotropy model [31]. Fe^{3+} ion occupying A site has a positive value of anisotropy constant while the other occupying B site has a negative value of it [32] and is twice as large as the positive value at A site. Since Mg^{2+} ions contributes insignificantly to the anisotropy constant, the ferrite of the composition $MgFe_2O_4$ (x = 0.0) is expected to have a negative value of K_1. Also, since Cr^{3+} ions have negligible anisotropy constant, the replacement of a part of Fe^{3+} ions by Cr^{3+} ions results an appreciable decrease in the anisotropy constant at B site. This, in turn, decreases the net value of anisotropy constant and thus, increases in the permeability with increasing Cr^{3+} ions concentration in $MgCr_xFe_{2-x}O_4$ ferrite system.

The initial permeability remains almost constant up to a frequency 1 MHz and increases thereafter. The complete resonance peak could not be observed in the frequency range employed in the present study and are expected to appear at frequencies >10 MHz. This effect may be attributed to the phenomenon of domain wall resonance due to domain rotation occurs in the microwave region [29]. The observed permeability spectra indicate that the expected resonance peaks are likely to appear at higher frequencies for all the samples. This is in accordance with the Globus model [33].

The magnetic loss factor ($\tan\delta$) exhibit high values at frequencies of about 1 kHz to 100 kHz, whereas it almost stabilize at very low values between 100 kHz and 8 MHz and increases thereafter for the spinel oxides $MgCr_xFe_{2-x}O_4$ at different sintering temperature (T_s). **Figure 9** shows the frequency dependence of magnetic loss factor at T_s = 1300°C as for representative one. The increase in loss factor at 10MHz may be attributed to the phenomenon of domain wall relaxation which involves the hindrance of domain wall motions of small grains by those of large grains where the latter type of grains occur in small number [34]. The critical relaxation frequency, f_c is not apparent in the present spectra and, presumably, lies beyond 10

MHz. Hence, it is difficult to determine the effect of substitution of Cr^{3+} ions on the relaxation frequency. The occurrence of high values of $\tan\delta$ near 1 kHz may be due to different types of relaxation process involved in the system [35].

Relative loss factor (RLF) is the ratio of the magnetic loss tangent to the initial permeability. The loss is due to lag of domain wall motion with respect to the applied alternating magnetic field and is attributed to various domain wall defects [36]. The values of RLF found in the present ferrite system are of the order of 10^{-3} to 10^{-4} at frequency 1 MHz. The Curie temperature (T_c), initial permeability (μ_i) and relative loss factor ($\tan\delta/\mu_i$) with Cr substitution in the spinel oxide $MgCr_xFe_{2-x}O_4$ are presented in **Table 2**. The RLF decreases with the increase of Cr content for samples sintered at 1300°C while for samples sintered at 1250°C and 1350°C it changes monotonically with Cr content. Loss factors are minimum for frequencies from 10 MHz to 13 MHz for all the samples at three different sintering temperatures. At high frequencies, losses are found to be lower. Evidently, the grain boundary can be modified by sintering and this has profound effects on the intragranular strength and magnetic properties. The initial permeability is therefore affected by the grain size, sintered density and anisotropy constant.

Figure 10 shows the frequency dependence of relative quality factor (RQF) for the samples sintered at 1300°C exhibiting maximum RQF values. The variation of the relative quality factor with frequency showed a similar trend for all the samples sintered at different temperatures. Q-factor increases with increasing frequency showing a peak and then decreases with further increase of frequency. It is seen that RQF decreases beyond 100 kHz i.e., the loss
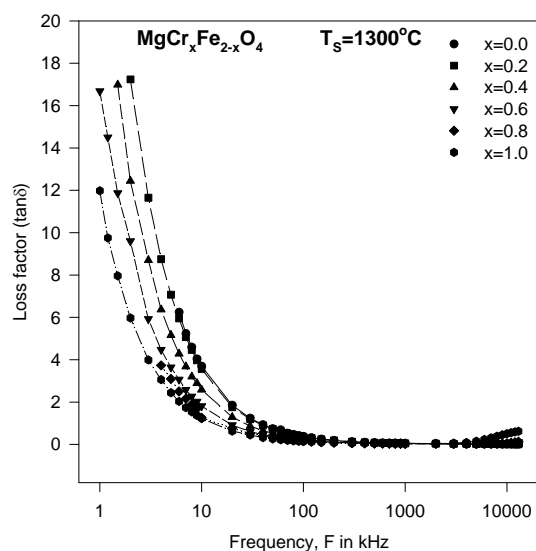
Figure 9. Frequency dependence of magnetic loss factor ($\tan\delta$) for the spinel oxides $MgCr_xFe_{2-x}O_4$ at sintering temperature 1300°C.

Table 2. Curie temperature (T_c), initial permeability (μ_i) and relative loss factor ($\tan\delta/\mu'$) of $MgCr_xFe_{2-x}O_4$ ferrite system.

Cr content (x)	T_c (K)	Sintering temperature, T_s = 1250°C		Sintering temperature, T_s = 1300°C		Sintering temperature, T_s = 1350°C	
		Initial permeability (μ_i)	Relative loss factor	Initial permeability (μ_i)	Relative loss factor	Initial permeability (μ_i)	Relative loss factor
0.0	733	18.12468	3.7×10^{-4}	19.79511	5.76×10^{-3}	72.21168	5.3×10^{-3}
0.2	657	18.51599	4.9×10^{-4}	23.02708	4.95×10^{-4}	64.79648	4.61×10^{-4}
0.4	583	26.85023	3.2×10^{-4}	31.22447	2.72×10^{-4}	46.96002	4.05×10^{-4}
0.6	468	37.18765	0.15×10^{-4}	37.08403	2.06×10^{-4}	48.15527	12.4×10^{-3}
0.8	400	42.21165	1.11×10^{-3}	45.13735	1.25×10^{-4}	78.53287	6.25×10^{-4}
1.0	317	47.05590	9.38×10^{-3}	49.06072	1.03×10^{-4}	81.48811	4.74×10^{-3}

tangent is minimum up to 100 kHz and then increases rapidly. This decrease may be due to the various defects in the domain that include non-uniform and non-repetitive domain wall motion. This happens at the frequency where the permeability begins to drop. This phenomenon is associated with the ferrimagnetic resonance within the domains [37] and at the resonance, maximum energy is transferred from the applied magnetic field to the lattice resulting the decrease in RQF. The peak corresponding to maxima in Q-factor shifts to lower frequency range as Cr content increases in the system. It is observed from **Figure 10** that sample with x = 0.4 sintered at 1300°C has the highest RQF value (6787.3364), which is probably due to the growth of less imperfection and defects in it than all other samples. Smaller grain size is competent to larger Q values. Though, the permeability being benefited, it appears that larger Cr content considerably decrease the quality factor of Mg ferrites.

4. Conclusions

The polycrystalline ferrites $MgCr_xFe_{2-x}O_4$ prepared in the solid state sintering method in air at 1300°C were characterized by X-ray diffraction technique. The diffraction measurements confirmed the single phase cubic spinel structure for all the ferrite specimens. Lattice parameter has been measured by using Nelson-Riley function and found to be decreased linearly with increasing Cr concentration obeying Vegard's law. The decrease in the lattice parameter may be due to the replacement of larger Fe^{3+} (0.645Å) ions from A site to B site by slightly smaller Cr^{3+} (0.615Å) ions. Curie temperatures (T_c) of all the samples have determined from the temperature dependent permeability measurement. The maximum value of the permeability μ' just below the Curie temperature is a manifestation of Hopkinson peak attributed to the minimization of anisotropy energy with temperature. The Curie temperature (T_c) decreased linearly with increasing Cr
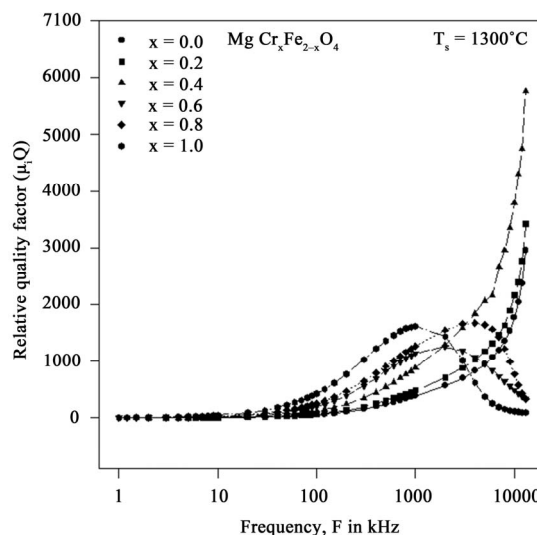
Figure 10. Frequency dependence of relative quality factor (RQF) for the spinel oxides $MgCr_xFe_{2-x}O_4$ sintered at 1300°C.

content may be due to the weakening of A-B exchange interaction. The internal energy required to offset the spin alignment decreases and Curie temperature is expected to fall.

Frequency stability of the real part of permeability (μ') increases with increasing Cr concentration and also with sintering temperature for all the samples. The increase in the permeability with increasing Cr content is connected with decreased density with the addition of Cr in the ferrites system. Quality factor increases with increasing frequency and decreases both with increasing Cr content and sintering temperature. The magnetic loss tangents decrease for larger substitution of Cr ions in the megahertz range. Hence, these materials may become suitable, particularly, for those applications for which the magnetic losses are of prime concern. Therefore, the increased substitution of Cr in the $MgFe_2O_4$ ferrites and various sinter-

ing condition has greatly influenced the magnetic properties of MgFe$_2$O$_4$ ferrites.

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