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Determination of Activation Energies from Complex Impedance Parameters of Microwave Sintered NiMgZn Ferrites

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ABSTRACT. The $\text{Ni}_{0.2}\text{Mg}_{0.8-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ ($x = 0.2, 0.4, 0.6$ & 0.8) ferrites are prepared via microwave double sintering solid state reaction method. The phase purity and structure are confirmed by diffraction studies. These investigations reveal that the lattice parameter as well as crystallite sizes are increasing with zinc content. In addition, the variation of complex impedance (Z^*) parameters as a function of temperature and frequency is studied. The activation energies of ferrites are evaluated from Arrhenius plots and observed to be decreasing with increase of x value.

Introduction. Spinel ferrites are used as well known electromagnetic materials such as due to their various electrical and magnetic properties [1-4]. Hiti [5] reported the variation of distinct ac-electrical properties with increase of zinc composition using conventional solid state reaction method. The microstructure and electrical properties of ferrites are analyzed by impedance (Z^*) properties [6]. This approach offers discussion on real (Z') and imaginary parts (Z'') of impedance providing the material properties. It also attributes the bulk and grain boundary effect from the relaxation behaviour of Z' and Z'' parameters [6]. The complex impedance (Z^*) is generally written as $Z^* = Z' - j Z''$. The real and imaginary parts are calculated using the equations: $Z' = Z \cos\phi$ and $Z'' = Z \sin\phi$, where ϕ denotes the phase [7]. In this study the authors discuss the behaviour of Z' , Z'' and relaxation time as a function of temperature and frequency. Also, the authors attempt to evaluate the activation energies from relaxation time.

Experimental Procedure. A series of $\text{Ni}_{0.2}\text{Mg}_{0.8-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ ($x = 0.2, 0.4, 0.6, 0.8$) are synthesized by microwave double sintering technique. The starting materials of NiO (99.5% purity, Sigma Aldrich), MgO (99.4% purity, Sigma Aldrich) and Fe_2O_3 (99.5% purity, Sigma Aldrich) are weighed according to the stoichiometric ratio and grinded (for 14hr) to make fine powder. The fine powder is pressed into cakes of 20 mm disk shape by applying 0.5 tons pressure in hydraulic press. The cakes are calcined at a temperature of 800°C for 30min. in microwave furnace (V.B. Ceramics, India). Later the calcined cakes are crushed into powder and are again grind for another 4hrs to make it into fine powder. The ferrite powder is uniaxially pressed into pellets (10mm diameter) applying 1 ton pressure. The pellets and powder samples are sintered at 1000°C for 30min in microwave furnace fitted with two magnetrons of 2.45GHz frequency with a power output of 2.2kW. The sintered samples are characterized using X-ray diffractometer (Bruker X-Ray Powder Diffraction Meter, $\text{CuK}\alpha$, $\lambda=0.15418\text{nm}$) at room temperature for structural identification. HIOKI 3532-50 LCR HiTESTER (Japan) is used for impedance analysis.

Results and Discussions.

Structural Properties. Fig.1 shows the diffraction pattern of Zn substituted NiMg ferrites. Phase purity and structure formation of spinel ferrites is confirmed. The peak positions are indexed in Fig.1 and are in consistent with standard JCPDS data file No. 08-0234. The crystallite size (D) is calculated using Debye-Scherrer formula [8, 9] and the corresponding values are changing between 22-50nm. The lattice constant ' a ' is increasing with increasing x value as the ionic radius of zinc (0.083 nm) is greater than that of magnesium (0.066 nm) and nickel (0.078 nm) [10]. X-ray density (D_x) is evaluated using relation [9]:

$$D_x = \frac{ZM}{Na^3} \quad (1)$$

where Z is the no. of molecules per unit cell ($Z = 8$), M is the molecular weight of the composition, N is Avogadro's number (6.023×10^{23}) and a is the lattice parameter.

The bulk density (D_b) is estimated by Archimedes principle. Both the densities are increasing with x . Further, porosity ($P\%$) is computed and found to be decreasing with increase of Zn^{2+} ions. The structural parameters are mentioned in Table.1.

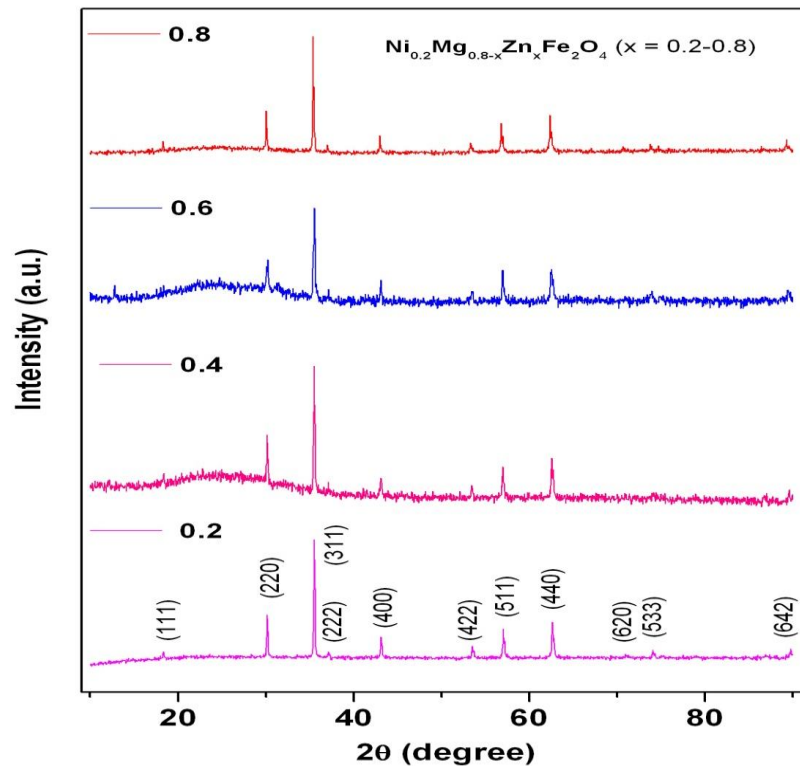


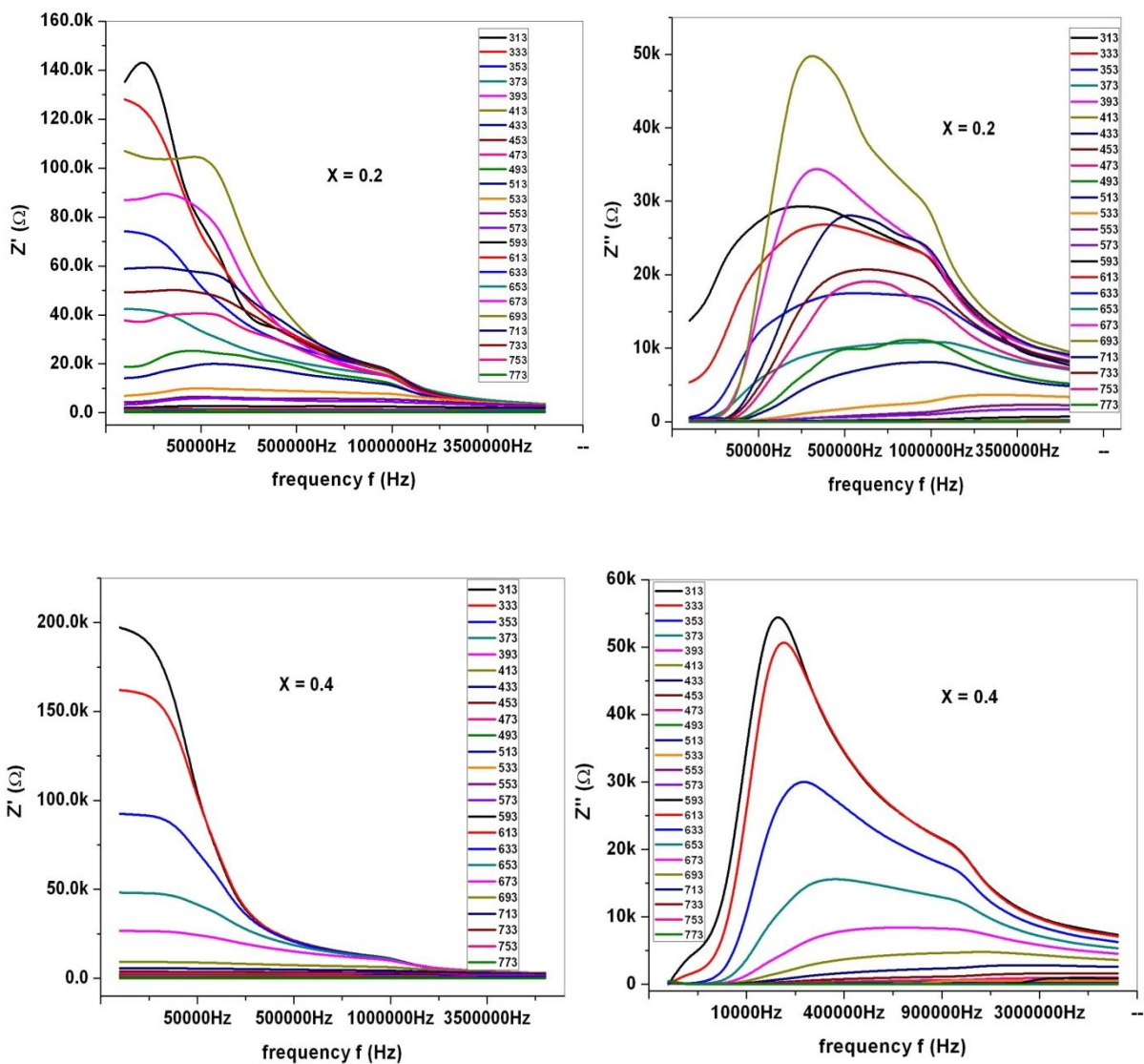
Fig. 1. XRD of $Ni_{0.2}Mg_{0.8-x}Zn_xFe_2O_4$ ($x = 0.2-0.8$).

Impedance Analysis. The frequency dependence of real (Z') and imaginary (Z'') parts at the selected temperatures (313K-773K) is depicted in Fig.2. It is seen from Fig.2 that Z' shows a constant trend at lower frequencies for $x = 0.2 - 0.8$. The real part is combined for all selected temperatures at higher frequencies. The space-charge effect [11, 12] is responsible for achieving larger Z' values at lower frequencies. But Z' values are decreasing with increase of temperature which can be attributed to smaller density of trapped charges [11]. Similarly, the frequency dependence of Z'' in the range of 313K-773K is shown in Fig.2. The relaxation peaks are moved to higher frequencies with increase of temperature from 313K-773K. It reveals that the NiMgZn ferrites show the temperature dependent

relaxation process. The Z'' versus frequency (with temperature) plots (Fig.2) provides the relaxation frequency.

Table 1. Structural and electrical parameters of NiMgZn ferrites.

x	0.2	0.4	0.6	0.8
a (Å)	8.383	8.394	8.401	8.412
D (nm)	21.6	27.4	32.5	49.7
D_x (g/cm ³)	4.850	5.015	5.186	5.350
D_b (g/cm ³)	4.525	4.739	4.955	5.183
Porosity (P %)	6.7	5.5	4.5	3.1
E_1 (eV)	0.37	0.34	0.32	0.30
E_2 (eV)	0.32	-	-	0.25



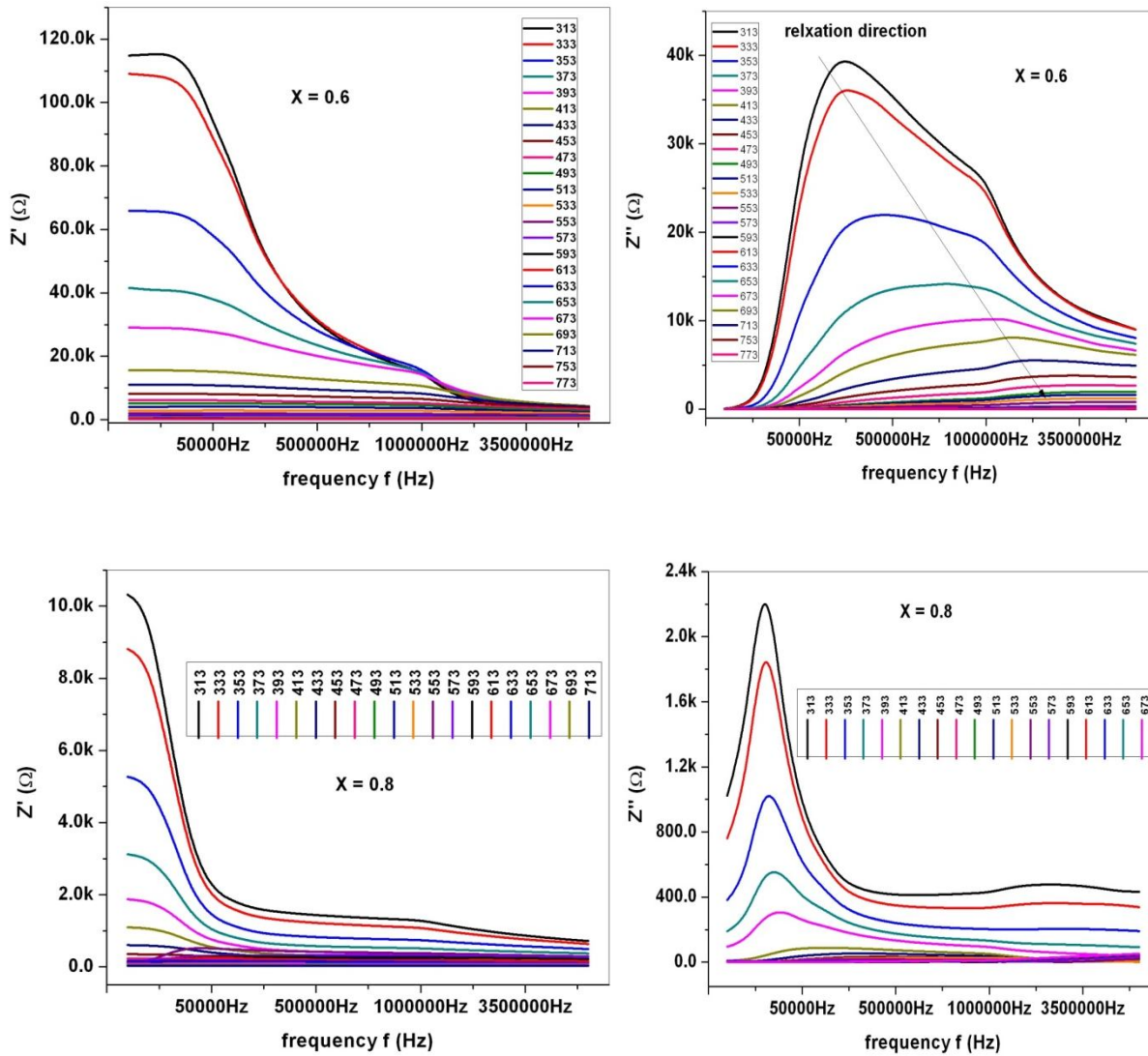


Fig. 2. Frequency dependence of Z' and Z'' parameters at selected Kelvin temperatures.

In addition, the relaxation time (τ) is calculated using the relation: $\tau = (2\pi f)^{-1}$ where f is the relaxation frequency. As the temperature goes on increasing the relaxation frequency also increased. Thus, the relaxation time is decreased with temperature. Similar trend is observed by Laurel et al [11]. Therefore, the variation of τ with T follows the Arrhenius law: $\tau = \tau_0 \exp(-E_a/K_b T)$, where τ_0 is a pre-exponential factor, K_b is Boltzmann constant and T is absolute temperature [13]. The Arrhenius plots ($\ln \tau$ versus $1/T$ plots) are drawn (Fig.3) to find the activation energies of ferrites. The plots reveal two linear slopes that belong to both extrinsic and intrinsic regions. These two slopes are taken at two regions i.e. region -I high temperature region ($>700K$) and region-II low temperature region ($<700K$). The slope change is usually occurred in ferrites due to either ferri – paramagnetic transitions or two hopping mechanism as reported in literature [14]. The estimated activation energies (E_1 & E_2) are reported in Table.2. It is observed from results that the energies at high temperature region are more than that of low temperature region. The conduction process is of extrinsic type at low temperature region. This can be attributed to the presence of impurities. Similar slope changes at two regions are reported in the literature [15].

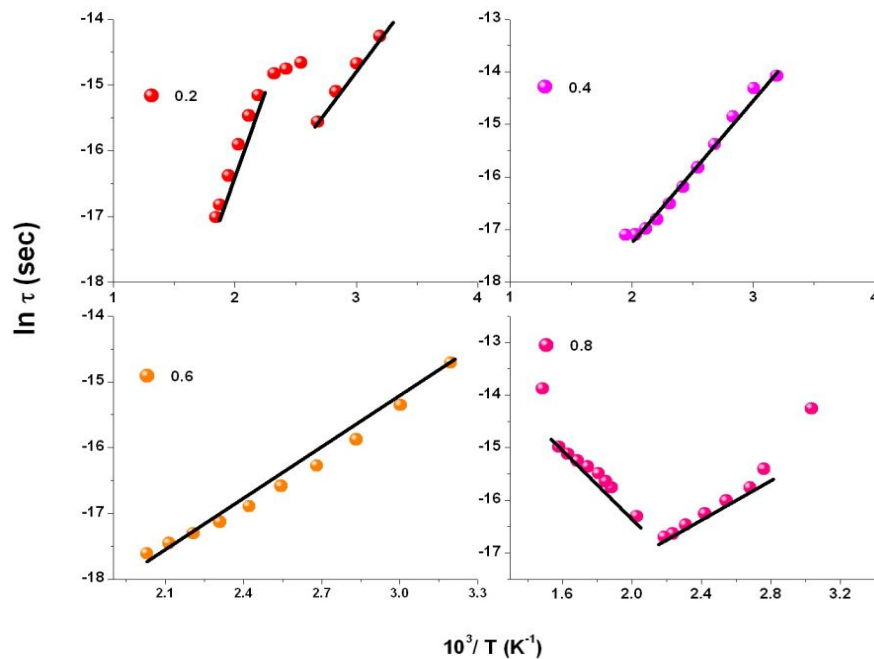


Fig. 3. Arrhenius plots ($\ln \tau$ Vs. $1/T$ plots) of NiMg ferrites from impedance relaxations.

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