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IMPEDANCE AND MODULUS STUDIES OF MAGNETIC CERAMIC OXIDE $Ni_{0.27}Cu_{0.10}Zn_{0.63}Fe_2O_4$

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Abstract- *The complex impedance spectroscopy is a very convenient and powerful technique that enables us to correlate the electrical properties of a material with its microstructure. This also helps to analyze and separate the contributions, from various components (i.e., through grains, grain boundary, interfaces, etc.) of materials in the wide frequency range. In the present work, the preparation of magnetic ceramic oxide* $Ni_{0.27}Cu_{0.10}Zn_{0.63}Fe_2O_4$ *is described by the solid state reaction method. The dielectric impedance properties were studied over the range of frequency between 20Hz-5MHz and in the temperature range of 303-498K, using the modulus formalism. The impedance plot showed a first semicircle at high frequency which was assigned to the grain intrinsic effect and a second semicircle, at lower frequencies, which corresponds to grain boundary polarization. A complex modulus spectrum was used to understand the mechanism of the electrical transport process, which indicates that a non-exponential type of conductivity relaxation characterizes this material. The study of the dielectric property and loss of ferrite materials, as a function of temperature, is important for microwave absorption applications.*

Keywords: NiCuZn ferrite, Temperature dependence, Impedance spectroscopy, Dielectric relaxation.

1. INTRODUCTION

With the latest development of satellite communications and consequent mobile applications, the miniaturization and high efficiency of devices for information and communication technology have been required, especially on multilayer components. This rapid development of wireless communication necessitates the design of new ceramics, sinterable at low temperature, and exhibiting good dielectric properties [1-4]. The physical properties of ferrites are controlled by the preparation conditions, chemical composition, sintering temperature and time, type and amount of substitutions [5]. NiCuZn come under the umbrella of the soft ferrites and chemically symbolized as MFe_2O_4. In general, the cation distribution in spinel lattice has the form: $(D_{1-x}M_x)[D_xM_{2-x}]O^{-2}_4$, where D and M are divalent and trivalent ions respectively, and x is called the degree of inversion. The round and square brackets denote the cations located at the center of tetrahedral lattice of oxygen (A) and those at octahedral (B) lattice respectively [6].

Low cost, easy manufacturing, and interesting electrical and magnetic properties make polycrystalline ferrite one of the most important materials in use today. As magnetic materials, ferrites cannot be replaced by any other magnetic material because they are relatively inexpensive, stable, and have a wide range of technological applications in transformer cores, high quality filters, high and very high frequency circuits, and operating devices [7-8]. The high electrical resistivity and consequently low eddy currents and dielectric losses make them very important material for technological and industrial applications. The electrical properties of these materials have been the subject of continuous investigation, which depend upon the preparation conditions, amount of doping element and preparation time etc.

It is well known that impedance spectroscopy is an important method to study the electrical properties of ferrites, since impedance of the grains can be separated from other impedance sources, such as impedance of electrodes and grain boundaries. One of the important factors, which influence the impedance properties of ferrites, is microstructural effect. Two semicircles are often obtained in Cole–Cole plot, when the grain boundary resistance is larger than that of grain. The distributions of relaxation times, which result in deviation from ideal semicircles, are attributed to various factors, such as disorder in the samples, and inhomogeneity in microstructures [9].

Ferrite, as a semiconductor, has attracted considerable attention in the field of technological applications in a wide range of frequencies, extending from microwave to radio frequency.

The classic ceramics method for preparing soft spinel [10] ferrite requires a high calcinations and sintering temperature, which induces aggregation of the particles. Many physical properties of polycrystalline ferrites are very sensitive to the microstructure. The grain (bulk) and grain boundary are the two main components that determine the microstructure. Thus, information about the associated physical parameters of the components that constitute the microstructure is important in understanding the overall properties of the materials [11].The concept of dielectric relaxation is associated with ionic/electronic conductivity in ceramic oxides under the influence of an ac field.

In the present work, the electrical properties of NiCuZn ferrites have been investigated by impedance spectroscopy. It is preferable to plot the complex impedance or conductivity for characterizing our ferrite materials when dealing with charge carrier systems. The impedance spectroscopy or ac conductivity technique enables us to evaluate and separate out the contributions to overall electrical properties due to various components such as grain, and grain boundary or polarization phenomenon in a material, in the frequency or time domains. Here, the principle of analysis is based on the fact that ac response of a sample to sinusoidal electrical signal, and subsequent calculation of the resulting transfer (impedance) with respect to the frequency of the applied signal. The aim of this work is to study the bulk and interface phenomena over a wide range of frequencies and at selected temperatures in order to get information about the relaxation times, and relaxation amplitudes of various processes present in the system, when a small perturbation signal is sent to the system over a wide range of frequencies. It is worth to note that various physical parameters and characteristic properties that influence the performance of a ferrite material can be obtained from the analysis of complex impedance spectra.

2. EXPERIMENTAL

Polycrystalline spinel ferrite with chemical formula $Ni_{0.27}Cu_{0.10}Zn_{0.63}Fe_2O_4$ was prepared by conventional solid state reaction technique. High purity 'AR' grade oxides, iron oxide (Fe_2O_3), nickel oxide (NiO), zinc oxide (ZnO) and copper oxide (CuO) were mixed together according to their chemical weights. The mixture of each composition was ground to a very fine powder, and calcinated at 1173K for 5h. Finally, the samples were ground to fine powder, pressed into disk shaped pellets, and sintered at 1573K for 5h. At the start and end of each heat treatment the samples were allowed to warm and cool slowly to room temperature at a rate of 10K/min. and 5K/min. respectively. The pellets were polished to make the opposing surfaces as parallel as possible. Silver paste coating was applied on the opposite faces of pellets, in order to make parallel plate capacitor geometry and the ferrite material as dielectric medium. The complex impedance measurements were carried out using a computer controlled impedance analyzer (Wayne kerr, Model no. 6500B) as a function of frequency (20Hz to 5MHz), and temperature (303K–498K).

3. RESULTS AND DISCUSSION

3.1 Complex impedance spectrum analysis

The complex impedance spectroscopy method [12] has been used to analyze the electrical and dielectric properties of a polycrystalline sample and their interfaces with electronically conducting electrodes in a wide range of frequencies (20 Hz–1MHz) at different temperatures (303–498 K). The dielectric properties of a material are often represented in terms of complex dielectric permittivity, ε*, complex impedance, Z*, and electric modulus, M*, which are related to each other as: $Z^* = Z' - jZ''$; $M^* = 1/\varepsilon^*(\omega) = j(\omega C_0)Z^* = M' + jM''$, where (Z', M') and (Z'', M'') are the real and imaginary components of impedance and modulus, respectively, $j = \sqrt{-1}$ is the imaginary factor and ω is the angular frequency, $\omega = 2\pi f$.

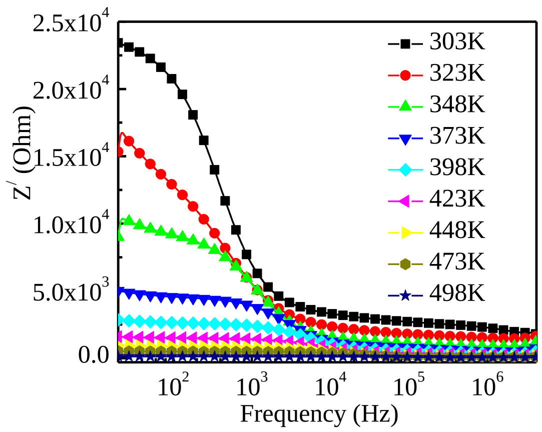

Fig.1: Z' vs frequency at several measurement temperatures of the NCZ ferrite.

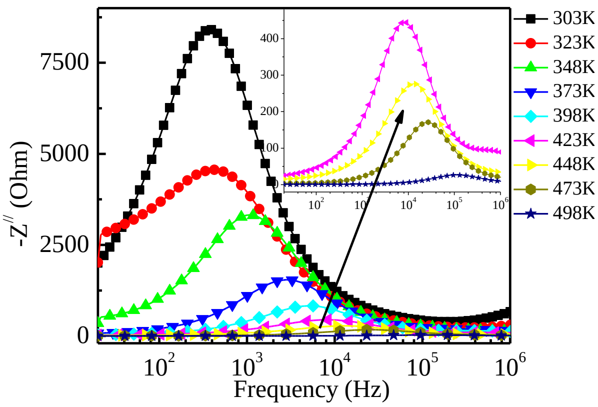

Fig.2: Z'' vs frequency at several measurement temperatures of the NCZ ferrite.

The variation of Z', Z'' and Z' versus Z'' of impedance at selected temperatures are shown in Figs.1–3. The Fig. 1 shows the variation of the real part of the impedance (Z') with frequency at different temperatures (333–498 K). The observed behavior indicates that Z' is related to the sample electric resistance, and decreases with the rise of the frequency and temperature. Therefore, the conduction should increase with the rise of the temperature. At higher frequencies, the Z' values at different temperatures present a limit to the same point, suggesting a possible release of space charge and a consequent lowering of the energy barrier properties [13–15]. Figure 2 represents the variation of the Z'' with frequency at different temperatures. This plot is suitable

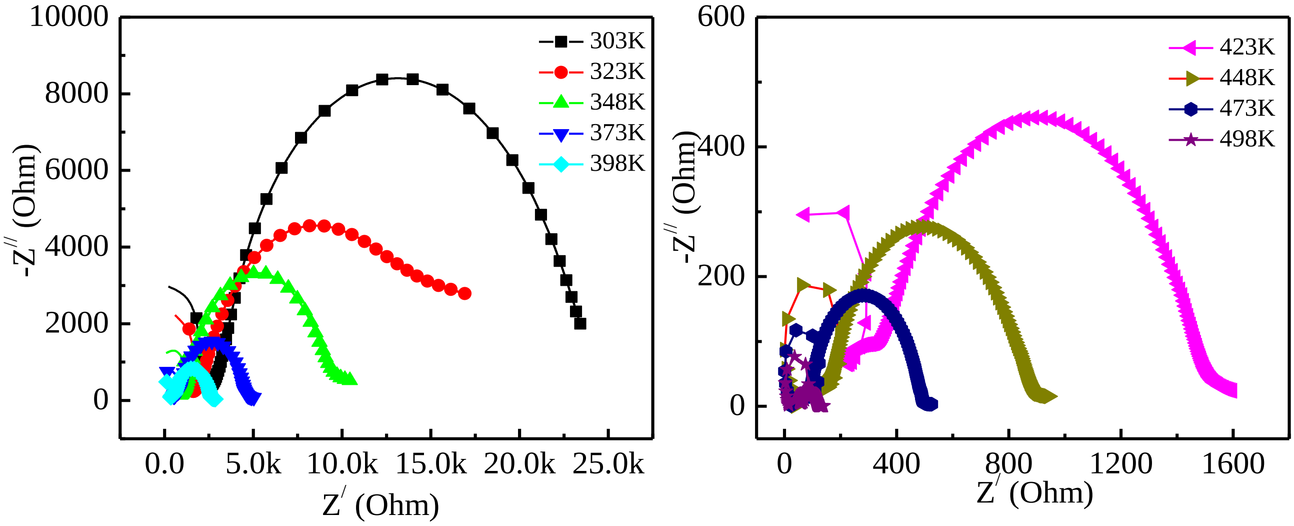

Fig.3: $Z^{/}$ vs $Z^{//}$ at several measurement temperatures of the NCZ ferrite.

to evaluate the relaxation frequency, if it exists, of the sample's most resistive component. It is observed that two peaks appear for all measurement temperature, indicating the presence of at least two dielectric relaxation processes. This spectrum presents some important features: (i) the appearance of two peaks, (ii) the position of the peaks shift to the higher frequency with the increase of the temperature, (iii) the absolute value of $Z^{//}$ decreases and shifts to the higher frequency side as the temperature increases, (iv) the first peak shows the typical peak broadening, and the second peak shows the typical peak symmetric, and (v) a decrease in the height of the peaks with increasing temperature. The merging of all of the curves above 1 kHz indicates a possible release of the space charge. Broadening of the $Z^{//}$ peaks with the rise in temperature indicates a non-unique (i.e., multiple) relaxation time scale, whose variance/distribution it is broadening at the higher frequency side after the position of the peak maximum [14,16].

The variation of the real versus the imaginary part of the impedance at selected temperatures (i.e., 303–498 K) is shown in Fig.3, which is characterized by the appearance of two semicircles for all temperatures.

The center of those semicircles are located below the real axis ($Z^{/}$), indicating a non-Debye type of relaxation process and the presence of a relaxation time distribution. This suggests the presence of several different dielectric entities that contribute to the dipolar moment. In all of the temperature ranges, these two depressed semicircles were observed, centered at the low and high frequency regions. The impedance spectra presenting a spike or depressed semicircle in the low frequency region are assigned to dielectric polarization between the sample surface and the electrode and are thus extrinsic to the sample. This is known as the Maxwell–Wagner effect [3].

The small diameter of the high frequency semicircle represents small bulk resistance, which decreases with the increase of the temperature. This behavior indicates a thermally activated conduction mechanism. In these types of materials (ceramic materials), these semicircles in the impedance formalism, are generally ascribed to grain (bulk) and grain boundary (interface) effects [17]. Electrically, these two semi-arcs can be represented by an equivalent circuit constituted by the parallel combinations of resistance, R, and capacitance, C, connected in series with another RC parallel connection. One branch is related with sample intrinsic characteristics, i.e., with the grain (bulk), and other with the interfaces, grain boundaries and surface-electrodes [9,11,18].

In the impedance formalism, the relative position of the semi-arc depends upon the resistance and capacitance values. The resistance and capacitance of the interfacial grain boundary is usually larger than the grain. Thus, the semicircle at high frequencies corresponds to the grain effect (bulk) and the second semicircle, at lower frequencies, to the grain boundary (conduction phenomenon). It is important to remember that the impedance spectra are reproducible within the experimental error. This fact excludes the possibility that the lower frequency loop might be due to other sources such as the contacts between the electrodes and material surfaces.

3.2 Dielectric analysis

The dielectric modulus formalism (M*), have been adopted to study the frequency and temperature dependence of the conductivity of ceramics, [19].

The electric modulus approach began when the reciprocal complex permittivity was discussed as an electrical analog to the mechanical shear modulus [20]. The usefulness of the modulus representation in the analysis of the relaxation properties has been demonstrated for polycrystalline ceramic [21–23].

In the sample, the frequency dependence of the real part of the dielectric modulus ($M^{/}$) as a function of frequency over a range of temperature is given in Fig.4. It was observed that the value of $M^{/}$ is very low (approaching zero) in the low frequency region. A continuous increase in the $M^{/}$ dispersion with the increase of the frequency shows a tendency to saturate at a maximum asymptotic value (i.e., $M_{\infty} = 1/e_{\infty}$), for all the temperatures. This supports the suggestion that the short range mobility of charge carriers is a conduction process [24]. These observations may possibly be related to a lack of restoring force governing the mobility of charge carriers under the action of an induced electric field. These features indicate that the electrode polarization makes a negligible contribution in the material [25].

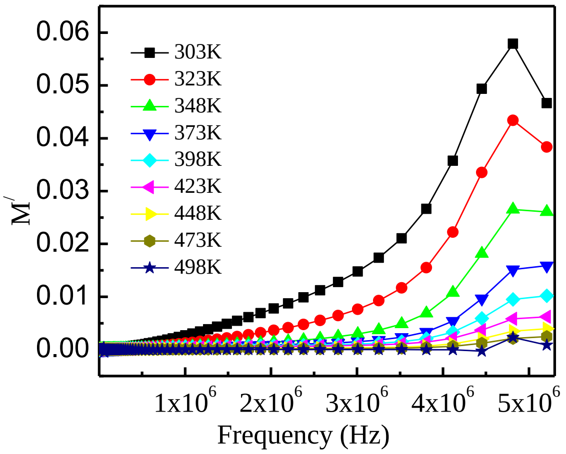

Fig.4: $M^{/}$ as a function of frequency of the NCZ ferrite.

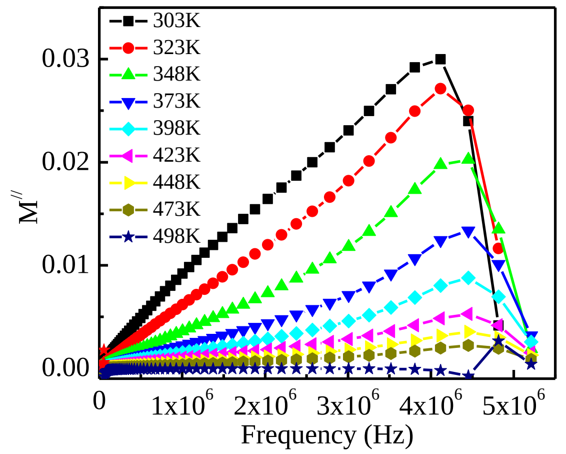

Fig.5: $M^{//}$ as a function of frequency of the NCZ ferrite.

The frequency dependence of the imaginary part of the electric modulus, $M^{//}$ (f), exhibits a maximum in Fig. 5. It may be noticed from Fig.5 that the position of the peak shifts toward higher frequencies as the temperature increases. The frequency region below the peak maximum determines the range in which charge carriers are mobile over long distances. At the frequency above peak maximum (high-frequency), the carriers are confined to potential barriers, being mobile over short distances. The region where the peak occurs is indicative of the transition from long-range to short-range mobility with increase in frequency.

This type of behavior suggests the existence of a temperature-dependent hopping type mechanism for electrical conduction (charge transport). The broadening of the peak shows the spread of the relaxation with a relaxation time constant distribution. Furthermore, the appearance of peaks in the modulus spectrum provides a clear indication of the conductivity relaxation. The frequency of the $M^{//}$ maximum (f_{max}) gives the mean value of the relaxation time distribution, $\tau_\sigma = 1/(2\pi.f_{max})$. This indicates that the electric entities responsible for the conduction and for the relaxation process, observed in M*, are the same [26–30].

The observed dielectric relaxation mechanism shifts to higher frequencies with the increase of the measurement temperature indicating an increase in the difficulty of the dielectric dipoles during depolarization. This relaxation, due to the modulus formalism characteristics, should be attributed to the intrinsic characteristics of the samples.

It is known that the conductivity and polarization phenomena in ferrites has been largely attributed to the presence of Fe^{2+} ions, which give rise to a heterogeneous structure, and the consequent electron exchange between Fe^{2+} and Fe^{3+}. Moreover, if nickel, copper are introduced into the ferrite composition, which is the present case, the contribution from the hole transfer between the Ni^{3+} and Ni^{2+}; Cu^{+} and Cu^{2+} ions should also be taken into consideration [31].

In the conduction process, the contribution of the holes is smaller than the electrons, due to their lower mobility. Therefore, the electron exchange between the Fe^{2+} and Fe^{3+} ions, which results in local displacements in the direction of the applied external electric field (while the hole exchange between Ni^{3+} and Ni^{2+}; Cu^{+} and Cu^{2+} is in the opposite direction), causes the dielectric polarization in ferrites [39]. Nevertheless, a charge exchange between Ni^{2+} and Fe^{3+} can also exist [32]. However the polarization of Fe^{2+}-Fe^{3+} is easiest and thus their number will be reflected in the dielectric constant value. The results of Fig.5 indicate that this number should be the same for all measurements. This is true if we assume that the maximum measurement temperature used (498 K) is not enough to promote the modification of the oxidation state of this ion.

The temperature dependence of the complex modulus spectrum ($M^{/}$ vs $M^{//}$) of NCZ ferrite is shown in Fig. 6. The appearance of an arc in the spectrum at a particular temperature confirms the single phase character of the materials. At higher temperatures, these arcs are not exact semicircle(s). They are in the form of two deformed semicircles (or their tendency) with their centers lying below the real axis. This indicates spread of relaxation with different (mean) time constant and hence again supports the non-Debye type of relaxation in the materials. The two semicircular arcs or their tendency in the complex modulus plots suggest the presence of both the grain and grain boundary contributions in the NCZ compounds. It is also observed that with the increase in temperature, intercept of the arcs on real $M^{/}$ axis shift towards the higher values of $M^{/}$ which indicates about the increase in capacitance. It supports the negative temperature coefficient of resistance (NTCR) type behavior of the materials since bulk capacitance (C_b) is inversely proportional to the bulk resistance (R_b). At higher temperatures and lower frequencies, tails are not fully evolved to its true shape which may be due to frequency and temperature limitations. The single semicircular arcs in Fig. 2 exhibit the dominant bulk (grain) contribution whereas the double semicircular arcs in Fig. 6 suggest the presence of both the bulk (grain) and grain boundary contributions. It is based on the fact that

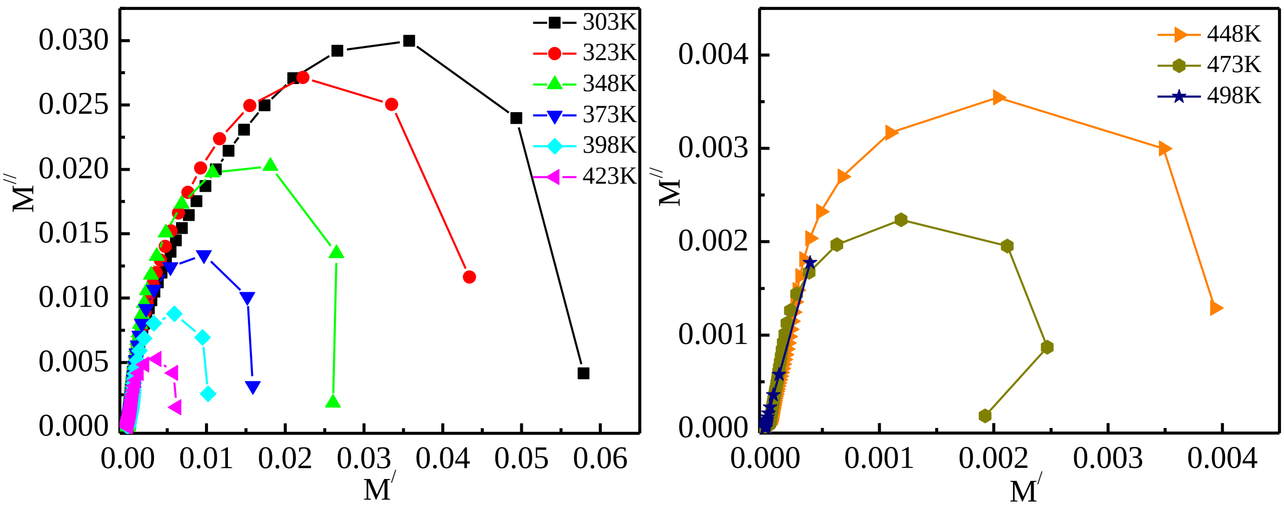

Fig.6: $M^{/}$ vs $M^{//}$ at several measurement temperatures of the NCZ ferrite.

the impedance plot highlights the phenomenon with the largest resistance whereas modulus plot picks out those of the smallest capacitance. With the huge difference (orders of magnitude) between the resistive values of grains and grain boundaries, it is difficult to obtain two full semicircles for grains and grain boundary on the same scale in the impedance plot. Complex modulus analysis is suitable when materials have nearly similar resistance but different capacitance [33].

4. CONCLUSIONS

The present work reports the results of our investigation on the dielectric, impedance and electrical modulus properties of the magnetic ceramic compound $Ni_{0.27}Cu_{0.10}Zn_{0.63}Fe_2O_4$. Single arc with single/double semicircle(s) (or tendency) obtained at a particular temperature corresponding NCZ composition in both the complex impedance and modulus plots suggests the single phase character of the materials. The complex impedance plots reveal two semicircles described by the grain (bulk) and grain boundary (interface) effects. The modulus plot shows non-Debye behavior and is asymmetric with respect to the peak maxima and the peaks are considerably broader on both sides of the maxima and suggest dielectric relaxation in the material. The interfacial polarization is a consequence of inhomogeneities in the material, which is dominated by grain boundaries.

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