

INVESTIGATION OF TEMPERATURE SENSITIVE ELECTRICAL PROPERTIES OF MANGANESE-ZINC FERRITES

By

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ABSTRACT

Keeping pace with facets of nanotechnology and its applications in the field of development of smart instrumentation to cater today's needs and future requirements of various sectors, sensor is the key portion of the measurement system, which responds directly to the physical variables that need to be measured. Therefore, one should opt for proper sensor of better characteristics, nanoparticle spinel Manganese-Zinc ferrites have been synthesized by co-precipitation method. The formation of the materials is confirmed by X-ray powder diffraction and FTIR absorption technology. The results of X-ray diffraction investigation confirm the formation of a single phase composition with the average particle size from 40 nm to 48 nm. Temperature dependent electrical properties of the compositions of $Mg_{1-x}Zn_xFe_2O_4$ nano ferrites were investigated for suitability of these materials as a sensing element for designing of sensors. The sensors developed, are employing thick film technology. Measure the DC electrical resistivity of the pelletized compositions and shows the semiconducting behavior, which is attributed to the electron hopping mechanism. This electrical conductivity exhibits the influence of magnetic ordering at curie temperature. The curie temperature values depict the compositional dependence. The electrical resistivity shows a negative temperature coefficient with temperature, hence the materials could be used to design temperature sensors. The results of implementation are interpreted in this paper.

Keywords: Electrical Properties, Sensing Element, DC Electrical Resistivity, Electron Hopping.

INTRODUCTION

During recent days, the state-of-art is used to develop smart instrumentation to cater needs of present day and future requirements of various sectors, such as industrial parameters measurement and controlling, biomedical, agriculture, food processing domestic appliances, R & D instrumentation, textile industries etc., Modern instrumentation exhibits the deployment of ubiquitous and most pervasive technologies to enhance the reliability and preciseness of results. To achieve these goals, instead of traditional sensor system, the use of intelligent sensor system is recommended for dedicated applications. The traditional sensor systems are based on the technologies of the early days. Therefore, the designers have to put more effort into the development of instrumentation. Deployment of the ferrites for sensor

based applications is a novel field for the researchers (Patil & Ladgaonkar, 2013; Patil et al., 2017). Electrical properties of the polycrystalline spinel ferrites reveal the suitability of materials' for sensor based applications. The possibility of preparing ferrites in the form of nanoparticles has opened a new and exciting research field, with revolutionary applications in the electronic technology. Polycrystalline ferrites exhibit interesting electrical properties, wherein the semiconducting behavior is realized. Measurement of DC conductivity is an important technique to explore the transport process in ferrites. The electrical properties, such as DC resistivity, dielectric polarization, mobility etc., are the intrinsic properties and these properties are found to be dependent on the chemical compositions, preparation conditions, substitution of divalent or trivalent cations in the

parent lattice, temperature, environmental conditions etc. These ferrite materials are highly resistive and its resistivity is mostly sensitive to the microstructure of the compositions. The microstructure usually develops in the sintering stage. Therefore, sintering conditions play a significant role in the electrical properties of ferrites. Thus, by controlling preparation condition, the electrical properties can be optimized. It is also reported that, the electrical properties are sensitive to the distribution of cations among tetrahedral (A) and octahedral (B) sites of the spinel structure (Patil & Ladgaonkar, 2013). Development of smart sensor module is the major objective of present research work. For this purpose, it is proposed to design own sensors of required characteristics. To develop the sensor, the sensing materials play a vital role. Therefore, the polycrystalline spinel nano ferrites can be synthesized, and used to develop sensors.

1. Review of Literature and Mechanism of Temperature Dependent Electrical Conduction

The exponential dependence of electrical conductivity of ferrites with temperature can be attributed to the semiconducting nature. However, the semiconducting nature of spinel ferrites is quite different than that of semiconductors. Because, in semiconductor, the conductivity is because of thermally generated charge carriers. However, in case of the ferrites, the electrical conductivity is due to thermally activated charge carriers in the ionic lattice, which can be explained on the basis of hopping model (Patil et al., 2017; Shinde et al., 2008). The activation energy for electrical conduction is considerably reduced if the crystal lattice intrinsically contains cations of one element in more than one valence state. According to the previous study, the conduction in the ferrite composition is due to electron hopping between Fe^{3+} and Fe^{2+} ions localized in the crystallographic octahedral (B) site (Fu & Hu, 2010). During this electron exchange, the valence states of two cations are exchanged. However, this conductivity is very low, which can be attributed to the lower mobility of the charge carriers. Cations are not free to leave the lattice sites. However, due to lattice vibrations, these cations come close enough to transfer the electrons as $Fe^{2+} \Leftrightarrow Fe^{3+} + e^-$. The number of electrons contributing

the conduction process depends upon the concentration of the Fe^{2+} ion in the octahedral site. Therefore, in ferrous ferrite the concentration of Fe^{2+} ions is same as that of Fe^{3+} ions in the octahedral site. Therefore, it favours the conduction process and results into low resistivity. Moreover, on substitution, the nickel ion is contributing electron hopping mechanism as $Ni^{2+} Fe^{3+} \Leftrightarrow Ni^{3+} + Fe^{2+}$ (El-Shabasy, 1997). However, this is featured with low mobility. Because of the thermal energy, the mobility of charge carriers increases significantly. Hence, electrical conduction significantly depends on temperature. In addition to electron hopping model, other models are also suggested by the investigators (Abbas & Chaudhry, 2002; Chavan et al., 2010). Polaron, the electron with strain field, hopping mechanism has been suggested (Abbas & Chaudhry, 2002; Chavan et al., 2010) to explain the conduction phenomenon, due to thermally activated mobility (Gopalan et al., 2009a; Murthy & Sobhanadri, 1976). Thus, the electrical conductivity of the ferrite compositions strongly depends on the temperature.

The temperature dependence of electrical resistivity obeys Wilson's relations (Shinde et al., 2010),

$$\rho = \rho_0 \exp(\Delta E/KT) \quad (1)$$

where, ΔE is the activation energy and K is the Boltzman constant. An expression 1 depicts the fact that, the ferrite compositions reveal semiconducting nature with a negative temperature coefficient of resistance. The graph of $\log \rho$ against $1/T$ is the straight line with discontinuities in the slope at certain temperatures, which can be attributed to the Curie points. Thus, the electrical conductivity is strongly influenced by the magnetic ordering (Bhise et al., 1996).

The temperature dependence of DC resistivity of the Mg-Zn-Cu ferrites was studied (Bachhav et al., 2013) and reported three significant regions in the graph of $\log \rho$ against $1/T$. The nature of graph is attributed to the effect of magnetic interaction on electrical conduction. Temperature dependent electrical resistivity of CuNi nanoferrites, synthesized by using citrate gel auto combustion technique, which was investigated by Kumar et al. (2014) and reported that, electrical conduction is due to electron hopping for lower temperature. Moreover, the

behaviour of resistivity for elevated temperature is attributed to the polaron hopping conduction mechanism. They reported that the activation energy in the paramagnetic region is more than that of at ferrimagnetic region (Vasambekar et al., 1999). The variations in the $\log \rho$ against $1/T$ curve depict the semiconducting nature with two regions referred to as ferri region and para region. They reported that, the substitution of Mg causes a decrease in the Curie temperature, which could be attributed to a decrease in the magnetic interaction. The substitution of rare earth element in spinel ferrites caused increased resistivity (Gopalan et al., 2009b; 2009c; Rama et al., 2012). This may be due to stable valence of the rare earth ions.

Thus, it is found that, to decide the applicability of the nanoferrites, the study of DC electrical properties and its temperature dependence is essential (Chaudhari & Ghatage, 2013). Therefore, electrical properties of the $Mg_xZn_{1-x}Fe_2O_4$ ferrites under investigation are studied and results are reported in this paper.

2. Experimental

Present investigation emphasizes the synthesis of polycrystalline ferrite materials suitable for typical sensor based applications using a co-precipitation method. The materials can be characterized by X-ray diffraction and FTIR to explore the structural details of the compositions. X-ray diffractograms of $Mg_xZn_{1-x}Fe_2O_4$ ferrite compositions $x = 0.20, 0.40, 0.60,$ and 0.80 are depicted in Figure 1. From X-ray diffractogram information, the structural details of composition under investigation are calculated (Patil et al., 2016, 2018). The particle size, for all compositions under investigation is estimated for employing Scherrer Equation obtained and it is within the range of 40 nm to 48 nm. The FTIR spectra of the compositions $Mg_xZn_{1-x}Fe_2O_4$, ferrites were obtained in the range from 400 cm^{-1} to 4000 cm^{-1} . The significant absorption bands reveals the confirmation of structural details and suitability of ferrite material for sensor base applications (Ladgaonkar et al., 2013; Patil et al., 2018). Figure 2 shows the IR absorption spectra of the composition $x=0.60$.

The measurement of electrical resistance (R) of the pelletized composition of the polycrystalline, $Mg_xZn_{1-x}Fe_2O_4$

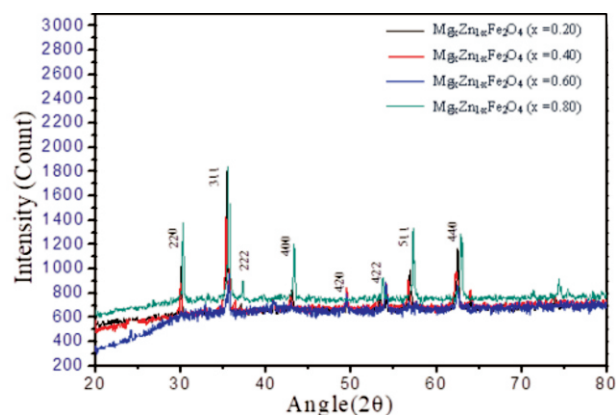


Figure 1. X-ray Diffract Grams of $Mg_xZn_{1-x}Fe_2O_4$ ($x=0.20, 0.40, 0.60,$ and 0.80)

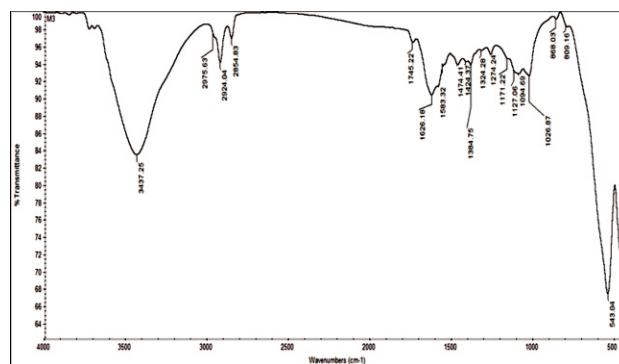


Figure 2. IR Absorption Spectra of the Composition $Mg_xZn_{1-x}Fe_2O_4$ ($x=0.60$)

($x = 0.20, 0.40, 0.60,$ and 0.80) spinel ferrites were carried out at room temperature up to 600 K using two probe method. An experimental arrangement is shown in Figure 3. The measurements are carried out using samples in pellet (disk) form. The electrode designed in the laboratory is depicted in Figure 3 and mentions the schematic of electrode. A silver paste is used to achieve Ohmic contacts. Highly precise digital meter, Tektronix DMM4050, having range $10\ \Omega$ to $1.2\ G\Omega$, with $10\ \mu\Omega$ resolution is used for resistance measurement. Automatic controlled electric furnace is used and temperature of the furnace is measured by Cromel-Alumel thermocouple.

3. Results and Discussion

3.1 Investigation of DC Electrical Resistivity of Pelletized Compositions

The DC electrical resistance 'R', of the pelletized compositions under investigation, is measured in the range from 300 K to 675 K. To explore the details regarding

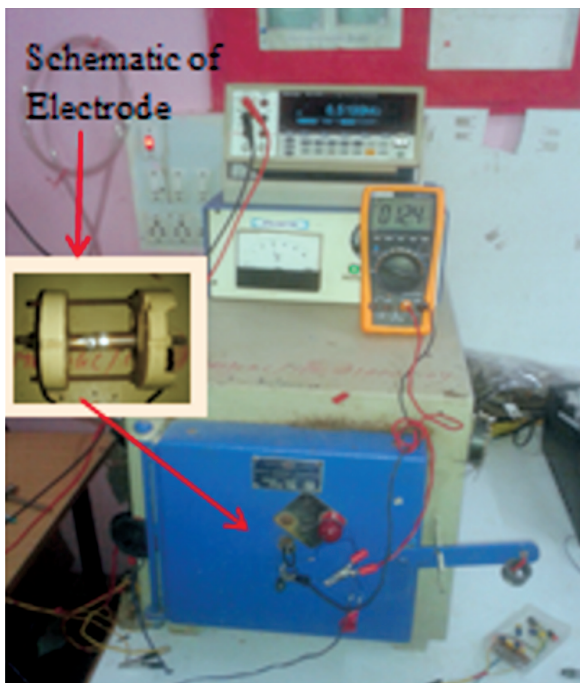
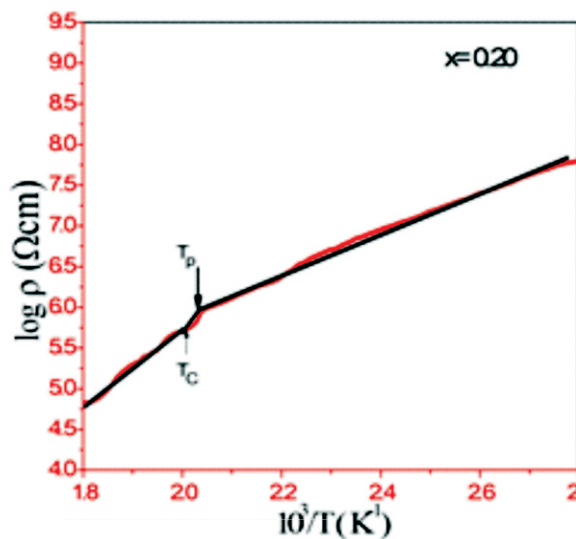


Figure 3. Experimental Setup to Measure Temperature Dependent Electrical Resistance

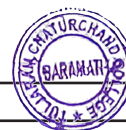
conduction mechanism, the values of resistivity ' ρ ', in Wcm, are obtained from resistance data and values of $\log \rho$ are plotted against inverse temperature ($1000/T$) and presented in Figure 4 (a-d) for the compositions of polycrystalline, $Mg_xZn_{1-x}Fe_2O_4$, spinel nanoferrites. On inspection of these figures, it is found that, the graphs of $\log \rho$ against $1/T$ are almost linear obeying Wilson relation. The figures reveal the semiconducting nature by exhibiting decrease of resistivity due to an increase in the temperature. As depicted in the Figure 4, the compositions of $Mg_xZn_{1-x}Fe_2O_4$ ferrites exhibit decrease in resistivity with increase in the temperature. The graphs of $\log \rho$ against inverse temperature ($1000/T$) are almost linear with two significant breaks at distinct temperatures. This decrease in resistivity with increase in the temperature can be attributed to the conduction mechanism, wherein conductivity is ensured due to thermally activated mobility of the charge carriers. It is known that, the zinc ions prefer to reside at one site, whereas magnesium ions preferentially distribute among A and B site. Moreover, valency of Mg ion remains stable. Therefore, the conduction is due to hopping of electrons between Fe^{3+} and Fe^{2+} ions at octahedral site (Gopalan et al., 2009). On inspection of

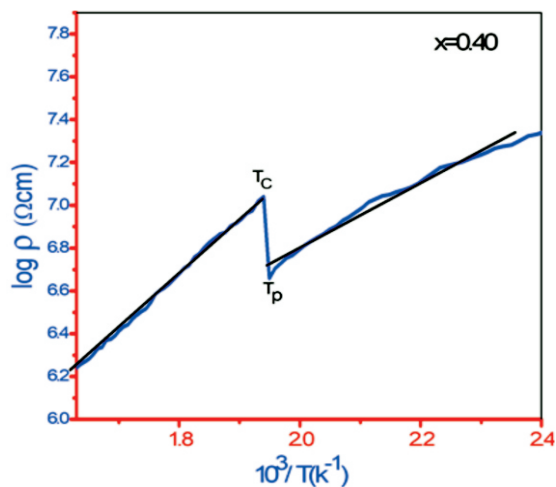
Figure 4 (a-d), it is found that, the linear relationship of $\log \rho$ against $1/T$ graph shows two significant breaks T_p and T_c for the compositions for $x = 0.20, 0.40,$ and 0.60 . Moreover, the composition for $x = 0.80$ depicts one break at a temperature T_c . Such two breaks are also reported (Bachhav et al., 2011; Varalaxmi & Sivakumar, 2010). These two temperatures, T_p and T_c , are called magnetic phase transition temperature and Curie temperature, respectively. The magnetic interaction is sensitive to the thermal energy and hence at a typical temperature, the disordering of interaction begins. Due to this disorder, an amount of energy required for activation of the hopping is increased and hence the graph depicts the change in slope. Moreover, at Curie temperature, the magnetic interaction vanishes. Therefore, the compositions become paramagnetic in which more amount of disorder is realized. This results into increase in the slope of the graph of $\log \rho$ against inverse of temperature (T). This reveals the fact that, electrical conductivity is strongly influenced by magnetic interaction. Therefore, the region of the graph before Curie temperature is called ferri region and that of after Curie temperature is called Para region. Thus, the graphs of $\log \rho$ against $1000/T$, Figure 4 (a-d) clearly depicts the ferri and para magnetic nature of the compositions.

The Curie temperature values are obtained from temperature dependent electrical resistivity data and are presented in Table 1. On inspection of this table, it is found

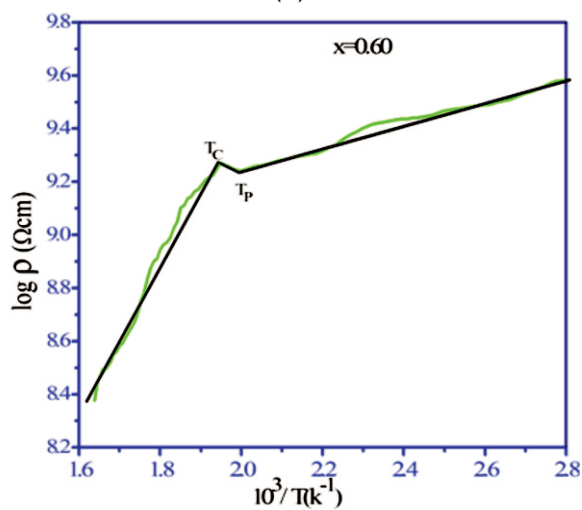


(a)

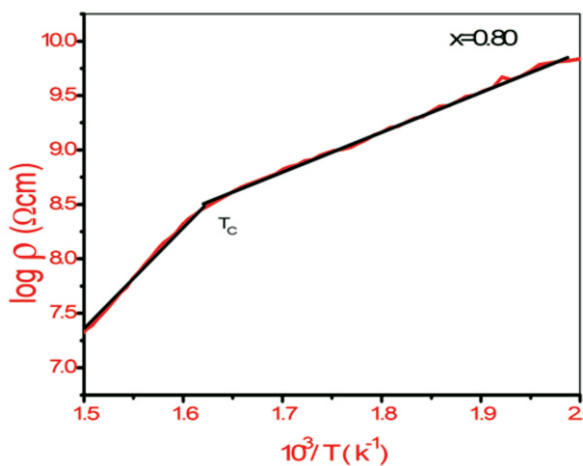




(b)



(c)



(d)

Figure 4. Graph of Log ρ Against $(1000/T)$ for $Mg_xZn_{1-x}Fe_2O_4$ (a) ($x = 0.20$); (b) ($x = 0.40$); (c) ($x = 0.60$); (d) ($x = 0.80$)

that, the Curie temperature values reveal compositional dependence. Pure zinc ferrite is paramagnetic in nature

X	Activation Energy DE (eV)		Curie Temperature (Tc) K
	Ferri Region	Para Region	
0.20	0.084	0.095	499
0.40	0.024	0.102	515
0.60	0.010	0.135	602
0.80	0.135	0.377	626

Table 1. The Curie Temperature and Activation Energy Values for the Composition $Mg_xZn_{1-x}Fe_2O_4$

(Totagi et al., 2015). Moreover, the occupancy of magnesium ion in B site displaces proportional amount of iron ion from B site to A site, which causes to increase in the magnetic interactions (Choudary et al., 2014). Therefore, increase in concentration of magnesium ion (x), decreases the concentration of Zinc ion. Therefore, magnetic interaction becomes more favorable. Thus, increase in the curie temperature, due to increase in magnesium ion, can be attributed to the increase in magnetic interactions (Ladgaonkar et al., 2000). Activation energies for both ferri region as well as para region are estimated from the slope of $\log \rho$ against inverse temperature, which are shown graphs and presented in Table 1. On inspection of the Table 1, the activation energies observed in present investigation were found to be close match with that of earlier reports (Li et al., 2007). From, this Table, it is also found that, the activation energy for para region is more than that of ferri region. This supports the statement that electrical properties are strongly influenced by magnetic ordering.

Conclusion

The $MgZn$ polycrystalline ferrite materials have been successfully synthesized by chemical routh and confirm the nature of material using X-ray diffraction and FTIR. The diffracto grams show the formation of single phase compositions. Using Debye-Scherer relation, the particles size of compositions is estimated and it is in the range from 40 nm to 48 nm. Temperature dependent electrical properties of the composition of $Mg_xZn_{1-x}Fe_2O_4$ nano ferrite, were investigated for suitability of these materials as a sensing element for the designing of sensor. The DC electrical resistivity of the pelletized compositions shows semiconducting behavior, which is attributed to the electron hopping mechanism. This electrical conductivity exhibits the influence of magnetic ordering at Curie temperature. The Curie temperature values depict the

compositional dependence. Keeping pace with the objectives of present research, the compositions of the ferrite under investigation, it is attempted to design the sensors on different substrates, Such as epoxy resin, glass and ceramic as sensing elements, employing thick film technology. The resistance of sensing element shows a negative temperature coefficient within the range of investigation.

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