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Electrical resistivity and dielectric studies of Pb^{4+} substituted nickel ferrites prepared by double sintering method

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ABSTRACT

The variation of a.c. resistivity (ρ_{ac}), dielectric constant (ε') and dielectric loss factor (ε'') of mixed Ni_{1+x} $Pb_xFe_{2-2x}O_4$ (where x = 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5) ferrites system has been studied as a function of temperature and frequency. The variation of a.c. resistivity (ρ_{ac}) with temperature shows unusual metal-semiconductor type transition for compositions x≤02 whereas for the compositions x≥0.3 shows usual behavior. The dielectric constant (ε') and dielectric loss (ε'') with temperature increases very slowly in the beginning till the temperature reaches to 600^0K and then increases very rapidly. The dielectric constant (ε') and dielectric loss (ε'') shows decrease with increase in log frequency shows dispersion in the low frequency range for all the compositions. The dielectric loss tangent (tan∂**)** appreciably increases with temperature and decreases with log frequenc**y.** These variations have been explained on the basis of Koop's phenomenological theory, concentration of Fe^{2+} and Fe^{3+} ions on octahedral sites and hopping frequency of electrons between Fe^{2+} and Fe^{3+} ions.

Keywords: Ferrites; Electrical properties; Dielectric properties

INTRODUCTION

Ferrites play a very important role in the electrical and electronic industries because of their interesting electrical and dielectric properties. These properties of ferrites depend upon chemical composition, methods of preparation, sintering temperature, cations distribution, porosity, grain size, etc.

Substituted nickel ferrites have been the subject of interest for large number of workers because of their large number of applications in microwave frequencies. The nickel ferrite ($NiFe_2O_4$ or NiO Fe_2O_3) is an inverse spinel structure to be a collinear ferrimagnet [1]. The addition of tetravalent ions like Ti^{4+},Ge^{4+},Si^{4+},Mn^{4+} and Pb^{4+} influence the structural, magnetic and transport properties of the ferrites system [2-8]. According to best of our knowledge, no information regarding Pb^{4+} substituted nickel ferrite on electrical and dielectric properties exist in the literature so far. Therefore, an attempt has been made to study the electrical and dielectric properties of Ni_{1+x} $Pb_xFe_{2-2x}O_4$ ferrite system.

METHODOLOGY

The six samples of $Ni_{1+x}Pb_xFe_{2-2x}O_4$ with x = 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5 were prepared by standard solid state reaction technique. The details regarding sample preparation, structural and IR studies have been given in our earlier paper [8].

The a. c. electrical resistivity (ρ_{ac}) and other parameters such as dielectric constant (ε′), dielectric loss (ε′′) and loss tangent (tan∂) were studied as a function of temperature at fixed frequency 1 KHz and frequency (100Hz-1MHz) at room temperature using LCR-Q Meter Bridge. For good electrical contact the pellets were polished and silver paste was applied on end surfaces. Resistivity measurements were carried out from room temperature to well above the Curie temperature. During the measurement sufficient time was allowed for sample to attained equilibrium temperature. The activation energy was calculated from the plot of logρ_{ac} versus reciprocal of temperature using the relation

$$\rho=\rho_0 \exp^{\Delta E/KT} \qquad [1]$$

where ρ is the resistivity at temperature T, ρ_0 is the temperature independent constant, k is the Boltzman's constant and ΔE is the activation energy.

Assuming the capacitance of the parallel plate arrangement the dielectric constant Є′ was calculated by using the relation

$$\varepsilon'=C.t/\epsilon_0.A \qquad [2]$$

Where C is the capacitance of the sample, t is thickness of the pellet, A is the area of cross-section of pellet and ϵ_0 is the permittivity of free space. The value of loss tangent (tan∂) was calculated by using the relation

$$\tan\partial = \varepsilon''/\varepsilon' \qquad [3]$$

$\tan\partial = 1/Q$ where Q is quality factor

RESULTS AND DISCUSSIONS

A.C. Electrical resistivity

The electrical resistivities as a function of temperature for Ni1+xPbxFe2-2xO4 ferrites in the form of Arrhenius plot are shown in the fig.1. and 2. From the fig.1 it can be seen that, the resistivity initially increases with increasing temperature within the temperature range 300-500^{0}K; thereafter it decreases with increase in the temperature. It is not possible to identify the slope changes at Curie temperature because of their anomalous thermal characteristics. This anomalous electrical behavior of resistivity with temperature that is increase in resistivity with increase in temperature is observed for Sn^{4+} and Zn^{2+} substituted nickel ferrites and has been explained on the basis of electron hopping and current due to electron in the conduction band [9]. Such a metal-insulator characteristics is reported in [10]. From the fig.2 it can be seen that the electrical resistivity decreases with increase in the temperature. It is possible to identify the slope changes at Curie temperature. This type of conventional behavior is observed for number of ferrites. The a. c. resistivity for different samples with x = 0.0 to 0.5 at room temperature are given in the table 1.

From the table 1 it is seen that the resistivity increases with addition of Pb^{4+} content it is because of the resistivity of Pb^{4+} larger (2.08×10^5 ohm-cm) as compared to Fe (0.98×10^5 ohm-cm) and Ni (0.69×10^5 ohm-cm) [ref introduction to ssp c kittel p149). The resistivity value for all the compositions lies in between 1.66×10^5 to $9.33 \times 10^7 \Omega$-cm.

The activation energy values for conduction are computed from the Arrhenius plots of $\log\rho_{ac}$ versus $10^3/T$ and are given in the table 1. It should be noted that activation energy increases with addition of Pb^{4+} concentration which can be effectively interfere with electron hopping process between Fe^{2+} and Fe^{3+} ions in octahedral sites and increase the activation energy for conduction [11]. The observed value of activation energy varies from 0.16 to 0.51ev and that high activation energy is accompanied by high resistivity as was shown by Smith and Wijn.

Fig.3 shows the variation of a.c. resistivity with log frequency at room temperature. From the figure 3 it can be seen that all the samples shows a decrease in a.c. resistivity with increase in the log frequency from 100 Hz to 1 MHz, which is normal behavior of ferrites. The conduction in the ferrites is through exchange of electrons between Fe^{2+} and Fe^{3+} on octahedral sites. The increase in the frequency of applied field enhances the hopping of charge carriers resulting increase of conductivity and decrease of resistivity. At higher frequencies a.c. resistivity decreases and remains constant because of the fact that hopping frequency can be no longer follow the frequency of the applied field leading to decrease the value of a.c. resistivity.

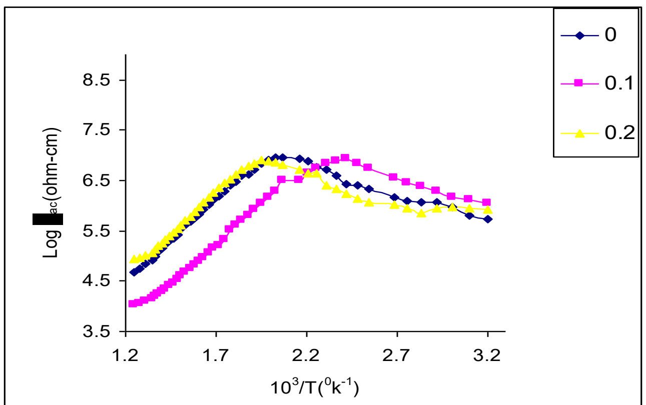

Fig.1 Plot of log ρ_{ac} versus $10^3/T$ ($^0K^{-1}$) for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

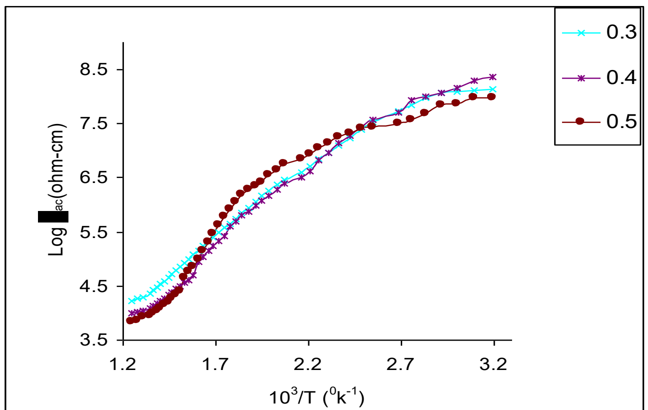

Fig.2 Plot of log ρ_{ac} versus $10^3/T$ ($^0K^{-1}$) for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

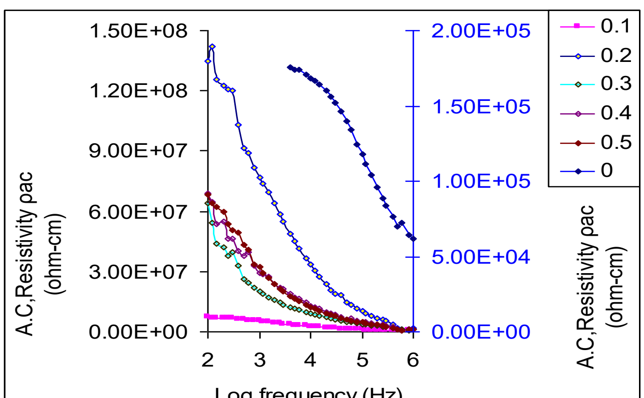

Fig.3.Plot of a.c. resistivity (ρ_{ac}) with log frequency for Ni_{1+x} $Pb_xFe_{2-2x}O_4$ ferrites

Table 1: Variation of Curie temperature, activation energy and a.c. resistivity for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

Composition (x)	Curie temperature (Susceptibility) Tc(^{0}K)	Activation energy		ΔE= (E_2-E_1) eV	Resistivity (ρ_{ac}) (Ohm.cm)
		Ferimagnetic region (E_1) eV	Paramagnetic region (E_2) eV		
0.0	-----	0.48	0.31	0.16	$1.69x10^5$
0.1	-----	0.58	0.31	0.26	$2.53x10^6$
0.2	850	0.63	0.32	0.31	$3.34x10^7$
0.3	830	0.52	0.004	0.51	$9.23x10^7$
0.4	820	0.30	0.006	0.29	$4.34x10^7$
0.5	790	0.96	0.49	0.47	$6.65x10^7$

Table 2: Variation of room temperature dielectric constant, dielectric loss, loss tangent and a.c. conductivity for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites at 10 KHz

Ferrite composition (x)	Dielectric constant (ε′)	Dielectric loss (ε″)	Dielectric loss tangent(Tan∂)	Conductivity (σ_{ac})(Ω^{-1}-cm^{-1})
0.0	212	1066	5.02	$5.92x10^{-6}$
0.1	100	71	0.71	$3.95x10^{-7}$
0.2	47	5	0.11	$2.99x10^{-8}$
0.3	58	19	0.33	$1.08x10^{-8}$
0.4	61	13	0.22	$2.30x10^{-8}$
0.5	62	14	0.23	$1.50x10^{-8}$

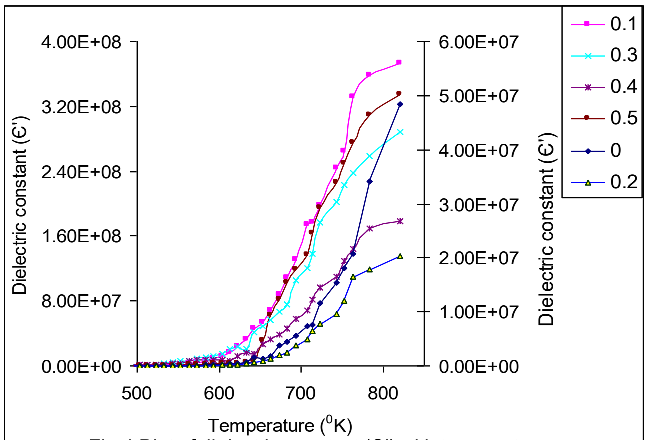

Fig.4 Plot of dielectric constant (Є') with temperature for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

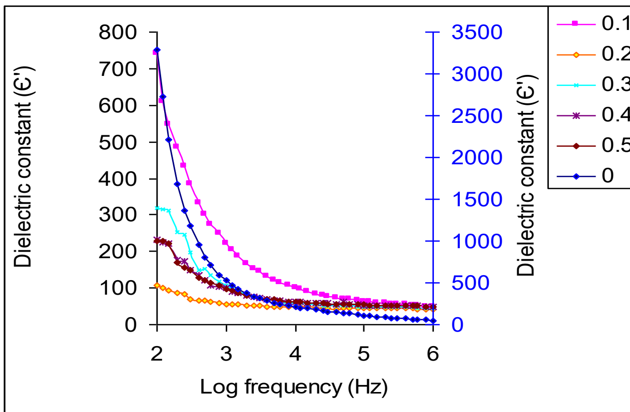

Fig.5. Plot of dielectric constant (Є') with log frequency for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

Dielectric constant

The variation of dielectric constant ε′ as a function of temperature at fixed frequency 1 KHz is shown in the fig.4. From the figure it is appear that the dielectric constant remains constant or increases very slowly in the beginning till temperature reaches to 600^{0}K and then increases very rapidly. Since the resistivity of ferrites increases with decrease in the temperature, an increase in the dielectric constant at high temperature is expected because the resistivity and dielectric constant are inversely related. Such behavior has been reported by S. S. Shinde et.al [12] in case of Co-Si ferrite system, Shaikh et. al [13] in case of Li-Mg-Zn ferrite and Bellad et al [14] in the case Li-Cd ferrite.

The variation of dielectric constant ε′as a function of log frequency for all the samples at room temperature is shown in the fig.5. From the figure it is appear that the dielectric constant for all the samples decreases rapidly with increase in the frequency in the beginning and then decreases very slowly or nearly remains constant for higher values of log frequency. It is also conclude that at constant frequency of 10KHz, dielectric constant decreases with increasing Pb^{4+} content up to x = 0.2 and thereafter it increases. This type of behavior is observed for number of ferrites. The dielectric behavior of our samples may be explained on the basis of that the mechanism of the polarization process in ferrites similar to that of conduction process. The electronic exchange between Fe^{2+}-Fe^{3+} gives the local displacement of electrons in the direction of applied electric field, which includes the polarization in ferrites. In normal dielectric behavior, the dielectric constant decreases with increasing frequency reaching a constant values depending upon the fact that beyond fixed frequency of electric field the electrons exchange does not follow the alternating electric field.

Dielectric loss

The variation of dielectric loss ε′′as a function of temperature for all the compositions at frequency 1 KHz is shown in the fig.6. It is observed from the figure that the dielectric loss almost remains constant or varies very slowly up to temperature 625^{0}K (approximately) and then increases very rapidly in the same way as the dielectric constant increases. This behavior of dielectric loss is in full agreement with that obtain by V. A .Ioffe et al [15] for spinel ferrites.

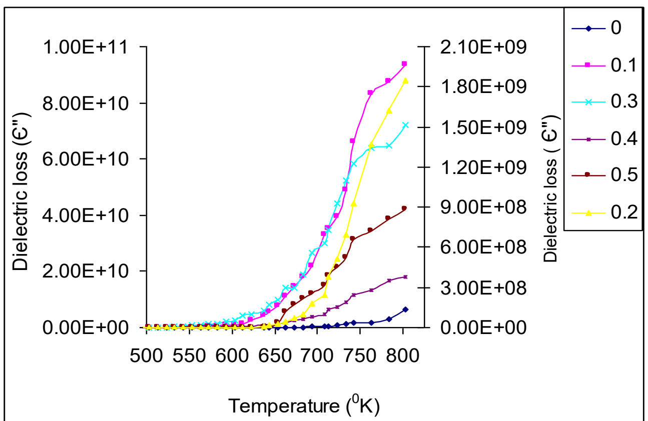

Fig.6 Plot of dielectric loss (Є") with temperature for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

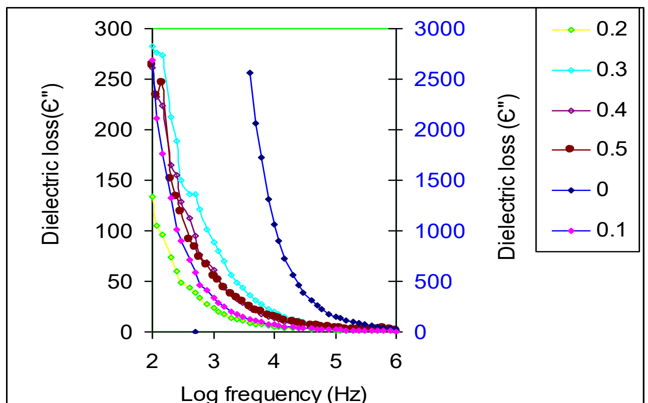

Fig.7. Plot of dielectric loss(Є") with log frequency for $Ni_{1+x}Pb_xFe2\text{-}2xO_4$ ferrites

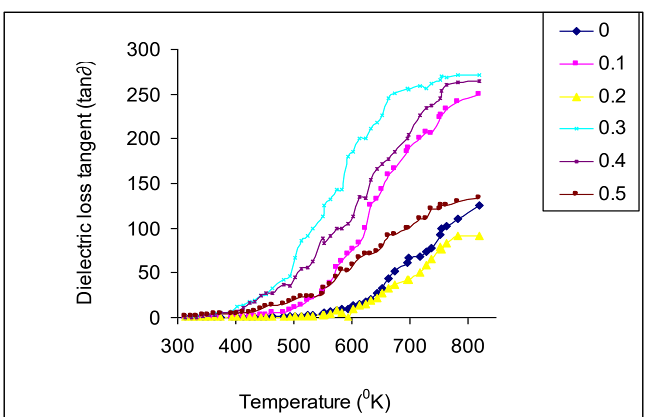

Fig.8 Plot of dielectric loss tangent (tan∂) with temperature for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ feriites

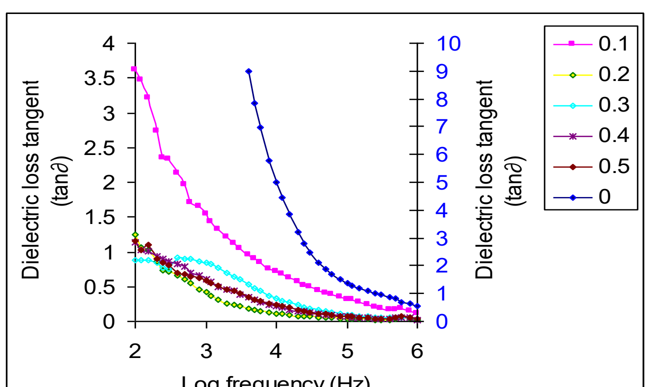

Fig.9. Plot of dielectric loss tangent (tan∂) with log frequency for $Ni_{1+x}Pb_xFe_{2-2x}O_4$ ferrites

The variation of dielectric loss ε′′as a function of log frequency for all the samples at room temperature is shown in the fig.7. The variation in dielectric loss ε′′shows a similar dispersion behavior as seen for ε′′ for log frequency. The decrement for dielectric loss ε′′ curve is very fast at low frequency while it decays with slow rate reaching a constant value at higher frequencies. This dielectric loss ε′′curve are attributed to domain wall resonance [16].

3.4. Dielectric loss tangent

The thermal variation of loss tangent (tan∂) for all the compositions at fixed frequency 1 KHz is shown in the fig.8. It can be seen from the figure that the loss tangent (tan∂) increases very slowly in the beginning and then increases rapidly with temperature. This behavior is in good agreement with general rule that is; loss tangent (tan∂) appreciably increases when the temperature is rise [17].

The variation of loss tangent (tan∂) with log frequency for all the samples at room temperature is shown in the fig.9. From the figure it is cleared that loss tangent (tan∂) decreases rapidly with log frequency in the beginning then decreases very slowly or remain nearly constant for higher values of frequencies. When hopping frequency become equal or nearly equal to that of externally applied electric field a maximum dielectric loss can be obtained [18]. Samples in the present series did not exhibits the peak in the plot of log frequency versus loss tangent (tan∂) indicating that the frequency at which maximum dielectric loss occurs is beyond high frequency.

CONCLUSION

1 The variation of ac. Resistivity (ρ_{ac}) with temperature shows unusual metal-semiconductor type transition for compositions x≤02 whereas for the compositions x≥0.3 shows usual behavior.

2 The dielectric constant (ε′), dielectric loss (ε′′) and loss tangent (tan∂) increases slowly in the beginning and then rapidly with temperature showing higher values for the composition x=0.0

3 The dielectric constant (ε′), dielectric loss (ε′′) and dielectric loss tangent (tan∂) decreases as frequency increases.

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