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RESEARCH ARTICLE

EFFECT OF EUROPIUM CONCENTRATION ON THE PHYSICAL PROPERTIES OF NI-ZN FERRITE SUBSTITUTED BY MN²⁺ ION.

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Abstract

The effect of the substitution by europium ions on the magnetic and electrical properties was studied for the ferrite samples $Mn_{0.5}Ni_{0.1}Zn_{0.4}Eu_xFe_{2-x}O_4$, ($0 \leq x \leq 0.15$). The samples were prepared using conventional ceramic method. Cubic spinel structure of the prepared samples was examined using XRD. The porosity, crystallite size and the lattice parameter of the prepared were also calculated. The obtained results show an improvement in the porosity of the prepared samples due to Eu^{3+} substitution. The temperature dependence of the initial magnetic permeability was also studied. The experimental results indicate that the substitution by Eu^{3+} improves the magnetic properties by increasing the Curie temperature of the substituted samples. Temperature and composition dependence of the dc resistivity were also studied. The obtained results show the sample of $x=0.025$ has high physical properties such as high resistivity, high magnetic permeability, highest Curie temperature and low porosity so it is useful in the technological applications.

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Introduction:-

Ni-Zn ferrites are attractive for microwave device applications owing to their large magnetic permeability in radio frequency region, low dielectric loss, high Curie temperature, chemical stability at relatively low frequencies and low cost. Rare earth ferrites have important applications in modern telecommunication. For these reasons, scientists are keenly interested in new methods for preparation of rare earth ferrites and for determining their characterization. Hence, they are important commercially. Evaluation of magnetic and electrical properties of ferrites and correlation of results may help in tailoring composites for various applications.

The study of resistivity throw light on the behaviour of the charge carriers under the influence of the ac field, their mobility and the mechanism of conduction in ferrite materials. The influence of small rare earth addition on the crystal structures and the physical properties of Ni-Zn ferrite were studied by many authors [1-4]. Also, it was found that for improving the magnetic and electrical properties of Ni-Zn ferrite, rare earth ions with a large ionic radius and with a stable valance state are the best substitution.

Rezescu et al. [2, 5] shows that the integration of rare earth cations into the spinel structure of Ni-Zn ferrite results in modification of basic electrical and magnetic properties. Their investigation showed a lowering of the Curie temperature and saturation magnetization due to the substitution of Fe^{3+} ion with lanthanides, formation of a plateau in the temperature dependent initial permeability curve and an increase in the specific DC resistivity. Additionally, microstructural parameters like grain size and porosity were also affected by the rare earth ions addition. The change in the magnetic properties as compared with the samples without rare earth ions has been discussed in terms of microstructural parameters.

The present work dealing with the effect of substitution by Eu^{3+} ion on the electrical and magnetic properties of Ni-Zn ferrite substituted by Mn^{2+} ion.

Experimental techniques:-

Samples with chemical composition $\text{Mn}_{0.5}\text{Ni}_{0.1}\text{Zn}_{0.4}\text{Eu}_x\text{Fe}_{2-x}\text{O}_4$, ($0 \leq x \leq 0.15$) were prepared by using standard ceramic technique. X - ray diffraction was formed using diffractometer of type X'pert graphics and identify with Cu K_α radiation. The porosity of the prepared samples was calculated according to the relation $P\% = 100 [1 - (d/d_x)]$ where d_x is the theoretical x- ray density and d is the apparent density of samples measured in toluene using Archimedes principle. The initial magnetic permeability (μ_i) was measured as a function of temperature at constant frequency and low magnetizing current. The value of μ_i was calculated using Poltinnikov's formula.

$$(V_s = K \mu_i \quad \text{where } K = 0.4 \pi N_p N_s I_p A \omega / l)$$

where V_s is the induced voltage in secondary coil, N_p , N_s are the numbers of turns of primary and secondary coils ($N_p = N_s = 15$), A is the cross-sectional area of the sample, ω is angular frequency and l is the average path length of the magnetic flux. The DC electrical resistivity (ρ) was measured using two-probe method.

Results and discussion:-

Structure analysis:-

X-ray diffraction patterns (XRD) for all the samples with composition $\text{Mn}_{0.5}\text{Ni}_{0.1}\text{Zn}_{0.4}\text{Eu}_x\text{Fe}_{2-x}\text{O}_4$, ($0 \leq x \leq 0.15$) are shown in Fig.(1). The crystalline phases were identified using ICDD powder diffraction card No.8-234 for Ni-Zn ferrite. Figure (1) shows that, the samples $x = 0.0$ and $x = 0.025$ have a cubic spinel structure with no extra lines corresponding to any other phases but for samples ($x \geq 0.075$) there is a secondary phase of EuFeO_3 with very small intensity. All the compositions show the presence of cubic spinel structure as a major phase.

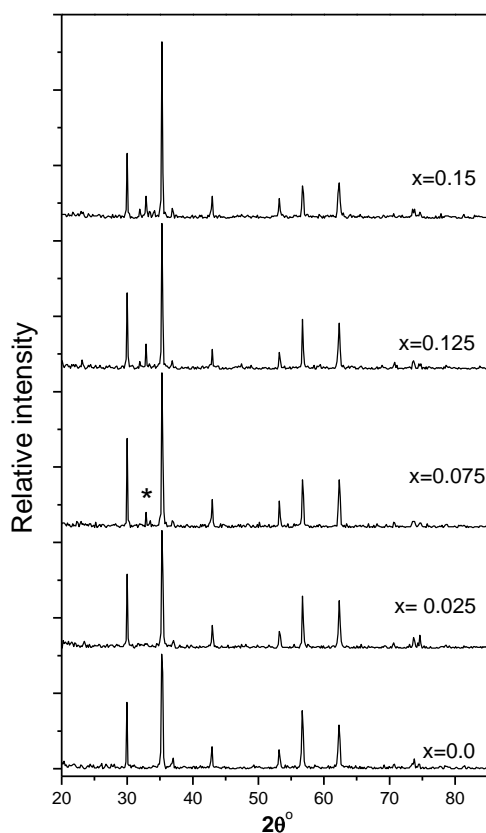


Fig. (1): X-ray diffraction patterns for the prepared system $\text{Mn}_{0.5}\text{Ni}_{0.1}\text{Zn}_{0.4}\text{Eu}_x\text{Fe}_{2-x}\text{O}_4$, ($0 \leq x \leq 0.15$).

The porosity of the prepared samples (P%) was calculated and shown in Fig (2). Fig.(2) shows that the substitution by Eu^{3+} ion improves the porosity of the samples where the porosity decreases as Eu^{3+} concentration decreases. The atomic weight of Fe^{3+} is less than that of Eu^{3+} , therefore the replacements of Eu^{3+} by Fe^{3+} caused a relative increase in apparent density (D) of the sample and consequently decrease in the porosity.

Figure (2) shows the variation the lattice parameters (a) with Eu^{3+} content. The figure shows that, the lattice parameter increase with increasing Eu^{3+} content. This increase is attributed to the large ionic radius of Eu^{3+} (0.95 Å) compared with Fe^{3+} ion (0.64 Å) [6,7].

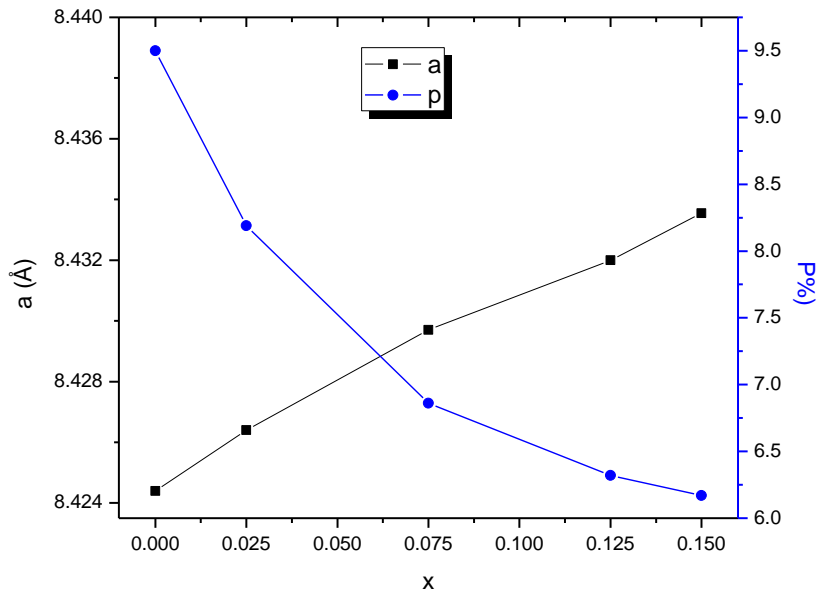


Fig.(2): Compositional dependence of the porosity and lattice parameter of the prepared samples.

The average crystallite size of the samples was calculated using Scherrer`s equation [8].

$$t = \frac{0.9\lambda}{h_{1/2}\cos\theta} \quad (1)$$

where $h_{1/2}$ is the peak width at half maximum and θ corresponding to the peak position. The calculated average crystalline size for all studied samples is shown in Table (1). The obtained results show that, as Eu^{3+} content increases the crystallite size decreases. The variation of lattice parameter will lead to strains which produce internal stress [9,10]. Such stress hinder the growth of grains, and the grains size of the samples decreases with Eu^{3+}

The initial magnetic permeability:-

The initial magnetic permeability (μ_i) for all the samples was studied as a function of temperature and the obtained results are shown in Fig. (3), the figure show that the initial magnetic permeability (μ_i) decreases gradually with increasing temperature and then drop at the Curie temperature T_C . The sharp decrease was observed near Curie temperature for the ferrite samples $x = 0.0$ and $x = 0.025$. This suggests a single phase formation of these ferrites. These results are agreement with the previous results of XRD patterns in which no secondary phase was observed. But for $x \geq 0.025$, a gradual decrease was observed near the Curie temperature, this is also agrees with XRD patterns.

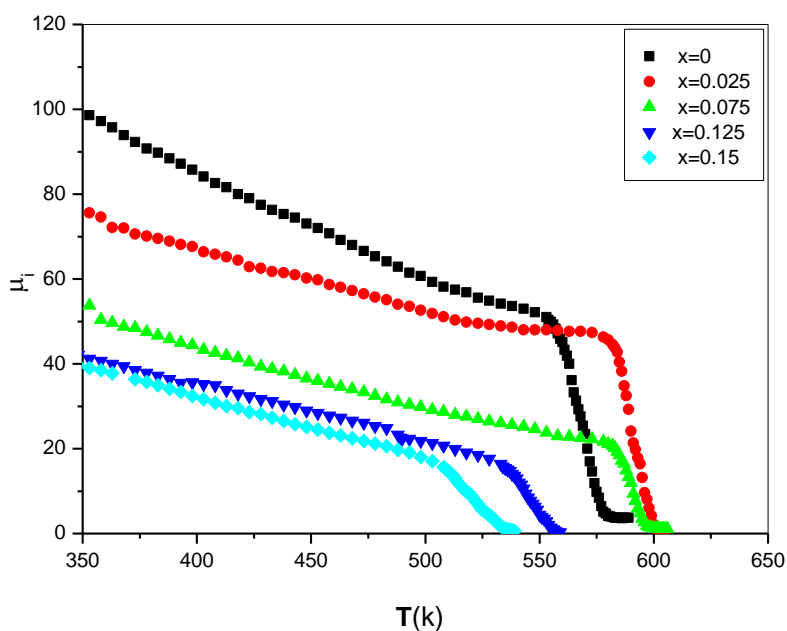


Fig.(3): The variation of initial permeability (μ_i) with temperature (T) of $Mn_{0.5}Ni_{0.1}Zn_{0.4}Eu_xFe_{2-x}O_4$.

The compositional dependence of the initial magnetic permeability (μ_i) at room temperature for all studied samples is shown in Fig. (4). This figure shows that the initial magnetic permeability (μ_i) decreases with increasing Eu^{3+} ions. This is due to the substitution of Eu^{3+} ions instead of Fe^{3+} ions, which has a magnetic moment ($3.63\mu_B$) respectively. This leads to decrease in the magnetization of the samples and then decreasing the initial magnetic permeability (μ_i).

Figure (4) illustrates that as the Eu^{3+} content increases, the Curie point T_C increases to $x = 0.025$ and then shifts towards lower temperature. The Curie temperature decreases as Fe^{3+} content decreases. The decrease of T_C in the samples with Eu^{3+} content is due to the appearance of secondary phase, which affects the magnetic order and helps the material to transfer to the paramagnetic state at lower temperature.

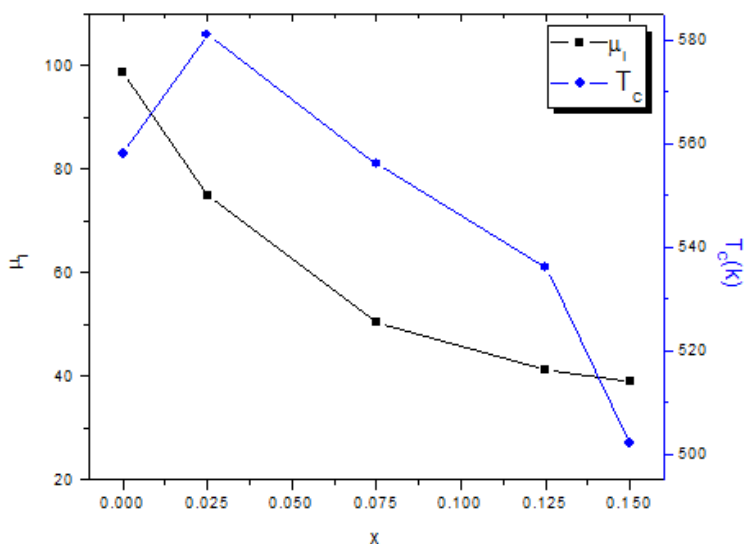


Fig.(4): The variation of the initial permeability (μ_i) and Curie temperature of the prepared samples.

Electrical resistivity

The effect of temperature on the DC electrical resistivity of the ferrite spinel system $\text{Mn}_{0.5}\text{Ni}_{0.1}\text{Zn}_{0.4}\text{Eu}_x\text{Fe}_{2-x}\text{O}_4$, ($0 \leq x \leq 0.15$) was studied. The obtained results, shown in Fig.(5), show that, as temperature increases the resistivity decreases. This decrease of resistivity in ferrite is due to the exponential increase of drift mobility of charge carriers with temperature. The DC resistivity of the studied ferrite was studied in the light of the formula [11].

$$\rho = \rho_0 \exp\left(\frac{E}{KT}\right) \quad (2)$$

where K is the Boltzmann constant and E is the activation energy of the conduction process obtained from the slope of $\text{Log } \rho$ and $1/T$ as shown in Fig. (6), for the sample $x = 0.025$. The temperature variation of resistivity exhibits two breaks and three distinct regions (I, II and III). Such a break was associated with a change in the slope which is attributed to the change of magnetic order and lowering the generation of charge carrier. Basazynski and Murthy et al. [12,13] confirmed this discussion and suggested that the change in the slope can be either linked with magnetic ordering or with a conduction mechanism. The first region is attributed to extrinsic conduction mechanism. This region extended from room temperature up to the first transition temperature (T_1) ≈ 400 K for all studied samples. The second transition temperature (T_C) is always attributed to the magnetic phase transition from ferrimagnetic to paramagnetic state [14]. The activation energies E_{ferri} for region II and E_{para} for region III and the transition temperature T_C between ferrimagnetic and paramagnetic region are given in Table (1). Similar results were previously observed for nickel manganese zinc ferrite [15].

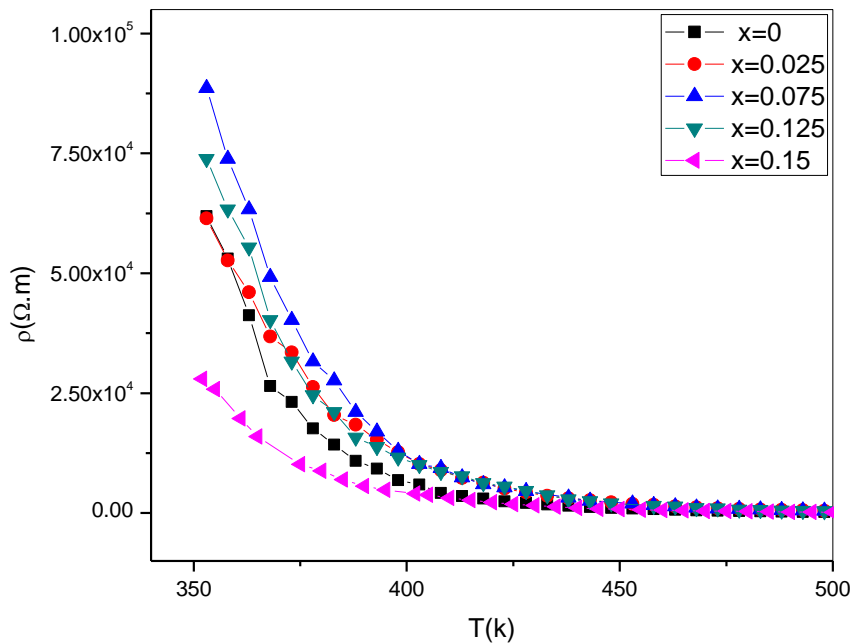


Fig.(5): Effect of temperature on the resistivity of the prepared samples.

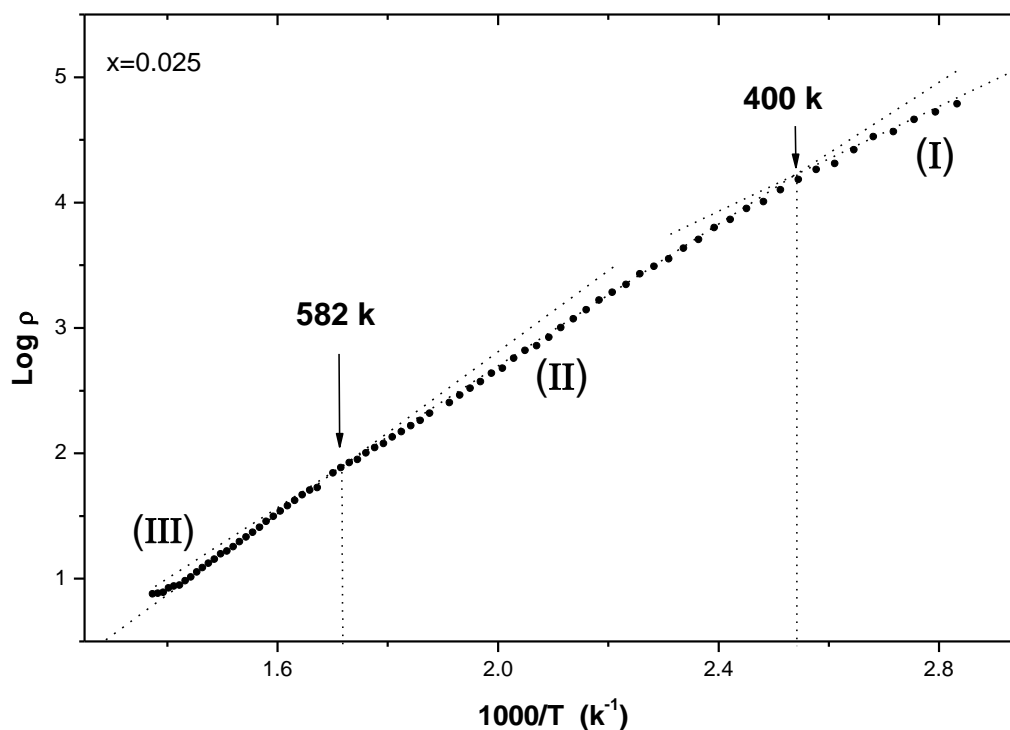


Fig.(6): The relation between $\text{Log } \rho$ and $1/T$ for $x = 0.025$ as an example of the prepared samples.

Table (1) shows the activation energies in region II and III which slightly decreased with change Eu^{3+} content. It is noted that the value of the activation energy increase going from region II to III. The change in activation energy is attributed to the change in conduction mechanism to polaron hopping mechanism [16, 17]. The behavior of our results is similar to that in previous work [15]. Table (1) shows also, the effect of composition on the DC electrical resistivity at room temperature which indicates that the DC resistivity slightly decreases with increasing Eu^{3+} content up to $x = 0.025$. The decrease in the resistivity may be attributed to the increase in the drift mobility of the charge carriers and the valence fluctuation between Eu^{3+} and Eu^{2+} [18]. For $x \geq 0.075$ the resistivity increases due to the appearance of the secondary phase at the grain boundaries which acts as scattering centers of the charge carriers.

Table 1: Show the effect of Eu^{3+} concentration on the activation energies, crystallite size and the electrical resistivity.

x	E_{para} (ev)	E_{ferri} (ev)	t(nm)	$\rho(\Omega.m)$
0	0.76	0.54	154	6.2×10^4
0.025	0.63	0.58	148	6.14×10^4
0.075	0.67	0.60	138	8.9×10^4
0.125	0.7222	0.5772	135	7.38×10^4
0.15	0.6325	0.5405	133.5	2.79×10^4

Conclusions:-

1. The lattice parameter of the prepared samples increases as the Eu^{3+} content increases due to its large ionic radius
2. The obtained results show an improvement in the porosity of the prepared samples due to Eu^{3+} substitution.
3. The substitution by Eu^{3+} ions improves the magnetic properties by increasing Curie temperature.
4. The obtained results show the sample of $x = 0.025$ has high physical properties such as high resistivity, high magnetic permeability, highest Curie temperature and low porosity so it is useful in the technological applications.

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