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Study of the Variation of Resistivity, Permeability and Curie Temperature of Rare Earth Metal lanthanum (la) Substitution on $Ni_{0.60}Zn_{0.40-x}La_x Fe_2O_4$(x=0.05, 0.10, 0.15) Ferrites.

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Abstract

In the present work, ferrites with compositions of $Ni_{0.60}Zn_{0.40-x}Re_xFe2O4$ where x=0.05, 0.10, 0.15 were prepared by conventional Solid State Reaction Method. The samples were pre sintered at 1000^0 C for 4 hours in air and sintered at 1250^0 C for 3 hours. The influence of Lanthanum (la) substitution on various properties of Ni-Zn ferrites have been studied in this work. Investigations were carried out by the measurements of AC resistivity, Permeability and Curie temperature of the sample. AC resistivity has been found to be decreased of the samples. The initial magnetic permeability remains constant up to 10 MHz thenceforth sharply fall to very low values at higher frequencies and again remain constant from 9 MHz to 120 MHz and onward due to Zn deficient of Ni -Zn ferrites with substituting of La. The sharp directress of permeability at $T = T_c$ indicates the samples good homogeneity. The T_C is found to increase with increasing Zn-deficient by substituting rare earth metal lanthanum (La).

Keywords: Conventional Solid State Reaction Method, Sintering; Resistivity; Permeability; Curie Temperature.

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1. Introduction

At the beginning of the industrialization, iron and its alloys were used as magnetic materials to serve the need of the electrical industry. With the advent of higher frequencies, the standard techniques of reducing eddy current losses, using lamination or iron powder cores, were no longer efficient or cost effective. This realization stimulated a renewed interest in "Magnetic Insulators" as first reported by S. Hilpert in Germany in 1909. By 1945 Snoek had laid down the basic fundamentals of the physics and technology of practical ferrite materials [1]. In 1948, the Neel theory of ferromagnetic provided the theoretical understanding of this type of magnetic material. Polycrystalline soft ferrites prepared from metal oxides are magnetic semi-conductors and have made important contribution, both technological and conceptual to the development of electronics. Soft ferrites still remain the best magnetic materials and cannot replaced by any other magnetic materials with respect to their very high frequency application because they are inexpensive, more stable, easily manufactured [2].The most important advances were made in ferromagnetism in the field of magnetic oxides (ferromagnetic). The advancement of high frequency ferrites was initiated by the work done by J. L. Snoek [3], who found that associated with excellent properties in the high frequency range, Mn-Zn and Ni-Zn ferrites provide a family of magnetic materials useful for radio and TV sets as well carrier telephony as cores of inductors transformer and so forth. The most popular combination are Ni-Zn [4], Ni-Cu-Zn [5-7], Mg-Zn [8] and Mg-Cu-Zn [9] a ferrites. Ferrite crystallizes with two magnetic sub-lattices i.e. tetrahedral (A) site and Octahedral (B) site based on Neel's model. Magnetic and electrical properties of ferrites are strongly dependant on distribution of cation in A and B sites and their valance state [10]. The properties of Ni-Zn ferrites can be tailored by substitutions them with different metals ions such as Co^{2+},Mg^{2+},Mn^{2+},Cu^{2+}etc. The addition of rare earth metal in Ni-Zn ferrite composition is known to play a crucial role in increasing the sintering density and lowering the sintering temperature. Various additives such as V_2O_5, Bi_2O_3, PbO, MoO_3, WO_3, E_2O_3 P_2O_5, La_2O_3, W_2O_3, LiO etc. having low melting point, facilitated to reduce the sintering temperature of these oxides materials due to liquid phase either due to the melting of the additives or due to the eutectic liquid phase formation between the additives and the ferrite. Amount of liquid phase increase with increasing amount of sintering aids which results in increased densification. However excessive amount of sintering additives' will deteriorate electromagnetic properties of the ferrite. So, optimum content of sintering aids is necessary to achieve good sinter ability as well as better electromagnetic properties. Till now researches have not yet been able to formulate a rigid set of rules for ferrites about a single property. Among these La_2O_3,Y_2O_3 and E_2O_3 were the most effective sintering for Ni-Zn ferrite the systematic research is still necessary for effects has been under taken to prepare Ni-Zn ferrite doped with La_2O_3,Y_2O_3 and Eu_2O_3. Scientists still continue their efforts to find out optimum parameters of ferrites, like high saturation magnetization, high permeability, high resistivity, etc.

2. Experimental

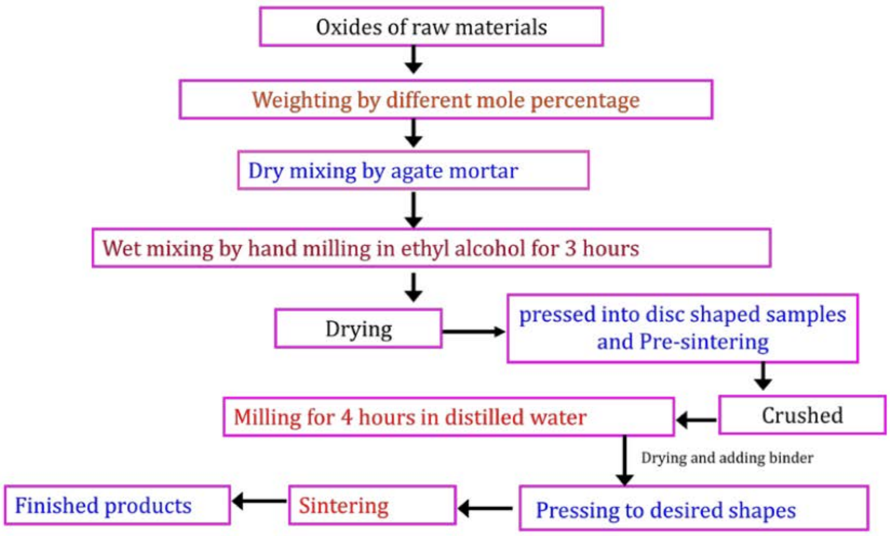
Conventional Solid State Reaction Method: In the solid state reaction method, the required composition is usually prepared form the appropriate amount of raw mineral oxides or carbonates by crushing, grinding and milling. The most common type of mill is the ball mill but instead of ball mill we performed it by hand millig. Milling can be carried out in a wet medium to increase the degree of mixing. This method depends on the solid state inter-diffusion between the raw materials. Solids do not usually react at room temperature over normal time scales. Thus it is necessary to heat them at higher temperatures for the diffusion length $(2Dt)^{1/2}$ to exceed the particle size, where D is the diffusion constant for the fast-diffusing species, and t is the firing time. The ground powders are then calcined in air or oxygen at a temperature above 1000^0C. For some time, this process is continued until the mixture is converted into the correct crystalline phase. The calcined powders are again crushed into fine powders. The tablet and ring shaped samples are prepared from these calcined powders using hydrostatic pressure. Pre sintering is carried out at temperature 1000^0C for (3) three hours and sintering at the temperature 1250^0 C for (4) four hours. The general solid state reaction leading to a ferrite MFe_2O_4 may be represented as

$MO + Fe_2O_3 \rightarrow MeFe_2O_4$ where M is the divalent ions.

Table 1: Compositional details of $Ni_{0.6}Zn_{0.4-x}La_xFe_2O_4$ [x = 0.05, 0.10, 0.15] ferrites

Content,x	NiO	ZnO	La_2O_3	Fe_2O_4
0.05	3.7161	2.3622	0.6754	13.2426
0.10	3.7280	1.6319	1.3551	13.2851
0.15	3.5952	1.6324	1.9603	12.8121

A flow chart of preparation of sample is presented below



As a whole the preparation procedure generally consists of four major steps:

- Preparing a mixture of materials with the cations in the ratio corresponding to that in the final product.
- Pre-firing the mixture to form ferrite.
- Converting the raw ferrite into powder and pressing the powder and pressing the powder into the required shapes.
- Sintering to produce a highly densified product.

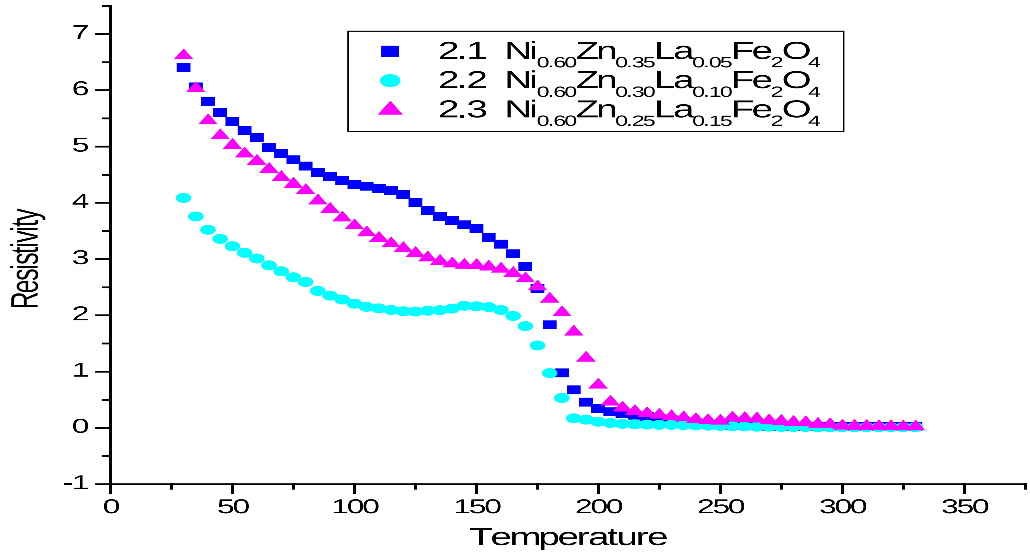


Figure 1: Temperature dependence of resistivity, as a function of Temperature of $Ni_{0.60}$ $Zn_{0.35}$ $La_{0.05}$ Fe_2 O_4, $Ni_{0.60}$ $Zn_{0.30}$ $La_{0.10}$ Fe_2 O_4, and $Ni_{0.60}$ $Zn_{0.25}$ $La_{0.15}$ Fe_2 O_4, sintered at 1250^0C

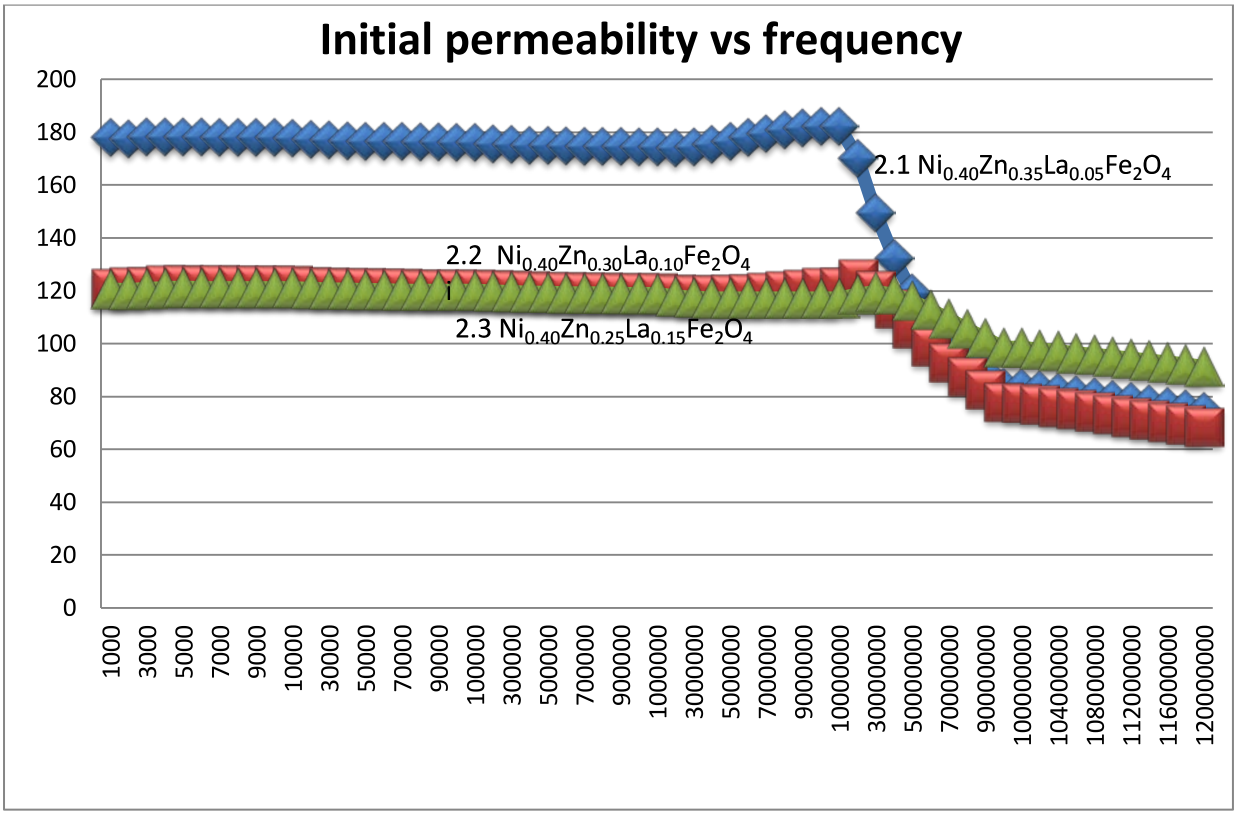


Figure-2: Frequency dependence of the real part of the permeability, as a function of frequency of $Ni_{0.60}$ $Zn_{0.35}$ $La_{0.05}$ Fe_2O_4, $Ni_{0.60}Zn_{0.30}La_{0.10}Fe_2O_4$ and $Ni_{0.60}Zn_{0.25}La_{0.15}Fe_2O_4$ sintered at 1250^0C

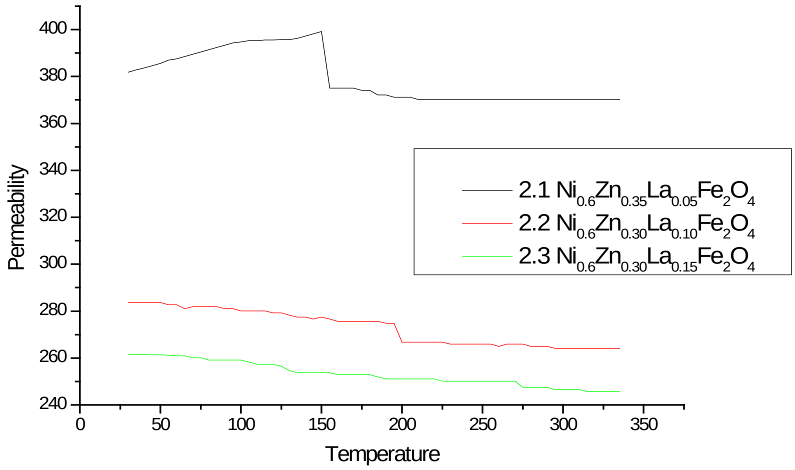


Figure 3: Temperature dependence of the real part of the permeability, as a function of Temperature of $Ni_{0.60}$ $Zn_{0.35}$ $La_{0.05}$ Fe_2O_4, $Ni_{0.60}Zn_{0.30}La_{0.10}Fe_2O_4$ and $Ni_{0.60}Zn_{0.25}La_{0.15}Fe_2O_4$ sintered at 1250^0C

Table 2: Data of Curie temperature (T_C) ($Ni_{0.60}$ $Zn_{0.40-x}$ $La_xFe_2O_4$)

Zn- deficient content , x	$T_s = 1250^0$C
	T_c^{0C}
2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$	145
2.2 $Ni_{0.60}Zn_{0.30}La_{0.010}e_2O_4$	200
2.3 $Ni_{0.60}Zn_{0.25}La_{0.15}Fe_2O_4$	275

3. Results and discussion

In figure -1 the rectangular blue sign indicates the sample 2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$ .The curve of sample 2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$ shows that with increasing the temperature from room temperature the resistivity decreasing up to 225^0C. After that with increasing the temperature the resistivity becomes constant. In figure-1 the circle shaped green sign indicate sample 2.2 $Ni_{0.60}Zn_{0.30}La_{0.10}Fe_2O_4$ which indicates that with increasing the temperature the resistivity decreasing In figure-1 triangular light red sign indicates sample 2.3 which shows that with increasing temperature the resistivity decreasing gradually. For sample 2.1, 2.2 and 2.3 the relation between the resistivity and temperature shows that the resistivity decreasing with increasing the temperature. Actually this is happen because sample 2.1, 2.2 and 2.3 is a ferrite semiconductor alloy.Figure-2 describes the change of permeability with the change of frequency. For sample 2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$ it is seen that the initial permeability is as same as up to a very high frequency 10 MHz i.e., initial permeability is constant up to 10 MHz thenceforth the initial permeability sharply decreases up to 100MHz after that it remains constant on wards. For sample 2.2 $Ni_{0.60}Zn_{0.30}La_{0.10}Fe_2O_4$ with creasing the frequency the initial permeability remains constant up to 30 MHz and then gradually decrease up to 100 MHz and then it remains constant onward. For sample 2.3 the same phenomena is seen as for sample 2.1 and 2.2 hence for sample 2.3 the frequency remain constant up to 60 MHz. Curie temperature Tc is the basic quantity in the study of magnetic materials. It corresponds to the temperature at which a magnetically ordered materials becomes magnetically disordered i.e. becomes paramagnetic. Curie temperature also signifies the strength of the exchange interaction between the magnetic atoms. In figure-3 The black line sign indicates sample 2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$ graph. With increasing the temperature the domain of the sample tends to align parallel to each other. When all the domain align parallel to each other completely then the last limit of ferromagnetic material reach which indicate the homogeneity of the sample and with further increase of temperature permeability of the sample sharply break down which is shown in figure-1 sample 2.1 $Ni_{0.60}Zn_{0.35}La_{0.05}Fe_2O_4$. At temperature 145^0C the permeability sharply breaks down and, this break down temperature is known as Curie temperature. The red line sign indicates the graph of sample 2.2 $Ni_{0.60}\,Zn_{0.30}\,La_{0.10}\,Fe_2O_4$. The sharply break down points of this sample 2.2 is 200^0C. The green line sign indicates the graph o the sample 2.3 $Ni_{0.60}\,Zn_{0.25}\,La_{0.15}\,Fe_2O_4$. The sharply break down points of the sample 2.3 is 275^0C.This Curie temperature indicates the single phase of the studied samples. After Curie temperature the domains of the ferromagnetic materials align in random directions as a result the ferromagnetic materials change into paramagnetic materials.

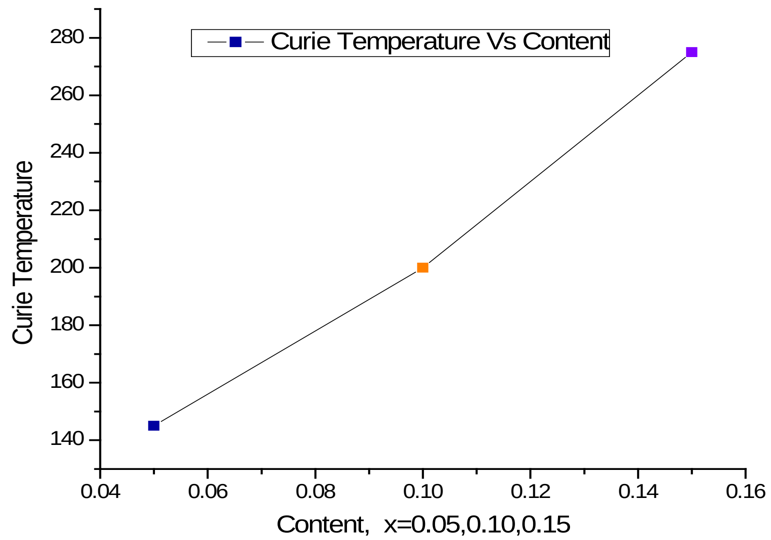


Figure 4: Curie temperature dependence of the content, as a function of Temperature of $Ni_{0.60}\,Zn_{0.35}\,La_{0.05}\,Fe_2O_4$, $Ni_{0.60}Zn_{0.30}La_{0.10}Fe_2O_4$ and $Ni_{0.60}Zn_{0.25}La_{0.15}Fe_2O_4$ sintered at 1250^0C

4. Conclusion

Effect of magnetic and electrical properties of Ni-Zn ferrites has studied thoroughly on the Lanthanum (La) substitution on $Ni_{0.60}Zn_{0.40-x}$ La_x Fe_2 O_4 (x=0.05, 0.10, 0.15) Ferrites. Temperature dependence resistivity was measured to identify the nature of the samples. Frequency dependant permeability was studied to fathom the frequency response initial permeability. Initially resistivity decreases very slowly at lower frequency region, while after a certain frequency it decreases sharply and at high frequency region it becomes almost independent of frequency. Temperature dependence permeability has been used to measure the Curie temperature of $Ni_{0.60}Zn_{0.40-x}$ La_x Fe_2 O_4 (x=0.05, 0.10, 0.15) Ferrites. Curie temperature of Ni-Zn ferrite increases with the decrease in Zn-content. There is much scope for further research in controlling the magnetic characteristics by changing composition and heat treatment certain important parameters like temperature dependence magnetization, anisotropy and magnetostriction can be study in detail for a better understanding of micro structure properties of the samples.

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