iJRASET

International Journal For Research in
Applied Science and Engineering Technology

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 6 Issue: IV Month of publication: April 2018

DOI: <http://doi.org/10.22214/ijraset.2018.4614>

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Nanocrystalline NiCuZn Ferrites For Microwave Absorber Applications

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Abstract: A series of Cu⁺² substituted Ni-Zn ferrite nano powders Ni_{0.5-x}Cu_xZn_{0.5}Fe₂O₄ with x = 0.01, 0.04, 0.06, 0.08, 0.12 mol% were synthesized by microwave hydrothermal method at a low temperature of 160 °C/ 30 min. As synthesized powders were characterized by X-ray diffraction, TEM and microwave sintered at 800°C/30 min. The sintered samples were characterized by XRD and SEM. The temperature variation of electrical resistivity of these samples was measured by two probe method. The electrical resistivity in these ferrites have been explained on the basis of hopping mechanism. The plots of resistivity versus 10³/T are show a transition near the Curie temperature. The activation energy in the ferromagnetic region is in general less than that in the paramagnetic region. Dielectric properties were measured in the frequency range of 20 Hz – 1MHz.

Keywords: Ferrites, Microwave-hydrothermal method, Microwave Sintering,, Electrical properties and Dielectric properties.

I. INTRODUCTION

This The development of microwave-absorbing materials continues to attract much attention. Among the candidates for such application, ferrites present an interesting material. The use of ferrite-based absorbers requires better performance at higher frequencies such as X-band. In this direction, many efforts were focused on studying the effect of ferrite content [1], chemical composition [2–7], and also copper addition. The aim of this paper is the development of microwave-absorbing materials based on ferrites using the microwave-hydrothermal method and investigation of their electrical properties were undertaken. The Ni-Zn ferrite is a well-known mixed inverse spinel, whose unitary cell is represented by the formula (Zn_xFe_{1-x})[Ni_{1-x}Fe_{1+x}]O₄ [8]. The addition of impurities induces changes in the defect structure and texture of the crystal [9], creating significant modifications in the magnetic and electrical properties of these materials. However, the synthesis route also plays a crucial role, so that samples of comparable crystallite size prepared by different processes show different electrical and magnetic properties. The aim of this work is to review the effects of Cu⁺² addition in NiZn ferrites by partial substitution of Ni²⁺ that has influence on resistivity, and dielectric properties.

II. EXPERIMENTAL

Pure (99.99%) Nickel nitrate [Ni (NO₃)₂.6H₂O], Copper nitrate [Cu (NO₃)₂.3H₂O], Zinc nitrate [Zn(NO₃)₂.6H₂O] and Iron nitrate[Fe(NO₃)₂.9H₂O] were dissolved in 50 ml of deionized water. The molar ratio of powders was adjusted so as to obtain the composition Ni(0.5-x)Cu_xZn_{0.5}Fe₂O₄ with x = 0.01(MH1), 0.04(MH2), 0.06(MH3), 0.08(MH4), 0.12(MH5). In sequence, the neutralizer NaOH (10M) was added keeping the co-precipitated solution with pH in ~ 9.45. Then the solution was treated with microwaves at 160°C/30 min. The crystal phase of the powder was analysed with powder X-ray diffraction (Philips (P analytical). The sizes and morphologies were characterized by a transmission electron microscope (TEM) at 200 kV with Hitachi H-800 (Hitachi High-Technologies Co. The pressed samples were microwave sintered at 800°C/ 30 min and characterized by XRD and SEM. The sintered samples were used for further studies. The temperature variation electrical resistivity measurements were carried out using a two probe method. The frequency dependence of dielectric constant (ε') and dielectric loss tangent (Tan δ) on the nanocrystalline NiCuZn ferrites has been measured in the frequency range of 20Hz to 1MHz by using LCR meter at room temperature.

III. RESULTS AND DISCUSSION

Figs.1 shows the X-ray diffraction pattern for the nano-powders of MH 1 to MH5, respectively. It is clear from the figures that single-phase spinel structure is seen in all the cases. The broad diffraction peaks observed in XRD pattern indicates the small particle size nature. The 311-reflection peak in the present ferrite (NiCuZn ferrite) is the major existing peak at 2θ = 35°. The average particle size of the synthesized powders was estimated from the X-ray peak broadening. Scherer equation and the calculated particle sizes for the present powders are in between 15-20 nm.

Fig. 1 X-ray diffraction pattern for the nano-powders of NiCuZn ferrites

In the present investigation, the particle size and morphology of the as synthesized powders were obtained using Transmission Electron Microscopy (TEM) technique. For the TEM observations, the powders were dispersed in ethanol by the ultrasonic machine. Figs.2. show the TEM micrograph for the as synthesized powders of the samples MH1, MH2, MH3, MH4 and MH5 respectively. It can also be seen from the figures that the nanocrystalline particles are well distributed. The particlesize has been calculated and presented in Table 1. From the selected area electron diffraction patterns of NiCuZn ferrite powders (Fig.2), it can be observed that the synthesized particles are crystalline in nature. The particle size estimated from TEM pictures for present nano-powder of ferrites varies from 20nm to 30nm. Thus, the particle size calculated from the XRD of the synthesized samples is nearly correlating with the particle size calculated from the TEM pictures.

Fig. 2. TEM pictures on Nanopowders of NiCuZn ferrites

All the sintered samples were characterized by using X-ray diffraction patterns. It was observed that the XRD patterns showed all samples have spinel phase, indicating the absence of any other impurity phase. The line widths observed in the XRD pattern are narrower than that of the line widths of the corresponding nanocrystalline powders. The Scherer's equation is used to estimate the particle size of samples. The average crystalline size found to be 32nm -50nm.

Fig. 3. XRD patterns on sintered NiCuZn ferrites

Fig.4represents the SEM pictures for the microwave sintered samples of the NiCuZn ferrites. It can be seen from the figures that the sintered powders are spherical in shape and the particle size have been estimated from the SEM pictures for all the samples and presented in the Table 2. It can also be observed from the figures that the surface of the sample shows voids and pores formed by the escaping gases during the microwave reaction. This type of porous network is typical of microwave sintered powders. These porous powders are highly friable which facilitate easy grinding to obtain finer particles.

Fig. 4 : SEM pictures on sintered NiCuZn ferrites

Using the XRD data, the lattice constant (a) values are calculated and are given in Table 1. It can be observed from the table that the lattice parameters for these ferrites increase with an increase of Cu^{+2} ion concentration. Since Cu^{+2} ions have larger ionic radius ($0.87 \text{ \AA}$) than those of Ni^{+2} ions ($0.83 \text{ \AA}$) [6], a partial replacement of latter by the former causes the expansion in unit cell dimensions, thereby increasing the lattice parameter.

The bulk density (d_{bulk}) of all the prepared ferrites has been measured accurately by the Archimedes principle. This method gives the density values with an accuracy of $\pm 0.01 \text{ gm/cc}$ and results are presented in Table1 for microwave sintered samples. X-ray densities ($d_{\text{x-ray}}$) have been determined using the value of lattice constant (a). The X-ray density ($\rho_{\text{x-ray}}$) for the ferrite and the porosity of the samples are given Table 1. It can be seen from the table that the values of porosity vary from 3% to 7%. Table 1 gives the electrical resistivity (ρ) values for the microwave sintered NiCuZn ferrites at room temperature. Similar to conventionally sintered sample, the resistivity of the samples increased one order of magnitude with an addition of Cu^{2+} to NiCuZn ferrite[6]. Hence, this increase of resistivity in microwave sintered samples may be due to the formation of Cu^{2+} as an insulating liquid film on the grain boundaries [7]. It can be seen from the table that the microwave-sintered samples possess higher resistivity as compared to that of conventionally sintered samples [6]. This is may be due to the formation of fine and uniform microstructures obtained present in these ferrites due to microwave sintering, which provides ample grain boundary area. There are few reports on the d.c. resistivity of NiCuZn ferrites in available literature. Nam.et.al. [7] have prepared $(\text{Ni}_{0.4}\text{Cu}_{0.1}\text{Zn}_{0.5})(\text{Fe}_2\text{O}_3)_{0.98}$ sample using oxide powders and then sintering conventionally at 1000°C . They have obtained d.c. resistivity values equal to $2.17 \times 10^8 (\Omega\text{-cm})$. Wang.et.al. [8] have prepared $\text{Ni}_{0.38}\text{Cu}_{0.12}\text{Zn}_{0.5}\text{Fe}_2\text{O}_3$ ferrite using the conventional sintering method and obtained a d.c. resistivity of the order of $10^7 (\Omega\text{-cm})$. Z.Yue.et.al. [9] have prepared NiCuZn ferrites using the sol-gel method and the values of d.c. resistivity obtained were about $4.3 \times 10^8 (\Omega\text{-cm})$. We compared the present values of d.c. resistivity with that of the reported values; it is found that there is fair agreement between them. In the present investigation, the sample having 8 mol% Cu and microwave sintered sample (MW4) possess a high value of resistivity.

Table 1: Preparation data on Nanocrystalline NiCuZn ferrites

Sample	Crystallize size Nm	Grain size (nm)	Lattice constant ($\text{\AA}$)	Bulk density d_{bulk} (g/cm^3)	$d_{\text{x-ray}}$ (g/cm^3)	ϵ' at 1MHz	ϵ'' at 1MHz	P_{dc} Ohm-cm $\times 10^9$
MH1	72	76	8.373	4.98	5.37	11	0.04	1.4
MH2	78	80	8.390	5.07	5.36	9	0.03	1.5
MH3	85	90	8.393	5.09	5.36	8	0.03	1.9
MH4	63	70	8.422	5.12	5.28	6	0.02	2.5
MH5	90	95	8.436	5.00	5.26	10	0.03	1.6

Fig. 5 shows the variation of electrical resistivity ($\log \rho$) with temperature ($1000/T$). The resistivity of the sample decreases with increasing temperature according to the relation $\rho = \rho_0 e^{E_g/kT}$, where E_g represents the activation energy which is the energy needed to release an electron from an ion for a jump to the neighboring ion, so giving rise to the electrical conductivity. K is Boltzmann constant and T is the absolute temperature. The activation energies E_f (for region I), E_p (for region II) from the slope of the graph and the transition temperature, T_{NO} obtained from temperature at which a change of slope occurs are obtained. It is found that the value of activation energy in the ferrimagnetic region is lower than that of the paramagnetic region. The lower activation energy in the ferrimagnetic region is attributed to the magnetic spin disordering [10] due to decrease in the concentration of current carriers [11] while the change in activation energy is attributed to the change in conduction mechanism [12].

Fig. 5 Temperature variation of $\log \rho$ on NiCuZn ferrites

Fig6 shows the variations of dielectric constant, and loss tangent with frequency in the range 20 Hz to 1 MHz for nanocrystalline NiCuZn ferrites. It can be seen from the figures that dielectric constant and loss tangent are decreasing with the increase in frequency. This can be explained on the basis of Maxwell–Wagner model [13-14]. According to this model the dielectric structure was composed of well conducting grains separated by the poorly conducting grain boundaries. The electrons reach the grain boundary through hopping and if the resistance of grain boundary is high enough then electrons pile up there and produce polarization. The decrease in dielectric constant with the increasing frequency can be explained on the basis of the fact that the space charge carriers require a finite time to line up their axes in the direction of an applied alternating field. Thus, if the frequency of the applied field increases, the electrons turn around their direction of motion more frequently. This reduces the possibility of electrons reaching the grain boundary and as a result polarization decreases resulting thereby in the decrease of dielectric constant with an increase in frequency. Further, the dielectric loss tangent arises, if the polarization lags behind the applied altering field and is caused due to the presence of impurities and structural in homogeneities. The low dielectric loss tangent values obtained in the present work are therefore attributed to the more structurally perfect and homogeneous ferrites.

Fig. 6 Frequency dependence of dielectric constant and $\tan \delta$ on NiCuZn ferrites

IV. CONCLUSIONS

A series of NiCuZn ferrites were prepared using microwave- hydrothermal method. The powders were sintered at were sintered at 800°C/30 min. The prepared samples were characterized using X-rays. Crystallite size was calculated with the Scherer formula. With increasing Cu²⁺ content, electrical resistivity increased up to 8 mol% and then decreases on higher addition. The dielectric constant and dielectric loss are no much affected by the incorporation of Cu²⁺.

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