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Effect of Sintering Temperature on Structural and Magnetic Properties of $Ni_{0.55}Zn_{0.45}Fe_2O_4$ Ferrites

Robiul Islam[1*], Md Obaidur Rahman[1], M. Abdul Hakim[2], Dilip Kumar Saha[2], Saiduzzaman[1], Saroaut Noor[3], Md Al-Mamun[2]

[1]The Department of Physics, Jahangirnagar University, Dhaka, Bangladesh; [2]Materials Science Division, Atomic Energy Centre, Dhaka, Bangladesh; [3]Department of Physics, Khulna University of Engineering & Technology, Khulna, Bangladesh.
Email: [*]robiulislam41@yahoo.com

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ABSTRACT

The effect of frequency and sintering temperature on initial permeability of $Ni_{0.55}Zn_{0.45}Fe_2O_4$ ferrites have been studied by using an impedance analyzer. The samples were prepared by conventional double sintering ceramic technique using oxide nanoparticles of grain size 30 - 50 nm. Single phase spinal structure has been confirmed for the prepared samples by X-ray diffraction. As the sintering temperatures increase from 1160˚C to 1300˚C, the permeability gradually increases. The increase of permeability is ascribed to the increase of density and grain size. Grain size is expected to grow with the increase of sintering temperature. Ferrites with large average grain size posses higher initial permeability. The Curie temperatures determined from temperature dependence of permeability of the samples sintered at different temperatures are found to be T_c = (321 ± 1)˚C and independent of sintering temperature. At T_s = 1300˚C, T_c is found to increase substantially which can be explained by the fact that Zn has evaporated from the surface layer.

Keywords: Nanoparticle; Sintering Temperature; Curie Temperature; Microstructure; Grain Size

1. Introduction

The Ni-Zn ferrites are one of the most versatile, reasonable cost magnetic materials for general use in both low and high frequency devices because of their high resistivity, low dielectric losses, high Curie temperature and chemical stability [1-4]. The magnetic properties of Ni-Zn ferrites are strongly dependent on their chemical composition, density, grain size, etc. [5-7]. The permeability increases with the substitution of Zn for Ni in Ni-Zn ferrites [8], but the Curie temperature slightly decreases at sufficiently high temperature due to Zn evaporation. It is observed that the permeability falls sharply at the ferri-paramagnetic phase transition *i.e.* the paramagnetic character. The sharp vertical drop of the permeability at Curie point indicates the degree of good homogeneity in the sample composition. Due to the remarkable behavior of magnetic and electric properties they are subjects of intense theoretical and experimental investigation for application purpose [9,10]. The X-ray diffraction patterns for the composition at different sintering temperatures clearly indicate their single phase and formation of spinal crystal structure [11]. This papers focuses on the effect of sintering temperature on the initial and complex permeability of Ni-Zn ferrites. A possible correlation between sintering temperature, grain size and density is also discussed.

2. Experimental

The mixed ferrites of composition $Ni_{0.55}Zn_{0.45}Fe_2O_4$ was prepared by the double sintering ceramic method. The raw materials for the preparation of the studied composition were oxide nanoparticle of grain size 30 - 50 nm. The oxide powders were weighed precisely according to their molecular weight. Intimate mixing of the materials was carried out using agate mortar for 5 hours and then ball milled in a planetary ball mill in ethyl alcohol media for 4 hours with stainless steel balls of different sizes in diameter. The slurry was dried and the dried powder was pressed into disc shape. The samples were pre-sintered at 900˚C for holding time 4 hr. The pre-sintered materials were crushed and ball milled for another 4 hours in distilled water to reduce it to small crystallites of uniform size. The mixture was dried and a small amount of saturated solution polyvinyl alcohol were added as a binder. The resulting powders were pressed uni-axially under a pressure of 1.75 ton·cm^{-2} and 1.2 ton·cm^{-2} in a stainless steel die to make pellets and toroids respectively. The pressed pellets and toroid shaped samples were then fi-

[*]Corresponding author.

nally sintered at 1160˚C, 1200˚C, 1250˚C and 1300˚C. Initial permeability were measured by an impedance analyzer at different sintering temperature 1160˚C, 1200˚C, 1250˚C and 1300˚C. The density was calculated by measuring the volume and weight of the cylindrical samples. X-ray diffraction (XRD) pattern of all the samples, recorded at room temperature with CuK_α radiation of wavelength λ = 1.54178 Å using Philips X' Pert PRO X-ray diffractometer. X-ray patterns confirmed the spinal phase for the samples.

3. Results and Discussion

The XRD patterns of the sample $Ni_{0.55}Zn_{0.45}Fe_2O_4$ at different sintering temperatures are shown in **Figure 1**. The main reflection were from plane (111), (220), (311), (222), (400), (422), (511) and (440) which are characteristic of spinel structure with a single phase [12]. The lattice parameter was determined by using the Nelson-Riley (N.R) extrapolation method. In this experiment we observed that, the lattice parameter remains constant with different sintering temperature [13], **Figure 2**.

Initial permeability as dependent on frequency and temperature of a magnetic material is an important parameter. **Figures 3** and **4** show the real part and imaginary part of permeability for different sintering temperature [14]. Here the real part of permeability μ' remains constant in a certain frequency range (1 KHz - 13 MHz), while at higher frequencies, after a small rise, it begins to drop due to resonance. We have found that the permeability increases with increasing sintering temperature. It is seen from **Table 1** that the grain size increases with increasing sintering temperature [15,16] because the density increases with increasing sintering temperature.

From **Figures 5** and **6** show the real and imaginary part of the complex permeability with variation of temperature [17]. It is observed that, μ' increases gradually with increase of temperature attaining a peak value just before T_c known as Hopkinson peak. The Curie temperature determined from temperature dependence of

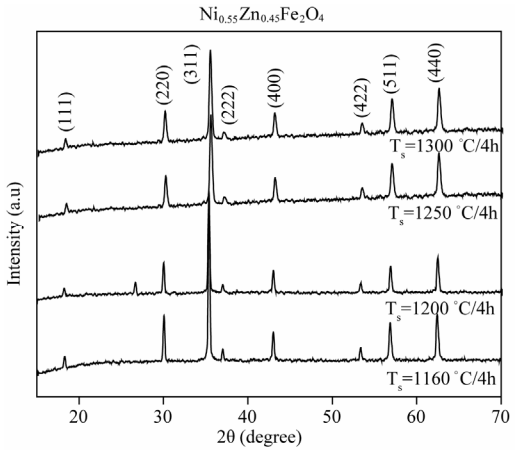

Figure 1. XRD pattern of Ni-Zn ferrites at different sintering temperature.

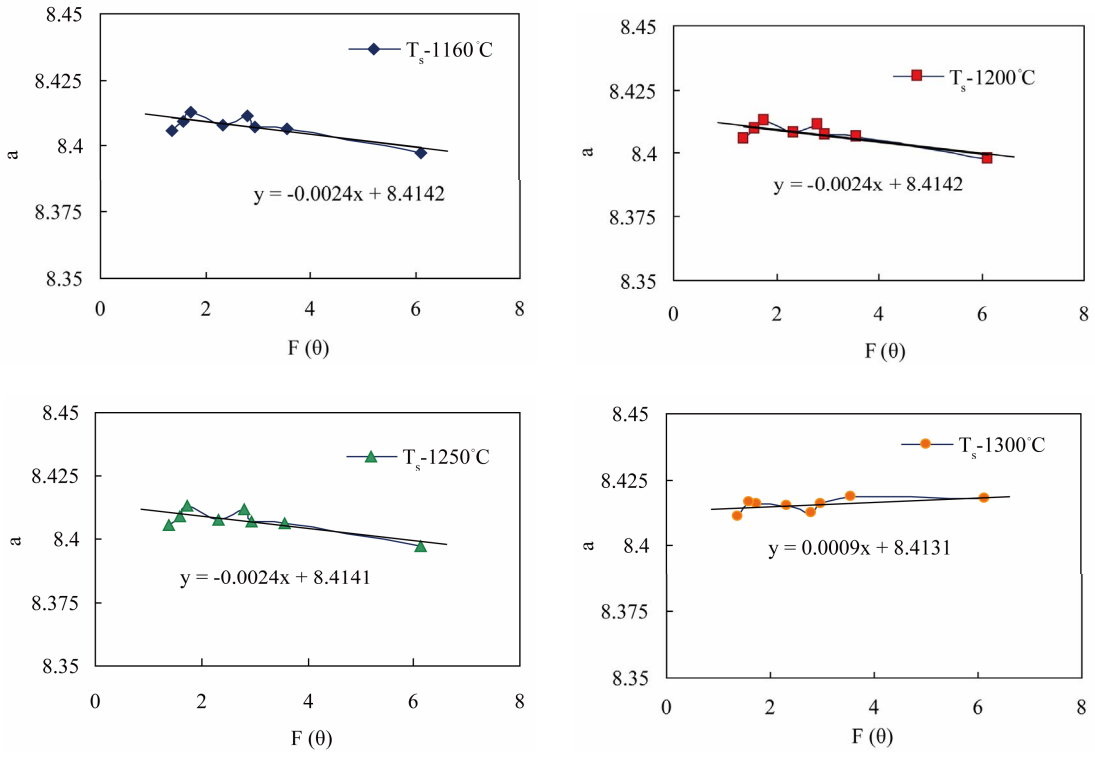

Figure 2. The variation of lattice parameter "a" as a function of F(θ).

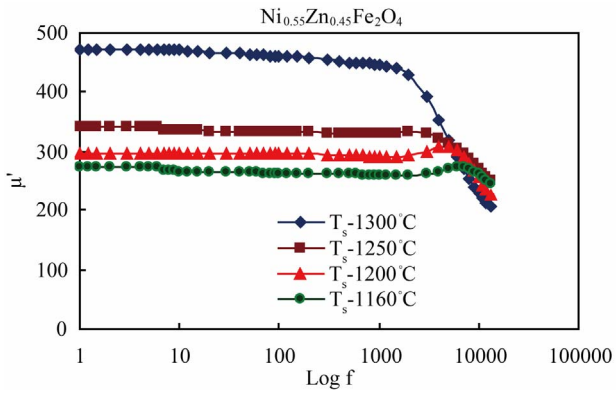

Figure 3. Real part of permeability vs frequency at various T_s.

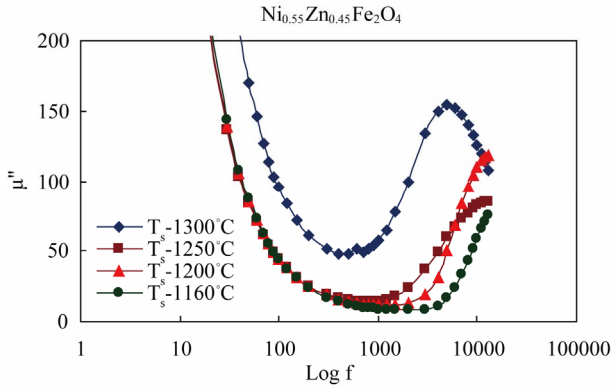

Figure 4. Imaginary part of permeability vs frequency at various T_s.

Table 1. Variation of Curie temperature and Lattice parameter with different sintering temperature.

Composition	Sintering temperature (T_s) °C	Curie temperature (T_c) °C	Lattice parameter a (Å)
$Ni_{0.55}Zn_{0.45}Fe_2O_4$	1160	321	8.414
	1200	320	8.414
	1250	321	8.414
	1300	323	8.414

Table 2. Variation of density and porosity with sintering temperature.

Sintering temperature (T_s) °C	ρ_{th} (kg/m^3)	ρ_B (kg/m^3)	Porosity P (%)
1160	0.005287	0.0049985	5.4567
1200	0.005287	0.0050637	4.2235
1250	0.005287	0.005074	4.0287
1300	0.005287	0.0048564	8.1445

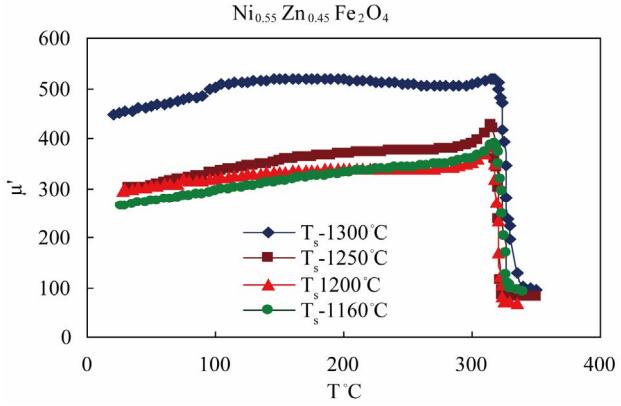

Figure 5. Real part of permeability vs temperature at various T_s.

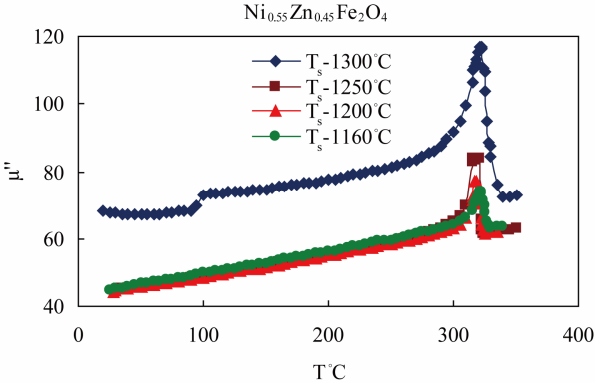

Figure 6. Imaginary part of permeability vs temperature at various T_s.

permeability of the samples sintered at different temperatures are found to be $T_c = (321 \pm 1)$°C. The T_c value is almost independent of T_s except for the sample sintered at 1300°C where a substantial increase of T_c is found.

The Scanning Electron Microscope (SEM) of $Ni_{0.55}Zn_{0.45}Fe_2O_4$ samples sintered at 1160°C, 1200°C, 1250°C and 1300°C are shows in **Figures 7(a)-(d)** respectively. The sintering temperature has a great influence on the microstructure. It was observed that the average grain size of the samples increased with increase sintering temperature [18].

The density and porosity of the composition are changed with different sintering temperature [8]. The density of $Ni_{0.55}Zn_{0.45}Fe_2O_4$ samples increases from 1160°C to 1250°C and above 1250°C the density begins to decrease as shown in **Figure 8**. On the other hand, porosity (P) of the samples decrease as increasing sintering temperature up to 1250°C, and above 1250°C the porosity increases which is also shown in **Table 2**.

It is known that the porosity of the samples come from two sources, intragranular porosity and intergranular porosity. The intergranular porosity mainly depends on the grain size [18]. At higher sintering temperatures the den-

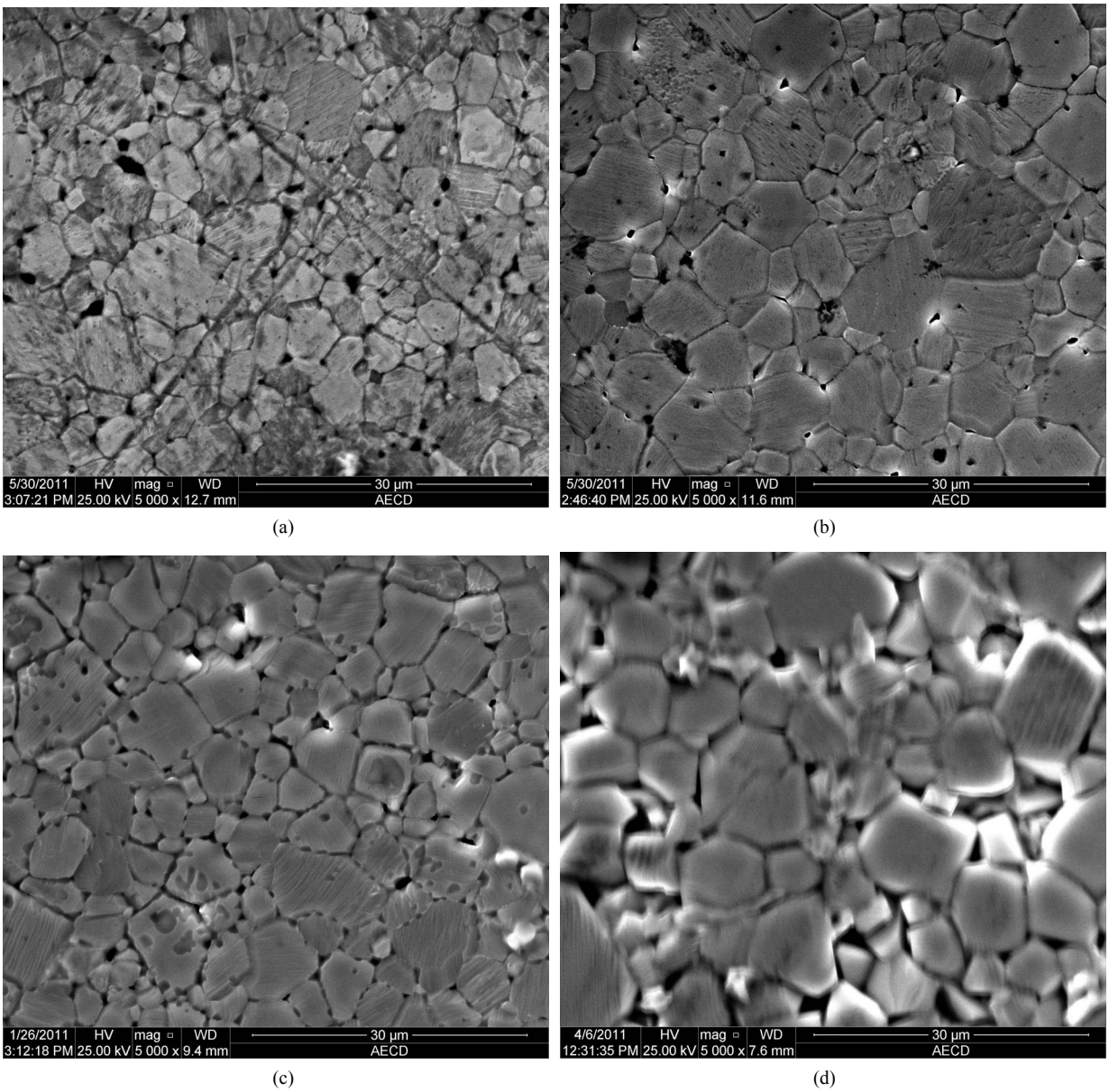

Figure 7. Microstructure of the samples at different sintering temperature. (a) 1160˚C; (b) 1200˚C; (c) 1250˚C; (d) 1300˚C.

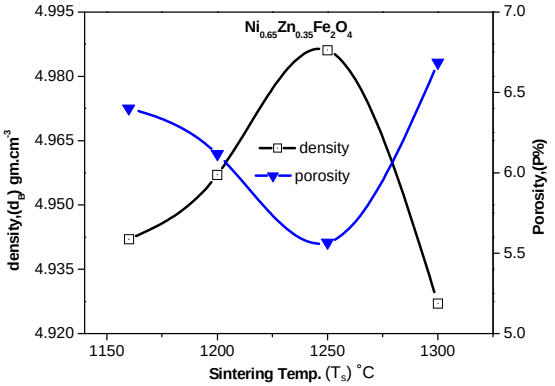

Figure 8. Variation of density and porosity with T_s for $Ni_{0.55}Zn_{0.45}Fe_2O_4$.

sity is decreased because the intragranular porosity is increased resulting from discontinuous grain growth.

According to sintering mechanism, we can explained the grain growth of the spinel ferrites. During the sintering process, a shrinkage of up to approximately 20% can occur and the samples produced an entirely dense material. But a product having a certain porosity. At higher sintering temperatures, the density is increased and the porosity is decreased (**Figure 8**). But at T_s = 1300˚C, the porosity is increased slightly.

During the sintering process, the thermal energy generates a force that drives the grain boundaries to grow over pores, thereby the pore volume and hence the materials becomes denser. At higher sintering temperature,

density is decreased because the intragranular porosity is increased. The crystal grain growth depends on grain boundaries migrating and larger crystal grains swallowing the small ones. During the growth the more different in size of crystal grains, the more beneficial for larger crystal grains to swallow smaller ones, so some pores inside the sintered ferrites are eliminated and the crystal growth improves.

When the grain growth rate is very high, pores may be left behind by rapidly moving boundaries. As a result in pores that are trapped inside the grains. This discontinuous grains growth rise with high temperature and the bulk density is reduced [10].

It is well known that the permeability of nanocrystalline ferrite is related to two different magnetizing mechanisms: spin rotation and domain wall motion, which can be described as, $\mu_i = 1 + \chi_W + \chi_{SPIN}$, where χ_W is the domain wall susceptibility, χ_{SPIN} is intrinsic rotational susceptibility. We also written, $\chi_W = 2\pi M_S^2/4\gamma$ and $\chi_{SPIN} = 2\pi M_S^2 D/K_u$ with M_s saturation magnetization, K_u is the total anisotropy, D the grain diameter and γ the domain wall energy. Thus, the domain wall motion is affected by the grain size and enhanced with the increase of grain size [19].

4. Conclusion

The X-ray diffraction confirmed the single phase cubic spinel structure of the samples. The initial permeability increases with increasing sintering temperature. Also the density is increased with increasing sintering temperatures. The Curie temperatures determined from temperature dependence of permeability of the samples sintered at different temperatures are found to be T_c = 321˚C ± 1˚C and independent of sintering temperature. But, at 1300˚C the Curie temperature is found to increase substantially which can be explained by the fact that Zn has evaporated from the surface layer. At higher sintering temperatures the density is decreased because the intragranular porosity is increased resulting from discontinueous grain growth. The lattice parameter is 8.414 Å and which is also independent of sintering temperatures.

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REFERENCES

[1] Y. Li, J. P. Zhao and J. C. Han, "Self-Propagation High Temperature Synthesis and Magnetic Properties of $Ni_{0.35}Zn_{0.65}Fe_2O_4$ Powder," *Bulletin of Materials Science*, Vol. 25, No. 4, 2002, pp. 263-266. doi:10.1007/BF02704117

[2] J. Smith and H. P. J. Wijn, "Ferrites," John Wiley and Sons, New York, 1995.

[3] B. D. Cullity and C. D. Graham, "Introduction to Magnetic Materials," John Wiley and Sons, Hoboken, 2009.

[4] E. Olsen and J. Thonstad, "Nickel Ferrites as Inert Anodes in Aluminium Electrolysis: Part I Material Fabrication and Perliminary Testing," *Journal of Applied Electrochemistry*, Vol. 29, No. 3, 1999, pp. 293-299. doi:10.1023/A:1003460220418

[5] M. M. Barakat, M. A. Henaish, S. A. Olofa and A.Tawfik, "Sintering Behaviour of the Spinel Ferrites System $Ni_{0.65}Zn_{0.35}Fe_{2-x}Cu_xO_4$," *Journal of Thermal Analysis and Calorimetry*, Vol. 37, No. 2, 1991, pp. 241-248. doi:10.1007/BF02055926

[6] O. F. Caltun, L. Spinu and Al Staneu, "Structural and Magnetic Properties of Ni-Zn-Cu Ferrites Sintered at Different Temperatures," *Journal of Optoelectronics and Advanced Materials*, Vol. 4, No. 2, 2002, pp. 337-340.

[7] A. M. Kumar, M. C. Verma, C. L. Dube, K. H. Rao and S. C. Kashyap, "Development of Ni-Zn Nano Ferrite Core Material with Improved Saturation Magnitizition and DC Resistivity," *Journal of Magnetism and Magnetic Materials*, Vol. 320, No. 14, 2008, pp. 1995-2000. doi:10.1016/j.jmmm.2008.02.129

[8] S. T. Mahmud, A. K. M. Akther Hossain, A. K. M. A. Hakim, M. Seki, T. Kawai and H. Tabata, "Influence of Microstructure on the Complex Permeability of Spinel Type Ni-Zn Ferrites," *Journal of Magnetism and Magnetic Materials*, Vol. 305, No. 1, 2006, pp. 269-274. doi:10.1016/j.jmmm.2006.01.012

[9] G. F. Dionne and R. G. West, "Magnetic and Dielectric Properties of the Spinel Ferrite System $Ni_{0.65}Zn_{0.35}Fe_{2-x}Mn_xO_4$," *Journal of Applied Physics*, Vol. 61, No. 8, 1987, pp. 3838-3910. doi:10.1063/1.338623

[10] A. K. M. A. Hossain, S. T. Mahmud, M. Seki, T. Kawai and H. Tabata, "Structural, Electric Transport and Magnetic Properties of $Ni_{1-x}Zn_xFe_2O_4$," *Journal of Magnetism and Magnetic Materials*, Vol. 312, No. 1, 2007, pp. 210-219. doi:10.1016/j.jmmm.2006.09.030

[11] B. P. Rao, A. M. Kumar, K. H. Rao, Y. L. N. Murthy, O. F. Caltun, I. Dumitru and L. Spinu, "Synthesis and Magnetic Studies of Ni-Zn Ferrite Nanoparticles," *Journal of Optoelectronics and Advanced Materials*, Vol. 8, No. 5, 2006, pp. 1703-1705.

[12] S. Zahi, "Synthesis, Permeability and Microstructure of the Optimal Nickel-Zinc Ferrites by Sol-Gel Route," *Journal of Electromagnetic Analysis and Applications*, Vol. 2, No. 1, 2010, pp. 56-62. doi:10.4236/jemaa.2010.21009

[13] S. Akhter, D. P. Paul, Md. A. Hakim, D. K. Saha, Md. Al-Mamun and A. Parveen, "Synthesis, Structural and Physical Properties of $Cu_{1-x}Zn_xFe_2O_4$ Ferrites," *Materials Science and Applications*, Vol. 2, 2011, pp. 1675-1681.

doi:10.4236/msa.2011.211223

[14] A. Verma, T. C. Goel and R. G. Mendiratta, "Frequency Variation of Initial Permeability of Ni-Zn Ferrites Prepared by the Citrate Precursor Method," *Journal of Magnetism and Magnetic Materials*, Vol. 210, No. 1-3, 2000, pp. 274-278.

[15] M. Sorescu, L. Diamandescu, R. Peelamedu, R. Roy and P. Yadoji, "Structural and Magnetic Properties of Ni-Zn Ferrites Prepared by Microwave Sintering," *Journal of Magnetism and Magnetic Materials*, Vol. 279, No. 2-3, 2004, pp. 195-201. doi:10.1016/j.jmmm.2004.01.079

[16] S. R. Murthy, "A Study of Ultrasonic Velocity and Attenuation on Polycrystalline Ni-Zn Ferrites," *Bulletin of Materials Science*, Vol. 24, No. 6, 2001, pp. 611-616. doi:10.1007/BF02704009

[17] J. L. Snoek, "Dispersion and Absorption in Magnetic Ferrites at Frequencies above one Mc/s," *Physica*, Vol. 14, No. 4, 1948, pp. 207-217. doi:10.1016/0031-8914(48)90038-X

[18] Z.-H. Yang, Z.-Q. Gong, H.-X. Li, Y.-T. Ma and Y.-F. Yang, "Synthesis of Ni-Zn Ferrites and Its Microstructure and Magnetic Properties," *Journal of Central South University of Technology*, Vol. 13, No. 6, 2006, pp. 618-623. doi:10.1007/s11771-006-0004-x

[19] M. Jalaly, M. H. Enayati, P. Kameli and F. Karimzadeh, "Effect of Composition on Structural and Magnetic Properties of Nanocrystalline Ball Milled $Ni_{1-x}Zn_xFe_2O_4$ Ferrites," *Physica B*: *Condensed Matter*, Vol. 405. No. 2, 2010, pp. 507-512. doi:10.1016/j.physb.2009.09.044

