INFLUENCE OF COMPOSITION AND SINTERING TEMPERATURE ON COMPLEX PERMEABILITY OF SPINEL TYPE Ni-Zn FERRITE

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ABSTRACT

The polycrystalline ferrites $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ with x=0.97, 1 and 1.03 were prepared by conventional ceramic technique, sintered at various temperatures. The influence of composition and sintering temperature on complex permeability of these samples were investigated. The relative quality factor (Q-factor) of the samples was also discussed. The single-phase cubic spinel structures of the samples have been confirmed from X-ray diffraction patterns. The lattice parameters of all the samples have been determined using the extrapolated Nelson-Relay function. Lattice parameter increases both with decreasing and increasing Fe_2O_3 content from the equimolar concentration. The Bulk density decreases with Fe_2O_3 content. Initial permeability increases with decreasing Fe_2O_3 content. Initial permeability also increases with increasing sintering temperature at first then decreases. The Q-factor decreases with increasing sintering temperature.

Keywords: Complex initial permeability, Lattice parameter, Quality factor.

1. INTRODUCTION

Ferrites are ceramic magnetic materials. The ferrites were developed into commercially important materials, at the Philips Research center (Snoek, 1933-1945). Ferrites possess the combined properties of magnetic materials and insulators. They form a complex system composed of grains, grain boundaries and pores. They have the magnetodielectric property of material which is useful for high frequency applications. Ferrites have very high electrical resistivity which means an applied alternating magnetic field will not induce eddy currents in a ferrite. This property makes ferrites almost ideal materials for high frequency applications, which was first investigated by Hilpert S. in 1909. The metal ions in a ferrite crystal occupied two different kinds of position called A sites and B sites. Neel made the basic assumption that the exchange force acting between an ion on A site and an ion on B site is opposite direction, as in an antiferromagnetic. There is thus a lattice of A ions spontaneously magnetized in one direction and a lattice of B ions magnetized in the opposite direction. However, the magnitudes of the A and B sublattice magnetization are not equal. The two opposite magnetic moments do not cancel and a net spontaneous magnetization results. Magnetic property of ferrites depends on the sublattice distribution of cations. Spinel type ferrites are commonly used in many electronic and magnetic devices due to their high magnetic permeability and low magnetic losses (Toshiyuki Suzuki et al., 2001; Giannakopoulou et al., 2002) and also used in electrode materials for high temperature applications because of their high thermodynamic stability, electrical resistivity and resistant to corrosion (Olsen et al., 1999; Augustin et al., 1993). The Ni-Zn ferrite is considered as the most versatile ferrites, due to their high resistivity and low eddy current losses. Ni-Zn ferrite has also especially been attractive for high frequency transformers and inductors. The structural and magnetic properties of these ferrites have been investigated by a number of researchers (Tsutaoka, 2003; Jingling et al., 2002; Ahmed et al., 2003; Hua Su et al., 2007; Robiul et al., 2012; Luis et al., 2012). The present paper focuses the influence of composition and sintering temperature on complex permeability of Ni-Zn ferrite.

2. EXPERIMENTAL

The $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ (for x=0.97, 1, 1.03) samples were synthesized using the solid state reaction technique. Powder of NiO (purity 99.9%), ZnO (purity 99.9%) and Fe₂O₃ (purity 99.9%) were used as raw

materials. The stoichiometric proportions of required powders were weighed first and then mixed thoroughly using ceramic mortar and pestle. The resultant powder was then ball milled for 5 hours to produce fine powder of mixed constituents. After ball milling the mixture was calcined at 1000° C for 9h. The calcined powders were then pressed into disk-shaped and toroid- shaped samples. The samples were sintered at various temperatures 1150°C, 1200°C, 1250°C, 1300°C and 1350°C for 3h. X-ray diffraction was carried out with an X-ray diffractometer with Cu-K_{α} radiation. From the X-ray diffraction pattern lattice parameter was determined by using Nelson-Riley function. Theoretical density, d_{th}, was calculated using the relation d_{th}=ZM/N_Aa³, where M is the molecular

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weight, N_A is the Avogadro's number, a is the lattice parameter and Z is the number of molecules per unit cell, which is 8 for the spinel structure. The bulk density was calculated by considering the cylindrical shape of the pellets and using the relation $d_B=m/V=m/\pi r^2h$, where m is the mass, r is the radius and h is the thickness of the pellet. Porosity of the samples was then determined by the relation $P=(1-d_B/d_{th})$. The frequency characteristics i.e. the initial permeability spectra of the toroid shaped samples were measured using an Agilent precision impedance analyzer (model no. 4294A) at room temperature in the frequency range 1 kHz to 100 kHz. The real part of complex permeability (μ') was calculated using relation, $\mu'=L/L_o$, where L is the self inductance of the sample and L_o is the inductance of the coil of same geometric shape of vacuum. L_o is determined using the relation, $L_o=\mu_oN^2(d_1-d_2)h/\pi(d_1+d_2)$, where μ_o is the permeability of the free space, N is the number of turns (here N=10), d_1 is the outer diameter and d₂ is the inner diameter of the sample and h is the thickness of the sample. The imaginary part of complex permeability (μ'') was determined using the formula $\mu''=\mu'\times D$. The relative quality factor (or Qfactor) was calculated from the Loss factor, tan δ (tan $\delta=\mu''/\mu'$) using the relation Q=1/ tan δ .

3. RESULTS AND DISCUSSIONS

3.1. Lattice parameters, Density and Porosity of the Samples

The X-ray diffraction patterns of the samples with x=0.97, 1 and 1.03 are given in Figure 1. The analysis of XRD patterns indicated that the studied Ni-Zn ferrite samples have spinel cubic structures with a single phase (Amer *et al.*, 2001). The sharp peak reveals that the samples are in good crystalline form. The measured lattice parameter, bulk density, theoretical density and porosity for different samples sintered at 1250°C are given in Table 1. From this table, one can clearly see that the lattice parameter increases with both increasing and decreasing Fe₂O₃ content from the equimolar concentration. This may be related to the cation distribution on the A-sites and on the B-sites because of covalence effects (Smit, 1959; Blasse, 1964). The bulk density decreases with Fe₂O₃ content where as porosity increases with Fe₂O₃ content. It is known that the porosity of ceramic samples results from the two sources, intra-granular porosity and inter-granular porosity (Sattar *et al.*, 2005). Thus, the total porosity could be written as P = P_{intra} + P_{inter}. The intra-granular porosity mainly depends on the grain size (Sattar *et al.*, 2005).



Figure 1: X-ray diffraction patterns for the compositions $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ with x =1, 0.97 and 1.03 sintered at 1250°C

Table 1: Lattice parameter, Molecular mass, Bulk density, Theoretical density and porosity for the samples(Ni_{0.42}Zn_{0.58}O)_x(Fe₂O₃)_{2-x} with x=1, 0.97, 1.03 sintered at 1250°C.

Composition	Lattice Parameter a (Å)	Molecular mass, M	Bulk density $d_B (g/c.c)$	Theoretical density, $d_{th}(g/c.c)$	Porosity P (%)
X=0.97	8.396	240.6966	4.9	5.40	9.73
X=1	8.393	238.2633	5.0	5.35	7.45
X=1.03	8.401	235.8299	5.1	5.28	3.67

3.2. Complex permeability

The real and imaginary permeability spectra of the samples with x=0.97, 1 and 1.03 sintered at 1150°C, 1200°C, 1250°C, 1300°C and 1350°C are `shown in Figures 2(a), 2(b),3(a), 3(b), 4(a) and 4(b) respectively. For all samples, with increasing sintering temperature, T_s the real part of initial permeability, μ' increases at first then decreases.



Figure 2: (a) The real and (b) imaginary permeability spectra for $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ samples with x=0.97 sintered at various temperatures in air



Figure 3: (a) The real and (b) imaginary permeability spectra for $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ samples with x=1 sintered at various temperatures in air



Figure 4: (a) The real and (b) imaginary permeability spectra for $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ samples with x=1.03 sintered at various temperatures in air



Figure 5: The real part of permeability spectra for $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ samples with x=1, 0.97 and 1.03 sintered at 1250°C in air

In case of samples $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ with x=1 and 1.03, μ' increases with increasing T_s up to 1250°C and above 1250°C μ' decreases. The $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ samples with x=1 also show similar behaviour except that μ' increases up to 1300°C. The μ' is almost independent up to 1000 kHz or even at higher frequency. The imaginary part of permeability, μ'' starts rising beyond this frequency which is indicative of dispersion or resonance. As sintering temperature increases, dispersion shifts to the lower frequency range. Figure 5 shows the real permeability spectra for samples with x=1, 0.97 and 1.03 sintered at 1250°C. From this figure it is observed that the initial permeability value increases with decreasing Fe₂O₃ content.

The relative Q factors of all the samples are shown in Figures 6(a, 6(b) and 6(c) respectively. It is observed that relative Q factor decreases with increasing sintering temperature. The relative Q factor is highest for all the samples sintered at 1150°C. The increase of real part of initial permeability with decreasing Fe₂O₃ content may be due to weakening of exchange interaction and reduction of anisotropic energy. Similar result also found by

Mahmud *et al.* There are some liquid phase developing within the samples at higher sintering temperature and the samples contain a number of pores within the grain that results sharp decrease in permeability. At higher sintering temperature pores cannot move so fast as compared with the grain growth. Therefore the pores cannot move to the grain boundary rather they are trapped within the grain. Similar behavior was observed by Guillaud 1957. It is found that the highest permeability of samples with x=1, x=1.03 for sintering temperature 1250°C and with x=0.97 for sintering temperature 1300°C. For a large grain, permeability should increase as it varies proportionally with grain diameter. Large grains are preferred to high permeability values because of the contribution of the domain wall motion (Belled *et al.*, 2000; Nakamura, 1997) studied the magnetic properties of Ni-Zn-Cu ferrites and showed that sintered density and the average grain size increased with increasing sintering temperature.



Figure 6: The variation of Q factor with frequency for samples $(Ni_{0.42}Zn_{0.58}O)_x(Fe_2O_3)_{2-x}$ with (a) x=0.97, (b) x=1 and (c) x=1.03

The permeability can be expressed as $\mu = 1 + \chi_{spin} + \chi_{dw}$, where χ_{spin} is the susceptibility due to the spin and and χ_{dw} is the susceptibility due to the domain wall motion (Globus, 1977). The dispersion of domain wall component depends on the square of the frequency and that of spin rotational component is inversely proportional to the frequency. The domain wall motion contribution starts to decrease at lower frequency and spin rotational component decreases at relatively higher frequencies (Valenzula, 1980). The permeability value for all the samples remain independent of frequency until resonance takes place, above which it starts decreasing sharply with simultaneously increase of imaginary part of the permeability. It is observed that as the permeability started to decrease, the resonance frequency f_r (i.e. the frequency at which μ' show peak) gets higher. This confirms the Snoek's relation stated as μf_r =constant (Snoek, 1948). Since the relative quality factor is highest for all the samples sintered at 1150°C may be used as a good material for inductors or transformers.

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4. CONCLUSIONS

The effects of composition and sintering temperature on the properties of Ni-Zn ferrites were investigated and the essential points of this study can be summarized as follows:

- The samples are characterized by X-ray diffraction, which confirms the single-phase cubic spinel structure.
- Lattice parameter increases both with decreasing and increasing Fe₂O₃ content from the equimolar concentration.
- The Bulk density decreases where as porosity increases with increasing Fe₂O₃ content.
- Initial permeability increases with decreasing Fe₂O₃ content. Real part of initial permeability also increases with increasing sintering temperature up to a specific limit then decreases.
- The relative Q factor decreases with increasing sintering temperature.

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REFERENCES

- Ahmed, M. A., Okasha, N., Salah, L., "Influence of yttrium ions on the magnetic properties of ferrites", J. Magn. Magn. Mater. 264, 241-250, 2003.
- Amer, M. A., Hiti, M. El, "Mössbauer and x-ray studies ferrites for Ni_{0.2}Zn_xMg_{0.8-x}Fe₂O₄ ferrites", J. Magn. Magn. Matter, 234, 118-125, 2001.
- Augustin, C. O., Provakaran, D., Srinivasan, L. K., "Fabrication and characterization of NiCr₂O₄ spinel", J. Mater. Sci. Lett. 12(6), 383-386, 1993.
- Belled, S. S., Watawe, S. C., Shaikh, A. M., Chougule, B. K., Bull. Mater. Ind. Acad Sci., 23, 2000.
- Blasse, G., Philips Res. Suppl. 3, 91, 1964.
- Giannakopoulou, T., Kompotiatis, L., Kontogeorgakos, A., Kordas, G., "Microwave behavior of ferrites prepared via sol-gel method", J. Magn. Magn. Mater. 246, 360-365, 2002.
- Globus, A., Pascard, H., Cagan, V. J., J. Physique 38, C1-163, 1977.

Guillaud, C., Proc. IEE 104B, 165, 1957.

Hilpert S., Ber. Deutsch. Chem. Ges. Bd. 2, 42, 2248, 1909.

Hua Su, Zhang Huaiwu, Tang Xiaoli, Jing Yulan, Liu Yingli, "Effects of composition and sintering temperature on properties of NiZn and NiCuZn ferrites", J. Magn. Magn. Mater. 310, 17-21, 2007.

Jingling S., Li, J., Geliang S., "Effects of La₂O₃ Gd₂O₃ on some properties of Ni-Zn ferrite", J. Magn. Magn. Mater. 250, 20-24, 2002.

Luis Z. F., Gualdi, Alexandre Jose, Eiras, Jose Antonio, Oliverira, Adilson Jesus Aparecido de, Garcia, Ducinei, "Influence of the sintering temperature on the magnetic and electric properties of $NiFe_2O_4$ ferrite", Materials Research, 15(3), 428-433, 2012.

Mahmud, S. T., Hossain, A. K. M. Akther, Hakim, A. K. M. Abdul, Seki, M., Kawai, T., Tabata, H., "Influence of microstructure on the complex permeability of Ni-Zn ferrite", J. Magn. Magn. Mater. 305, 269, 2006.

- Nakamura, T., "Low-temperature sintering of Ni-Zn-Cu ferrite and its permeability spectra", J. Magn. Magn. Mater. 168, 285-291, 1997.
- Olsen, E., Thonstad, J., "Nickel ferrite as anodes in aluminium electrolysis: Part II Material performance and long-term testing", J. Appl. Electrochem. 29(3), 301-311, 1999.
- Robiul I, Rahman Md. Obaidur, Hakim M. A., Saha D. K., Saiduzzaman Saiduzzaman, Noor, Saroaut, Mamun, Md. Al, "Effects of Sintering Temperature on Structural and Magnetic Properties of Ni_{0.55}Zn_{0.45}Fe₂O₄ Ferrites", Materials Sciences and Applications, 3(5), 326-331, 2012.
- Sattar, A. A., Sayed, H. M. El, Shokrofy, K. M. El, Tabey, M. M. El, "Improvement of the Magnetic Properties of Mn-Ni-Zn Ferrite by the Non-magnetic Al³⁺-Ion Substitution", J. Appl. Sci. 5(1) 162, 2005.

- Smit, J., Wign, H. P. J, Ferrites, John Willy & Sons, Pub. Co. New York, 143, 1959.
- Snoek J. L., New developments in Ferromagnetism, 139pp, 1933-1945.
- Snoek, J. L., "Dispersion and absorption in magnetic ferrites at frequencies above one Mc/s", Physica, 14, 207, 1948.

Suzuki Toshiyuki, Tanaka Termitsu, Ikemizu Karou, "High density recording capacity for advanced particulate media", J. Magn. Magn. Mater. 235, 159, 2001.

- Tsutaoka, T., "Frequency dispersion of complex permeability in Mn-Zn and Ni-Zn spinel ferrites and their composite materials", J. Appl. Phys. 93, 2789, 2003.
- Valenzula, R., "A sensitive method for the determination of the Curie temperature in ferrimagnets", Journal of Material Science, 15, 3173-3174, 1980.