

A COMPARATIVE STUDY OF MAGNETIC COLE-COLE LIKE PLOT AND DIELECTRIC COLE-COLE PLOT IN POLYCRYSTALLINE NICKEL ZINC FERRITE

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ABSTRACT

Polycrystalline nickel zinc ferrite of composition $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ has been synthesized by using the conventional ceramic technique. The dielectric constant ϵ' and dielectric loss ϵ'' were found to increase as the sintering temperature increased and to decrease as the frequency increased. Meanwhile, the real part of magnetic permeability μ' and the imaginary part μ'' showed dependence on the microstructure and frequency in a manner very similar to that shown by ϵ' and ϵ'' above. A dielectric cole-cole diagram can be obtained by plotting the dielectric loss ϵ'' against the dielectric constant ϵ' . The cole-cole diagram is generally used for studying the dielectric polarization characteristics by following the variation of dielectric loss ϵ'' with dielectric constant ϵ' . From this work, it is possible to use magnetic cole-cole like plots to deduce magnetization polarization mechanisms associated with magnetic domain wall movement and magnetic spin rotation as it is possible to deduce interfacial polarization and dipolar polarization from dielectric cole-cole plots.

Keywords: Zinc ferrite; dielectric

INTRODUCTION

Polycrystalline Ni-Zn ferrites have been extensively used in a number of electronic devices due to their large permeability in a high frequency, remarkably high resistivity, mechanical hardness, reasonable cost and chemical stability [1]. The high electrical resistivity and good magnetic properties make this ferrite an excellent core material for power transformation in electronic and telecommunication applications [2, 3]. Nickel zinc ferrite is a soft magnetic material having low magnetic coercivity and high resistivity values [4]. The electrical and magnetic properties of Ni-Zn ferrite are sensitive to microstructure (grain size, grain boundary, and pores), which is governed by the preparation process [5]. For several decades, the most common method for preparation of ferrites is by the conventional ceramic technique and many of the

findings on their excellent properties were obtained using samples produced by this technique. Although the magnetic and dielectric properties of Ni-Zn ferrite have reported [6, 7], but there is no report on the comparative studies of magnetic and dielectric properties of Ni-Zn ferrite and no report on magnetic cole-cole like plot and dielectric cole-cole plot. In the present work, the nickel zinc ferrite was prepared by the conventional technique. The effect of sintering temperature and frequency on the magnetic, dielectric and structure of the nickel zinc ferrite samples were discussed.

EXPERIMENTAL METHOD

Samples of $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ was prepared by a conventional ceramic technique. The starting materials used in the preparation were nickel oxide (99.99%), zinc oxide (99.9%) and iron oxide (99.945%). The chemicals were weighed according to the required stoichiometric proportion. The oxides were mixed and grind with mortar and pestle. The mixture was pre-sintered at 1240 °C. The powders were milled using ball milling for 3hour. The powders were mixed with polyvinyl alcohol (PVA) as a binder and zinc stearate as a lubricant and then pressed into toroidal and pellet shape. The samples with toroidal shape for the magnetic measurement and the pellet shape are for dielectric measurement. Finally, the samples were sintered at 1260 °C, 1300 °C and 1340 °C. Their density was measured using Archimedes method. The ferrite phase formation was confirmed by the powder X-ray diffraction technique. The microstructure was examined using JEOL JSM-6400 scanning electron microscope (SEM). The dielectric and magnetic measurements were carried out with an HP 4291A Impedance Analyzer in the frequency range 1 MHz to 1.8 GHz.

RESULTS AND DISCUSSION

The x-ray diffraction patterns of $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ prepared by the conventional ceramic processing sintered at different temperatures are shown in Figure 1. The XRD patterns confirm the formation of the single-phase cubic spinel structure with no extra lines corresponding to any other crystallographic phase or unreacted ingredient. All peaks observed match well with standard pattern of Ni-Zn ferrites. Also, the intense sharp peaks show a good crystalline single phase.

Table 1: density for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$

Sintering temperature (°C)	Density (g/cm ³)
1260	4.30
1300	4.37
1340	4.59

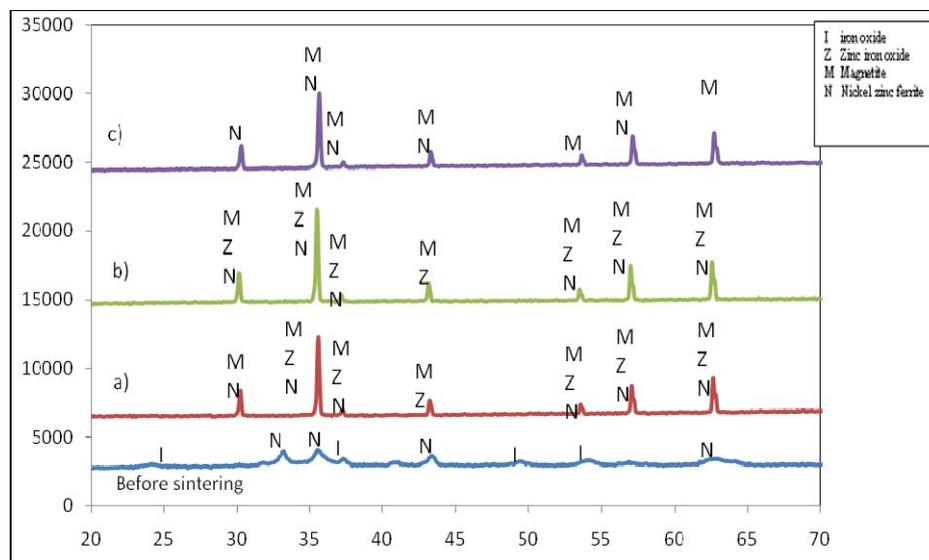


Figure 1: the XRD patterns of $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ sintered at a) 1260 °C, b) 1300 °C, c) 1340 °C prepared by the conventional ceramic technique

Table 1 present the density for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$. From the table, it is observed that the density is increased with sintering temperature. The lowest sintered density was obtained at 1260 °C and the highest sintered density was obtained at 1340 °C. On the other hand, the porosity decreased when the sintering temperature increase. During sintering, the grain size increased, this caused less porosity in the sample and higher density. The sintered density measurement has an important contribution to the permeability. The highest density was obtained for the sample sintered at higher sintering temperature due to the grain growth and low porosity.

The microstructure plays an important role in ferrites properties. The complex permeability ferrites depend not only on the chemical composition but also on the microstructure such as grain size and porosity. Figure 2 presents SEM images of the samples sintered at 1260 °C, 1300 °C, and 1340 °C. SEM images shown that the grain size increases and pores size decreases with sintering temperature. During sintering, three major changes occur because of decomposition or phase transformation [10]. The grain size increase and the pores size and shape changes usually decreases. Samples sintered at 1260 °C shown the lowest average grain size, 2.054 μm while samples sintered at 1340 °C shown highest average size, 2.33 μm . The higher grain size for the sample sintered at 1340 °C may be attributed to the formation of Fe^{2+} ions, which accelerate the growth rate of the grains [8, 9]. When the sintering temperature increased, the average grain size and density increased indicating that the microstructure become more compact, grain boundaries to be diminished and reduced the discontinuity between the grains.

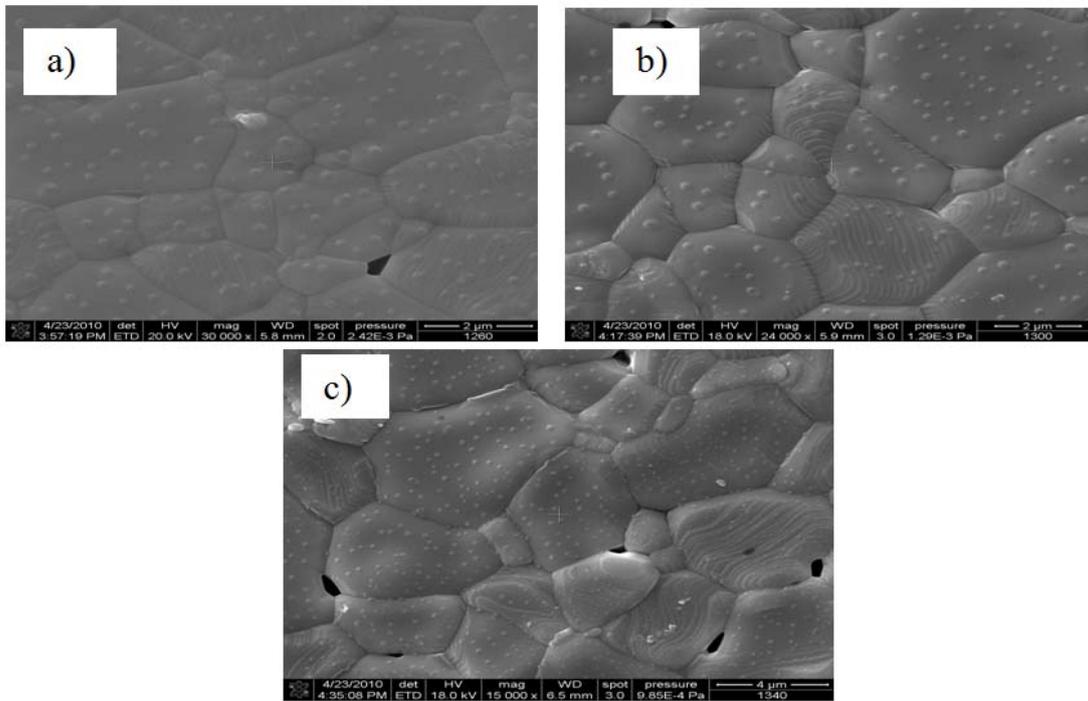


Figure 2: Scanning electron micrographs of $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ sintered at a) 1260 °C, b) 1300 °C, c) 1340 °C

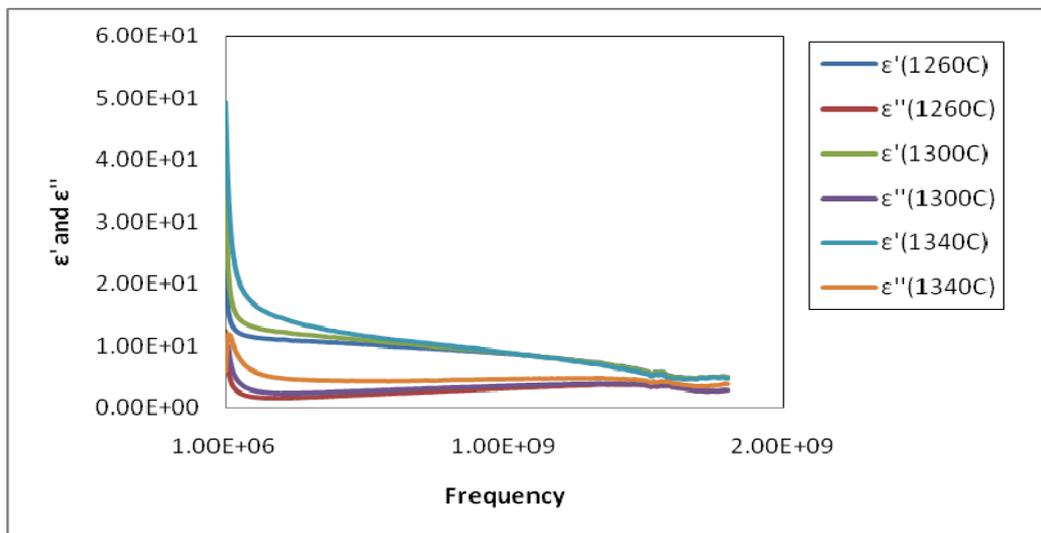


Figure 1: complex permittivity for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ at sintering temperature 1260 °C, 1300 °C and 1340 °C

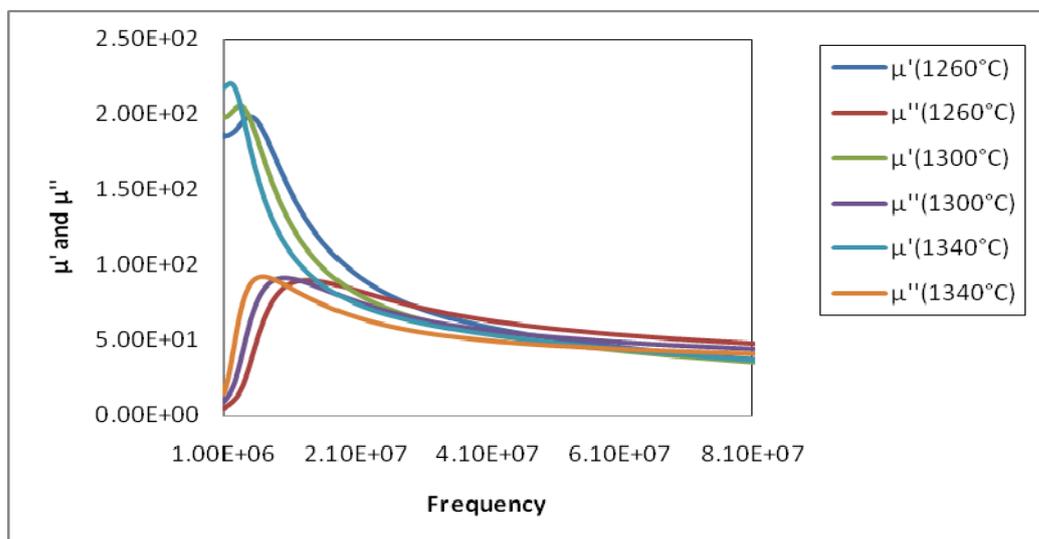


Figure 4: complex permeability for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ at sintering temperature 1260 °C, 1300 °C and 1340 °C

Figure 3 shows the complex permittivity for samples sintered at 1260 °C, 1300 °C and 1340 °C in a frequency range from 1 MHz to 1.8 GHz at room temperature. From the figure above, it is shown that the dielectric constant ϵ' and dielectric loss ϵ'' decrease with increasing frequency and increase with increasing sintering temperature. The polarization in ferrites is similar to the conduction [10]. The conductivity in ferrite occurs as a result of electron hopping between ions of the same element (Fe) that exist in different valences on equivalent lattice sites. Occurrence of ions in different valences is highly dependent on sintering time and temperature. Increasing sintering temperature will increase Zn loss and produce some cation vacancies and unsaturated oxygen. The excess electrons on oxygen bonded with neighbouring Fe^{3+} ions give rise to Fe^{2+} . Thus, electron hopping between the $\text{Fe}^{2+} \longleftrightarrow \text{Fe}^{3+}$ ions is produced. Electron hopping is the main responsible mechanism for conductivity in ferrites [10]. Hence when the sintering temperatures increase, the dielectric constant will also increase due to the increase in electron hopping. When the frequency increases, the dielectric constant decreases because at high frequency electron exchange between Fe^{2+} and Fe^{3+} ions does not follow the applied field, which causes a decrease in the contribution of interfacial polarization. Thus, the dielectric constant decreases at high frequencies. Figure 4 shows the frequency dependence of the real part of permeability (μ') and imaginary part of permeability (μ'') in the range of 1 MHz-1.8 GHz for the samples. The permeability of polycrystalline ferrites is related to two different magnetizations: spin rotation and domain wall motion [10]. The larger grain tends to contain more domain walls. As the number of domain walls increases, the contribution of wall movement in magnetization increases. Thus, the domain wall motion is more dominant compared to spin rotation. The microstructure studies reveal that at higher sintering temperature the average grain size increases. Higher sintering temperature will create bigger grain size and have more domain walls so the real part of permeability (μ') will increase. The imaginary part of

permeability (μ'') also depends on grain size. By increasing the grain size, will have more domain walls. The larger grain increases the eddy current loss. In other words, when the bigger grain sizes the domain walls can move easily and induced larger eddy current [10]. So, when the sintering temperatures increase the loss will also increase due to the increasing eddy current loss.

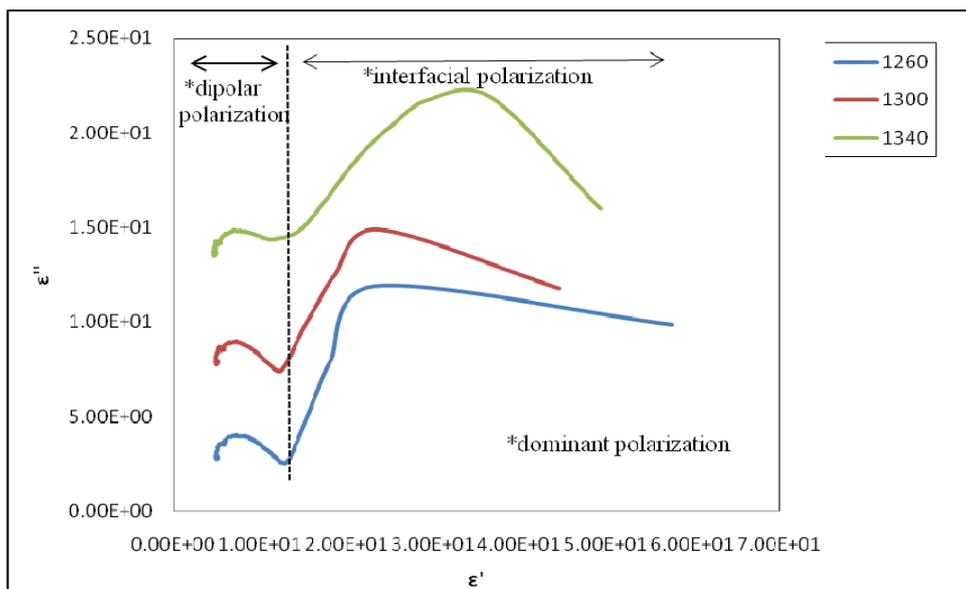


Figure 5: Dielectric cole-cole plot for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ at sintering temperature 1260 °C, 1300 °C and 1340 °C

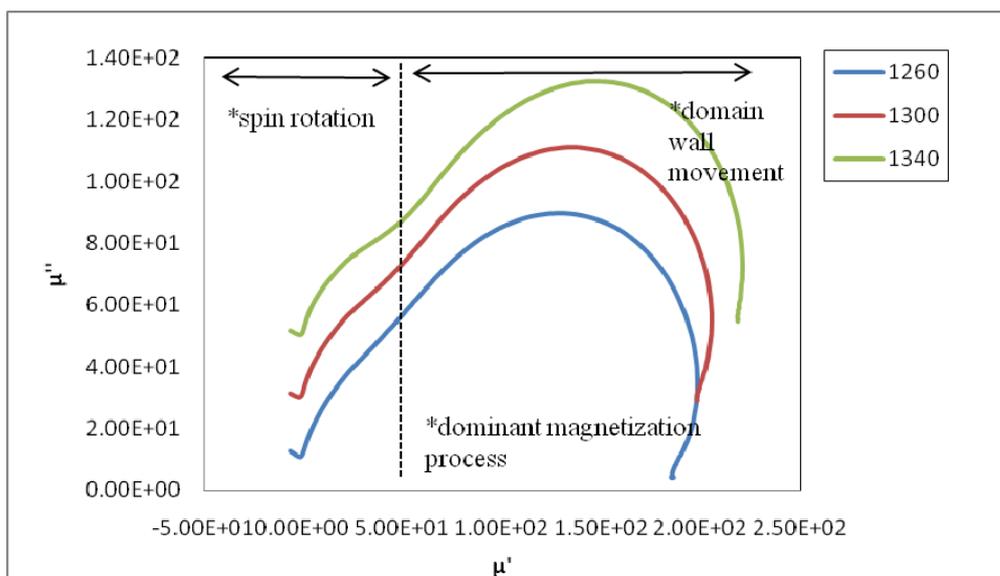


Figure 6: Magnetic cole-cole plot for $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ at sintering temperature 1260 °C, 1300 °C and 1340 °C

Figures 5 and 6 represent the dielectric cole-cole plot and magnetic cole-cole like plot for three sintering temperature 1260°C, 1300°C and 1340°C. Because of the lack information on the magnetic cole-cole like plot so in this paper, the dielectric cole-cole plot will be use as the guided to interpret the magnetic cole-cole like plot. From the Figure 5 and 6, it shows that the shape between this two graph approach to have similar shape and these two kinds of plots reveal dielectric and magnetic polarization. From Figure 5, as the frequency increase from right to the left, the dielectric constant will decrease and dielectric loss will also decrease. At low frequency, the motion of charges in grains is interrupted at grain boundaries and causes localized charge accumulation at the interface which results in the interfacial polarization. The polarization can be influences by the grain size so the bigger grain means easier electron movements and higher contact between the adjacent grains. Thus, the electrons can be easily flow towards the grain boundaries. Therefore the dielectric polarization increases by increasing the sintering temperature. As the frequency increase the electron exchange between Fe^{2+} and Fe^{3+} ions does not follow the applied fields and the charges have enough energy to overcome the barrier at the grain boundary, which cause a decrease in the contribution of interfacial polarization and the dipolar polarization will takes places and resulting in decrease polarization. As the guided from dielectric polarization mechanism, it is possible to deduce magnetization polarization mechanism related to magnetic domain wall movement and magnetic spin rotation. From the Figure 6, as the frequency increase from right to the left, the real part of permeability will decrease and increase with increasing sintering temperature. At low frequency, the magnetization polarization influences by the contribution of domain wall motion. Larger grains contain more domain walls. Increasing in the number of domain walls will increase the contribution of domain wall motion in magnetization. As the frequency increase, the domain wall will continues to move until at certain frequency where the domain wall cannot follow the alternating magnetic field and the contribution of spin will take places.

CONCLUSION

The $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ferrite successfully prepared by the conventional ceramic method and was confirmed by X-ray diffraction (XRD). The SEM images prove that the microstructure, grain size and pores have the contribution to the magnetic and dielectric properties. As the sintering temperature increase the grain size will increase. Bigger grain size can influence to the dielectric polarization and magnetization polarization. From the dielectric cole-cole plot and the magnetic cole-cole like plot, we can say the plots reveal the mechanism of polarization. The dielectric polarization is contributed by the interfacial and dipolar polarization. Meanwhile magnetic cole-cole like plots are probably due to magnetic domain wall movement and magnetic spin rotation

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REFERENCES

- [1] K. Ishino, Y.Narumiya, *Ceram. Bull.* **66** (1987) 1469
- [2] P.I.Slick, in:E.P.Wohlfath(Ed),*Ferrogmagnetic Materials*,vol 2,North-Holland Pub.Co.,Amsterdam,1980,p.196
- [3] T.Abraham, *Am.Ceram.Bull.Soc.Bull.***73** (1994) 62-65
- [4] P.S. Anil Kumar, J.J.Shontri, S.D.Kulkani, C.E.Deshpande, S.K.Date, *Mater.Lett.* **27** (1996) 293-296
- [5] J.J.Shontri, S.D.Kulkani, C.E.Deshpande, A.Mitra, S.R.Sainkar, P.S.Anil Kumar, S.K.Date, *Mater.Chem.Phys.***59** (1999)1-5
- [6] G.Ranga Mohan,D.Ravinder, A.V.Ramana Reddy,B.S.Boyanov, *Materials Letters* **40**(1999) 39-45
- [7] T.Nakamura, *J.Mag.Magn.Mater.* **168** (1997) 285
- [8] T.Kodama.T.Itoh, M.Tabata, Y.Tamaura, *J.Appl.Phys.* **69** (1991) 5915
- [9] Y.H.Han, J.J.Suh, M.S.Shin, S.K.Han, *J.Phys.IV France* **7** (1997) C1-111-C1-112.
- [10] J. Tania, Magnetic, Dielectric and Microstructure Properties of Nickel Zinc Ferrite Prepared by Conventional and Co-precipitation Techniques, (2009) Master Thesis, Universiti Putra Malaysia