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## DIELECTRIC BEHAVIOUR OF NIOBIUM DOPED Ni-Zn FERRITES

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Polycrystalline Ni-Zn ferrites with the formula Ni<sub>0.65</sub> Zn<sub>0.35</sub> Fe<sub>2</sub> O<sub>4</sub> + x Nb<sub>2</sub> O<sub>5</sub> with x values ranging from 0.0 to 1.5 wt% in steps of 0.3 wt% have been prepared by conventional ceramic technique to investigate and understand the influence of pentavalent niobium additions on the dielectric behavior of Ni-Zn ferrites as a function of composition and frequency. Dielectric constants of these ferrites have been found to be of the order of  $10^6$ . The dielectric constant of any sample containing niobium addition is higher than the undoped one. However, the dielectric constant is found to decrease by increasing the concentration of the additive. The dielectric loss has been found to be smaller for the samples containing niobium. The dependence of dielectric constant and the variation of tan  $\delta$  with frequency exhibit normal behavior up to 4 MHz beyond which they experienced dimensional resonances. The results are explained on the basis of Koop's two layer model and Maxwell-Wagner polarization theory.

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### 1. Introduction

Ferrites are very attractive soft magnetic materials for high frequency applications due to their excellent electric and magnetic properties. The properties of ferrites are highly sensitive to composition and impurity levels. Several investigators have tried to improve the basic properties of ferrites by doping the ferrite with ions of having different valence states [1,2,3,4]. Additions of pentavalent ions in Mn-Zn ferrites have been reported to improve the core losses, even though the reasons for such improvements have not been established so far [5,6]. But, because of the higher n-type electrical conductivity of Mn-Zn ferrites, the upper limit for use of these materials in power electronics has been limited to around 1 MHz. Alternately, Ni-Zn ferrites have been termed as good candidates for devices that use operating frequencies beyond of 1 MHz. However, it seems that only little attention has been made to understanding the influence of high valent ions in Ni-Zn ferrites. Thus, an attempt has been made to understand the influence of pentavalent niobium ions on various electrical and magnetic properties of Ni-Zn ferrites. The present paper reports and discusses the dielectric behaviour of such ferrite as a function of composition and frequency.

# 2. Experimental details

Polycrystalline Ni-Zn ferrites with chemical formula  $Ni_{0.65} Zn_{0.35} Fe_2 O_4 + x Nb_2 O_5$  where x values vary from 0.0 to 1.5 wt% in steps of 0.3 wt% have been prepared by conventional ceramic

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technique described elsewhere [7]. Sintering of the samples was carried out at  $1250^{\circ}$  C for 4 hours in air atmosphere followed by natural cooling. X-ray pattern of the basic composition Ni<sub>0.65</sub> Zn<sub>0.35</sub> Fe<sub>2</sub> O<sub>4</sub> confirms single phase cubic spinel structure. Characterization of this composition was done further by measuring lattice constant, Curie temperature and saturation magnetization which compare well with the respective parameters of the same composition reported earlier [8]. Dielectric measurements were made using a HP 4192A Impedance Analyzer.

## 2. Experimental details

The variation of dielectric constant, determined at 10 kHz, versus the concentration of the niobium additive, x, is shown in Fig. 1. Dielectric constants of these ferrites have been found to be of the order of 10<sup>6</sup>. All the doped samples have resulted in higher dielectric constants if compared to undoped sample. However, the dielectric constant is found to decrease with the increasing of the concentration of niobium.



Fig. 1. Variation of dielectric constant of  $Ni_{0.65} Zn_{0.35} Fe_2 O_4 + x Nb_2 O_5$  ferrite versus the concentration of niobium addition (at 10 kHz).

Variation of dielectric loss tangent (tan  $\delta$ ) versus the concentration of niobium addition is shown in Fig. 2. The dielectric loss is found to be small for doped samples compared to the undoped sample and it experienced weak variation with the amount of addition concentration in the studied range.



Fig. 2. Variation of dielectric loss of  $Ni_{0.65}$  Zn<sub>0.35</sub> Fe<sub>2</sub> O<sub>4</sub> + x Nb<sub>2</sub> O<sub>5</sub> ferrite versus the concentration of niobium addition (at 1 MHz).

The variation of dielectric constant versus AC field frequency, determined at different frequencies in the range 10 kHz-10 MHz for the sample added with 0.3 wt% Nb<sub>2</sub> O<sub>5</sub> is shown in Fig. 3. The dielectric constant has been observed to decrease with increasing frequency but the dispersion in all the cases showed an unusual dielectric behavior similar to that observed by Brockman et al. [9] in the lower megahertz region. The dielectric constants of all the samples remain of the order of  $10^4$  in the lower megahertz region while showing flux reversal accompanying dielectric peaks at around 4 MHz.



Fig. 3. Variation of dielectric constant versus frequency for the sample added with 0.3 wt% Nb<sub>2</sub>O<sub>5</sub>.

The variation of tan  $\delta$  versus frequency for the sample doped with 0.3 wt% Nb<sub>2</sub>O<sub>5</sub> is shown in Fig. 4. The dielectric loss is minimal up to 2 MHz and thereafter it increases and exhibits a peak at 4 MHz. All the samples show similar dielectric loss peaks in the MHz region.



Fig. 4. Variation of dielectric loss versus frequency for the sample added with 0.3 wt% Nb<sub>2</sub>O<sub>5</sub>.

The dielectric behavior in ferrites can be explained on the basis of the assumption that the mechanism of dielectric polarization is similar to that of conduction. Many scientists established a strong correlation between the conduction mechanism and dielectric constant of ferrites [10,11]. In the studies, they have explained the dielectric behavior on the basis of number of available  $Fe^{2+}$  ions in the octahedral sites. The electronic exchange such as  $Fe^{2+} \Leftrightarrow Fe^{3+}$  results in a local displacement of

electrons, which determines the polarization, and thus the dielectric constant of ferrites. The pentavalent niobium ions are expected to enter in the spinel lattice and the accompanying changes could be resulted are in microstructure and conduction mechanism. It is reported that pentavalent additions by entering into lattice modify the valence states of iron while producing more  $Fe^{2+}$  ions to maintain the charge balance [12]. The observed higher values of dielectric constants for the niobium added samples are in accordance with this. However, subsequent additions influence the microstructure and decrease the values of the dielectric constants.

The dielectric dispersion curve can be explained on the basis of Koop's two layer model and Maxwell-Wagner polarization theory. To interpret the frequency response of dielectric constant in ferrite materials, Koops [13] suggested a theory in which relatively good conducting grains and insulating grain boundary layers of ferrite material can be represented with the behavior of an inhomogeneous dielectric structure, as discussed by Maxwell [14] and Wagner [15]. Since an assembly of space charge carriers in the inhomogeneous dielectric structure described requires finite time to line up their axes parallel to an alternating electric field, the dielectric constant naturally decreases, if the frequency of the reversal field increases. This is in agreement with the observed dielectric dispersion up to 2 MHz.

#### **3. Conclusions**

The results indicate that the pentavalent niobium additions in Ni-Zn ferrites provide higher dielectric constants and lower dielectric losses. The frequency response is normal up to 2 MHz followed by flux reversal accompanying dielectric peaks around 4 MHz. Microstructural and structural modifications due to the addition are used to explain the results. In addition, chemical analyses to estimate the  $Fe^{2+}$  concentration and micrographs to understand the grain and grain boundary structures on these samples would consolidate the conclusions further.

#### References

- [1] A. Lakshman, K. H. Rao, R. G. Mendiratta, J. Magn. Magn. Maters. 250, 92 (2002).
- [2] A. R. Das, V. S. Ananthan, D. C. Khan, J. Appl. Phys. 57, 4189 (1985).
- [3] J. Neamtu, M. I. Toacsen, D. Barb, J. Phys. IV France Suppl. C1 7, 79 (1997).
- [4] O. F. Caltun, L. Spinu, A. Stancu, IEEE Trans. Magn. 37, 2353 (2001)
- [5] B. Ramesh, J. Magn. Soc. Jpn. 22, S32 (1998)
- [6] Y. Tamamoto, A. Makino, T. Nikaidou, J. Phys. France Suppl. C1 7, 123 (1997).
- [7] B. Parvatheeswara Rao, K. H. Rao, J. Appl. Phys. 80, 6804 (1996).
- [8] B. Parvatheeswara Rao, P. S. V. Subba Rao, K. H. Rao, IEEE Trans. Magn. 33, 4454 (1997).
- [9] F. W. Brockman, P. H. Dowling, W. G. Steneck, Phys. Rev. 77, 85 (1950).
- [10] K. Iwauchi, Jpn. J. Appl. Phys. 10, 1520 (1971).
- [11] L. I. Rabinkin, Z. I. Novikova, in Ferrites, Minsk, 1960, p. 146.
- [12] G. C. Jain, B. K. Das, R. B. Tripathi, R. Narayan, IEEE Trans. Magn. 18, 776 (1982).
- [13] C. G. Koops, Phys. Rev. 83, 121 (1951).
- [14] J. C. Maxwell, A Treatise on Electricity and Magnetism, Clarendon Press, Oxford (1982), p. 328.
- [15] K.W. Wagner, Ann. Physik 40, 817 (1983).