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Effect of Si on the dielectric properties of $Ni_x Zn_{1-x} Fe_2O_4$ as a function of composition and frequency

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Nickel-zinc ferrites of composition $Ni_xZn_{1-x}Fe_2O_4$ (*x*=0.66, 0.77, 0.88, 0.99) were prepared by the usual ceramic technique. The dielectric properties have been studied as a function of composition and frequency. It is observed that the values of dielectric constant $\in'$ and dielectric loss factor tanδ for samples prepared in the present work are much lower and of reduced price than those commercially obtained Ni-Zn ferrites. These low values are attributed to the presence of Si in Fe_2O_3 powder. It has been found that the ceramic grain growth was suppressed by Si, which results in a decrease in the dielectric constant and dielectric loss factor. The dielectric constant $\in'$ and loss tangent tan δ also decrease as the frequency of applied *ac* electric field increases.

Keywords: Ferrites, Silicon, Semiconductor materials, Dielectric properties

1 Introduction

Nickel-zinc ferrite is a versatile technological material due to its low-eddy current losses and high-frequency applications. It is well-known fact that the properties of ferrite materials are strongly influenced by their composition. The selection of an appropriate composition and additive is, therefore, the key action to obtain improved quality ferrites[1-4]. Various substitutions have been incorporated to achieve desired electrical, mechanical and magnetic properties. Dielectric behaviour is one of the most important properties of ferrites which markedly depend on the preparation conditions, type and quantity of additives. Many researchers[5-9] studied the influence of composition and sintering conditions on the dielectric properties of different ferrite systems. It was, therefore, felt appropriate to use iron oxide with 0.5 wt% of Si additive to prepare Ni-Zn ferrites and analyze the effect on dielectric properties. The main objective of this study was to produce commercial ferrites by using an economically viable manufacturing process and selecting technically feasible compositions.

2 Experimental Details

$Ni_xZn_{1-x}Fe_2O_4$ ferrites (x = 0.66, 0.77, 0.88 and 0.99) were prepared in polycrystalline form by high temperature solid-state reaction method. The X-ray diffraction (XRD) patterns confirm the phases[10]. In the present work, the dielectric properties of $Ni_xZn_{1-x}Fe_2O_4$ ferrites have been measured by using capacitance bridge method. Air-dried silver epoxy electrical contacts were deposited on the flat surfaces of sintered pellets and the dielectric measurements were carried out in frequency area 100, 300, 500, 700 and 900 kHz. This was repeated for different temperatures from 25ºC (room temperature) to 180ºC.

3 Results and Discussion

The dielectric properties of polycrystalline materials are, generally, determined by a combination of various factors like method of preparation, sintering temperature, grain size, additive used, the ratio of Fe^{3+} / Fe^{2+} ions and *ac* conductivity[2,11]. In the present work, low cost Fe_2O_3 with 0.5 wt% of Si as an additive has been selected to be introduced into this system to study the effect of dielectric constant and dielectric loss tangent by using the formula:

$$\in' = Cd/\in_o A \quad \ldots(1)$$

where C is the measured capacitance, d the thickness of the sample, A the area of the capacitor's plate and $\in_o$ is the permittivity of free space (i.e. 8.85×10^{-12} F/m).

The angle δ between the vector for the amplitude of the total current and that for the amplitude of charging current is called the loss angle and is less than 90º. The tangent of this angle is the loss tangent (dissipation factor) which is used as a measure of dielectric loss and given as:

$$\tan\delta = \epsilon''/\epsilon' \quad \ldots(2)$$

where ϵ' is the measured dielectric constant of dielectric material in the capacitor and it is the real part of complex permittivity and ϵ'' is the loss factor being the imaginary part of complex permittivity.

3.1 Dependence of dielectric properties on composition

Dielectric dispersion in ferrites can be explained on the basis of space charge polarization, which is a result of the presence of higher conductivity phases (grains) in the insulating matrix (grain boundaries) of a dielectric causing localized accumulation of charge under the effect of an electric field. The stoichiometry composition is an important requirement for obtaining low dielectric constant and dielectric loss[6]. The effect of Si containing iron oxide influences the dielectric behaviour of ferrites. The electric hopping between Fe^{2+} and Fe^{3+} ions (*n*-type) and hole hopping between Ni^{3+} and Ni^{2+} (*p*-type semiconductor) on B-sties are responsible for electric conduction and dielectric polarization[11-13]. The values of dielectric constant ϵ' and dielectric loss tanδ as shown in Figs 1 and 2, are found to decrease with increase in Ni content *x* as polarization in ferrites has largely been attributed to the presence of Fe^{2+} ions which give rise to heterogeneous spinal structure. Since Fe^{2+} ions are easily polarizable, the larger the number of Fe^{2+} ions the higher would be the dielectric constant[14-16]. But in the present paper, the presence of Si in iron oxide powder lowers the values of dielectric constant ϵ' and dielectric loss tan δ. Addition of Si has a great influence both on magnetic and dielectric properties of the sintered bodies[14] and has low dielectric constant. As a result, the dielectric constant of the composition with Si addition should decrease as observed. It was found that Si suppressed the grain growth and caused the grain size to decrease, so that the grain boundary was enhanced and this also contributed to the decrease of the dielectric constant and dielectric loss.

3.2 Variation of dielectric constant ϵ' and dielectric loss tangent tanδ with frequency

The variations of the dielectric constant and dielectric loss tangent as a function of frequency for $Ni_xZn_{1-x}Fe_2O_4$ ferrites at room temperature are shown in Figs 3 and 4. It is shown that both the dielectric

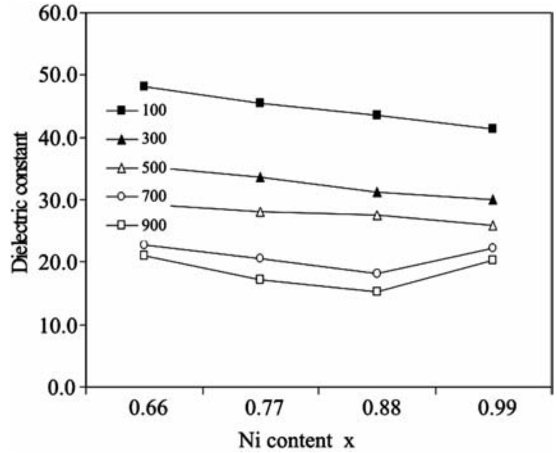

Fig. 1 — Plot of dielectric constant (ϵ') versus Ni concentration, *x* for Ni-Zn ferrite

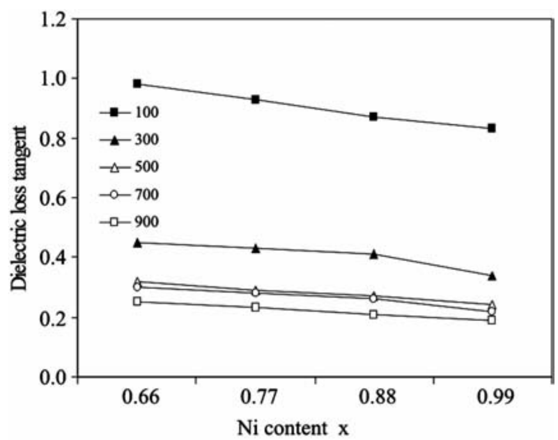

Fig. 2 — Plot of dielectric loss (tanδ) versus Ni concentration, *x* for Ni-Zn ferrite

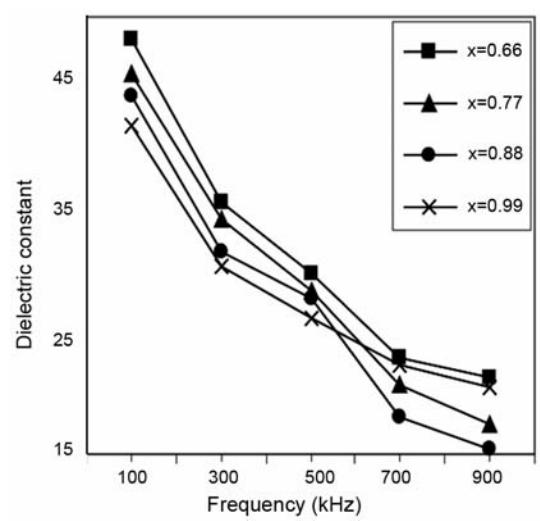

Fig. 3 — Plot of dielectric constant (ϵ') versus frequency for Ni-Zn ferrite

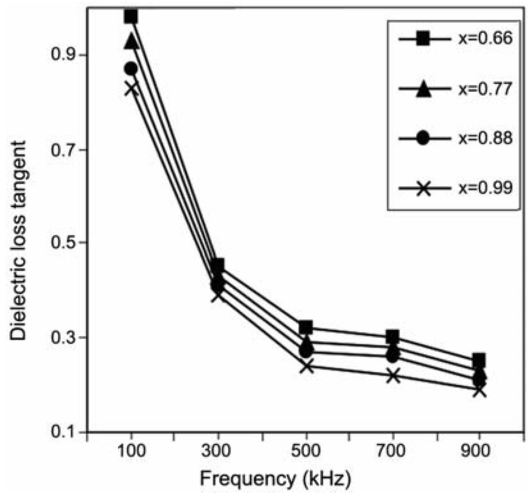

Fig. 4 — Plot of dielectric loss (tanδ) versus versus frequency for Ni-Zn ferrite

constant and dielectric loss decrease as the frequency increases and is the normal behaviour of ferrites[7,11,16]. The frequency of the externally applied field increases gradually, though the number of ferrous ions present in the ferrite material also increases, as a result, the dielectric constant decreases. The reduction occurs because beyond a certain frequency of the externally applied electric field, the electronic exchange between ferrous and ferric ions, i.e. $Fe^{2+} \Leftrightarrow Fe^{3+}$ cannot follow the alternating field[16,17]. Whereas the decrease of dielectric loss by increasing frequency is due to the reason that there is a strong correlation between the conduction mechanism and dielectric behaviour of ferrites. The conduction mechanism in ferrite is considered due to hopping of electrons and holes in *n* and *p*-type, respectively. As such, when the hopping frequency is nearly equal to that of the externally applied electric field, a maximum loss may be observed. The low values of $\in'$ and tanδ as the frequency increases could be explained on the basis of Maxwell-Wagner model[18].

4 Conclusions

$Ni_xZn_{1-x}Fe_2O_4$ ferrites were prepared using low cost iron oxide with 0.5 wt% of Si as an additive. This Si additive has great influence on dielectric properties of sintered bodies. The ceramic grain growth was suppressed by this addition resulting in a decrease in $\in'$ and tan δ in ferrite composition. Dielectric constant and loss also decrease with increasing frequency.

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