Dielectric behavior of Cu–Zn ferrites with Si additive

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(Received 14 September 2013; revised manuscript received 8 November 2013; published online 25 March 2014)

Since ferrites are highly sensitive to the additives present in or added to them, extensive work, to improve the properties of basic ferrites, has been carried out on these aspects. The present paper reports the effects of composition, frequency, and temperature on the dielectric behavior of a series of $Cu_xZn_{1-x}Fe_2O_4$ ferrite samples prepared by the usual ceramic technique. In order to improve the properties of the samples, low cost Fe_2O_3 having 0.5 wt.% Si as an additive is selected to introduce into the system. The dielectric constant increases by increasing the Cu content, as the electron exchange of $Cu^{2+} <=> Cu^{+}$ is responsible for the conduction and the polarization. However, the addition of Si could decrease the dielectric constant as it suppresses the ceramic grain growth and promotes the quality factor at higher frequencies. Dielectric constant ε' and loss tangent $\tan\delta$ of the mixed Cu–Zn ferrite decrease with increasing frequency, attributed to the Maxwell–Wagner polarization, which increases as the temperature increases.

Keywords: Cu–Zn ferrite, dielectric behavior, Si additive

PACS: 75.30.Hx, 75.47.Lx, 75.50.Cc **DOI:** 10.1088/1674-1056/23/5/057502

1. Introduction

When ferrite compositions are modified with varying amounts of impurities, the properties of the ferrite are affected. By studying the substituted Cu–Zn system, it was found that the rare earth oxide plays an important role.[1–5] Limited research was reported in literature for the subject. However, in view of the enormous advantages obtained by the addition of Si, the formation of Cu–Zn ferrite needs the attention of researchers to explore further the area of research.

The present work reports the influence of Si, containing low-cost iron oxide, on the dielectric properties of the Cu–Zn ferrite system.

2. Experimental details

$Cu_xZn_{1-x}Fe_2O_4$ ferrites ($x = 0.66$, 0.77, 0.88, 0.99) were prepared in polycrystalline form by the high temperature solid-state reaction method. The diffraction patterns of the $Cu_xZn_{1-x}Fe_2O_4$ ferrites verified the well defined major phase of Cu–Zn ferrite, with a few peaks of zinc silicate (Zn_2SiO_4) as an additive. The details of the methods of preparation and structural study were given in an earlier publication.[6] In the present work, the dielectric properties of $Cu_xZn_{1-x}Fe_2O_4$ ferrites were measured by using the capacitance bridge method. Air-dried silver epoxy electrical contacts were deposited on the flat surfaces of the sintered pellets, and the dielectric measurements were carried out in frequencies of 100 kHz, 300 kHz, 500 kHz, 700 kHz, and 900 kHz. This was repeated for different temperatures from 25 °C (room temperature) to 180 °C.

3. Results and discussion

The dielectric properties of polycrystalline materials are generally determined by a combination of various factors such as the method of preparation, the sintering temperature, the grain size, the additive used, the ratio of Fe^{3+}/Fe^{2+} ions, and the alerting-current (ac) conductivity.[7,8] In the present work, low cost Fe_2O_3 with 0.5 wt.% of Si as an additive has been introduced into the system to study the effects on the dielectric constant and the dielectric loss tangent with the formula

$$\varepsilon' = \frac{Cd}{\varepsilon_0 A}, \tag{1}$$

where C is the measured capacitance, d is the thickness of the sample, A is the area of the capacitor's plate, and ε_0 is the permittivity of the vacumn (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 the charging current is called the loss angle and is less than 90°. The tangent of this angle is the loss tangent (dissipation factor) which is used as a measure of the dielectric loss and given as,

$$\tan\delta = \varepsilon''/\varepsilon', \tag{2}$$

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

3.1. Dependence of dielectric properties on composition

The composition dependencies of ε' and the dissipation factor $\tan\delta$ are shown in Figs. 1 and 2 and can be explained by using the assumption that the mechanism of the dielectric polarization is similar to that in the conduction process.[9] It has been concluded that the electron exchange between Fe^{2+}

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and Fe^{3+} results in the local displacement of charge in the direction of an electric field, which is responsible for the polarization found in ferrites.[10,11] The magnitude of exchange depends on the concentration of Fe^{2+}/Fe^{3+} ion pairs present on the *B* site.[12] For the present ferrite, the Cu^{2+} ions are the majority ions and the probability of the formation of Cu^{2+} ions increases. Hence, the exchange mechanism between Cu^{2+} and Cu^{2+} is more probable. Therefore, the electron exchange of $Cu^{2+} <=> Cu^{2+}$ is responsible for conduction and the polarization, as the incorporation of Cu may increase the concentration of Fe^{3+}/Fe^{3+} ion pairs due to the oxidation from Cu^{+} to Cu^{2+}. Accordingly, the conductivity and the dielectric constants increase with increasing Cu content. However, in our case, the presence of Si in iron oxide powder lowers the dielectric constant ε' and the dielectric loss $\tan\delta$. The addition of Si has a great influence on both the magnetic and dielectric properties of the sintered bodies[13] and leads to a low dielectric constant. As a result, the dielectric constant of the composition with Si addition should decrease as observed. It was found that Si suppresses the grain growth and causes the grain size to decrease, so the grain boundary is enhanced, and this also contributes to the decreases of the dielectric constant and the dielectric loss and promotes the quality factor at higher frequencies.[14]

3.2. Variation of dielectric constant and dielectric loss tangent with frequency

The variations of the dielectric constant and the dielectric loss factor with frequency are illustrated in Figs. 3 and 4, respectively. Both ε' and $\tan\delta$ decrease as the frequency increases, which is a normal dielectric behavior in ferrites.[15–18] The decreases in ε' and $\tan\delta$ take place when the jumping frequency of the electric charge carriers cannot follow the alternation of the applied ac electric field beyond a certain critical frequency.[7] The decrease of the dielectric loss with increasing frequency is due to the fact that there is a strong correlation between the conduction mechanism and the dielectric behavior of ferrites. The conduction mechanism in ferrites is considered to be the hopping of electrons and holes in n-type and p-type, respectively. So, 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 ε' and $\tan\delta$ as the frequency increases could be explained on the basis of the Maxwell–Wagner model. The Maxwell–Wagner polarization in these ferrites is directly related to the Verway type conduction with jumping of electrons from Fe^{2+} to Fe^{3+}. This clearly shows that electronic and ionic conductions significantly contribute in the polarization process. At a lower frequency, the grain boundaries of lower

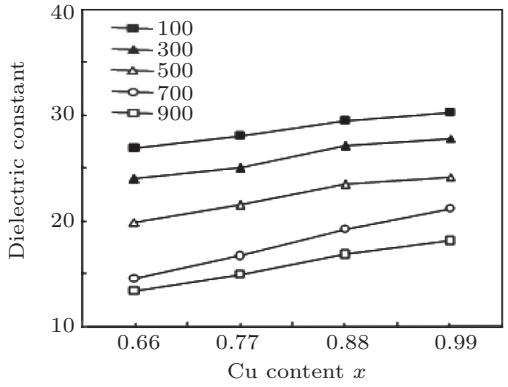


Fig. 1. Dielectric constant ε' versus Cu concentration x for Cu–Zn ferrite.

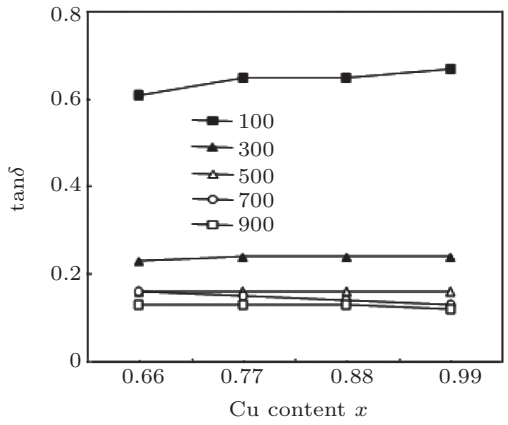


Fig. 2. Dielectric loss $\tan\delta$ versus Cu concentration x for Cu–Zn ferrite.

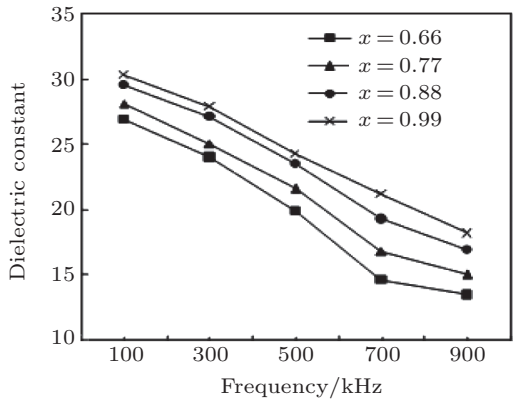


Fig. 3. Dielectric constant ε' versus frequency for Cu–Zn ferrite.

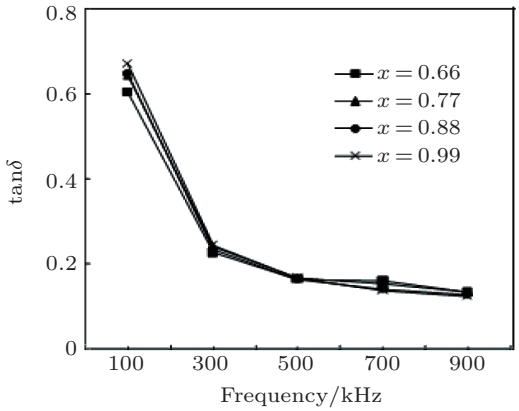


Fig. 4. Dielectric loss $\tan\delta$ versus frequency for Cu–Zn ferrite.

conductivity are effective, while at a higher frequency, the ferrite grains of moderate conductivity are prominent. It was reported in the literature that the dielectric constant is directly proportional to the grain size.[12] In our composition, the grain size decreases with the increase in Si, this leads to the decrease in the dielectric constant.

3.3. Variation of dielectric constant and dielectric loss tangent with temperature

The dielectric constant ε' and dielectric loss tangent $\tan\delta$ are found to depend on the variations of external factors such as temperature. The dependences of ε' and $\tan\delta$ on the temperature of the investigated system are shown in Figs. 5 and 6, respectively, for $x = 0.66$–0.99. It has been found that both ε' and $\tan\delta$ show an appreciable increase when the temperature is increased, the temperature dependencies of ε' and $\tan\delta$ may be due to the polarization effect.

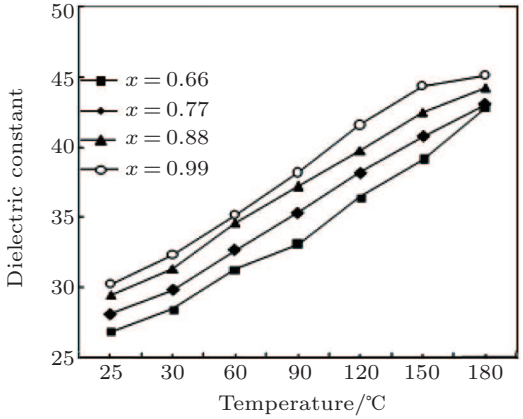


Fig. 5. Dielectric constant ε' versus temperature for Cu–Zn ferrite.

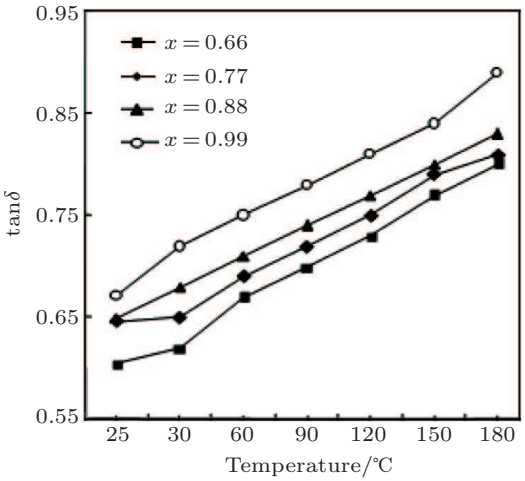


Fig. 6. Dielectric loss $\tan\delta$ versus temperature for Cu–Zn ferrite.

It is assumed that the mechanism of dielectric polarization is similar to that of conduction and it is concluded that the electron exchange interaction results in a local displacement of the electron in the direction of an electric field, which determines the polarization of the ferrites. The space charge polarization is governed by the number of space charge carriers. With the rise in temperature, the number of carriers increases, resulting in an enhanced built up space charge polarization and as a result increases in dielectric properties.

4. Conclusion

The main objective of this study is to produce commercial ferrites by using an economically viable manufacturing process and selecting technically feasible compositions. Therefore we have selected low cost iron oxide with 0.5 wt.% of Si as an additive. This Si additive has great influences on the dielectric properties of the sintered bodies. The ceramic grain growth is suppressed by this addition, resulting in decreases in ε' and $\tan\delta$ in the ferrite composition. The dielectric constant and the loss also decrease with increasing frequency and increase with increasing temperature.

Acknowledgment

Deep thanks to Professor Dr. Ghazanfar Abbas for helpful discussion and every possible support.

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