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DIELECTRIC BEHAVIOUR IN $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ FERRITES

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A series of samples of the type $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ (x = 0, 0.1, 0.2, 0.3, 0.4 and 0.5) were prepared by means of a ceramic technique. The existence of a single phase was confirmed in X-ray studies. The dielectric constant K and dielectric loss D were measured at different temperatures at a frequency of 1 kHz. The pronounced increase in the dielectric constant at the transition temperature is due to the phase transformation from the ferrimagnetic to the paramagnetic state. The decrease in the dielectric loss with temperature is due to the reduction in the relaxation time of the hopping electrons and holes. The Tawfik constant was calculated in the ferrimagnetic state and was found to be correlated to the dielectric constant.

The polycrystalline ferrites are good dielectric materials. The dielectric properties of ferrites depend on many factors, including the sintering temperature and the preparation conditions. Ferrites have low conductivity, which is one of the reasons for their microwave applications.

Many investigators have studied the dielectric properties of Ni–Zn ferrites [1], Mn–Zn ferrites [2, 3], Mn–Mg ferrites [4] and Li–Ni ferrites [5]. The measurements [4, 5] were carried out at room temperature in the frequency range 10^2 to 10^5 Hz. It was found that ε, ε and $\tan\delta$ decrease with increasing Ni content in Li–Ni ferrites [5]. The value of the measured dielectric constant ε was found to be of the order of 10^5. These results were explained on the basis of the model published in [5]. The same value of the dielectric constant was found for Mn–Zn ferrites [6]. The reported value for the dielectric constant at 10^3 Hz was 17, while the a.c. conductivity was $10.2 \cdot 10^{-9}\ \Omega^{-1} \cdot cm^{-1}$ for $Co_{0.49}Zn_{0.51}Fe_2O_4$ [7].

The aim of our investigation was to study the effect of Cu addition on the dielectric behaviour of Ni–Zn ferrites.

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Experimental

A series of samples in the system $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ (x = 0, 0.1, 0.2, 0.3, 0.4 and 0.5) were prepared by means of the usual ceramic technique. The samples were pressed into discs, sintered at 1200° for 8 h and slowly cooled by switching off the furnace. X–ray diffraction showed that the prepared samples were cubic spinels (single phase). The discs were polished and covered by silver paste as a contact material. The dielectric constant K and the dielectric loss D of the prepared samples were measured at 1 kHz by using a PLCG bridge of type BM 591.

Results and discussion

Temperature dependence of dielectric constant of $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$

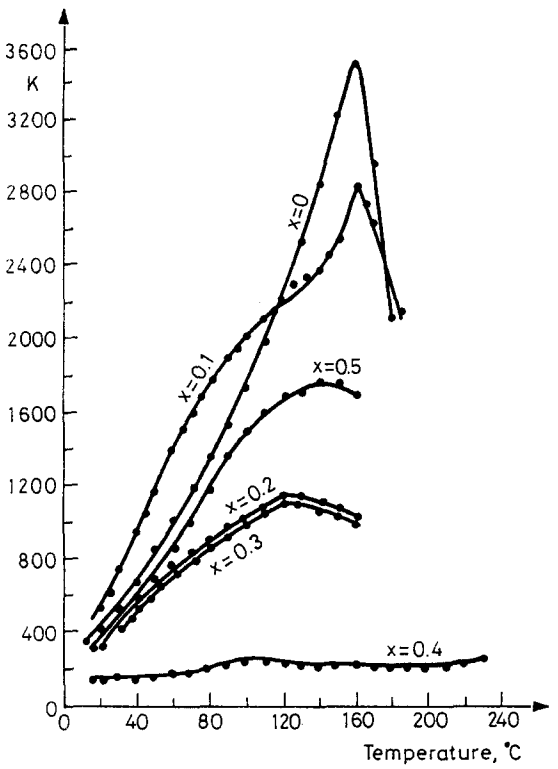

Fig. 1 Variation of dielectric constant with temperature for $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ ferrites

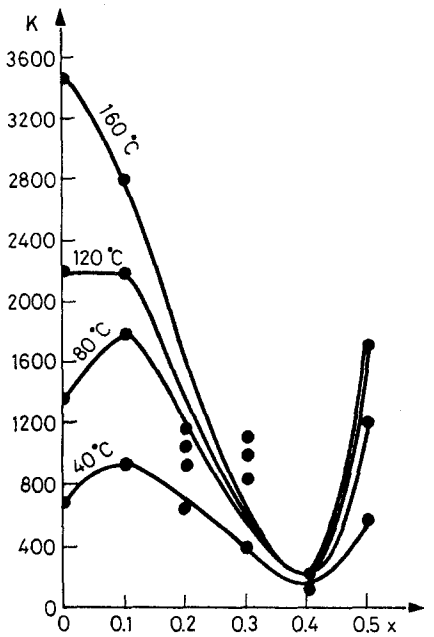

Fig. 2 Variation of dielectric constant as a function of x at different temperatures

The variation of dielectric constant K with temperature for ferrites $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ is shown in Fig. 1. It can be seen as a general trend for all the samples that K increases with increasing temperature up to 160° for $x = 0$. This temperature is shifted to lower values with increasing x, and K then decreases with increasing temperature for all samples. The pronounced increase in the dielectric constant of our samples is due to the transition of the crystal structure from the ferrimagnetic to paramagnetic state. This transition of the crystal structure increases the number of iron(II) ions at the octahedral sites which take part in the electron exchange interaction $Fe^{2+} \rightleftarrows Fe^{3+}$, and hence are responsible for the maximum polarization, as previously confirmed by DTA [8].

Composition dependence of dielectric constant

Figure 2 shows the effect of x on the dielectric constant K. It was observed that K decreases with increasing x, to reach a minimum value at $x = 0.4$, and then increases for higher Cu addition. This can be explained on the basis of a dielectric polarization mechanism. Previous workers [9] explained the composition dependence of the dielectric constant by using the

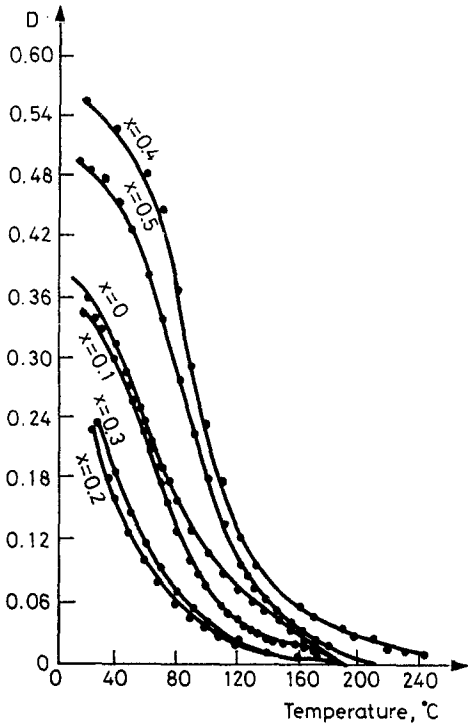

Fig. 3 Variation of dielectric loss D as a function of temperature for $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ ferrites

assumption [10] that the mechanism of dielectric polarization is similar to that of conduction. They observed that the electron exchange between Fe^{2+} and Fe^{3+} results in a local displacement of electrons in the direction of the electric field which determines the polarization of the ferrites. The reduction in the dielectric constant with increasing Cu addition is due to the decrease in the number of iron(II) ions at the octahedral sites which play a dominant role in the dielectric polarization. This reduction is due to the creation of lattice vacancies [8]. For additions higher than 0.4, the increase in the dielectric constant is attributed to the further creation of iron(II) ions caused by the substitution of Cu^{2+} instead of Fe^{3+}. The shift in the transition temperature to lower values as in Fig. 1 confirmed this substitution.

Variation of dielectric loss as a function of temperature

Figure 3 shows the effect of temperature on the dielectric loss D. It is noticed that the dielectic loss decreases with increasing temperature for all

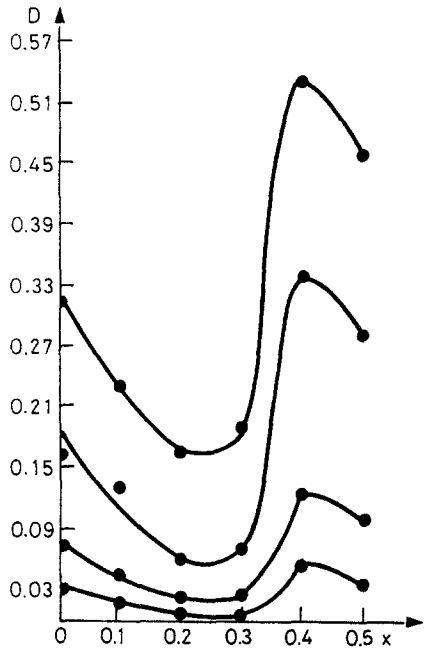

Fig. 4 Variation of dielectric loss D as a function of x at different temperatures

the prepared samples. The dielectric loss mechanism for ferrites $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ has been explained in terms of electron hopping between Fe^{2+} and Fe^{3+} over the octahedral sites for n–type ferrites and in terms of hole hopping for p–type ferrites. The increase of temperature increases the hopping frequency and then decreases the relaxation time. Therefore, the reduction in the relaxation time reduces the dielectric loss.

Variation in dielectric loss (D) with Cu addition (x)

The effect of x on Ni–Zn ferrites is shown in Fig. 4. The dielectric loss D decreases to a minimum at $x = 0.3$ and then increases roughly up to $x = 0.4$. It can justifiably be assumed that the jumping or hopping frequency for Ni–Zn ferrites is far from that of the external electric field. Therefore, for higher additions more oxygen vacancies are generated, which may approximate the hopping frequency with the frequency of the applied field (1 kHz), giving rise to the dielectric loss (D).

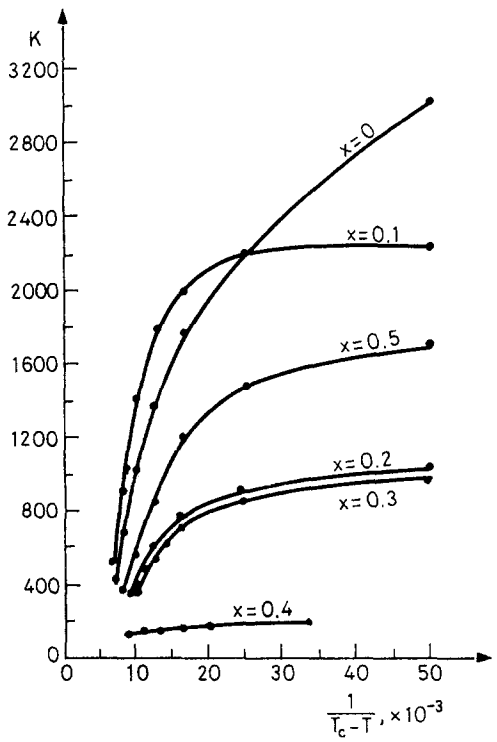

Fig. 5 Variation of dielectric constant with 1 /*T-Tc* for different *x* near the transition temperature in the ferrimagnetic state

Dependence of the dielectric constant on $1/T_c$–T

The dependence of K on $1/T_c$–T for the different compositions is shown in Fig. 5. It is noticed that the dielectric constant increases roughly with $1/T_c$–T in the ferrimagnetic state, where T_c is the transition temperature and T is the measured temperature. From the slope A of the line near the transition temperature, which is plotted against the composition x in Fig. 6, a minimum in A was obtained at $x = 0.3$, where A is the Tawfik constant and is determined from Fig. 5. An explanation is proposed similar to that for the composition dependence of the dielectric constant of $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$. A strong correlation between the dielectric behaviour and Tawfik constant A is concluded in the ferrimagnetic state.

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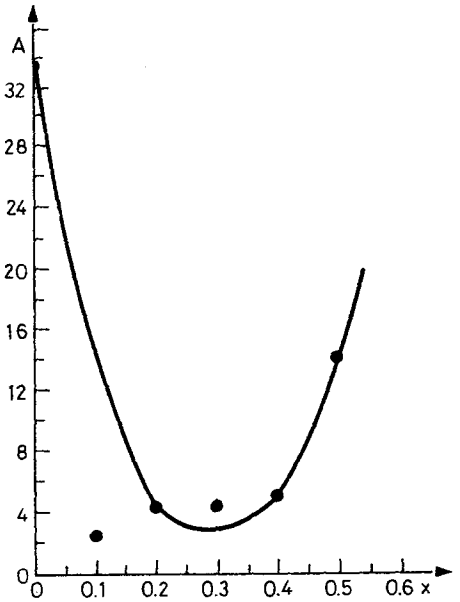

Fig. 6 Variation of Tawfik constant A as a function of x in the ferrimagnetic state

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Zusammenfassung — Mittels einer Keramiktechnik wurde eine Reihe von Proben des Types $Ni_{0.65}Zn_{0.35}Cu_xFe_{2-x}O_4$ mit $x = 0$, 0.1, 0.2, 0.3, 0.4 und 0.5 hergestellt. In Röntgendiffraktionsuntersuchungen wurde die Existenz von Single-Phasen bestätigt. Bei verschiedenen Temperaturen und einer Frequenz von 1 kHz wurde die Dielektrizitätskonstante K und der dielektrische Verlust D gemessen. Die ausgesprochene Erhöhung der Dielektrizitäts-konstante bei der Umwandlungstemperatur kann der Phasenumwandlung vom ferromagnetischen in den paramagnetischen Zustand zugeschrieben werden. Die Abnahme des elektrischen Verlustes mit steigender Temperatur kann durch kleinere Relaxationszeiten der springenden Elektronen und Löcher erklärt werden. Für den ferromagnetischen Zustand wurde auch die Tawfik-Konstante berechnet, die in Korrelation mit der Dielektrizitäts-konstante gefunden wurde.