

SYNTHESIS, MICROSTRUCTURE AND AC ELECTRICAL CONDUCTIVITY OF COPPER SUBSTITUTED NICKEL-ZINC FERRITES

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ABSTRACT

A series of nickel-zinc ferrites with composition $\text{Cu}_x\text{Ni}_{0.3-x}\text{Zn}_{0.7}\text{Fe}_2\text{O}_4$, with $x=0.00, 0.05, 0.15$ and 0.25 have been synthesized via the solid state ceramic route in order to carry out ac electrical measurement. Formation of single phase spinel structure have been confirmed by using X-ray diffraction. Microstructural features of sintered samples were obtained with scanning electron microscope. The micrographs showed that increasing copper substitution in nickel-zinc ferrite increased grain growth. The bulk density of these samples also increased as the copper content increased. AC conductivity, σ_{ac} has been measured in the temperature range $300\text{ K} - 473\text{ K}$ and frequency range 10^{-2} Hz to 1 MHz and discussed as a function of frequency and temperature. Analysis of the results shows that ac conductivity increases as the temperature of the sample and frequency of the applied ac electric field increase, but the dispersion of ac conductivity decreases with increasing the temperature for all the samples.

INTRODUCTION

In this work, the oxide-mixing technique was chosen as synthesis method as it is the most commonly used technique in ceramic syntheses. In addition, the results obtained could be easily transferred or translated to meet the industrial need. There are many researchers studied the magnetic properties of nickel-zinc ferrites but for the nonmagnetic properties such as electrical conductivity and dielectric properties are seldom reported. The electrical properties are important for ferrite not only from the application point of view but also from the fundamental point of view. Evaluation of a.c electrical conductivity reveals a wealth of information as regards the usefulness of these materials for various applications. Moreover, the study of AC electrical conductivity sheds light on the behaviour of charge carriers under the AC field, their mobility, and the mechanism of conduction. The conductivity studies on nickel zinc ferrites were carried out by several researchers (Elhiti 1996; pal et al 1996; Abdeen 1998).

MATERIALS AND METHODS

Synthesis of the samples

A series of polycrystalline spinel ferrites with chemical composition $\text{Ni}_{0.3x}\text{Cu}_x\text{Zn}_{0.7}\text{Fe}_2\text{O}_4$, with $x = 0.00, 0.05, 0.15$ and 0.25 were prepared using the

conventional ceramic method. Firstly, NiO, ZnO, CuO and Fe₂O₃ were mixed in suitable proportions for 16 hours in a ceramic ball mill using distill water as the mixing medium. After that, the mixture was dried in oven and pre-sintered at 1050°C for 10 hours in air. The mixture were reground again for 6 hours using plenatary micro mill and the final fine powders were pressed in disk-shaped samples. Then, the green samples were sintered at 1150°C for 10 hours and slowly cooled to room temperature.

Measurements

All the samples were structurally characterized by using Philips X'pert, high resolution X-ray diffraction system and Philips XL 30 Environmental Scanning Electron Microscope (SEM) was employed to examine the microstructural features. Electrical measurements were carried out using the dielectric spectrometer system consisting of a Chelsea Dielectric Interface (CDI) and Frequency Response Analyzer (FRA) at various frequencies ranging from 10⁻² Hz to 10⁶ Hz.

RESULTS AND DISCUSSION

X-ray analysis

The X-ray diffraction patterns of the sintered specimens are shown in Figure 1. It can be noted that the sharp peaks indicate a high crystallinity of the samples. All the samples were single phase with a cubic spinel structure. This proved that the preparation of Ni_{0.3-x}Cu_xZn_{0.7}Fe₂O₄ had been successfully performed.

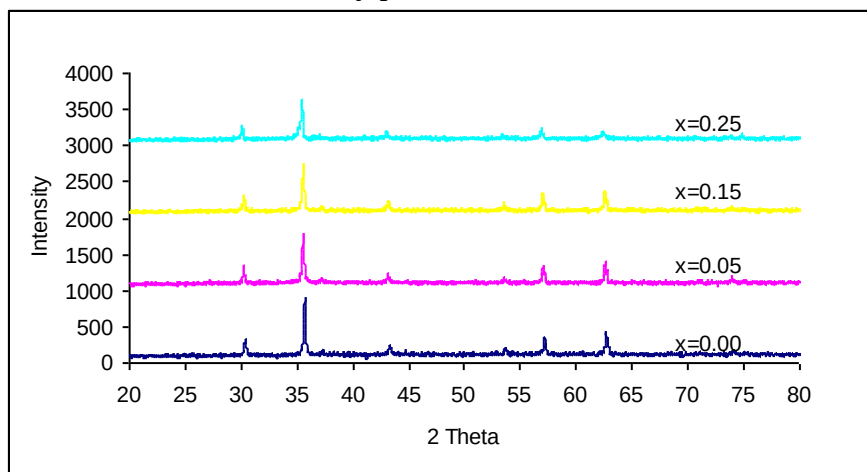


Figure 1: X-ray diffraction patterns for the system Cu_x Ni_{0.3-x} Zn_{0.7} Fe₂ O₄

Microstructural features

The SEM micrographs of the samples are shown in Figure 2. After substituted with copper in Ni-Zn ferrite and increase the molar ratio of copper (x=0.0 to x=0.25), there are grain growth as seen from the micrographs.

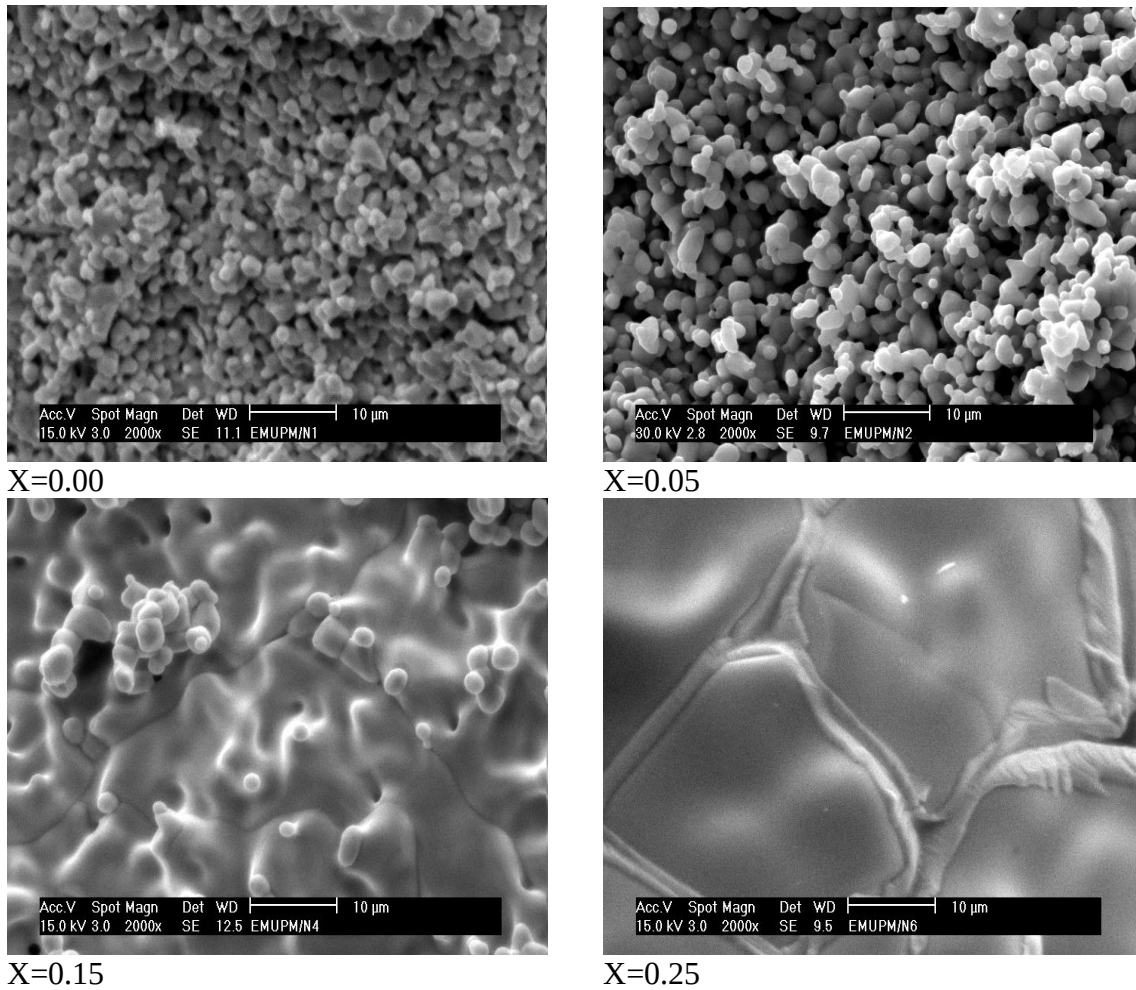


Figure 2: SEM Micrograph of $\text{Cu}_x\text{Ni}_{0.3-x}\text{Zn}_{0.7}\text{Fe}_2\text{O}_4$ ferrite system for $x=0.0, 0.05, 0.15,$ and 0.25 .

Table 1: Density of $\text{Cu}_x\text{Ni}_{0.3-x}\text{Zn}_{0.7}\text{Fe}_2\text{O}_4$ ferrite system for $x=0.0, 0.05, 0.15,$ and 0.25 .

Copper content, x	Density (gcm^{-3})
0.00	4.35
0.05	4.59
0.15	5.03
0.25	5.07

The substitution of nickel by copper has also affected the bulk density. Table 1 shows the bulk density of unsubstituted ferrite, $x=0.0$, was 4.35 gcm^{-3} as against 5.07 gcm^{-3} of copper substituted ferrite with $x=0.25$. The increase is attributed to the increase in grain size and reduction of the pores in the ferrite system, as seen in scanning electron micrographs in Figure 2.

AC conductivity

AC conductivity can be explained based on the basis of the assumption that σ can be expressed as (Joncher 1983)

$$\sigma = \sigma_{dc} + \sigma_{ac}$$

where the first term is a temperature dependent term represent the dc electric conductivity which is related to the drift mobility of the free charge carriers, and the second term which is frequency and temperature dependent term which is related to the bound charge carriers. The first term is predominant at low frequencies and high temperature, while the second term is predominant at high frequencies and low temperatures. The frequency dependence of the second term σ_{ac} can be written as

$$\sigma_{ac} = A\omega^n$$

where A is a constant having the units of the conductivity and the n is a temperature constant.

Frequency dependence

The a.c. electrical conductivity of nickel zinc ferrites has been carried out at different frequencies (0.01Hz to 1 MHz) from 300K to 473 K. It was observed from Figure 3 that the a.c. electrical conductivity increases with increasing frequency. The hopping of electron between Fe^{2+} and Fe^{3+} ions on the octahedral sites is responsible for conduction in ferrites. Hole hopping between Ni^{2+} and Ni^{3+} on B site also contribute to electric conduction in ferrites. The frequency dependence can be explained with the help of Maxwell-Wagner two layer model or heterogeneous model of the polycrystalline structure of ferrites (Koops 1951). According to this theory two layer formed dielectric structure. The first layer consists of ferrite grains of well conducting (ferrous ions), which is separated by a thin layer of poorly conducting substances, which forms the grain boundary. These grain boundaries are more active at lower frequencies, hence the hopping frequency of electron between Fe^{2+} and Fe^{3+} is less at lower frequencies. As the frequency of the applied field increases, the conductive grains become more active by promoting the hopping electron between Fe^{2+} and Fe^{3+} ions, thereby increasing the hopping frequency. Thus, we observed a gradual increase in conductivity with frequency.

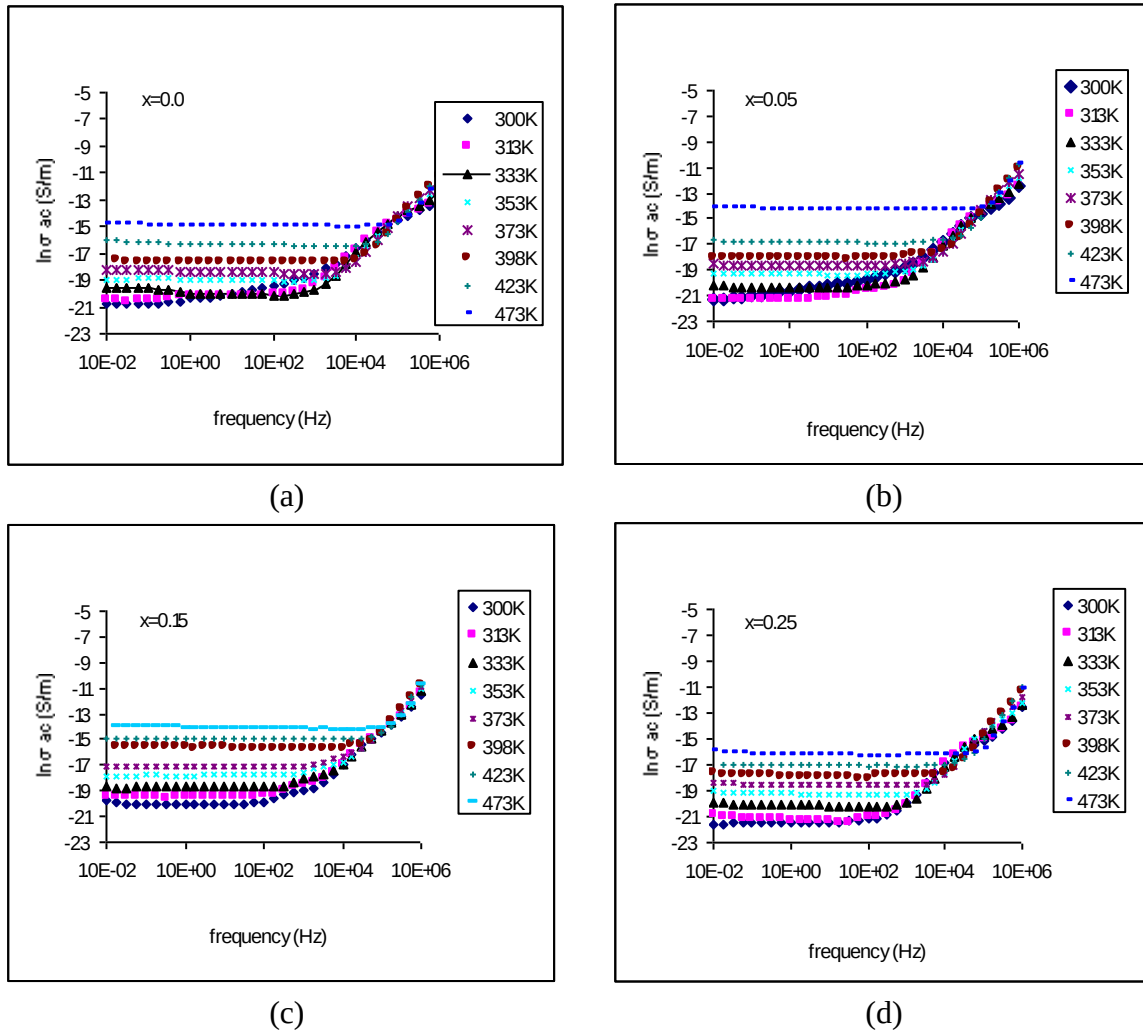


Figure 3: The variation of ac conductivity with the frequency at different selected temperatures for all studied samples.

Temperature Dependence

The effect of temperature on the a.c. electrical conductivity of ceramic nickel zinc ferrite samples were studied in the range 300K to 473K. The variation pattern of conductivity for different compositions are shown in figure 4. It can be seen from the figure that the conductivity increases with increasing temperature for all ceramic nickel zinc ferrites. At low frequencies the variation is very minimal but at higher frequencies the variation is noticeable. The influence of temperature on conductivity can be explained by considering the mobility of charge carriers responsible for hopping. As temperature increase the mobility of hopping ions also increase thereby increasing conductivity. The electrons involved in hopping are responsible for electronic polarization in these ferrites.

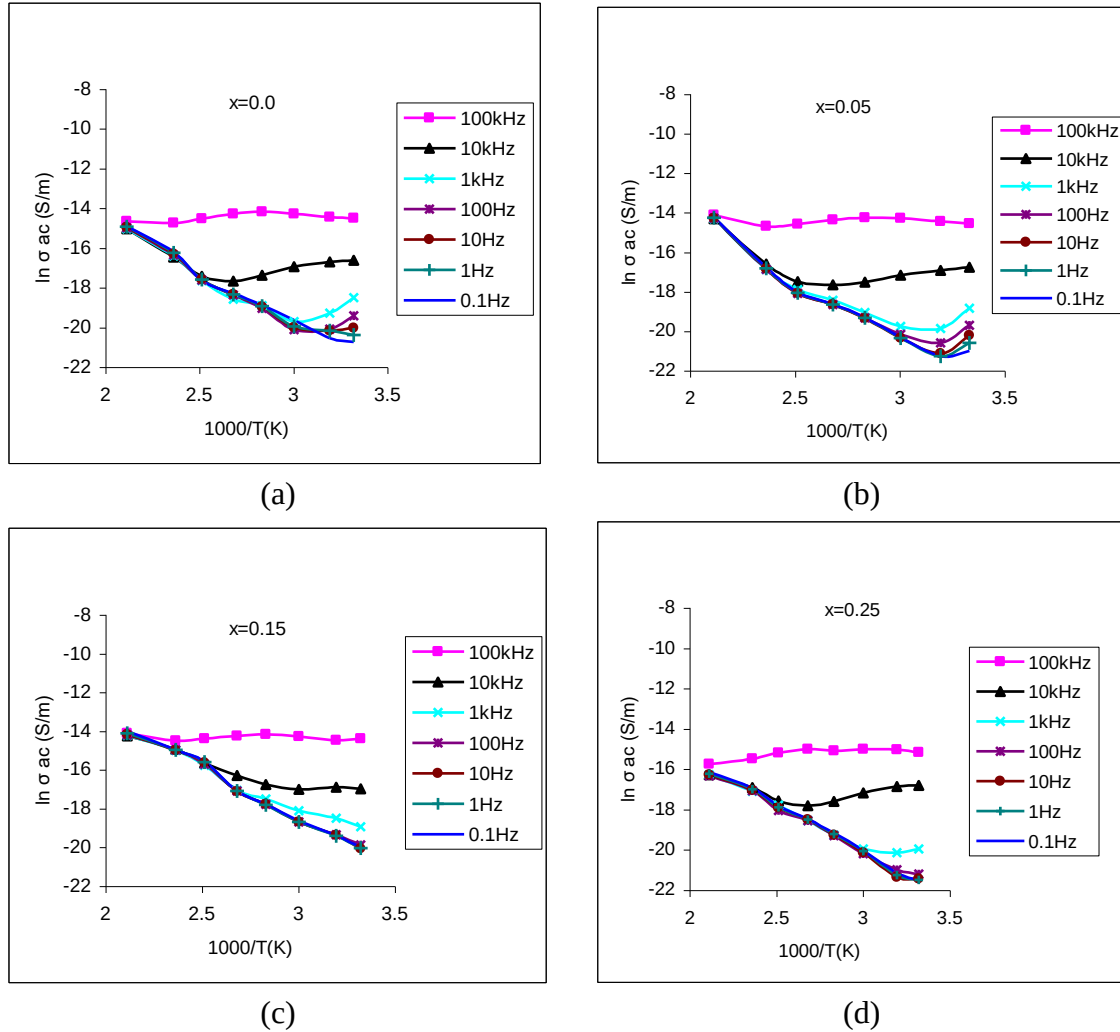


Figure 4: The variation of ac conductivity with the temperature as $10^3/T$, at different selected frequencies for all studied samples.

CONCLUSION

After substituted with copper grain growth was observed and the density of the Nickel Zinc ferrites also increased. AC also electrical conductivity increases as the temperature of the sample and frequency of the applied ac electric field increase. But the dispersion of ac conductivity decreases with increasing the temperature for all the samples. At relatively high temperature, the ac conductivity seems to be constant.

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