

Electrical conductivity and magnetic transition phase in ferrites

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The dielectric relaxation frequency, f_x , which is associated with bulk polarization processes in Ni-Zn and Mn-Zn polycrystalline ferrites, was investigated by using impedance spectroscopy. The behavior of f_x showed a clear maximum as a function of the temperature. This maximum agrees with Curie temperature, which was determined by an independent magnetic technique. These results show that impedance spectroscopy can be effectively used to study the ferri-paramagnetic transition.

Keywords: Impedance spectroscopy; ferrites; dielectrical relaxation frequency

Utilizando espectroscopía de impedancias, se realizó un estudio de la frecuencia de relajación dieléctrica, f_x , asociada con los procesos de polarización en los granos de ferritas policristalinas de Ni-Zn y de Mn-Zn. El comportamiento de dicha frecuencia como función de la temperatura, muestra claramente un máximo. Este máximo coincide con la temperatura de Curie, la cual fue medida mediante una técnica magnética independiente. Estos resultados muestran que la espectroscopía de impedancias puede ser empleada de manera efectiva para el estudio de la transición ferri-paramagnética.

Descriptores: Espectroscopía de impedancias; ferritas; frecuencia de relajación dieléctrica

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1. Introduction

Impedance Spectroscopy has been used to investigate the electrical conductivity as a function of the frequency because it allows to distinguish the different polarization mechanisms in polycrystalline materials. Since they have characteristic relaxation times, it is possible to get separately the different contributions from the impedance such as grains (or bulk), grain boundaries, electrodes, etc. [1]. Impedance Spectroscopy is particularly used in highly resistive ceramic systems. Since electrical and magnetic properties depend on the microstructure, this technique can be useful to investigate it.

2. Experimental

Ni-Zn and Mn-Zn ferrites were obtained by solid state reaction from the binary oxides: NiO, ZnO, MnO and Fe₂O₄. These powders were milled in an agata mortar for 24 hours and pressed in toroidal shape at 3 tons/cm². The toroids were calcinated at high temperature in an oxygen atmosphere during 12 hours. Zn_xNi_(1-x)Fe₂O₄ (with $x = 0.65$), were sintered at 1180°C and Zn_xMn_(1-x)Fe₂O₄, at 1150°C. X-rays powder diffraction revealed a single spinel phase.

Complex impedance measurements were carried out as a function of frequency in the 5 Hz–13 MHz range, from room temperature to 200°C. An HP4192 impedance Analyzer was used.

Measurements of the initial permeability were done as a function of the temperature by a method based on the Faraday law of electromagnetic induction for toroidal samples of ferromagnetic compounds [2].

3. Results

Cole-Cole plots exhibited two semicircles, one of them (low frequencies) was associated with the grain boundaries response and the other one (high frequencies), with the grains of the material. Figure 1 shows an example of these responses at room temperature. Associations were done taking account of the values of the corresponding capacitances calculated as $C = 1/2\pi f_x R$, where R is the diameter of semicircles and f_x is the relaxation frequency.

The conductivity mechanism can be represented by an equivalent circuit built with two RC elements in parallel, connected in series; one for modeling the grains and another one for the grain boundaries, as showed in Fig. 2.

In order to see more clearly the frequency relaxation at each temperature, imaginary impedances values were plotted as a function of f_x (Fig. 3). Frequency relaxations as a function of the temperature are presented in Fig. 4 for the grain boundaries and it is observed a normal linear behavior as expected. However, when the plot corresponding to the grains is analyzed (Fig. 5), it exhibits a clear maximum at a characteristic temperature.

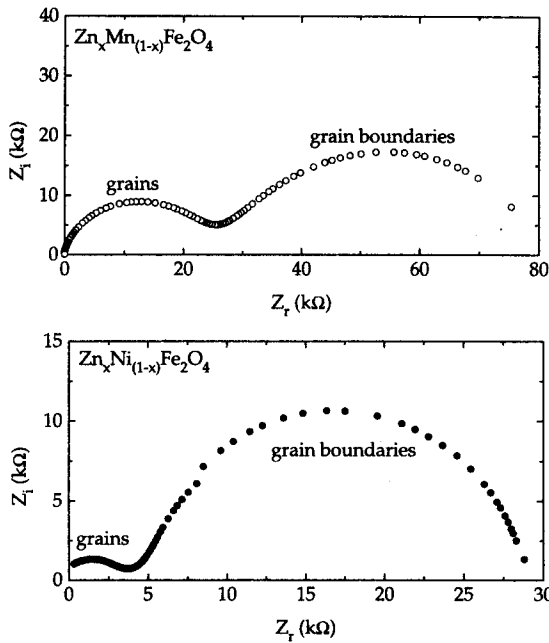


FIGURE 1. Complex impedance plots at room temperature.

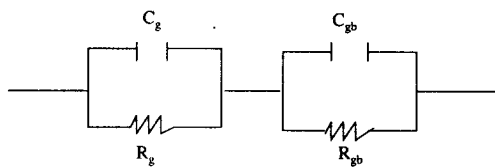


FIGURE 2. Equivalent circuit which models the conductivity mechanism. Indexes g and gb are referred to the grain and grain boundaries respectively.

In the other hand the initial permeability as a function of temperature was measured and results appear in Fig. 6. By analyzing these results, an agreement of the f_x maximum with curie temperature is observed. This is an evidence that the electrical conductivity mechanisms are related to the ferri-para magnetic transition in ferrites and they can be considered as another signal of the second order-transition phase since this transition involves an electronic structure change. In the case of the grain boundaries, the change can not be observed because they do not present the order-disorder transition.

4. Conclusion

Since the frequency relaxation of the grains in ferrites show a maximum at Curie temperature, this behavior can be used to characterize the ferri-para magnetic transition and it can be effectively made by impedance spectroscopy.

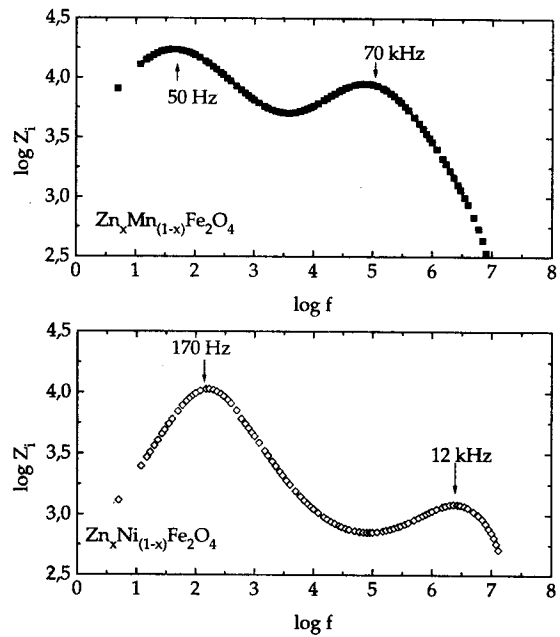


FIGURE 3. Imaginary impedance as a function of frequency at room temperature.

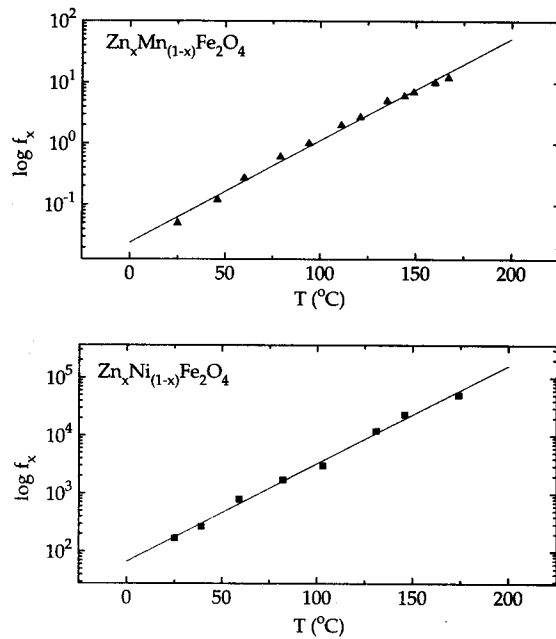


FIGURE 4. Behavior of f_x as a function of temperature in grain boundaries.

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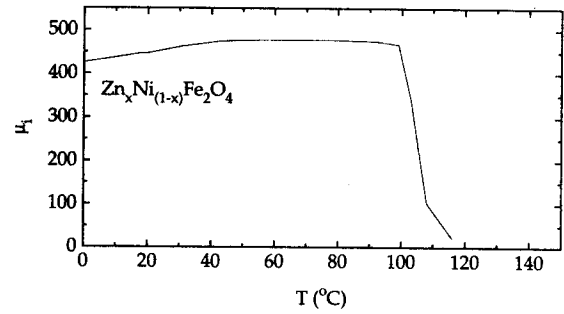
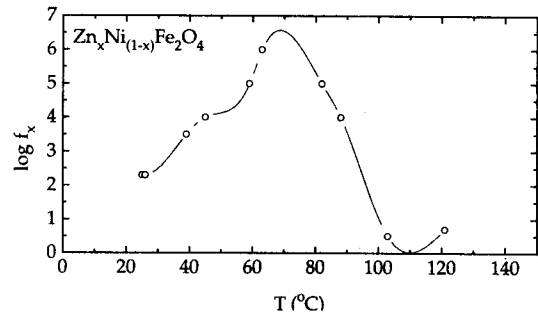
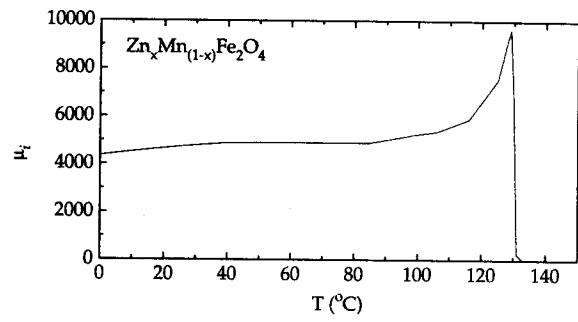
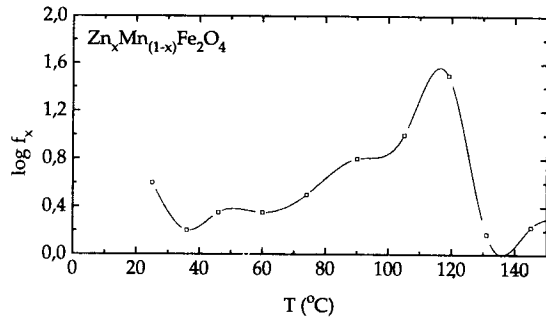
FIGURE 5. Behavior of f_x as a function of temperature in grains.

FIGURE 6. Initial permeability as a function of temperature.

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