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DIELECTRIC POLARIZATION OF SEMICONDUCTOR Co-Zn FERRITE

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An oscillographic study was done in Co-Zn ferrite which gave some light about the generated polarization with the a.c. field in playing dielectric hysteresis loop. It was noticed that polarization increased with temperature, which was attributed to the rapid hopping of exchange electrons between $Fe^{2+} \rightleftharpoons Fe^{3+}$ at the B sites leading to more orientation in the direction of the electric field. The polarization increased also with Zn additions, the theoretical dielectric constant which was estimated from hysteresis loop coincided the measured values. The dissipation factors for different zinc additions were estimated from the area of hysteresis loop and a pronounced increase of the dissipation factor above 100°C is noticed for the different compositions.

Key Words: Dielectric Hysteresis Loop; Spontaneous Polarization; Dielectric Constant; Semiconductor Co-Zn Ferrite; Oscillograph; Dissipation Factor; Ferroelectricity

Introduction

In 1947, the first piezoelectric transducer employing polarized ferroelectric barium titanate ceramics appeared. The art of producing and applying barium titanate ceramic transducers was developed broadly in the following ten years. Since 1957, barium titanate has been increasingly replaced with lead titanate zirconate. Solid-Solution ceramics offer higher piezoelectric coupling, wider operating temperature range, and a choice of useful variations in engineering parameters. The key physical property of barium titanate, the lead zireonates, and other piezoelectric ceramics is ferroelectricity[1-6]. Ferroelectrics have electronics and ionic displacements producing net spontaneous polarization, which can be preferentially oriented by an applied field.

These materials have reached a dominant position for sonic and the lower range of ultrasonic piezoelectric transducers. The polycrystalline ferrites are very good dielectric materials. The dielectric properties of ferrites are dependent on several factors including the method of preparation the sintering temperature and the sintering atmosphere. In the process of preparation of ferrites in the polycrystalline form, the ferrite powder is sintered under slightly reducing conditions. As a consequence the divalent iron produced in the bulk of the materials forms high conductivity grains separated by low conductivity layers so that the ferrites behaves as an inhomogeneous dielectric material. Therefore, the ferrites possess, dielectric constants as high as 10^5 at low frequency.

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In this paper, we have discussed primarily the new method of calculating the dielectric constant and dielectric loss of Co-Zn ferrite by displaying the dielectric hysteresis loops using Sawyer and Tawer circuit and compared it with the measured values using RLC bridge.

Experimental Procedure

The ferrite samples used had the compositional formula $Co_{1-x}Zn_xFe_2O_4$ where x ranged from 0 to 0.9. They were prepared by the well-known double sintering process starting with highly pure ferric, cobalt and zinc oxides. The final sintering temperature for specimens of different compositions was 1200°C. The details of the method of preparation are given in an earlier publication[7].

The polarization (p) and the electric field (E) hysteresis loops were observed by Sawyer Tower circuits at 50 HZ[8]. Specimen were prepared by lapping the pallet faces flat and parallel and painting with Ag electrodes the resulting dielectric polarization recorded. The capacitance of the sample was measured using LRC-SORTESTER Aplap 4912 Bridge.

Results and Discussion

Hysteresis Loops of Co-Zn Ferrite

The effect of Zn additions on Co ferrite is the main study of this new phenomena in ferrites. An oscillographic study was carried and the effect of Zn additions on the hysteresis loop is illustrated in Fig. 1.

Temperature Dependence of Induced Polarization for Co-ZnFe$_2$O$_4$

The induced polarization is given as follows:

$$Q = C \cdot V \qquad P = \frac{Q}{A} = \frac{C \cdot V}{A},$$

where C is the capacitance of the capacitor included in the circuit, A is the area of the sample surface and V is the Voltage in the sample indicated by the oscilloscope.

The dielectric polarization of $Co_{1-x}Zn_xFe_2O_4$ ($y = 0.0 \rightarrow 0.8$) was estimated from hysteresis loop and the results of the observation was indicated in Fig. 2. The polarization at the sample surface was increased with temperature. The behaviour of these results is explained on the bases that the thermoelectric power for $Co_{1-x}Zn_xFe_2O_4$ sample is negative indicating that the electrons are the majority of carriers at room temperature[9-10]. Let the applied field E make an angle ϕ with one of these preferred directions of polarization of a crystallite, then the effective field strength is $E \cos \phi$, so that a larger field strength is needed to line up all the domains of the crystallite. If they are lined up, the saturation polarization has a component $p_s \cos \phi$ in the direction of the applied field. After the domains of all crystallites are lined up along the directions of preferred polarization that make the smallest angles with the applied field, then the polarization p_s of the individual domains will rotate slowly toward the direction of the applied field strength E.

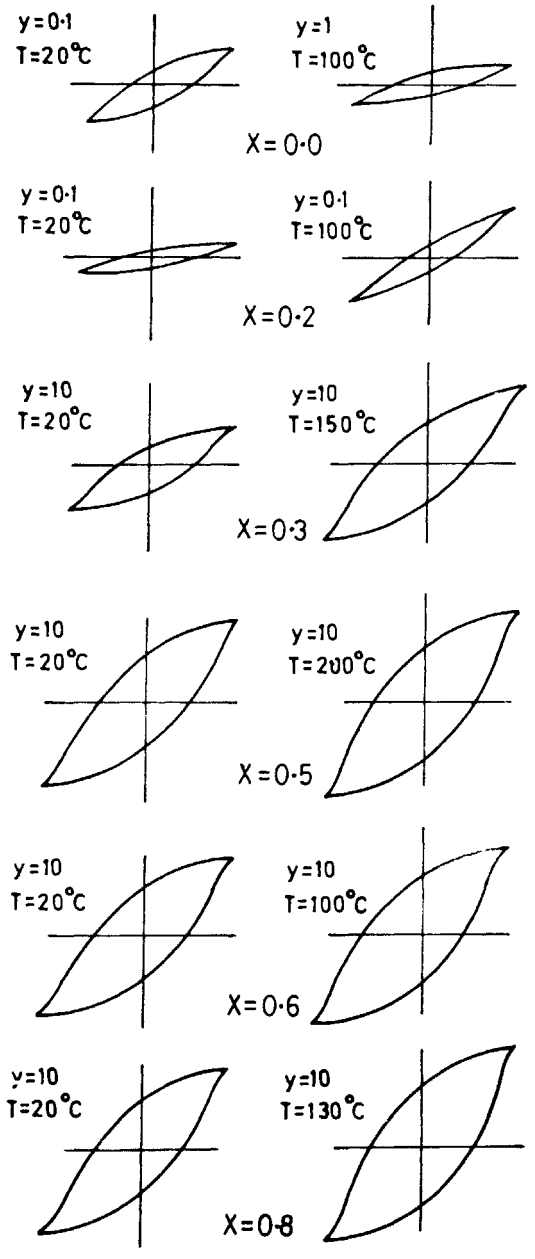

Fig 1 Hysteresis loops of $Co_{1-x}Zn_xFe_2O_4$

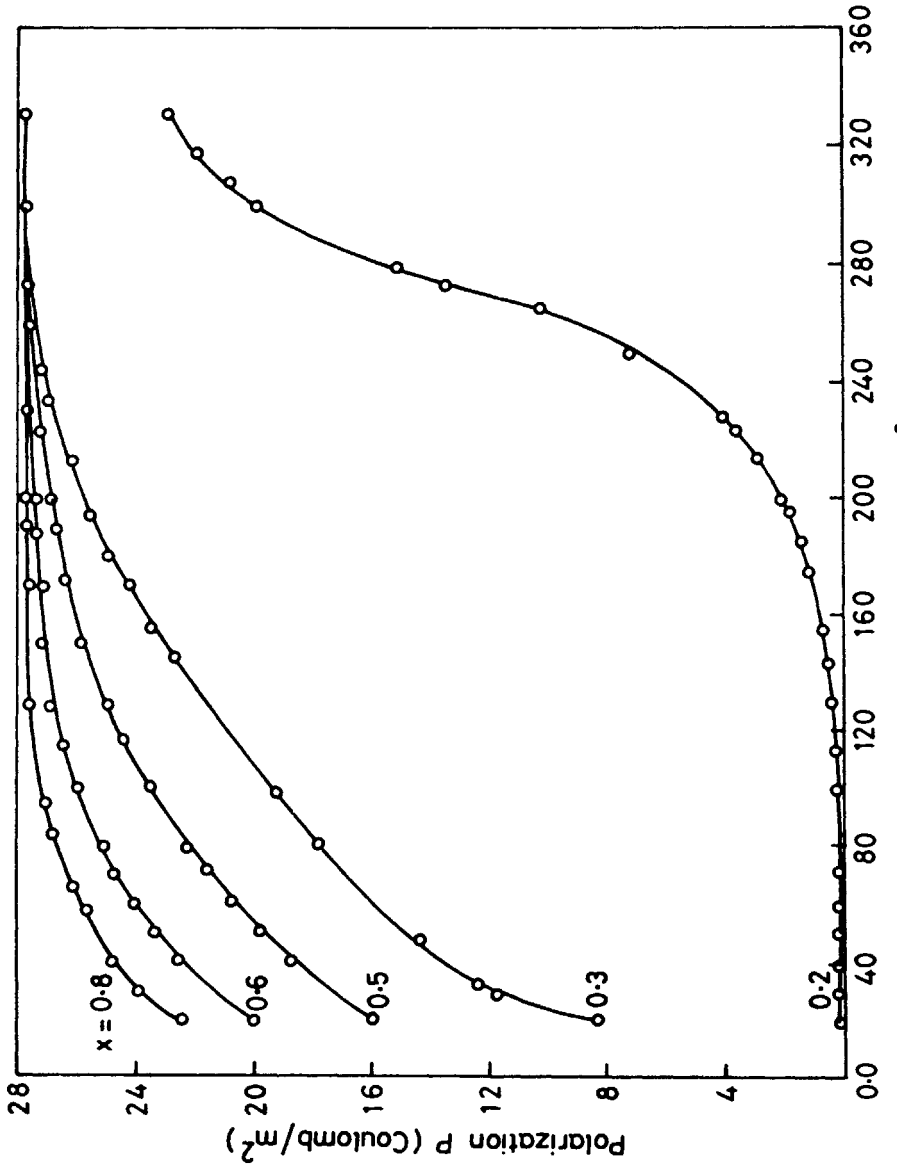

Fig 2 Temperature dependence of the induced spontaneous polarization at different values of x

The electronic charge carriers increased in magnitude with temperature due to activated electron hopping between Fe^{2+} and Fe^{3+}. The number of ferrous ions on the octahedral sites which take part in the electron exchange interaction $Fe^{2+} \rightleftharpoons Fe^{3+}$ and hence are responsible for the polarization, is a maximum in the case of rising temperature; therefore, a comparatively high value of the dielectric constant is expected with increase the temperature. Thus the number of ferrous ions on the octahedral sites plays a dominant role in the mechanism of conduction and dielectric polarization of ferrites.

Effect of Zn Additives on the Polarizations of Co Ferrite

Th polarization induced at the sample surfaces under applied a.c. field (310)V/cm, is indicated in Fig. 3. It is obvious that the induced polarization increased with Zn additions for $Co_{1-x}Zn_xFe_2O_4$. Our results are explained as follows:

Zinc ferrite is a normal ferrite and cobalt ferrite is an inverse one. So it is interesting to see the magnetic properties of the mixed ferrite. Assuming that Zn^{2+} occupies tetrahedral sites only, the mixed cobalt zinc ferrite has the cation distribution as $(Zn_x^{2+}Fe_{1-x}^{3+})Co_{1-x}^{2+}Fe_{1+x}^{3+}O_4$. The spontaneous magnetization

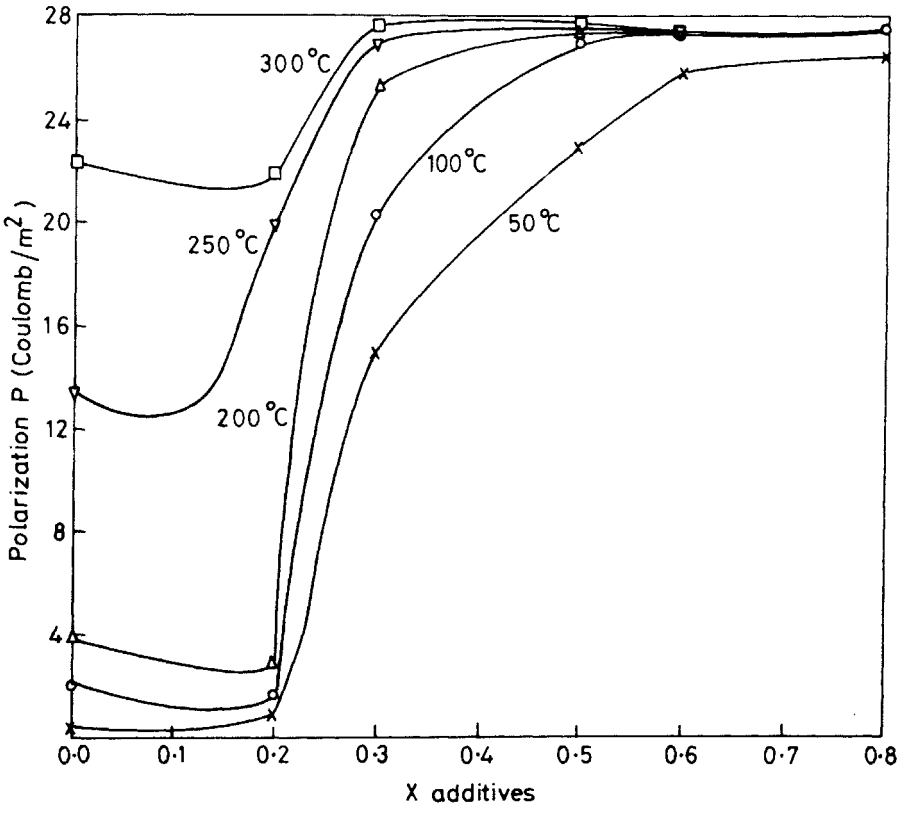

Fig 3 The dependence of the induced spontaneous polarization on Zn additives at different temperatures

of a ferrite material is given by the difference $(M_A - M_B)$ of the magnetizations of the sublattices A and B. Ferromagnetic substances can be magnetized by small magnetic fields because of the strong electron exchange interactions. The magnetic moment has increased with the increase of Zn content caused a pronounced increase of saturation magnetization with Zn additions. The mechanism of the dielectric polarization is similar to that of conduction because the electron exchange interaction $Fe^{2+} \rightleftharpoons Fe^{3+}$ results in a local displacement of the electrons in the direction of electric field which determine the polarization of ferrite. Cobalt ferrite ($CoFe_2O_4$) has the inverse spinel structure in which the tetrahedral sites are occupied by the ferrous ions and the octahedral sites are occupied half by Fe^{3+} and half by Fe^{2+}. The addition fo Zn ions which substitute the tetrahedral sites, leading to excess Co ions or Fe^{3+} at the octahedral

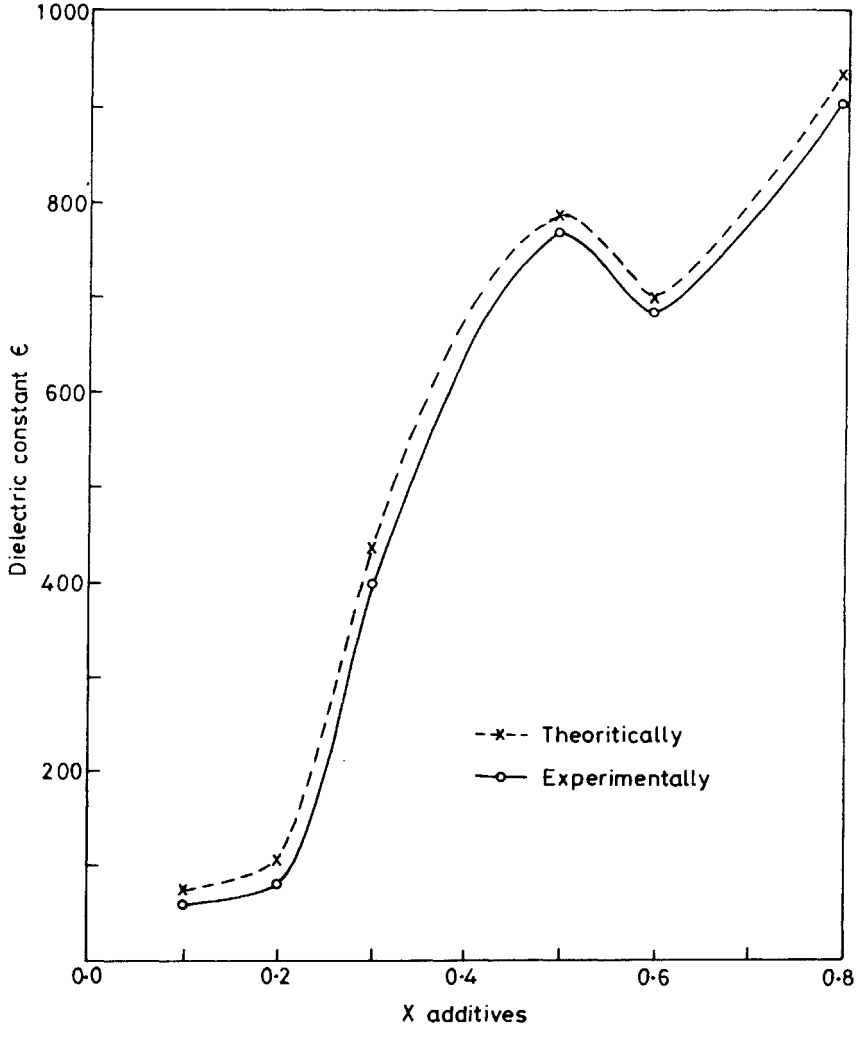

Fig 4 The dependence of dielectric constant on the Zn additives at room temperature

sites. The excess Co^{2+} at the B sites generates electrons. The number of ferrous ions on the octahedral sites whcih take part in the electron exchange interaction $Fe^{2+} \rightleftharpoons Fe^{3+}$ and hence are responsible for the polarization is a maximum in the case of $Co_{1-x}Zn_xFe_2O_4$. Thus the number of ferrous ions on the octahedral sites plays a dominant roles on the mechanism of conduction and dielectric polarization, and magnetization[11,12] which confirm the behaviour of dielectric polarizations of Zn additions.

Composition Dependence of Dielectric Constant

Fig. 4 shows the variation of the measured dielectric constant K with zinc additions at room temperature for $Co_{1-x}Zn_xFe_2O_4$. It can be seen that the dielectric constant increase with Zn additives indicating that the behaviour of dielectric constant of the sample is the same as the dielectric polarization.

From the dielectric hysteresis loop measurements, the dielectric constant was estimated using the formula

$$C = \frac{\varepsilon A}{4\pi d}, \qquad \ldots (1)$$

where C is the capacitance of the sample in *pf*, ε dielectric constant, A area of the sample surface in cm^2 and d is its thickness. It is known that

$$Q = C \cdot V = \left(\frac{\varepsilon A}{4\pi d}\right) \cdot V \times 10^{-12}$$

and

$$\varepsilon = \frac{4\pi Qd}{V \cdot A} \times 10^{12}, \qquad \ldots (2)$$

where C is the capacitance in (farad), Q is the generated charge on the sample surface in (coulomb), and V is the a.c. applied voltage, on the sample relative to its capacitance.

$$P = \frac{Q}{A}, \qquad \ldots (3)$$

where P is the induced polarization in (coulomb/m^2) and A is the area of the sample surface in (m^2). It is noticed that the calculated dielectric constant is in good agreement with the measured values which confirms the behaviour of dielectric hysteresis loop with the variation of Zn additions.

Temperature Dependence of Dissipation Factor

The dissipation factor was estimated from the area of the dielectric-hysteresis loop using the gravemetric technique. The loops is plotted on a paper out from the screen of the oscilloscope, then carefully cut from the paper and accurately weighed. The area could be obtained by comparison with a reference

area taken from the same paper of known weight. Gravimetry provide a simple technique which gives the area with an error of ± 2% depending on the personal skill for cutting the loops.

Fig. 5 shows the effect of temperature on the dissipation factor for different zinc additions $x = 0.2, 0.3, 0.5, 0.6$. A pronounced increase of the dissipation factor above 100°C is noticed for the different compositions. These results are explained using the assumption that the mechanism of dielectric polarization is similar to that of conduction. Previous workers observed that the electron exchange $Fe^{2+} \rightleftharpoons Fe^{3+}$ results in a local displacement of electrons in the direction of electric field[13,14]. The increase of temperature give rise to more hopping of exchange electrons at the octahedral sites in an *n*-type semiconductors. The hopping acceleration due to the temperature increase

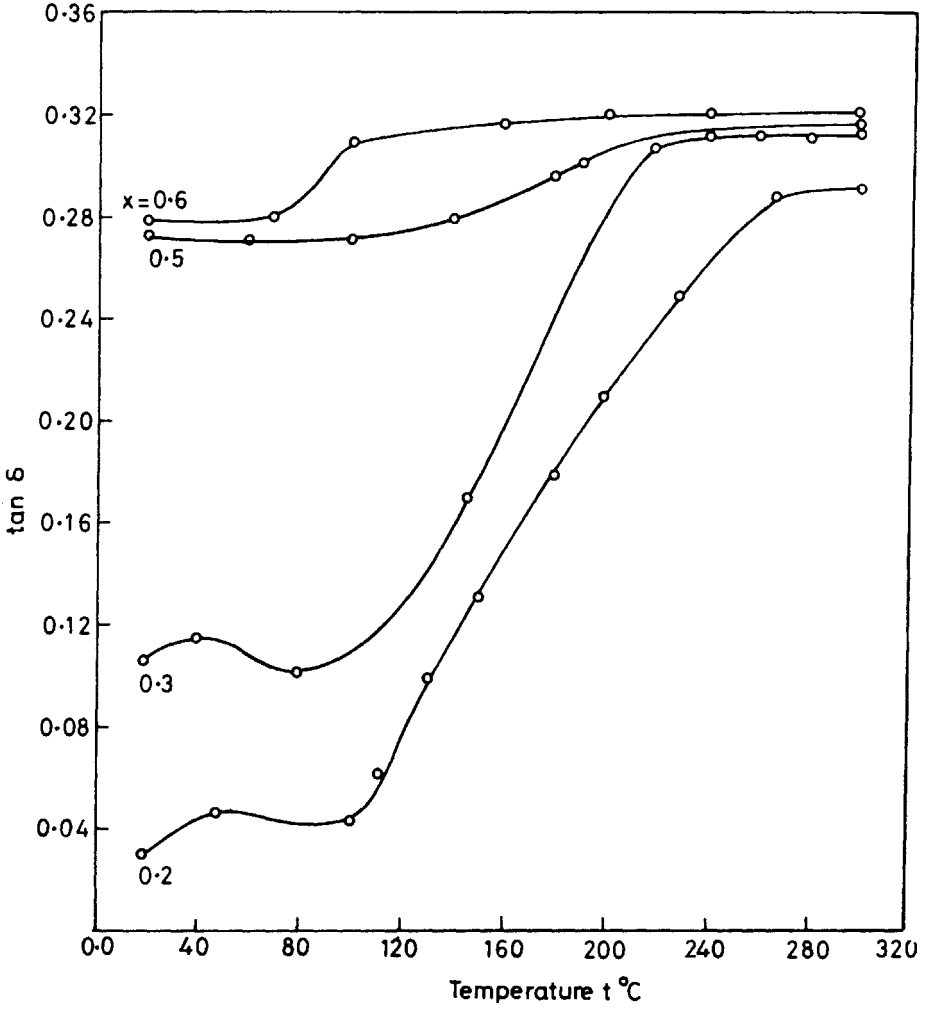

Fig 5 Temperature dependence of dissipation factor at different values of x

caused an increase of conductivity, consequently an increase of dielectric loss. Our explanation is in a good agreement with previous work which stated that the increase of temperature increased the drift mobility of electrons[15].

Acknowledgement

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