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Ferroelectric Aspect and Phase Transitions of Solid Solution of Cadmium Acetate with Aluminium (III) Oxide

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ABSTRACT: The behavior of variation of dielectric constant with temperature of solid solution of Cadmium Acetate $Cd(C_2H_3O_2)_2$ with Aluminium (III) Oxide Al_2O_3 and other dielectric parameters have been measured between the temperatures 35°C to 100°C using the capacitance bridge model ZENITH-FM89A and Q meter at the frequency of 2000 Hz. In the measurement, it have been observed that the compound has lower value of dielectric constant (ε = 1580) below 36ºC, which rises upto a value of 5600 at the moderate temperature of 45ºC. After this temperature, the dielectric constant of compound decreases upto the value of nearly 3521 at the temperature of 75ºC and a high peak at the temperature of 76°C (ε = 7000) in the heating cycle curve with some fluctuations.. When the variation of dielectric constant was studied in cooling cycle the peak was observed at 62ºC(ε = 6500), above and below this temperature dielectric constant decreases with some intermediate fluctuations and by increasing aluminium oxide content in the mother compound, its quality factor also increases with a little effect on dielectric constant. The cooling cycle curve does not follow heating curve because of the relaxor behaviour of the compound. The results have been explained on the basis of crystal structure changes and the possibility of free internal rotation of acetate groups within the crystal lattice at elevated temperature. One of the advantages of $Cd(C_2H_3O_2)_2$ with Al_2O_3 is that by varying the Aluminium Oxide content, one can control temperature coefficient of resonant frequency τ_f without affecting the other properties. This is important for many ferroelectric applications because τ_f of near about low value is not always required. A non-zero τ_f is often preferable to compensate for frequency variation due to the effect of temperature change on the resonator housing and dielectric support structure. Solid solution of Cadmium Acetate with Aluminium Oxide suggests for its valuable applications as dielectric material with excellent properties useful in ceramics engineering and communication system.

KEYWORDS: Polarization, Quality Factor, Dielectric Constant, Phase Transition, Curie temperature, Resonant Frequency and Ceramics Engineering.

INTRODUCTION

The dielectric constant is a solid state property which is technically important and is also helpful in understanding basic crystal physics. Combined with other information like the refractive index and the absorption frequency it throws light on the bonding in crystals. In theoretical studies of lattice dynamics, the dielectric constant forms one of the input parameters. Measurement of dielectric constant and loss as a function of frequency and temperature helps in understanding the polarization mechanism, process of conduction, influence of impurities and phase transition. AC conductivity obtained from the dielectric properties combined with the data on DC conductivity yields useful information on defect formation and nature of conduction. [1].

The dielectric constant measured in the frequency independent region is taken as static or low frequency dielectric constant ε_s (sometimes referred to as infrared dielectric constant ε_{ir}). As the frequency is increased further, the value remains unaffected till the strong resonance absorption frequency is approached in the infrared region. Beyond the resonant frequency, since the ions cannot follow the field, the polarization due to electronic contribution alone persists. Hence the dielectric constant in this region is termed as high frequency dielectric constant (ε_∞). Under the influence of static field, the dielectric constant is treated as a real number. The system is assumed to get polarized instantaneously on the application of the field. When the dielectric is subjected to alternating field, the displacement cannot follow the field due to inertial effects and spatially oriented defects. The dielectric constant is then treated as a complex quantity ε(ω). The variation of real and imaginary parts of the complex dielectric constant with frequency ω is given by the Debye equations[2]

$$\varepsilon(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega)$$

where $$\varepsilon'(\omega) = \varepsilon(\infty) + \frac{\varepsilon(s)-\varepsilon(\infty)}{1+\omega^2\tau^2}$$

and $$\varepsilon''(\omega) = \varepsilon(s) - \varepsilon(\infty)\frac{\omega\tau}{1+\omega^2\tau^2}$$

where τ is the relaxation time. ε' is identified with the measured dielectric constant ε and ε'' is a measure of the average power loss in the system. The loss is expressed in terms of the phase angle δ as

$$\tan\delta = \varepsilon''/\varepsilon$$

The term ''quality factor'' is more commonly associated with microwave resonators. Quality factor, or Q, is a measure of the power loss of a microwave system. The name quality factor is used for the reciprocal of the tan δ. One should carefully distinguish this quantity from the Q-factor of a resonator which is defined as[3]

$$Q = 2\pi[(\text{maximum energy stored per cycle})/(\text{Average energy dissipated per cycle})]$$

The temperature coefficient of resonant frequency τ_f is the parameter which indicates the thermal stability of the resonator. The τ_f indicates how much the resonant frequency drifts with changing temperature. The electronic device with microwave resonators requires τ_f values as close to zero as possible. Microwave circuits will normally have some low characteristics τ_f, so the resonator components which go into them are required to compensate for the inherent drift. For this reason, the τ_f values of resonators required are typically non-zero but with some low finite value. The origin of τ_f is related to linear expansion coefficient α_L which affects the resonator dimensions and its dielectric constant variation with temperature. Mathematically the relationship is[4]

$$\tau_f = -\alpha_L - \tau_{E/2}$$

where τ_E is the temperature coefficient of the permittivity and α_L is the linear thermal expansion coefficient of the dielectric material which is usually positive.

Polarization is the result of ordering of the electrically charged particles under the action of external field. Macroscopically, it shows up as an increase in the capacitance of a condenser in the presence of a dielectric. Microscopically, it is described in terms of the induced electric moment. The electric moment acquired by an atom or a molecule under the influence of an electric field is proportional to the applied field. The proportionality constant is known as the polarizability (α). It is the measure of the ability of the material to respond to the field. The polarization *P* is defined as the electric moment per unit volume of the dielectric and is given by $P = N\alpha E$ where N is the number of particles (atoms, ions or molecules) in unit volume and E the applied field. In general, the electric moment may be induced by different mechanisms. In a most general case the total polarizability α may be expressed as

$$\alpha = \alpha_e + \alpha_a + \alpha_d + \alpha_s,$$

where α_e, the electronic polarizability, arises due to the deformation of the spherical distribution of negative charge around the nucleus. In addition to the deformation of the electronic cloud, the positively and negatively charged ions are displaced. This leads to the atomic polarizability α_a. If the material consists of permanent dipoles, they align along the direction of the field and an additional factor α_d will be added. The fourth component α_s is the polarizability arising out of the space charge[5].

EXPERIMENTAL

The compound has been procured from Sigma-Aldrich (India), Bangalore. The chemical was grinded into the fine powder in an agate mortar, avoiding direct sunlight and preferably the most of the sample preparation was done at night. The pellets were prepared with compression machine (Flextural Testing Machine CAT No.AIM-313, S.No.91070 AIMIL Associated, India), having pressure range 0-15 tonne wt/cm^2. A suitable die was used having rectangular Cross-Sectional area of the piston =2.5cm^2. The polishing of the pellets has been done to obtain smooth parallel surface to be used for electrode formation polishing of the crystal introduces electrical charges inside the material. These charges and strains are to be removed, which we did by the process of annealing of the sample. In this process the pellets were kept in a suitable furnace at nearly 2/3 of their melting points for sufficient times (generally 8-10 hours). The most of the irreproducibility was removed by annealing and therefore this process was necessarily done. The electrodes were formed using colloidal silver paints. The sample holder loaded with pellet is kept into the furnace such that it lies very near to the middle part of the furnace. A good quality thermometer, precisely calibrated is used to record the temperature. This thermometer is adjusted with the help of stand in such a way that it touches the metallic part of sample holder to record the exact temperature of sample. The usual substitution method i.e. with and without the specimen in suitable sample holder is used[6, 7]. The sample holder was directly fastened to the capacity measuring unit (ZENITH-FM89A) and Q meter. There are some process from which we have done the measurement of the quality factors of dielectrics.. The practical effects include noise, crosstalk, coupling losses, transmission line delay and impedance mismatch. Inadequate accounting of these effects may lead to significant uncertainty in the measured Q-factor. The determination of the complex permittivity and other electromagnetic properties require precise measurements of the resonant frequencies and Q-factors. For some measurement techniques, these parameters have to be measured in the presence and absence of the test sample, for other techniques only once in the presence of the test sample[8].

RESULTS AND DISCUSSION

The solid solution of Cadmium Acetate $Cd(C_2H_3O_2)_2$ and Aluminium (III) Oxide Al_2O_3 has excellent dielectric property such as dielectric constant of nearly $\varepsilon = 5500$, quality factor $Q_f = 28000$GHz and temperature coefficient of resonant frequency $\tau_f = 100$ ppm/°C which is very much useful in ceramics engineering and communication system. The nature of dielectric constant with the variation of temperature of solid solution of Cadmium Acetate $Cd(C_2H_3O_2)_2$ and Aluminium (III) Oxide Al_2O_3 and other dielectric parameters have been measured between the temperatures 35°C to 100°C using the capacitance bridge model ZENITH-FM89A and Q meter at the frequency of 2000 Hz in which percentage of Al_2O_3 is from 20 to 35. In the measurement, it have been observed that the compound has lower value of dielectric constant ($\varepsilon = 1580$) below 36ºC, which rises upto a value of 5600 at the moderate temperature of 45ºC. After this temperature the dielectric constant of compound decreases upto the value of nearly 3521 at the temperature of 75ºC and a high peak at the temperature of 76°C ($\varepsilon = 7000$) in the heating cycle curve with some fluctuations.. When the variation of dielectric constant was studied in cooling cycle the peak was observed at 62ºC($\varepsilon = 6500$), above and below this temperature dielectric constant decreases with some intermediate fluctuations. The curve for the cooing cycle is shown in the Fig.2. The cooling cycle curve does not follow heating curve because of the relaxor behaviour of the compound. The results have been explained on the basis of crystal structure changes and the possibility of free internal rotation of acetate groups within the crystal lattice at elevated temperature[9,10].

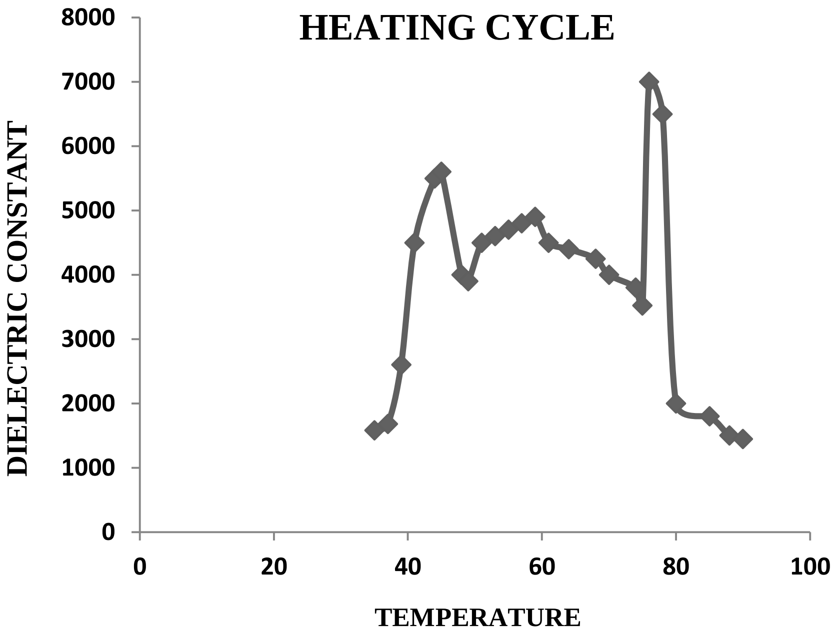

Fig 1. Temperature Variation Of Dielectric Constant Of $Cd(c_2h_3o_2)_2$ + Al_2o_3

Fig.3 shows the variation of τ_f as a function of aluminium oxide contant cadmium acetate .One of the advantages of Cadmium Acetate $Cd(C_2H_3O_2)_2$ with Aluminium Oxide Al_2O_3 is that by varying the Aluminium Oxide content, one can control temperature coefficient of resonant frequency τ_f without drastically affecting the other properties. A non-zero τ_f is often preferable to compensate for frequency variation due to the effect of temperature change on the resonator housing and dielectric support structure(11).

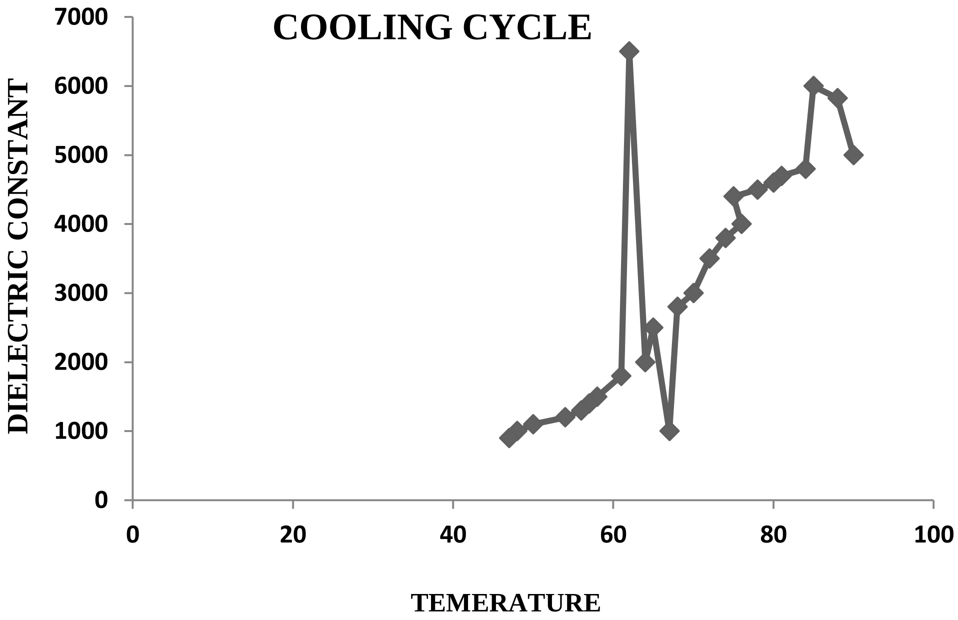

Fig 2. Temperature Variation Of Dielectric Constant Of $Cd(C_2H_3O_2)_2 + Al_2O_3$

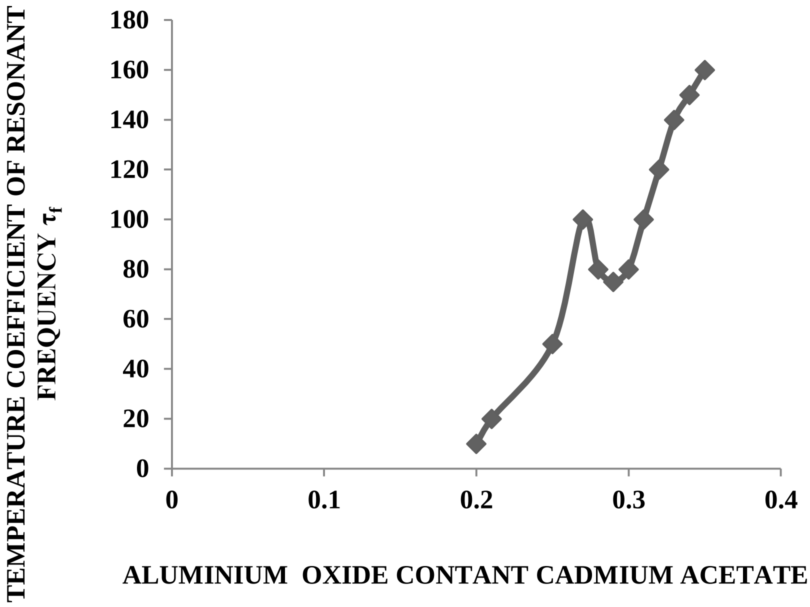

Fig . 3: Variation Of Temperature Coefficient Of Resonant Frequency T_f With Aluminium Oxide Contant Cadmium Acetate

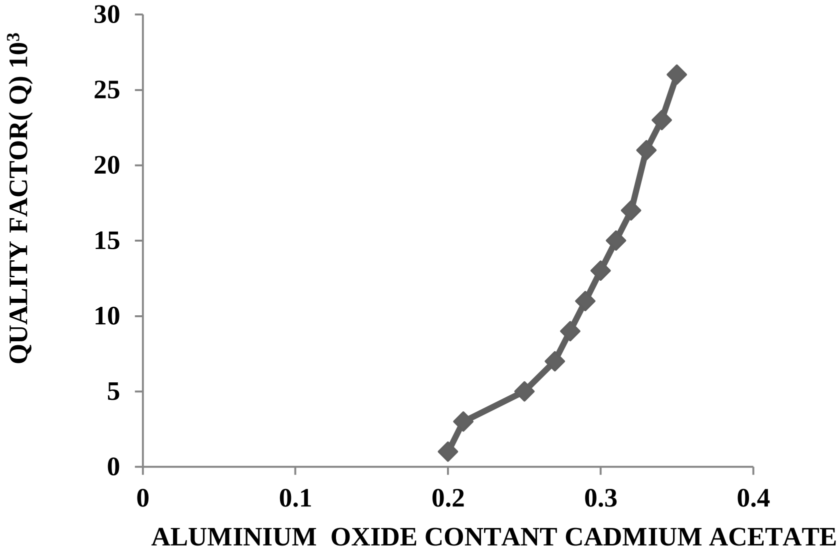

Fig.4: variation Of Quality Factor With Aluminium Oxide Contant Cadmium Acetate

Fig.4 shows the variation of quality factor as a function of Aluminium Oxide content Cadmium Acetate. By increasing Aluminium Oxide content in the mother compound, its quality factor also increases with a little effect on dielectric constant. In cadmium acetate, improvement in ordering increases the quality factor and the Q_f appears to increase with larger grains produced by longer sintering times, it is not the grain size itself that is controlling the Q_f. The effect of annealing the Cadmium Acetate on the dielectric properties was found that slow-cooled ceramic has a much higher quality factor as compared to rapidly cooled ceramic. Rapid cooling from the sintering temperature yielded a disordered structure having a low Q_f value. Solid solution of Cadmium Acetate $Cd(C_2H_3O_2)_2$ and Aluminium (III) Oxide Al_2O_3 is an important dielectric material with excellent properties which suggests for its valuable applications as dielectric material with excellent properties useful in ceramics engineering and communication system[12, 13].

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