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ORIGINAL ARTICLE

FERROELECTRIC PHASE TRANSITION IN SODIUM DIHYDROGEN ORTHOPHOSPHATE

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Abstract:

Ferroelectric transition and variation of dielectric constant with temperature in a compound is very important for the better application of the materials. In the present work we have studied the variation of dielectric constant with temperature of Sodium Dihydrogen Orthophosphate. During our investigation it is found that the temperature variation of the dielectric constant in heating cycle shows a very sharp jump in the dielectric constant, where the compound attains value from its value of 5600 to 42000 at the temperature of 42C (Fig. 1 & 2) which again falls in temperature range of 46^0*C to 51C. The cooling curve has nearly same nature, but has moderately low value of dielectric constant and also the peaks are less sharp. Sharp rise in the dielectric constant have been explained on the basis of activated polarization process and ferroelectric transition at* 46^0*C while the initial fluctuations in dielectric constant between* 27^0*C to* 46^0*C is due to the structural imbalance occurred by the elimination of water molecule from the compound. The occurrence of broad peak in is explained on the basis of relaxor systems which are generally due to heterovalent disorder. In the cooling cycle slight deviation in result have been explained on the basis of hysteresis and prolonged heating (annealing) of the specimen, as the annealing also shifts the transition temperature.*

KEYWORDS:

Ferroelectricity, Phase Transitions, Dielectric constant and Crystal Structure.

INTRODUCTION

The dielectric properties of amorphous solids are very important to the material scientists. Anomalous dielectric properties of amorphous materials discovered on ceramic specimen around 1943 by different group of scientists worldwide. The ferroelectric activities in amorphous crystals were reported in 1945-46 by Von Hippel and Co-workers [1] and independently by Wul and Goldman [2-3]. Owing to the extraordinarily structure sensitive character of the phenomenon of feroelectricity, it was only logical that, after the discovery of ferroelectricity in $BaTiO_3$, considerable effort would be expanded on a search for other ferroelectric materials with in the same family. This search was definitely fruitful, as it led to the discovery of ferroelectric activity in $PbTiO_3$, $KNbO_3$ and $KTaO_3$.These compounds have curie points scattered over a wide range of temperatures, while the room-temperature symmetry is characterized by very different distortions from the original perovskite cubic lattice.

The dielectric properties of single crystals were investigated extensively by Merz, [4] subsequently Cross [5] and a number of other authors extended and improved the results obtained by Merz. It is clear from the temperature dependence of dielectric constant of $BaTiO_3$ like crystals that, it is different corresponding to its different phase. As in the orthorhombic phase, where the crystal generally consists of a

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mixture of domains with different orientations of polar axis, one might at first expect that the measured values of the dielectric constant fluctuate around a certain mean value.

The structure of the compound under investigation shows its property resembling the spacific phase changes of the Disodium Hydrogen Orthophosphate [6]. It is also observed that in some ferroelectric crystals the dielectric response has a broad peak as a function of temperature; such crystals are also termed as relaxor systems. The origin of this behaviour is still controversial but it is most likely due to heterovalent disorder [7, 8].

The variation of dielectric constant and phase transitions in the compound Sodium Dihydrogen Orthophosphate have been explained on the basis of activated polarization process and ferroelectric transition at 46 ^{0}C while the initial fluctuations in dielectric constant between 27 ^{0}C to 46 ^{0}C is due to the structural imbalance occurred by the elimination of water molecule from the compound. The occurrence of broad peak , which are generally due to heterovalent disorder has been explained on the basis of relaxor system, as it is reported [6,8,9] in the case of disodium hydrogen ortho-phosphate and for tungsten- bronze type compound [10-13]. In the cooling cycle slight deviation in result have been explained on the basis of hysteresis and prolonged heating (annealing) of the specimen, as the annealing also shifts the transition temperature. The temperature sensitivity of the specimen suggests for its valuable applications as smart materials and the variation of dielectric constant with temperature gives the idea for its applications in the ceramics.

EXPERIMENTAL

The compound has been procured from E. Merk (India), Mumbai. The chemical was grinded into the fine powder in a 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 0-10 tonne wt/cm^2. A suitable die was used having rectangular cross-sectional area of the piston = 2.33cm^2.

The polishing of the pellets has been done to obtain smooth parallel surface to be used for electrode formation. The compound being very much soluble in water, therefore, instead of colloidal graphite, silver paint electrodes were used 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 [14]. The sample holder was directly fastened to the capacity measuring unit (Zenith M 92A).

RESULTS AND DISCUSSION:

We have selected the solid form of Sodium Dihydrogen Orthophosphate sample for the measurement.The chemical formula of the compound is NaH_2PO_4.2H2O. Its molecular weight is 156.01. The solid is colourless orthorhombic and has index of refraction 1.4629. The density of crystalline form is 1.91. The compound being very much soluble in water, therefore, instead of colloidal graphite, silver paint electrodes were used. The temperature variation of the dielectric constant in heating cycle shows a very sharp jump in the dielectric constant, where the compound attains value from its value of 5600 to 42000 at the temperature of 42C (Fig. 1) which again falls in temperature range of 46 to 51C. The cooling curve has nearly same nature, but has moderately low value of dielectric constant and also the peaks are less sharp (Fig.2). The temperature peaks are given in the following data.

Temperature peaks in Dielectric constant

Heating cycle C	Cooling cycle C
42^0C (42,000)	31^0C (3730)
52^0C(dip)(9700)	33^0C (9800)
	49 ^{0}C (1,16,000)

The fluctuation in the dielectric constant of sample in lower temperature (27^0C and 41 °C) range is perhaps due to the elimination of two molecules of water from the sample. Actually the elimination of water molecules from the crystal not only perturbs the physical measurements but also creates crystallographic defects, as observed in case of barium nitrate8. After the elimination of water molecules the crystal have its ferroelectric phase transition at 42°C, which can be interpreted in terms of Curie-Weiss law[12].

The transitions are of Ist order and has some hysteresis. The dielectric anisotropy in the antipolarized phase is not known. The decrease of with temperature about 45C is a commonly exhibited phenomena. The nature of increase of below 42 degree and its decrease about 45 clearly indicated $NaH_2PO_4.2H_2O$ has ferroelectric transition around this temperature. The spontaneous polarization measurement may be done to confirm the phase transition. The cooling curve has the anomaly of transition points, which may be due to prolonged heating (annealing) of the specimen, as the annealing also shifts the transition temperature.

The dielectric constant of the multidomain crystal of the compound is detected by a temperature measurement of the compound. The theoretical values obtained by the calculation of the free energy function determined from other data may say critically the nature of the dielectric constant of such a compound.

The curve for the cooing cycle is shown in the Fig.2. The cooling cycle of the compound shows a much profiled first order transition at the Curie temperature 49°C. The cooling does not follow heating curve because of the temperature relaxation of the compound.

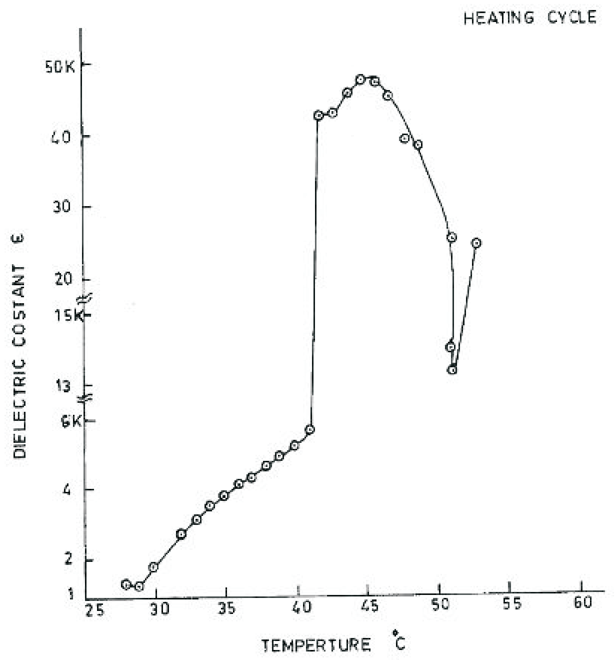

FIG. 1: VARIATION OF DIELECTRIC CONSTANT OF $NaH_2PO_4 \cdot 2H_2O$ PELLET WITH TEMPERATURE, p = 5 TONNE / 2.33 cm^2

Such a characteristics of a ferroelectric compound as a very sharp change of dielectric constant resembles that of Sodium nitrite measured along (010) and (101) as a function of temperature [15-16]. Above 49°C the dielectric constant of Sodium Dihydrogen Orthophosphate a Curie-Weiss law approximately (Fig.2) and the dielectric constant decreases inversely as the difference between the temperature and Curie temperature as it

is observed in case of Barium Titanate [17]. Actually ferroelectric transition in the sample develops due to structural phase change nearly at around 35°C. Around this temperature dipole moment develops due to the displacement of activated silver ions from its position, which causes the crystal change, similar as it is in case of Barium Titanate reported by Miss Megaw [18]. Similar explanation has been given for the variation of dielectric constant near Curie temperature in the case of Lead Nitrate [6] Barium Nitrate [8].

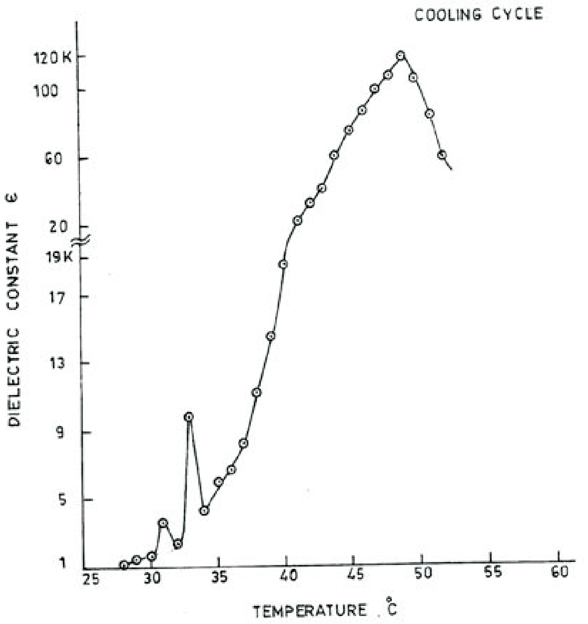

FIG. 2: VARIATION OF DIELECTRIC CONSTANT OF $NaH_2PO_4 \cdot 2H_2O$ PELLET WITH TEMPERATURE, p = 5 TONNE / 2.33 cm^2.

CONCLUSION:

In this work we have concluded that the change in temperature changes the structure causing the phase change in the compound and dipoles are created due to the active sodium ion get sufficient kinetic energy to face hindrance. The rotation and alignment of the dipoles shows the ferroelectricity in the compound. The presence of broad peak is explained similar to the relaxor systems, which are generally due to heterovalent disorder. The temperature sensitivity of the specimen suggests for its valuable applications as smart materials and the variation of dielectric constant with temperature gives the idea for its applications in the ceramics and as smart

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