

## On the changes in the electrical conductivity of thermo-electret and magneto-electret states of some solid dielectrics

K. D. RAY CHAUDHURI AND S. D. CHATTERJEE

*National Professor S. N. Bose's Laboratory,  
Indian Association for the Cultivation of Science, Calcutta 700032*

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The paper reports on the time-variation of electrical conductivity of the thermo-electret and magneto-electret states of carnauba wax, and nitrobenzene respectively. In either case, the following results have been obtained

- (a) An initial diminution in the values of electrical conductivity which is probably associated with the anisotropy in molecular crystals induced by superimposed electric or magnetic fields.
- (b) An augmentation in the values of electrical conductivity due to auto-ionization of aligned dipoles.
- (c) Restoration of a dynamical state of equilibrium between the two competing processes of spontaneous generation of carrier electrons and their subsequent trapping at suitable centres.

### 1. INTRODUCTION

Systematic studies of different aspects of thermo-electret phenomena like manifestation of surface charge, spontaneous charge reversal associated with hetero-charge and homocharge (Gemant 1935), polarization and depolarization current (Gross 1949), variation of thermal conductivity (Groetzinger 1936), optical anisotropy (Phillipova 1958), magnetic anisotropy (Chatterjee *et al* 1955, Polovikov *et al* 1964), dielectric constant (Chatterjee *et al* 1955) etc. have been made by different workers. But, curiously enough, very little work has been reported so far on the measurement of electrical conductivity of the electrets. In the present paper, we wish to report the time-variation of the electrical conductivity of thermo-electret and magneto-electret states. Two typical polar dielectrics, viz., carnauba wax and nitrobenzene have been selected for the purpose.

### 2. EXPERIMENTAL TECHNIQUES

The thermo-electrets and magneto-electrets were prepared in the usual way by the simultaneous application of field and temperature (Ray Chaudhuri *et al* 1973, Bhatnagar 1964, Chatterjee *et al* 1970). The d.c. electric field applied for the preparation of thermo-electrets was of the order of 10 KV/cm. The

magnetic fields used for the preparation of magneto-electrets of carnauba wax and nitrobenzene were 10 kilogauss and 28 kilogauss respectively. The fields were applied at the melting point (or freezing point) of the dielectrics.

The measurement of electrical conductivity of different polarized samples was carried out by a Tera Ohmmeter (TESLA BM 283).

### 3. RESULTS AND DISCUSSION

#### (i) Thermo-electret State

Figure 1 represents the time-variation of specific conductance of thermo-electret of carnauba wax. It is seen that immediately after the withdrawal of electric field, conductivity becomes less than that of the untreated control specimen. However, with passage of time, it increases and assumes maximum value on the tenth day. From then on, the specific conductance decreases amidst many pulsations, having an average value substantially higher than that of the untreated control specimen. Figure 2 illustrates the time-variation of specific conductance in the case of solidified nitrobenzene (273°K) after the removal of the electric field. Here, also conductivity at first decreases but shows a rapid rise above the corresponding normal value of the specimen. However, this apparent rise in the value of specific conductance is not permanent. Indeed, soon after reaching the maximum it declines gradually till the original value of the untreated control specimen is attained.

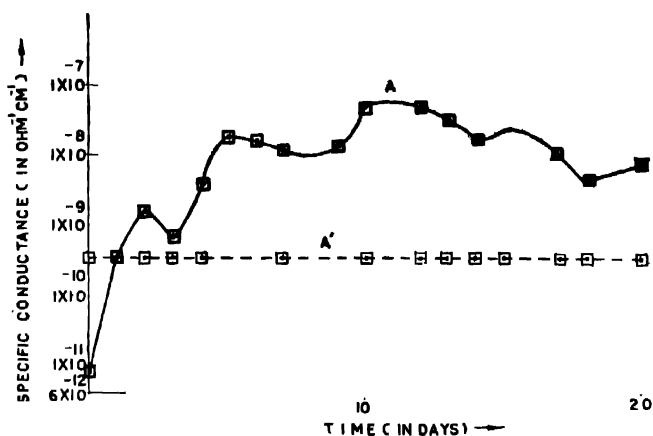


Fig. 1. Dependence of specific conductance (*A*) of carnauba wax thermo-electret on time. *A'* represents the value of specific conductance of the unpolarized control specimen.

To explain the results depicted in figures 1 and 2, one finds that the material under investigation, namely carnauba wax and nitrobenzene are polar dielectrics. The chief polar constituents of carnauba wax are myricil alcohol ( $C_{30}H_{61}OH$ )

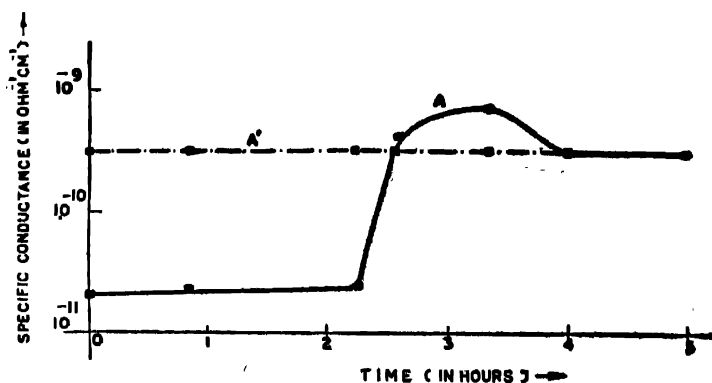


FIG. 2. Dependence of specific conductance ( $A$ ) of nitrobenzene thermo-electret on time.  $A'$  is the value of specific conductance of the unpolarized control specimen.

and cerotic acid ( $C_{26}H_{52}O_2$ ) (Koonce *et al* 1944). The presence of cerotic acid has been verified by micro-combustion analysis. The results of combustion analysis are the following :

$$C = 79.25\%$$

$$H = 12.88\%$$

$$O = 7.87\%$$

The dipolar characteristic of the molecular is due to the presence of  $O_2$ . It has a predominantly open chain structure. Nitrobenzene is the most familiar dipolar liquid being almost exclusively used in Kerr cells.

Evidently since both carnauba wax and nitrobenzene are polar compounds, the strong electric field applied during the formation of thermo-electrets tends to orient the dipoles in the direction of the field. However, at any temperature above absolute zero the random motion of thermal agitation works against orderly alignment. In a liquid state, the jostling of neighbouring molecules culminates in rapid disorientation. In general, however, the degrees of freedom of dipole molecules diminish considerably when a liquid dielectric solidifies. Thus, in solidified nitrobenzene or carnauba wax the dipole molecules remain aligned even after the removal of the external field. The initial diminutions in the values of conductivity as exhibited in figures 2 and 3, may perhaps be ascribed to dipolar orientation. Indeed, the lattice structure of the polarized dielectrics may be so modified that the directional motion of electrons decreases. As a result, the transport of electric charge through the thermo-electrets gets retarded and their conductivity becomes lower than the value of untreated control specimen as shown in figures 2 and 3. An analogous case is perhaps the diminution of conductivity of a strain gauge under applied stress.

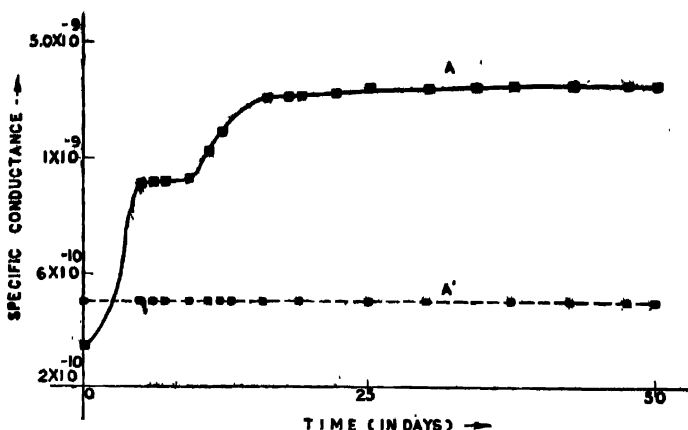


Fig. 3. Dependence of specific conductance ( $A$ ) of carnauba wax magneto-electret on time.  $A'$  is the value of specific conductance of unpolarized control specimen.

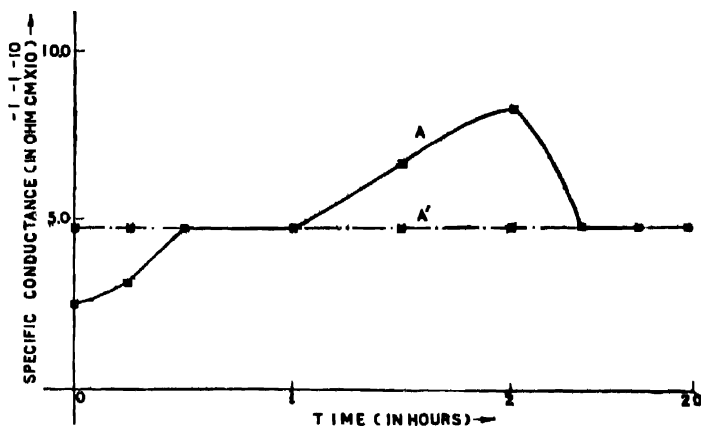


Fig. 4. Dependence of specific conductance ( $A$ ) of nitrobenzene magneto-electret on time.  $A'$  is the value of specific conductance of the unpolarized control specimen.

The subsequent rises in the values of conductivity of carnauba wax and nitrobenzene thermo-electrets can be ascribed to enhanced polarizability of molecules in strong electric fields. To discuss this effect in greater details let us consider two polar molecules at a distance  $d$  apart and with their dipole moments  $\mu$  pointing in the same direction. The electric field in this case is  $2\mu/d^3$ . Again, the energy of each dipole is

$$-\mu \cdot \mathbf{F} = -\mu(2\mu/d^3), \quad (1)$$

where  $\mathbf{F}$  = electric field.

Hence, the interaction energy for two dipoles oriented in the same direction along a common axis is  $-(2\mu^2/d^3) \times 2$ . Similarly, for the arrangement in which

the dipoles point at right angles to the line joining them and in opposite direction the energy is  $-(\mu^2/d^3) \times 2$ . Since the potential energy decreases with decreasing distance the molecules are attracted to one another for these relative orientations of the dipoles. It may easily be seen that the electric field has a large value in the neighbourhood of an ion or a dipole. Thus, the field strength at a point  $3\text{\AA}$  from an electronic charge is  $1.6 \times 10^8$  volts/cm, while at a distance of  $3\text{\AA}$  from a doublet of  $\mu = 3$  Debye ( $3 \times 10^{-18}$  e.s.u.) with a  $2\text{\AA}$  separation, it is  $0.7 \times 10^8$  volts/cm.

Usually, in the derivation of the expression  $\mu = \alpha F$  for the dipole induced in a molecule of polarizability  $\alpha$ , under a field strength  $F$ , the assumption is made that  $F$  is small. Sutton (1946) envisaged the possibility of a *hyperpolarizability* under such a large field, while applications, assuming a law such as

$$\alpha = \alpha_0 + \beta F^2 \quad (2)$$

were actually made by Matossi & Mayer (1948) and by Senftleben & Gladisch (1947). It has been suggested that, superimposed on the classical polarization is non-classical one involving electrons in  $\pi$ -type orbitals as in the case of nitrobenzene. Coulson *et al* (1952) investigated the dependence of molecular polarizability upon field strength. It was shown that the static polarizability increases with high field strengths. This effect does not normally become important in macroscopically attainable fields, but it may be a significant factor with fields such as those existing in the vicinity of an ion or dipole. As already pointed out that fields of the order of  $10^8$  v/cm do occur in the neighbourhood of an ion or dipole. However, for sufficiently large values of  $F$ , as was shown by Lanczos (1930) and Oppenheimer (1928) for an atom the probability of a molecule becoming ionized is so great that the whole concept of polarizability loses its significance. Probably, the first evidence of auto-ionization of molecules in the field of aligned dipoles was obtained by Chatterjee & Bhadra (1955) who detected an abnormal increase in the dielectric constant of carnauba wax electret. Later on, the present authors (1969) also detected the phenomenon in the case of nitrobenzene thermoelectret.

To interpret the results depicted in figures 2 and 3, we find that the increase of conductivity is due to the free electrons liberated from polarized molecules during the state of high field perturbation. It is equally likely that the electrons are transported from the valency band to the conduction band leaving requisite number of holes behind, which culminates in enhanced conductivity of the specimen.

The increase of conductivity of carnauba wax persists for a longer time, which may be attributed to the fact that the storage temperature is sufficiently lower than the melting point of carnauba wax. The rises and falls in the values of conductivity are due to the simultaneity of the processes of the release and

trapping of electrons at suitable sites within the volume of the material. Indeed, trapping of carriers in an organic crystal is a well-established phenomenon while detrapping is a source of generation of carriers. The traps are depleted by field emission and subsequently refilled. Trapping has been substantiated by Kallmann & Rosenberg (1955).

In the case of nitrobenzene, however, the operating temperature is 273°K which is very near its freezing point (278.7°K). And so, the influence of thermal agitation preponderates and forces the aligned molecules more easily into a state of disorder. During this process, the previously released electrons and holes recombine or are trapped at suitable sites. Thus, the initial value of conductivity of untreated control specimen of solidified nitrobenzene is quickly restored.

(ii) *Magneto-electret State*

Figure 3 shows the time-variation of conductivity of the magnetically treated specimen of carnauba wax. It may be noted that specific conductance exhibits an initial decrease followed by a rapid rise above the value of the untreated control specimen. Unlike the case of carnauba wax thermoelectret where the pulsating variations are rather frequent, the specific conductance in this case soon attains a saturation level. Figure 4 illustrates the time-variation of conductivity in the case of solidified nitrobenzene after the removal of magnetic field. It should be noted that a preliminary diminution in the value of conductivity is observed soon after the withdrawal of the magnetic field. However, with the passage of time, the equivalent value of conductivity of the untreated control specimen is gradually resumed which appears to persist for a while even afterward. Next, it begins to rise to higher positive values. However, this apparent rise is not permanent. As a matter of fact, after reaching maximum, it declines till the original value of the untreated control specimen is again attained. It is interesting to note that such variation is analogous to that obtained as the after-effect of electric field on the conductivity of solidified nitrobenzene. To explain magneto-electret state of solid dielectrics, Khare & Bhatnagar (1969) postulated the existence of two types of charges (a) a distribution of polarization called *idiocharge* arising due to dipole orientation, and (b) a distribution of polarization called *isocharge* originating due to the trapping of ions in macroscopic inhomogeneities. Evidences of the orientations of dipole molecules in liquids due to magnetic fields were obtained by Chatterjee (1936). He found that aromatic compounds like nitrobenzene, toluene etc. showed an increase in the coefficient of viscosity, while straight chain aliphatic compounds like normal propyl alcohol, normal butyl alcohol etc. showed a diminution of viscosity in a magnetic field. McMahon (1956) also detected an anisotropic increase of dielectric constant of nitro-wax under magnetic field. He attributed this to the diamagnetic anisotropy of molecules in nitro-wax. The present authors (1970) also observed a

variation of dielectric constant of nitrobenzene magneto-electret. It may reasonably be assumed that the strong magnetic field applied during the formation of magneto-electrets of carnauba wax and nitrobenzene produces orientations of the molecules comprising the dielectrics and consequently their lattice structure becomes altered. The initial diminutions in the values of conductivity as exhibited in figures 3 and 4 are ascribed to the slowing down of the directional motion of electrons due to the enhanced scattering by the newly formed lattice structure of the dielectrics. The manifestation of subsequent conductivity of carnauba wax magneto-electrets can be satisfactorily explained as in the case of corresponding thermo-electrets by the so-called electron emission mechanism.

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