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Condcution Mechanism In Polyethylene Terephthalate Under Electric Field And Temperaure

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Abstract

The electrical conductivity of polymer film (polyethylene terepthalate) film of thickness 100 microns has been investigated as a function if applied electric field and temperature under polarized and unpolarized conditions. The Ln J versus $E^{1/2}$ plots shows curvature at low field and linearity at high field region which signifies Schottky mechanism of conduction is operative. The activation energy is calculated from $\mathrm{Ln}\,\sigma$ versus 1/T plot within the low and high temperature region. As per Arrhenius plots, the decrease in activation energy with temperature and electric field suggests the delocalization of charge carriers as a result of increase in temperature and electric field.

Keywords: Poole- Frankel, polyethylene terepthalate, Arrhenius plot

INTRODUCTION

Polyethylene terepthalate is a thermoplastic polymer resin of the polyester family and is used in synthetic fibre; beverage, foods and other liquid containers; transforming application; and engineering resins often in combination with glass fibre. Depending on its processing and thermal history, polyethylene terepthalate may exist both as an amorphous and as semicrystaline polymer. The semicrystalline material might appear transparent (particle size < 500 nm) or opaque and white (particle size up to a few micron) depending on its crystal structure and particle size. Because of its high mechanical strength polyethylene terepthalate film is often used in tape applications,

such as the carrier of magnetic tape or backing for pressure sensitive adhesive tapesand also used as substrate in thin film and solar cell. With the advancement of science and technology polymer materials are used in spacecraft, electronic and telecommunication, medical science, even replacing of human organ. So, it is the main task to the researcher is to investigate the electrical, mechanical, thermal and chemical properties of polymer. Among the properties, the combined effect of electric and thermal energy on electrical conducting polymer is important one.

The study of electrical conduction in polymer is of considerable significance from two major points of view-firstly, for its own sake, because charge transfer characteristics are of fundamental interest and secondly for the information studies can provide the nature of electrical contacts, which may have a great influence on the measured electrical properties. The charge storage property of polymer can be considerably modified with the help of suitable dopants Khare *et.al*[1]. The broadest method that can be applied to enhance the conductivity of polymeric material in the formation of molecular aggregates or charge transfer complexes.

In conducting polymers, the effect of temperature and biasing voltage influence conductivity and in many semiconductor, transition temperature, pressure effects the conductivity such phase transition are found to be interesting phenomena. Electrical conduction in polymers has been investigated by several workers but a little has been reported about the nature of conduction both for polarized & unpolarized condition.

An attempt has been made to study the effect of electric field and thermal aging on conduction in polymer film having thickness 100 μm in polarized and unpolarized case for a range of temperature 293K and 373K and applied field to 0 to 1.45 x 10^7 V/m. The d.c. conductivity in polymer always lower than the a.c. conductivity at low temperature Syed *et.al*[2] . and is few order of magnitude less than a.c. conductivity and its variation with temperature is also different. It is expected because d.c. conductivity is determined by the most difficult transition in complete percolation path between the electrodes while a.c. conductivity is determined by easiest local movement of charges. But in the present study, due to more practical applications, d.c. conductivity is employed.

EXPERIMENTAL

The sample is cut circularly from the supplied sheet slightly larger than the contact surface area of the electrode having area 5.067×10^{-4} sq.m to avoid edge effect .The sample is placed between the electrodes under light constant pressure in the sample holder, such that the sample holder containing sample is connected in series with 1MΩ resistor. Then the sample holder is placed inside the thermostat bath Ultra Thermostat (U-10). Using the rotatory magnet attached in the thermometer the required temperature is adjusted in contact thermometer. The temperature is read out by the check thermometer. The required temperature is indicated by the single-lamp of the thermostat relay. All the measuring instrument are properly calibrated. The method for conductivity measurement was the same as that reported earlier, Karimi N

A *et.al*[3]. Now, different potential across the sample are applied by the power supply unit EHT-II. The value of potential across the sample is increased from 0 to 1450V by an interval of 50V, for a constant temperature. The potential drop across the 1MΩ resistor is recorded by the digital multimeter. The same operation is repeated for each different values of temperature from room temperature to 100°C by an interval of 5^0C. In order to the uniformly heating the sample is kept at constant temperature for 30 minutes for each consecutive reading.

After this operation the sample holder, containing sample is cooled down to room temperature and the sample is removed from the sample holder. A fresh sample of same type is replaced in the sample holder and heated up to 65^0C for 30 minutes for uniformly heating. At this constant temperature (i.e. 65°C) a potential of 1000 volt is applied across the sample for 1 hour. Then the sample holder along with the sample is cooled down to room temperature in presence of electric field (polarizing field). At room temperature the applied potential is removed and the sample is short circuited for 5 minutes to eliminate the stray of charges. This is the condition of electrically and thermally polarization. Again different values of potential are applied across the sample for a constant temperature. The potentials drop across the 1MΩ resistor are recorded as earlier.

This operation is repeated for different temperature. Hence, the conductivity was calculated after polarization. The current density J and the conductivity σ are calculated from the relations (3) and (4) and necessary graphs are plotted for different applied voltage and temperature from Ln σ and I/T curve the value of activation energy (E_a) for different regions are calculated from the relation

$$\sigma = \sigma_0 \exp\left(-\frac{E_a}{kT}\right)$$

where σ_0 is per-exponential factor and k is Boltzmann's constant.

The value of β is given by.

$$\beta = \text{slope} \times kT$$

Finding the slope from Ln J and $E^{1/2}$ the values of β are calculated for different sample at different applied voltage and temperature.

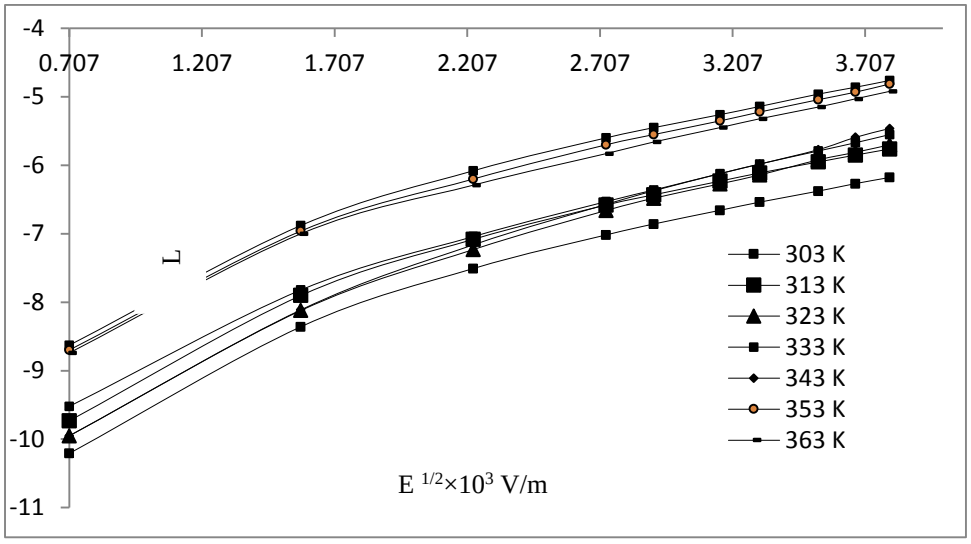

Figure -1. Plot of Ln J versus E ½ for unpolarized sample.

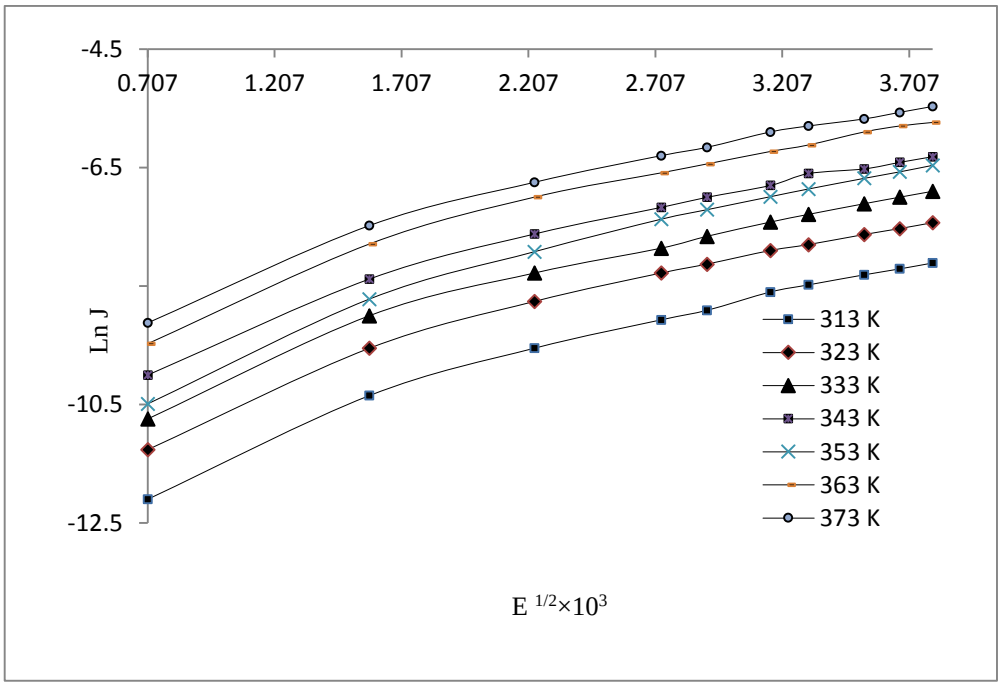

Figure -2. Plot of Ln J versus E ½ for polarized sample.

DISCUSSIONS

Ln J versus E½ plots: The plots of Ln J versus E½ for different samples before and after polarized are shown in figure 1 & 2. Such plots are considered for studying the conduction mechanism in the sample at high field region. These curves show the

curvature at low field and linearity at high field region. The straight portion of the plots at higher field shows different mechanism of conduction in the sample. The slope of the straight portion of the plot at high field is found out. In unpolarized case, the slope is greater than unity, while in polarized case, the slope is almost less then unity. The theoretical values of Schottky and Poole-Frenkel constants and their experimental values at different temperature for different samples are shown in table. When the sample is polarized, the curve, shows straight line at high field region i.e. above 2 x 10^6 V/m and show curvature at low field region (below 2 x 10^6 V/m).

Law field conduction: Figure shows the current voltage characteristics at different temperature of different samples, which is divided into low and high field regions. The plot of Ln J versus $E^{1/2}$, fig show that there is curvature in low field and straight line in high field. For the intermediate field $J \alpha E^{1/n}$, having n>1. The samples under the action of applied field are just to that in vacuum diode-space charge limited current, Mott and Gurney[4]. And also according to Aldert[5], this non-linearity of the plots is due to the space charge build up.

High field conduction:The plots of LnJ versus $E^{1/2}$ figures for different samples and different temperatures, show straight line in high electric field region, in which, $J\alpha E^{½}$ the case of hot electrons i.e. energy required by the field is greater than the thermal energy. The Schottky and Pool-Frenkel mechanism of conduction are considered Lamb[6], Ballan and Widdowson[7] as the dominant process of conduction in insulators or semiconductors at high field. Heavy reliance has been made on the measured slopes of Ln J versus $E^{1/2}$ for the interpretation of the experimental data.

It was found experimentally that, emission current increases with increasing field strength at the cathode, which is contradictory to that independence of current to the field strength at the cathode in thermionic emission. Schottky showed that a lowering of the work function due to an increase in the applied field was responsible for such behaviour. Hence due to high field Schottky emission of electrons may occur from the metal contact at the negative potential into the conduction band of the insulator. This mechanism corresponds to thermal activations of electrons over the metal-insulator interface barrier with added effect that the applied field reduce the height of the barrier. Considering the origin of the surface barrier, Schottky argued that, there are two reasons (i) polarization field and (ii) an image field. The Schottky effect is associated with the barrier at the surface of a metal and insulating material, whereas the Poole-Frenkel emission associated with the barrier in the bulk of material. In both effect the restoring force is due to coulomb interaction between the escaping electron and a positive charge they differ in that, the positive image charge is mobile with Schottky emission and fixed for Poole-Frenkel barrier. Hence lowering of barrier is greater for Poole-Frenkel than that for Schottky effect.

The current density for Schottky (S) and Poole-Frenkel (PF) process follows the relation Lamb D R (1967)

$$\mathbf{J = J_0 exp}\left(\frac{\beta E^{\frac{1}{2}}}{kT}\right) \qquad \text{... ... ...} (1)$$

Where $\mathbf{J_0 = AT^2 \exp(-\frac{\varphi}{kT})}$ *(Schottky mechanism)*

$\mathbf{J_0 = \frac{\sigma_0 V}{d}}$ *(Poole- Frenkel mechanism)*

T = absolute temperature, k = 1.36x 10^{-23} J/K Boltzmann constant

σ_0 = low field conductivity, V = applied field and d = thickness of the sample

$$\boldsymbol{\beta(s)} = \sqrt{(\frac{e^2}{4\pi\varepsilon_0\varepsilon})} \quad \textit{(Schottky constant)} \qquad \text{..... (2)}$$

$$\boldsymbol{\beta(pf)} = \sqrt{(\frac{e^2}{\pi\varepsilon_0\varepsilon})} \quad \textit{(Poole- Frenkel constant)} \qquad \text{..... (3)}$$

ε_0 = permittivity of free space and ε = permittivity of the material.

From equation (1), it is suggested that the plot of Ln J versus $E^{1/2}$ should be linear having slope $^{\beta}/_{kT}$. The value of β was calculated from such plot (putting k=1.38x10^{-23} J/K) for all temperature and samples.

The theoretical values of β (s) and β (p_F) were calculated from equation (2) and (3) using e=1.6×10^{-19} C, ε =3.0, ε_0 =8.854x10^{-12} F/m.

Calculated value of β from the slope of LnJ versus $E^{1/2}$ plots, compared with theoretical value of β_S and β_{PF}.

Table 1. Unpolarized sample

Temp. (K)	Slope *(of the order of 10^{-3})*	Experimental value *(of the order of 10^{-24})* β	Theoretical value *(of the order of 10^{-24})* β_S	β_{PF}
292	1.377	5.55	3.509	7.019
303	1.338	5.601	3.509	7.019
313	1.025	5.768	3.509	7.019
323	1.412	6.298	3.509	7.019
333	1.274	5.881	3.509	7.019
343	1.513	7.165	3.509	7.019
353	1.271	6.196	3.509	7.019
363	1.219	6.112	3.509	7.019
373	1.254	6.458	3.509	7.019

Table 2. Polarized sample

Temp. (K)	Slope *(of the order of 10^{-3})*	Experimentalvalue *(of the order of 10^{-24})* β	Theorityical value *(of the order of 10^{-24})* β_S	β_{PF}
292	1.635	7.065	3.509	7.019
303	0.913	4.073	3.509	7.019
313	0.863	3.967	3.509	7.019
323	0.878	4.158	3.509	7.019
333	1.011	4.925	3.509	7.019
343	0.873	4.373	3.509	7.019
353	0.882	4.543	3.509	7.019

Table shows the value of β is maximum and very near to the value of β_{PF} at temperature range from room temperature to 373 K for the sample before polarization. When it is polarized the value of β is high only at 313K, and for all the range of temperature 323K to 373K it coincides with β(s).

It is clear from the table that in unpolarized sample, the Poole-Frenkel mechanism of conduction is operative whereas in polarized sample Schottky mechanism of conduction is operative, i.e Poole Frenkel mechanism of conduction changes to Schottky mechanism of conduction when the sample is polarized. Hence Schottky emission is best observed for most samples in the present investigation. Thus the value of β is smaller in polarized condition than in unpolarized condition.

Temperature dependence of conductivity: The sample shows two peaks at 333 K and 353K in figure, which is close to those reported Ranjit Singh[8], in current thermogram of polyester (PET, ICI), carefully analysis reported in determination of transition temperature of the sample by finding the breaks in Ln σ versus 1/T plots.

The temperature dependence of conductivity can also be understood from then activation energy, which can be calculated from the relation.

$$\sigma = \sigma_o \exp\left(-\frac{E_a}{kT}\right) \quad \text{.....(4)}$$

Where σ_o = pre exponential factor, E_a = activation energy, T = absolute temperature, k = Boltzmann constant

On basis of cluster model Perepechko[9], it may be supposed that the low temperature transition, which is weakly pronounced, is associated region with unfreezing of segmental motion of disorder in the sample. The high temperature transition which is

more distinctly pronounced to be characterized by change in TCσ is apparently associated with the unfreezing of segmental motion inside the more ordered region. In polarization sample, only one peak is observed such that, this peak is shifted by 10K with low current from the unpolarized sample which may be due to the depletion of excess charge, creating a lower deriving field and charges may get swept less quickly towards the electrodes.

Polarization dependent electric conductivities: The electrical conductivity σ_u of unpolarized sample (virgin) was measured at different temperature and field. Next the sample was polarized by keeping it at constant voltage and at constant temperature for 1hour then allowed to cool down to room temperature under the same electric field. After polarization the values of electrical conductivity (σ_p) was measured at different temperature and field. The electrical conductivities were compared for sample when it was virgin i.e. unpolarized. This new technique has been employed for the study of polymer made electrets. This is the basic tool to identify and to evaluate the dipole orientation or forming the injection charges through contacts. This depends upon weather the sample receives the charge of same sign or opposite to that of electrode i.e. homocharges or hetrerocharges to which the surface of sample was in contact during polarization.

It was observed in our case that electrical conductivity of unpolarized sample (σ_u) is generally greater in almost all samples than (σ_p) the electric conductivity after being polarized.

It is well known that there are different mechanism Pillai[10] for generation of change in polarized sample such as.

(i) polarization due to the polar side group/dipole alignment.

(ii) ion trapped in defects on dislocation in crystalline region.

(iii) space charge builds up due to the migration of ions over microscopic distances.

(iv) the space charge injection from electrodes.

The first two mechanisms give a volume polarization with heterocharges where the polarization takes place with the sign of heterocharges due to dipole alignment or charge separation within dielectric i.e. dielectric absorption. The third gives non-uniform homocharge or heterocharge depending upon electrode-dielectric work function. It was observed in our case that electrical conductivity of unpolarized (σ_u) i.e. virgin sample is generally greater in almost all samples than (σ_p) the electrical conductivity after being polarized. On the other hand the current density (J) was found for different applied electric field both for virgin and polarized samples. From the plots of Ln J versus $E^{1/2}$, the value of β was calculated and compared their theoretical values i.e. Schottky (β_s) and Poole-Frenkel constant (β_{PF}). Schottky effect is associated with barrier at the surface of the metal and the polymer sample. The effect

of barrier height by applied field and hence the surface barrier was explained by Schottky, who gave two reasons for the effect such as (a) the polarizing field and (b) an image field. It was found that the values of β is higher for the unpolarized sample. But after the sample is polarized the value of β is always smaller and coincides with the theoretical values of β_s i.e. Schottky constant, evident with all samples. This result further supports that the space charge injection takes place from electrodes i.e. homocharge injection through connecting electrodes.

CONCLUSION

The plots of Ln J versus $E^{1/2}$ show curvature at low field region but straight line in high field region for both polarized and unpolarized samples. Poole-Frenkel mechanism of conduction changes to Schottky mechanism of conduction for sample at high temperature. The slope of Ln J versus $E^{1/2}$ at high field region at room temperature for the unpolarized sample shows Poole-Frenkel mechanism of conduction. From the above discussion/observation it can be concluded that the sample will useful in many electronic devices.

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