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A.C. Conductivity Measurement on Compressed Pellet of Pyrene

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Abstract

Dielectric constant, the dielectric loss and A.C. conductivity have been measured on compressed pellets of Pyrene from low frequency upto 30MHz and also its temperature variations are studied.

The observed conductivity has been found to satisfy the relation:

$$\sigma_{ob} = \sigma_0 \mathrm{Exp}\left[-Eg/2kT\right] + A.\omega^n \mathrm{Exp}\left[-E(T,\omega)/kT\right]$$

The results have been discussed with reference to hopping of carriers and coherent band model.

Keywords: A.C. Conductivity, Pyrene, Electrical Conductivity, Dielectric constant, Hopping, Band model.

Introduction

The Electronic conduction mechanism in Organic solids is a problem, which is seeking appropriate explanation even yet today. The measurements in the last 4, 5 decades did not conclude reproducible results. The experiments insisted on fake compensation plot to include the varying results. This compensation diagram could use the varying values of activation energy (E_{act}) and pre-exponential factor (σ_0) of the Arrehenius plot [1-5]. A serious objection about non-reproducibility of the results was indicated by a series of measurements on ultra pure organic crystals by workers in our laboratory [6]. Highly pure zone refined [7] materials were subjected to single crystals production under ideal conditions. The long-range order single crystals with highly precise methodology could yield highly reproducible and high value of activation energy [8].

The straight Arrehenius plot even up to low temperature yielding highest value of activation energy ever recorded, was so encouraging to force us to re-investigate the entire results obtained by eminent workers world wide [9-10]. We have serious objections about the different models and theories proposed to justify the experimental results obtained by different group of workers. We conclude that if the crystal is ultra pure and has long-range order and all necessary conditions (annealing of the crystal, reduction of surface states, use of inert ambient and non injecting electrodes, properly shielded measuring setup and use of noise free cable), are met 'Band structure' exists for organic solids also [11]. Our group has reported highest values of mobility comparable to Silicon of the order of several cm^2/v-sec, for pretty number of solids [12].

Such results need theoretical calculations for a series of organic solids. The pure and defect free organic solids can now be prepared in single crystal and their film forms for measurements for nearly all types of organic solids [13]. Band structure calculations for such solids are needed in view to obtain the parameters for such compounds and to be compared with the experimental results.

Compressed pellets of Pyrene is used for the measurement of ac and dc conductivities on the basis of which the conduction mechanism in Pyrene has been described. The ac method of conductivity measurement for the compressed pellets (amorphous or fine crystalline) has been used, both for organic and inorganic semiconductors to eliminate the contact resistance by capacitative short circuiting [9,14]. The general trend was to study the variation of capacity and the conductivity from low frequencies upto higher frequencies. The experimental dielectric conductivity includes all dissipation processes including the ohmic conductivity due to actual transport process and the experimental dielectric constant includes all polarization process.

Experimental

The compound Pyrene has been procured from NBS Biological, 14 Tower Huntingdom Cambs England. The specimen has been grinded into the fine powder in a agate mortar, avoiding direct sunlight and preferably the most of the sample preparation work was done at night. Pellets were prepared with the compression machine (Flexural 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-section of the piston - 2.33cm^2.

The pellets were polished to obtain smooth parallel surfaces and colloidal silver paint was used to form the electrodes. The sample was annealed at 2/3 of its melting point in a furnace for about 8-10 hours. The acrylic sample holder is used for high temperature measurement. To measure the dielectric loss and dielectric constant of the sample, this sample, holder is directly fastened to Q-meter. The sample holder has shielding envelope to avoid any electrical pickup. For our calculation we used the formula derived by Gupta and Misra, of our laboratory [10].

Results and Discussion

The frequency dependence of the conductivity from dc to 30 MHz of Pyrene has been obtained at different temperatures (shown in Fig.1). It is supposed that for a small interval of frequency, activation energy for hopping is constant [15] and the equation $\sigma \alpha \omega^n$ gives the value of n. The values of n so calculated for the frequency ranges at different temperatures have been given in Table 1.

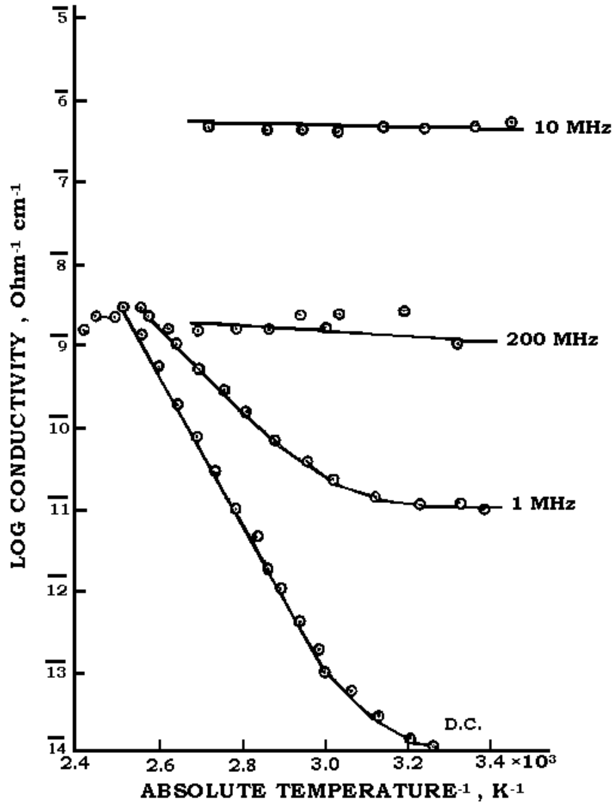

Figure 1: Log conductivity vs. inverse temperature plot for pyrene, C.P., P=5000kg wt/2.33cm^2.

Table 1: Values of n in the relation $\sigma \alpha \omega^n$ for the compounds.

Compound	Temp. °C	dc to 10^3	Frequency (Hz)						
			10^3	10^4	10^5	10^6	10^7	relaxation	10^8
Pyrene	30	0.8	0.8	0.8	0.9	1.0	1.1	1.2 (16 MHz)	1.0
	70	0.7	0.9	-	1.1	0.7	0.7		0.5
	90	0.2	0.6	1.1	0.9	0.8	0.7		0.5
	120	0.1	0.5	1.0	0.7	0.7	0.6		0.4

The observed conductivity can be represented by a general formula-

$$\sigma_{ob} = \sigma_0 \mathrm{Exp}[-Eg/2kT] + A.\omega^n \mathrm{Exp}[-E(T,\omega)/kT] \tag{1}$$

where the first part is dc band conductivity (in general sense), Eg/2 is the activation energy of band conduction, k is Boltzman constant, T is absolute temperature and σ_0 being the pre-exponential factor. At low frequencies, as the temperature increases the exponent n decreases and becomes very small (0.2 or 0.1) at higher temperature. It thus suggested that, as temperature increases the conduction mechanism is dominated by intrinsic band conduction. At higher frequencies and low temperatures, the observed value of n=1 shows that in these situations hopping mechanism predominates.

The temperature variation of conductivity at different frequencies (shown in Fig. 2) shows that the activation energy is highest for dc and it decreases as the frequency increases, (Table-2).

Table 2: Electrical conductivity parameters of Pyrene compressed pellets, P = 5000 kg.wt./2.33 cm2, graphite electrodes.

Name of compound	Activation Energy (E_{act}) eV.		σ_0 ohm^{-1}cm^{-1} (dc)	$\sigma_{20°C}$ ohm^{-1}cm^{-1} (dc)
Pyrene	dc	ac		
	1.8		6.2x10^{9}	5.0x10^{-15}
		1.4 (1 KHz)		
		0.6 (100 KHz)		
		02 (10 MHz)		

It is found that as the temperature is increased, the ac conductivity seems to approach the dc conductivity asymptotically at all frequencies. Consequently the frequency dependence is found to decrease with increasing temperature. This conclusion is justified from the σ (ω) vs 1/T plots at different frequencies as it is also found in case of phathalocynanine (pcpb) [16]. The analysis of data obtained for selected compound, in context of equation (1) tells us that at higher frequencies second part of this equation becomes dominating; with increasing temperature at reasonably higher frequency, although exponential part increases due to increase in temperature but as the power of ω decreases, the net effect is a slow increase with temperature; therefore a low apparent activation energy than dc is observed[17-18]. At very high frequencies it is just possible that increase in the exponential part with temperature may be compensated by an equal decrease in the ω^n and therefore the conductivity may found to be unaffected with temperature (Fig. 2). Above discussion reflects the opinion that the hopping model is more applicable [11] for some organic solids at low temperature and high frequencies whereas the intrinsic band model is useful at high temperatures and low frequencies [19]. Similar explanation has been suggested for the ac conductivity of Anthrone [20].

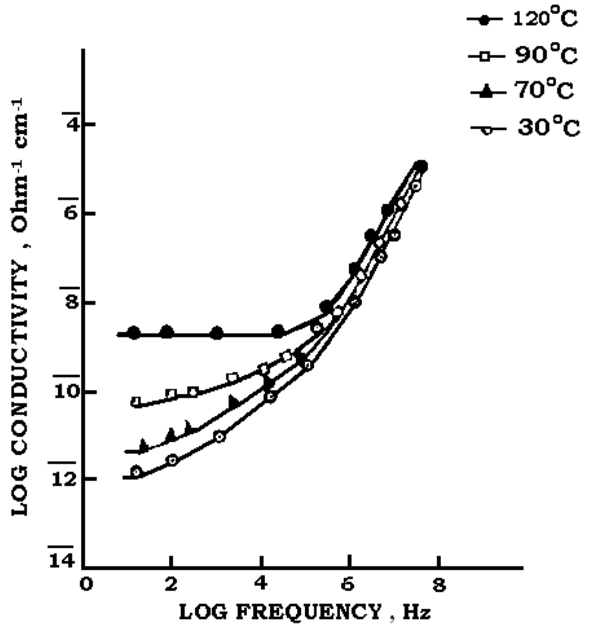

Figure 2: Log conductivity vs. log frequency plot for pyrene, C.P., P=5000 kg wt/2.33cm^2.

The results of our sample can also be explained on the basis of two phase R-C model network [21]. According to which the conductivity of sample in the frequency range dc to 10^5 Hz is true value of its conductivity because at these frequencies the interparticle contact capacity (C_c) in highly compressed pellet is greater than particle capacity (C_S)[22,9], being in series does not contribute to the total capacity. From the Fig.1, it is apparent that the conductivity of the sample (reciprocal of resistivity, R_p) shows an increment in its value with frequency approaching a value of 10^{-7} ohm^{-1} cm^{-1} at 10^8 Hz. It can be explained easily if we replace the simple equivalent network by more complex network including a frequency dependent term in resistance representing a dipolar loss [23].The apparent increase in conductivity is undoubtedly due to the some short of dipolar effect. An explanation for the relaxation at high frequencies and polarization at lower frequencies, which is due to the presence of air gap between specimen and electrode, is given by Miliotis and Yoon [24]. The above may also be reason for the high frequency loss observed in such a non-polar substance like Pyrene. The conducting properties of Pyrene and its variation with temperature and frequency have important applications in the treatment of grapheme/epoxy nanocomposites.[25]

Conclusions

Our results show that the activation energy is higher for dc which decreases with increase in frequency. It is also found that ac conductivity approaches to dc conductivity asymptotically at all frequencies as the temperature increases. Frequency dependent is found to decrease with increasing temperature. At low frequency and higher temperature the conduction mechanism is due to band conduction while at high frequency & low temperature the conduction mechanism is dominated by hopping.

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