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TRANSPORT PROPERTIES IN 2,6-DIAMINO ANTHRAQUINONE

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Current density–voltage characteristics were obtained from 2,6-diamino anthraquinone samples using ohmic aluminium electrodes. Results showed that at low voltage the conduction process was ohmic, while at high voltage space-charge-limited conduction controlled by a single dominant trap level was presented. Thickness dependence measurements proved that the trapping sites were located at a discrete energy level. The transition voltage, V_t, between ohmic and space-charge-limited conduction was approximately proportional to the square of the sample thickness and was found to be temperature independent. The temperature dependence of ohmic and space-charge-limited current densities have been investigated. The results were interpreted in terms of extrinsic nature of ohmic conduction. Traps with density $\approx 2\times 10^{24}$ m^{-3} located at 0.50 ± 0.03 eV below the conduction band edge have been observed.

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1. Introduction

The study of the temperature dependence of ohmic and space-charge-limited (SCL) currents in semiconductors and insulators is capable of providing considerable insight into the mechanism of charge transport and carrier trapping in these materials. The group of organic compounds known as anthraquinone is convenient for such a study. Anthraquinone (AQ) and its derivatives are a class of organic compounds which fall under the group of disperse dyes [1]. These compounds are of interest because they can promote photo-tendering of textiles. AQs derivatives are widely used as commercial dyes for both natural and synthetic fibers. The dark and photoconduction of AQs have been investigated [1–4] trying to explain the origin of the observed conductivity of such compounds in terms different modes of carrier generation.

In this paper we report measurements of ohmic and SCL currents in 2,6-diamino anthraquinone (2,6-DAAQ), with charge carriers injected from the electrodes and generated in the bulk towards understanding of the conduction mechanism in the 2,6-DAAQ compound.

2. Experimental details

The 2,6-DAAQ used in these measurements was purchased from Alderich Chemical Company Inc. (USA) and was purified by vacuum sublimation. The original powdered material was compressed in a die into pellets of 1–4 mm thickness and which were fitted with evaporated film electrodes of aluminium to serve as ohmic contacts for electrons to this materials [1]. Silver paste contacts with the electrodes have been used for studying the electrical measurements. The specimens were mounted onto an electrically heated copper disc, the temperature of which could be held constant to within 1 K over the range of 300 K to 373 K. The temperatures were measured by means of chromel/alumel thermocouple mounted in close proximity to the specimen of interest. The current flowing through a specimen was determined using a conventional dc technique and Keithley 610 electrometer. When the voltage was applied, the currents were allowed to increase to a steady value within few seconds. For consistency the currents were allowed to stabilize for 2 minutes before a reading was taken. All samples used were measured by means of this technique.

3. Results and discussion

The Seebeck effect in 2,6-DAAQ was investigated by measuring the electromotive force ΔV which developed across the specimens when a temperature difference ΔT was established across them. The experiment was carried out to establish the sign of the majority of carriers. Results varied slightly from a specimen to a specimen, but invariant electrons were observed to be the majority of carriers over the entire temperature range of 300–375 K. The smooth curve of $Q = \Delta V/\Delta T$ versus T is shown in Fig. 1. Recently Narasimharaghavan et al. [1], from their photoconduction studies, have shown that the AQs are n-type semiconductors with electrons being the majority of charge carriers. Figure 2 shows a typical set of current density–voltage (J–V) characteristics appropriate to the temperature indicated. The curves are characterized by two different slopes, which are approximately 1.0 and 1.9 for the low voltage and high voltage, respectively. Hence 2,6-diamino anthraquinone, in common with other organic materials, exhibits ohmic conduction at low applied voltage and space-charge-limited (SCL) conduction governed by a discrete traps at high voltage [5]. The ohmic behaviour in the low voltage region is due to the negligible injection of carriers from the contact and the initial current is governed by the thermally excited free carriers. Hence, within the ohmic region, the current density J_Ω is given by [6]

$$J_\Omega = ne\mu V/d, \tag{1}$$

where n is the concentration of thermally generated electrons, e — the electronic charge, μ — the mobility, d — the sample thickness and V — the dc applied voltage.

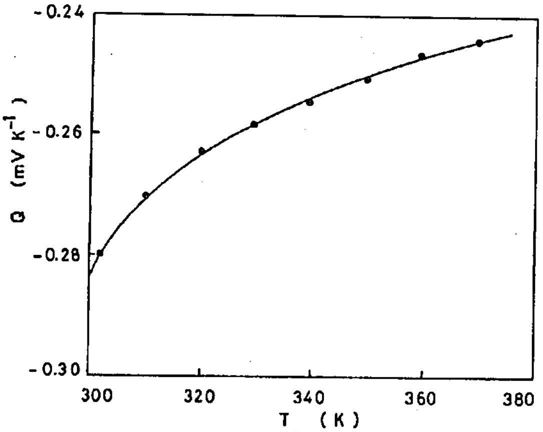


Fig. 1. Temperature dependence of the Seebeck coefficient for n-type 2,6-DAAQ.

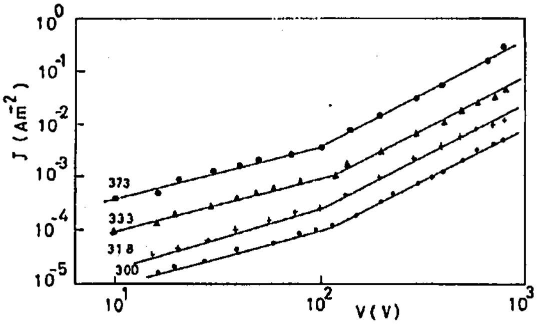


Fig. 2. Current density–voltage curves of different temperatures. Temperatures are given in K.

Within the SCL conduction region, the discrete trap distribution results in a current density, J_{SCL}, an expression given by [5]

$$J_{\mathrm{SCL}} = \frac{9}{8}\Theta\varepsilon\mu\frac{V^2}{d^3}, \tag{2}$$

where in addition to the previously defined symbols, ε is the permittivity and the trapping factor Θ is the fraction of the total number of charge carriers which are free.

Following the theory of Lampert [5], Eq. (2) is valid for the injection of one type of carrier only in the presence of a single discrete trapping level. Both J_{Ω} and J_{SCL} are thermally activated, their activaction energies being contained in n and Θ, respectively. According to Eqs. (1) and (2) the V_{t} voltage, at which the current passing through a specimen converts from being ohmic to SCL, can be defined as:

$$V_{\mathrm{t}} = \frac{8}{9}\frac{end^2}{\varepsilon\Theta}. \tag{3}$$

Clearly, V_t has a thermal activation energy equal to the difference between activation energies for ohmic and SCL conduction. Thus, V_t is temperature dependent unless the two activation energies are identical.

It can be seen from Fig. 2 that the voltage $V_t = (210 \pm 10)$ V, at which the transition from ohmic to SCL behaviour takes place, is independent of the temperature, it implies that the sample is extrinsic [7].

For n-type material containing N_d donors and N_a acceptors per unit volume (a partly compensated specimen), the density of free electrons responsible for ohmic conduction is given by [7]

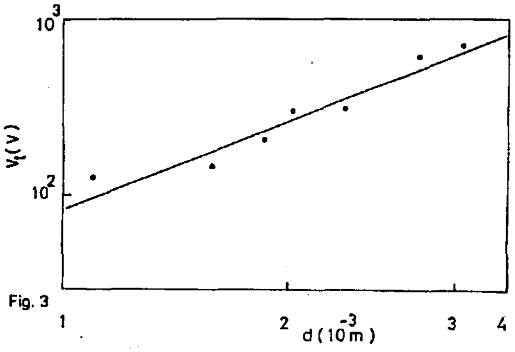
$$n = \frac{(N_d - N_a)}{N_t} N_c \exp(-E_t/kT), \tag{4}$$

where N_c is the effective density of states in the conduction band, E_t — the energy below the conduction band edge at which the traps are located with the state density N_t, k — Boltzman's constant and T — the absolute temperature.

The electron trapping factor, Θ, for SCL conduction is [8, 9]

$$\Theta = \frac{N_c}{N_t} \exp(-E_t/kT). \tag{5}$$

The measurements of the sample capacitance of a known thickness yielded a permittivity of $\approx 2.05 \times 10^{-11}$ F m^{-1} (or a relative permittivity of ≈ 2.30) and this value was used in all the following calculations. These measurements were performed at 100 KHz using Tesla BM 507 impedance meter. Using the set of Eqs. (3), (4) and (5), the donor excess was found to be $(N_d - N_a) \approx 7 \times 10^{15}$ m^{-3}. The dependence of V_t on d as predicted by Eq. (3) allows further verification of the model. In Fig. 3 the dependece of V_t on d is shown. The slope of this plot is ≈ 1.9 which is in good agreement with the expected linear dependence of V_t on d^2 from Eq. (3).



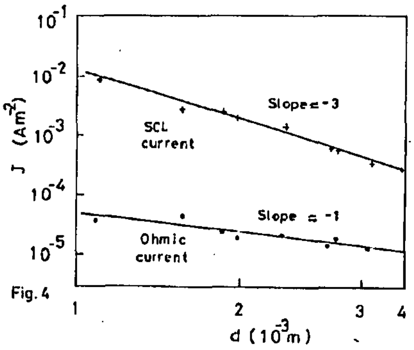


Fig. 3. Dependence of transition voltage V_t on thickness d.

Fig. 4. Current densities vs. thickness sample for ohmic and SCL conduction. (Temperature 300 K. Applied voltage 25 V and 500 V for ohmic and SCL currents, respectively).

A distingushing characteristic of single-carrier SCL current by a single trapping level is the thickness dependence given by Eqs. (2) and (5). In Fig. 4 the dependence of current density versus sample thickness for both ohmic and SCL regions is shown. It is evident that ohmic conduction and one carrier SCL currents dominated by a single set of traps are observed in our material. More information about material is obtained from the temperature dependence of the SCL and ohmic current densities. The applied voltage of 600 V and 40 V were chosen so that the data for both SCL and ohmic regions could be conveniently shown on the same figure. For all the samples studied, the thermal activation energies for ohmic and SCL conduction are identical within an experimental error. Following Schmidlin and Roberts [10], the data shown in Fig. 5 is associated with extrinsic behaviour and a discrete set of traps level, E_t, located at (0.50 ± 0.03) eV below the conduction band edge. Good straight lines are obtained, thus indicating that lattice scattering is dominant. Terefore, the product of mobility and the state density is relatively weakly dependent on temperature in agreement with the prediction of the band model.

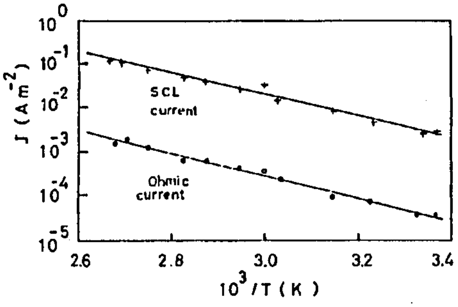


Fig. 5. Variation of current density with reciprocal temperature for ohmic and SCL currents.

The intercept on the $\log J$ axis for SCL conduction is given by $\log(\mu N_c/N_t)$ and immediately yields the trap density, N_t, provided that μ and N_c are known. Assuming the effective density of states in the conduction band to be of the order of the density of molecules (approximately 10^{27} m^{-3}) and using electron mobility to be $\approx 1 \times 10^{-4}$ m^2 V^{-1} s^{-1}, then the value of N_t was found to be 2×10^{24} m^{-3}. The nature of traps in 2,6-DAAQ is not clear at the moment, however structural imperfection is a very likely candidate.

4. Summarizing remarks and conclusions

Current density–voltage measurements showed ohmic and space-charge-limited conduction at lower and higher voltage levels, respectively. The slopes of the space-charge-limited $\log J$ versus $\log V$ characteristics were approximately 1.9, while those of $\log J$ versus $\log d$ characteristics were approximately -3. These results are entirely compatible. The transition voltage, V_t, between the ohmic and space-charge-limited conduction was approximately proportional to the square of the sample thickness, also in accordance with the theory. The thermal activation energies for the ohmic and space-charge-limited regions were approximately identical, it implies that a sample is extrinsic. The value for each region has been found to be ≈ (0.50 ± 0.03) eV, which suggests a trap located at this energy distance from the conduction band edge.

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