Isothermal Transient Current Studies in Cellulose Acetate Films

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Synopsis

Step voltage transient current studies have been made in cellulose acetate films as a function of field and thickness. A logarithmic plot (Scherr–Montroll plot) of the transient current vs. time gives a knee at a time t_T, which is interpreted as the transit time of the charge carrier. The value of the carrier mobility has been estimated to be 3.9×10^{-9} $cm^2 \cdot V^{-1} \cdot s^{-1}$ in cellulose acetate film. The carrier mobility in iodine-doped (2% w/w) cellulose acetate film has also been determined from Scher–Montroll plot and is found to be 3.3×10^{-7} $cm^2 \cdot V^{-1} \cdot s^{-1}$.

INTRODUCTION

The determination of carrier mobility is of fundamental importance in understanding the conduction mechanism in dielectrics, but, due to the involvement of high impedance, the mobility measurement in dielectrics usually encounters many difficulties. The study of transient currents in insulators gives valuable information about the charge carrier generation, transport, and recombination. Mobility can be deduced from transient current techniques such as the time of flight measurements.[1,2] In these measurements, charges are strongly injected under high fields by energetic electrons or photons. However, a simpler technique is to inject carriers from electrodes under the influence of a step voltage.[3–5] In principle, from these techniques, it is possible to measure directly the transit time and hence the charge carrier mobility.

A widespread characteristic of transient current measurements of carrier mobility, particularly in case of low-mobility materials, is the occurrence of appreciable dispersion in the transit pulse.[6] The anomalous transit time dispersion may result in a considerable smearing of the transit edge and makes the identification of transit time difficult on the observed current traces. In this case, a featureless current trace is generally obtained. Such a type of behavior has been encountered in the study of amorphous semiconductors,[7–9] polymers,[2,10,11] and As_2Se_3.[12] Various theories[13–20] have been put forward to account for the anomalous dispersive charge transport behavior observed in these materials. As discussed in these theories, the transit time, even in such cases, can be identified by plotting the current and time on the logarithmic axes.

In this paper, we report the results of step voltage current transient studies carried out on cellulose acetate films. A logarithmic plot (Scher–Montroll plot) of the transient current vs. time has been given, from which the transient time of the carrier front between electrodes has been identified. Also, the depen-

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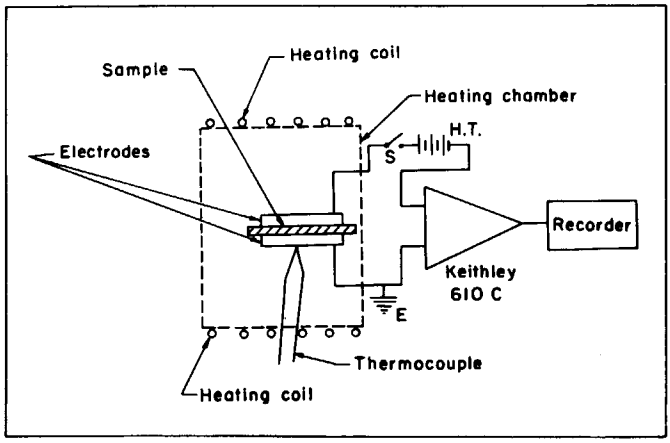

Fig. 1. Schematic diagram of the experimental setup.

dences of transient currents on thickness and field have been investigated. The transient currents in iodine-doped cellulose acetate samples have also been investigated.

EXPERIMENTAL

The films of cellulose acetate and cellulose acetate:iodine (2% w/w) were prepared by the method described elsewhere.[21] The films of 2.5 × 1.5 cm size and 25–100 μm thickness were employed in the present investigations. The surfaces of the films were chemically cleaned with ethanol and deionized water and then dried. The films were heated at 50°C for 12 h to remove the residual solvent.

The transient current studies were made in a way as shown in the schematic diagram of Figure 1. All measurements were performed on a sandwich cell using copper electrodes. The transient current was measured with a Keithley 610C Electrometer in conjunction with a DigiGraphic 2000 X-Y/t Recorder. The

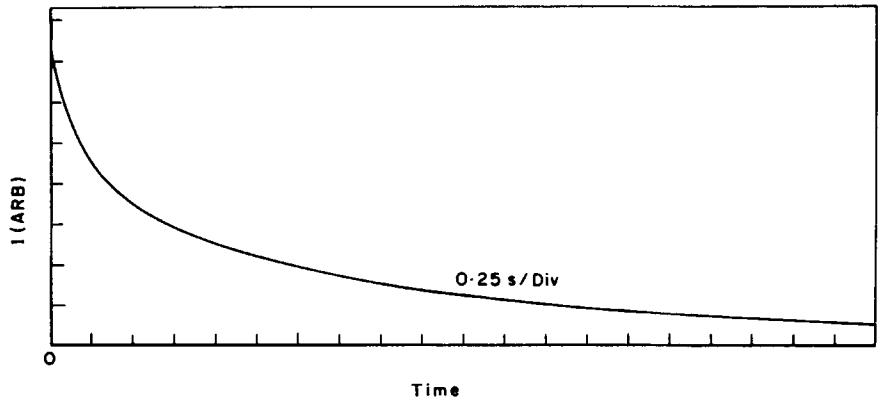

Fig. 2. A typical step voltage transient current trace for cellulose acetate film at field = 3.2×10^5 V·cm^{-1}.

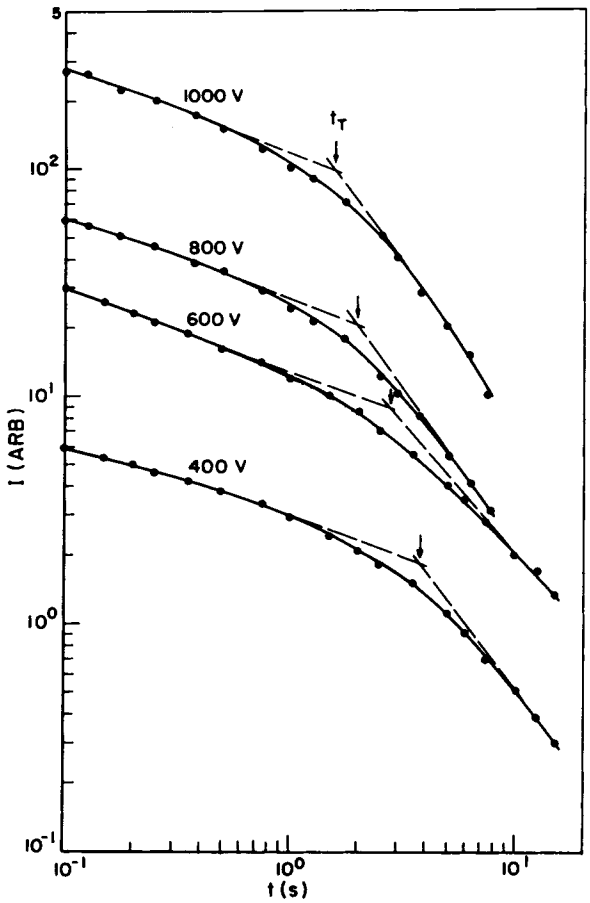

Fig. 3. Transient current vs. time plots on logarithmic axes at different voltages for cellulose acetate film illustrating the effect of applied voltage on t_T.

response of the recorder is 1/3-s full scale maximum and input resistance is 1 M Ω. In the present studies, a field range of 10^5–4×10^5 V·cm^{-1} was employed. The magnitude of the initial currents varied between 10^{-8} and 10^{-7} A for pure cellulose acetate samples. The measurements were performed for +Ve step voltage mode.

All the measurements were taken in laboratory atmosphere at temperature 26°C and relative humidity 50–60%.

RESULTS AND DISCUSSION

Current traces at various fields are obtained after the application of a step voltage on the cellulose acetate films sandwiched between two plane parallel copper electrodes. These current traces are found to be entirely featureless, and no characteristic time can be identified on these traces. A typical current-trace shape at an applied field of 3.2×10^5 V·cm^{-1} is shown in Figure 2. As may be seen from Figure 2, there is no "flat part" or any kind of peak in the current transient, which may correspond to the characteristic time.

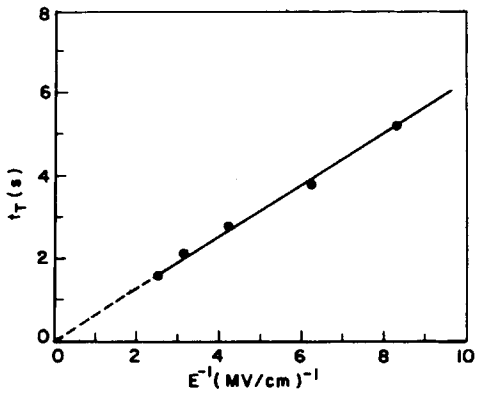

Fig. 4. Dependence of the transient time t_T, on the applied field in cellulose acetate film.

In Figure 3, we have replotted the current traces obtained at various fields in logarithmic scales. In these units, the transient current appears essentially as two approximately linear lines of different negative slopes, thus indicating an algebraic time dependence of the current. The intersection of the tangents to the initial and final part of the current trace clearly defines a characteristic time. As may also be observed from Figure 3, the characteristic time decreases with the increase of applied field. The plot of the characteristic time (t_T) vs. inverse of the applied field is found to be a straight line (Fig. 4). It shows that t_T varies inversely with the magnitude of the applied field in the range of the field studied.

The dependence of t_T on the sample thickness is also studied. Figure 5 shows the plot of t_T vs. sample thickness at a fixed field 10^5 V·cm^{-1}. t_T is found to be proportional to the specimen thickness for the range of thicknesses used in the present study.

From Figures 4 and 5, it seems clear that the characteristic time obtained from the plot of the current trace in logarithmic scales corresponds to the transit time of the charge carriers injected at the source electrode. The carrier mobility (μ)

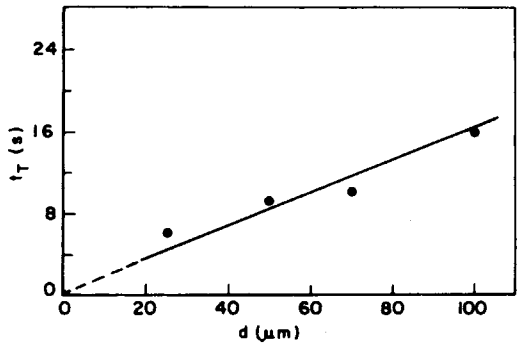

Fig. 5. Dependence of the transit time t_T, on the sample thickness in cellulose acetate film at field = 10^5 V·cm^{-1}.

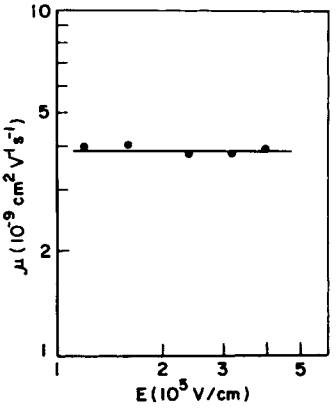

Fig. 6. Electric field dependence of the carrier mobility in cellulose acetate film.

may hence be evaluated in the usual manner from the relation

$$\mu = d/Et_T \tag{1}$$

where d is the sample thickness and E the applied field.

The values of the carrier mobility, after using expression (1), are plotted in

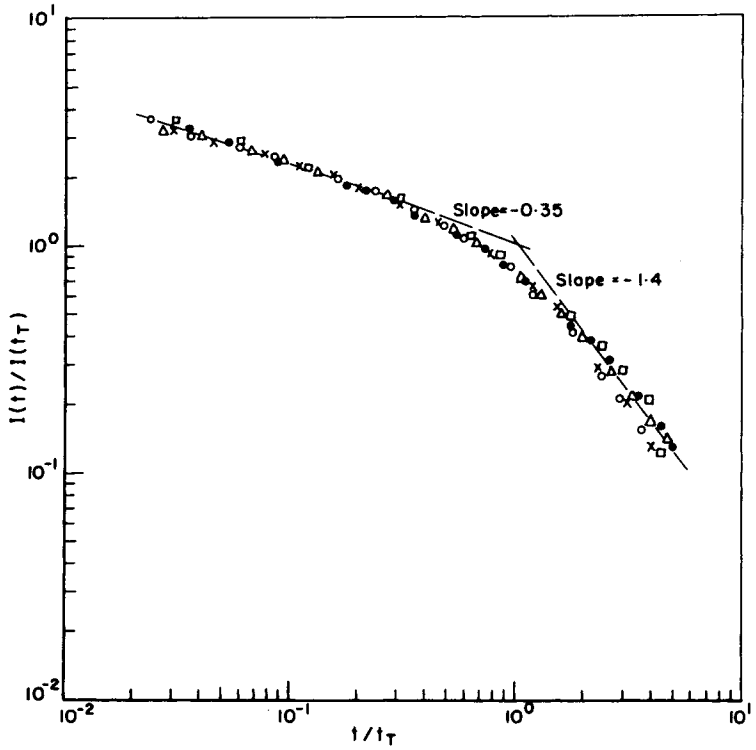

Fig. 7. Scher–Montroll plot (log i vs. log t) normalized by $I(t_T)$ and time t_T for cellulose acetate films. 25 μm: (×) 1000 V; (○) 800 V; (●) 600 V; (△) 400 V; 50 μm: (□) 500 V.

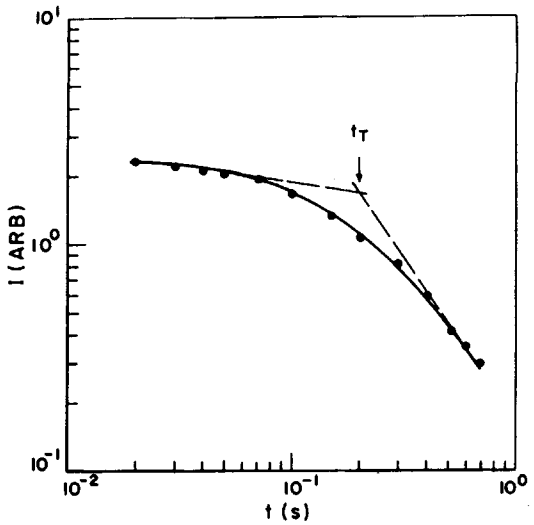

Fig. 8. Scher–Montroll plot of transient currents in iodine doped (2% w/w) cellulose acetate film at field 10^5 V·cm^{-1} and $d = 50$ μm.

Figure 6 as a function of applied field. From Figure 6, it may be observed that the carrier mobility is independent of the applied field in the range studied. The average mobility found from Figure 6, is nearly 3.9×10^{-9} cm^2·V^{-1}·s^{-1} and is in good agreement with the value 3.5×10^{-9} cm^2·V^{-1}·s^{-1} obtain from steady-state current-voltage measurements.[22]

For analyzing the featureless transient currents, Scher and Montroll[13] have suggested phenomenologically that log I vs. log t curve shows a knee at time t_T, which is interpreted as the transit time of the charge carrier. Scher–Montroll[13] analysis is based on the hopping mechanism and the distribution function of hopping waiting time is assumed to be

$$\Psi(t) = At^{-(1+\alpha)} \qquad (0 \leq \alpha \leq 1) \tag{2}$$

As α is constant with respect to applied field and specimen thickness, the universality of the current transients shape is expected on normalized current and time axes. The universality of the transient currents in cellulose acetate is demonstrated in Figure 7.

The preceding discussion of the present results, on the basis of Scher–Montroll[13] model, at first instance, reveals that the charge transport in cellulose acetate may be described by the hopping mechanism. However, as discussed by other workers,[14–20] if the carrier transport is controlled by traps distributed in the gap and characterized by a distribution function of eq. (2), the current traces will exhibit similar features. From this, the trap-limited mechanism also looks probable in cellulose acetate. Since trapping effects are likely to occur in the bulk of polymers,[23] the traps may be thought to play a predominant role in charge transport process. However, it is not yet clear whether the conduction process in cellulose acetate involves hopping or trapping. Further investigations like the temperature dependence of transient currents and thermally stimulated currents are needed to identify the exact conduction mechanism. Work in this direction is in progress.

Transient current studies were also performed on the iodine-(2% w/w) doped cellulose acetate films. The Scher–Montroll plot of the transient current in iodine-doped sample at an electric field of 10^5 V·cm^{-1} is shown in Figure 8. Taking the time corresponding to the knee in the Scher–Montroll plot as transit time, the mobility is estimated to be 3.3×10^{-7} cm^2·V^{-1}·s^{-1}. This value is larger by about 2 orders of magnitude than of the undoped cellulose acetate and is in reasonably good agreement with the value $\sim 10^{-7}$ cm^2·V^{-1}·s^{-1} obtained from steady-state current voltage measurements.[22]

CONCLUSIONS

(i) Scher–Montroll plots of the step voltage transient currents in cellulose acetate films have been given, from which the transit time of the carriers has been identified.

(ii) Carrier mobilities in pure and iodine-doped cellulose acetate have been found to be 3.9×10^{-9} and 3.3×10^{-7} cm^2·V^{-1}·s^{-1} respectively, at room temperature.

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