Space Charge Behavior of Polyimide Film

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Abstract: Space charge distributions in polyimide films strongly depend upon electric field, temperature, water content and so on. When a low dc field was applied, positive and negative homo space charges were observed near the respective electrodes. When the applied field was higher than 30 MV/m, additional positive and negative hetero space charges were also observed. The amounts of these charges depended upon applied field. The temperature dependence of the amounts of homo space charge showed a thermally-activated process. After drying by the heat treatment, the homo space charges were much reduced at 296 K and at 333 K. This suggests that the charge injections from the electrodes are strongly affected by absorbed water. The hetero space charges were not affected by drying. After short-circuiting, the homo space charges quickly decreased but the hetero space charges were very stable.

INTRODUCTION

Polyimide (PI) is widely used as a high-temperature insulating material because of its good insulating, mechanical and thermo stable properties. Although the electrical properties of PI were investigated by many researchers, the results were scattered depending on the experimental conditions such as temperature, sample thickness, electrode materials, humidity, etc [1-5]. Various conduction models such as ionic hopping conduction [1-3], thermally activated injections [1, 3, 4] and thermally assisted tunnelling [5] were reported. The high field properties, however, are not so clear still now. On the other hand, PI is a hydrophilic polymer and its dielectric properties largely depend upon absorbed water or humidity [6, 7]. In this paper, we studied the space charge distributions under dc high fields and the effect of absorbed water were discussed.

EXPERIMENTAL

Kapton 500H films with thickness of 125 μm were used. Some of samples were kept at 423 K (150°C) for 24 hour in order to remove absorbed water. The space charge distributions were measured by the pulsed electro-acoustic (PEA) method (Five Lab, PEANUTS) [8]. The measurements were carried out under dc fields for 90 minutes and after short-circuiting for 30 minutes at 296 K (23°C) or 333 K (60°C). The strength of applied field was mainly 50 MV/m. The electrodes were the grounded Al plate and the semi-conductive (SC) layer connected to a high-voltage dc source.

RESULTS AND DISCUSSIONS

Field Dependence

Figure 1 shows space-charge distributions in PI films at 296 K under the short-circuit condition after 90 minutes charging at dc fields between 10 and 50 MV/m. Negative and positive charges were injected from the Al cathode and SC anode, respectively, forming homo space charges [9]. When an applied field was larger than 30 MV/m, positive and negative hetero space charges were also observed in the bulk. The amounts of these four space charge components increased with applied field.

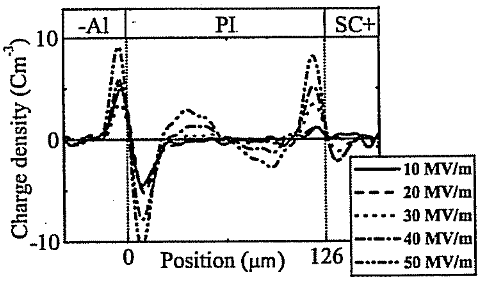

Fig. 1 Space charge distributions at various fields (296 K, positive polarity, short-circuited after 90 minutes charging).

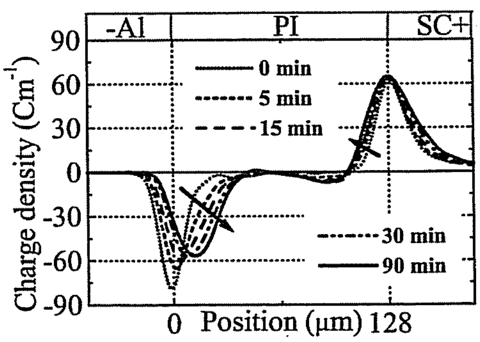

Fig. 2 Temporal change of space charge distribution under the field of 50 MV/m at 333 K.

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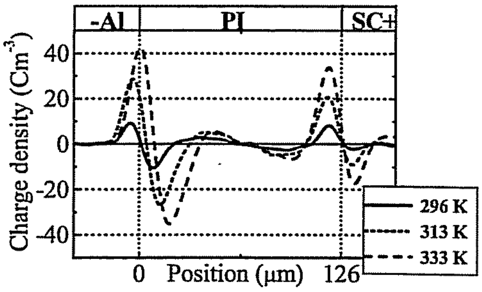
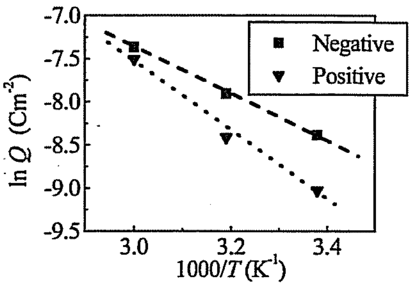
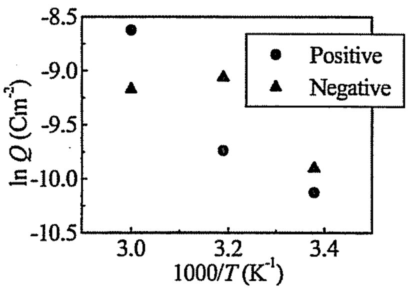


Fig. 3 Space charge distributions by 50 MV/m charging at 296, 313 and 333 K (after short-circuited, positive polarity).



(a) homo space charge



(b) hetero space charge

Fig. 4 Temperature dependences of (a) the amounts of homo and (b) hetero space-charges.

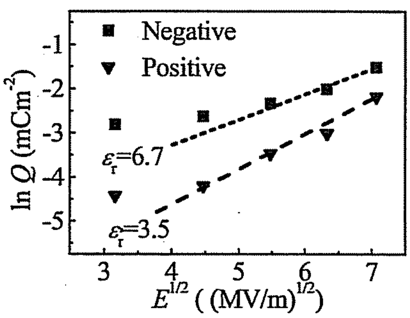


Fig. 5 Shocttky-plot of negative and positive homo space charge amounts at 296 K with fitted lines of dielectric constants ε_r = 6.7 and 3.5, respectively.

The temporal change of space charge distribution under the applied field of 50 MV/m at 333 K (Fig. 2) suggests that negative and positive charges are injected from the Al cathode and SC cathode, respectively, and migrate into the bulk. The migration of homo space charge is prevented by the adjacent hetero space charge which gradually increases with time.

The space charge distributions were similar when the polarity of applied field was reversed. Although Sessler *et al.* reported the dependence of steady-state current densities upon electrode materials such as evaporated Au, Al and Ag [4], the difference in charge injection between Al and SC was not so remarkable. The reason of this discrepancy may be due to the temperature and therefore water content in the sample.

Effects of Temperature

Figure 3 shows the space charge distributions after charging at 50 MV/m at 296, 313 and 333 K. The amount of homo space charge Q increased with temperature T, suggesting the thermally activated injection.

By assuming that the amount of homo space charge is proportional to the injection rate, the apparent activation energies of the negative and positive charge injections were roughly estimated from Fig. 4 (a) as 0.23 and 0.34 eV, respectively. The hetero charges also increased with temperature but the dependences were not clear as shown in Fig. 4 (b). This is partly because the adjacent large homo space charge overlaps and masks the hetero one.

Similarly, we can estimate the apparent dielectric constant ε_r from the plot of amounts of negative and positive homo space charges Q with respect to the square root of applied field E. The fitted lines in Fig. 5 correspond to $\varepsilon_r = 3.5$ for hole injection and 6.7 for electron injection, respectively. These values are similar to or larger than that of PI (3.4) reflecting the effect of water.

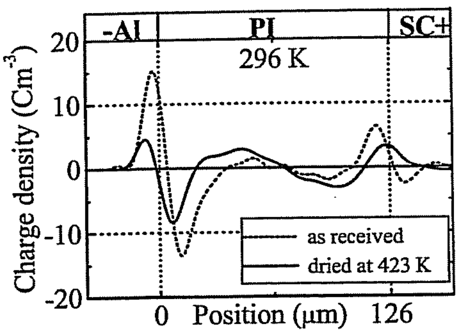
Thus, the temperature and field dependences can be explained by the Schottky injection

$$Q \propto \exp\left(\frac{\phi - \beta_S \sqrt{E}}{kT}\right),$$

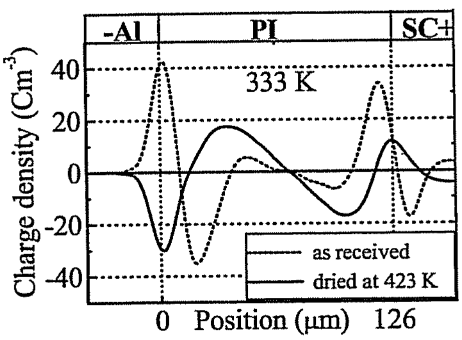
where ϕ is injection barrier, $\beta_S = \sqrt{e^3/(4\pi\varepsilon_r\varepsilon_0)}$, e elementary charge, ε_0 the vacuum permittivity.

Effects of Humidity

For examining the effects of absorbed water, dried samples were prepared by keeping PI films at 423 K (150°C) for 24 hours. The positive and negative homo

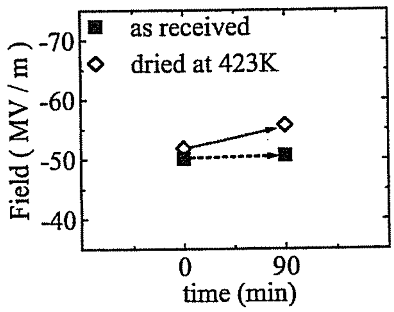


(a) 296 K

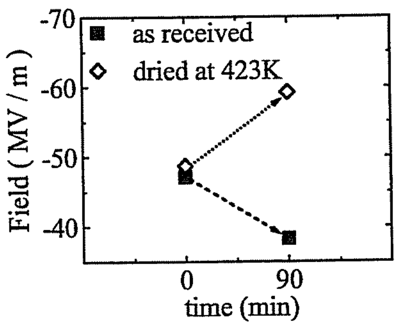


(b) 333 K

Fig. 6 Effects of absorbed water; space charge distributions at (a) 296 K and (b) 333 K after short-circuited (90 min. charging, positive polarity).



(a) 313 K



(b) 333 K

Fig. 7 Fields near the Al cathode at (a) 313 K and (b) 333 K of non-treated (open square) and heat-drying PI films (solid circle) .

space charges at 296 K (Fig. 6 (a)) were largely reduced by drying and disappeared at 333 K (Fig. 6 (b)). The reduction of homo space charges by drying may suggest that water absorbed in PI film assists the charge injection. Therefore, we concluded a thermally-activated and water-assisted charge injection from the experimental results of space charge distribution in non-dried PI films.

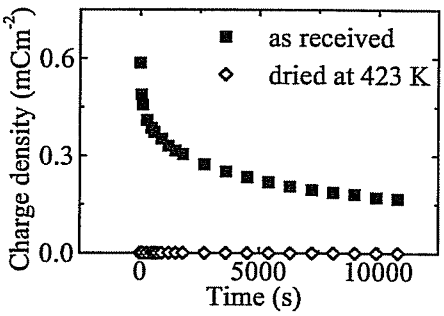
In contrast, the hetero space charge did not show any big change by the drying. This may suggest that the absorbed water does not play an important role in the behavior of the bulk carriers. Otherwise, it was reported that absorbed water in a thick PI film (> 50 μm) was removed only from the surface region of PI film but not from the inside part by the drying [7]. Hetero space charges may be affected by remaining water after drying.

The absorbed water affects the space charge distributions, and therefore the field distribution in a sample. The field near the cathode in a non-dried sample slightly increased at 313 K or decreased at 333 K, but that in a dried sample increased both at 313 K and at 333 K as shown in Fig. 7. The cathode field at 333 K was enhanced about 20 % of the applied field. Although the homo space charges quickly decreased after short-circuiting, the hetero space charges decayed very slowly (Fig. 8). Thus, the field distribution changes depending on the environmental conditions such as humidity and temperature. We should take into account this fact in practical use.

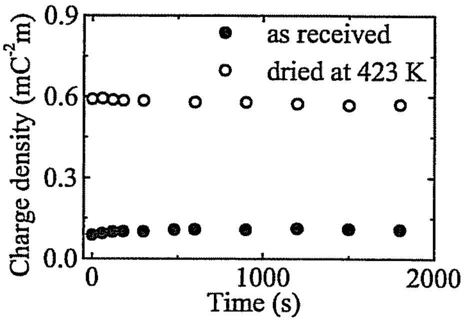
The estimations of activation energy and dielectric constant were very rough. They should be estimated from temporal change in charge density and the fields near the electrode at least. Other experimental results such as transient charging and discharging currents are also necessary for discussing the mechanisms more precisely. These experiments are being carried out.

SUMMARY

Under high fields, positive and negative charges injected from the electrodes, forming homo space charges in a polyimide (PI) film. When the applied field was higher than 30 MV/m, positive and negative hetero space charges were also observed. The injections of negative and positive carriers were reduced by removing absorbed water, but the hetero space charges were not affected by water content.



(a) negative homo space charge



(b) positive hetero space charge

Fig. 8 Decay of (a) negative homo and (b) positive hetero space-charges near the Al cathode at 333 K.

The polyimide is widely used as a high temperature insulating material but the water content strongly affects the space charge or the field distribution at high temperatures, at 333 K (60°C) for instance. We should take into account the environmental condition in practical use.

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