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The Effect of Space charge on Isothermal and non-isothermal Currents in Polyethylene Naphthalate

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Abstract

This paper presents an experimental results obtained by using three investigation methods. The temperature effect on space charge behavior, in semi crystalline polyethylene naphthalate (PEN), under electrical strength, was analyzed. The Anomalous charging current, which is generally attributed to space charge, was observed. The non-isothermal current exhibited two peaks one located at 130°C near the glass transition temperature T_g, and the other, which was associated to pre-melting of crystallites, at 150°C. The DSC analyze showed a T_g at 128°C and an endothermic peak at 150°C. From these results it can be concluded that the trapped space charge can not release from the sample and requires an increase of temperature. Above T_g, the molecular motion facilitates the de-trapping of the charge.
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1. Introduction

Due to their excellent electrical properties, polymers were widely used electrical insulation field. However, in particular conditions, especially under high electrical field, their insulating properties may be affected, as the space charge composed of trapped charge carriers, can have a drastic effect on dielectric properties, which influence tremendously the reliability of apparatus and systems where they are integrated. Indeed, this charge can increase the local electrical field and subsequently cause breakdown of the insulator.

The aim of this work was to make a correlation between the trapping of space charge and transient currents, by comparing the results obtained by three techniques. The material used in this study was the polyethylene naphthalate (PEN) which gives high performance films with excellent thermal mechanical and electrical characteristics.

Measurements were done in a large temperature-range including the glass transition temperature T_g so as to analyze the effect of polymer state, glassy or rubbery, on electrical properties.

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2. Material

Polymers are currently used as dielectrics in a wide range of electro-technological applications. Since the polyethylene naphthalate (PEN) gives high performance films with physical, thermal and chemical characteristics better then those of polyethylene terephthalate (PET), it is used as a substitute for the latter in a large number of applications, particularly in film capacitors.

The PEN film of 25 µm thickness, and 1.36 g/cm^{3}density was provided by Dupont de Nemours Luxembourg. The PEN sample was prepared with gold evaporated electrodes, 2 cm in diameter, on both sides of the sample. The chemical structure of PEN is reported in Figure1.
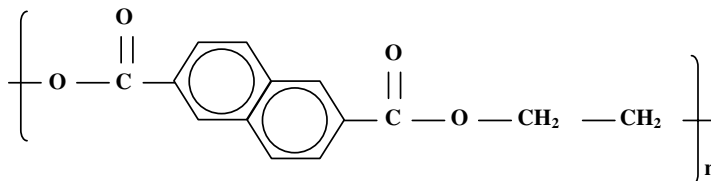
Fig 1 : Chemical structure of PEN.

3. Experimental and method

3.1. Transient current technique

A DC voltage was applied to a sample at a constant temperature, and the charging current was measured up to three hours. The voltage was then removed and the discharging current of the short-circuited sample was recorded for three hours too, according to the experimental setup [1]. The experimental cell was connected to a vacuum pump and a gas cylinder. This allowed to control the nature of the gas inside the cell, and to remove the moisture from the sample. A temperature controller connected to the heating element was located inside the cell. Hewlett Packard stabilized power supply was used to deliver DC voltages up to 4000V. The current was measured by Keithley 617 programmable electrometer driven by a microcomputer. A high voltage relay switches from polarization when the sample was exposed to the voltage, to depolarization when the sample was grounded.

A new technique combining isothermal charging and discharging with non-isothermal discharging measurement proposed by Neagu et al [2], was used.

In the second step of the experiment, the sample was heated at a constant rate of 2.4°C/min, from a charging temperature T_c to a final temperature T_f = 180°C. The current recorded was called "final thermally stimulated discharging current" or "non-isothermal discharging current".

3.2. DSC Analyze

The thermal analysis of PEN samples was obtained by using the TA instruments (Differential Scanning Calorimeter, DSC Q10 t). DSC thermograms were recorded from 30 to 300°C by a constant heating rate of 10°C/mn under a nitrogen atmosphere to avoid thermal degradation. The obtained data was analyzed by TA Universal Analysis Software.

4. Isothermal charging and discharging currents

Isothermal charging current, at different temperatures between 100 and 150°C, under a DC field of 72×10^6 V/m, were presented in figure 2. These results showed an anomalous behavior. Indeed, instead of monotonous decrease, a rise after few minutes of polarization [3], was observed, which was generally attributed to space charge effect.
Figure 2: Isothermal charging current in PEN at different temperatures Several works have reported the existence of these peaks with different polymers, like polyethylene (PE) [4-5], polyethylene terephthalate (PET) [6] and polypropylene (PP) [7]. In PEN very few [8] references cited this behavior. The charging current peak is similar to that given by Many and Rakavy [9] relating to transient SCLC(space charge limited current). It appears at time t_p :

$$t_p = 0.786 . \frac{d}{\mu E} \tag{1}$$

where E is the applied field, d the sample thickness, μ the carrier mobility and ε the permittivity of the polymer. This time was related to the transit time of a space charge between electrodes.

Figure 3 shows typical discharging current measurement subsequently to the charging current displayed in Figure 2. It can be noticed that, the discharging current present a monotonous decrease, suggesting a dependence on time given by the Curie Von Schweidler law [9]:

$$J = A \, t^{-n} \tag{2}$$

with n an exponent close to unity. Neagu [10] suggested that the isothermal discharge current described by this power law for the whole glass transition temperature range, is due to the space charge released from the deeper traps existing in the amorphous volume.

On the other hand, it should be noticed that the absorption current is not the mirror image of the discharging one. This suggests that the dipolar mechanism was not the predominant process. Mechanisms of dipolar origin are generally predominant at short time and low field [9].
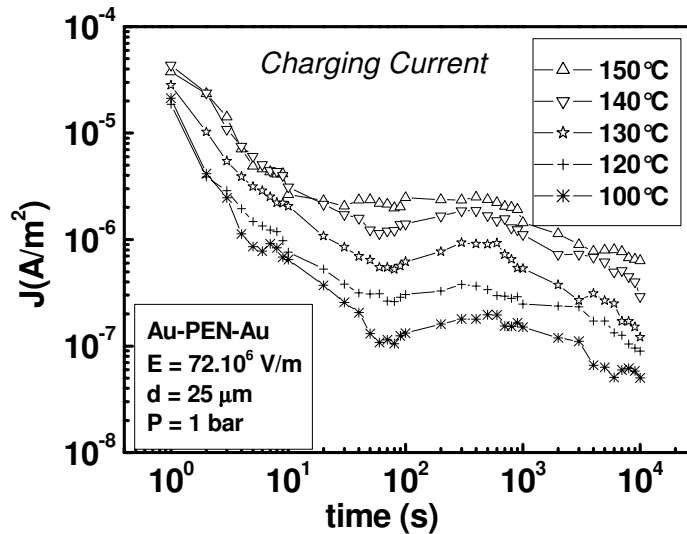

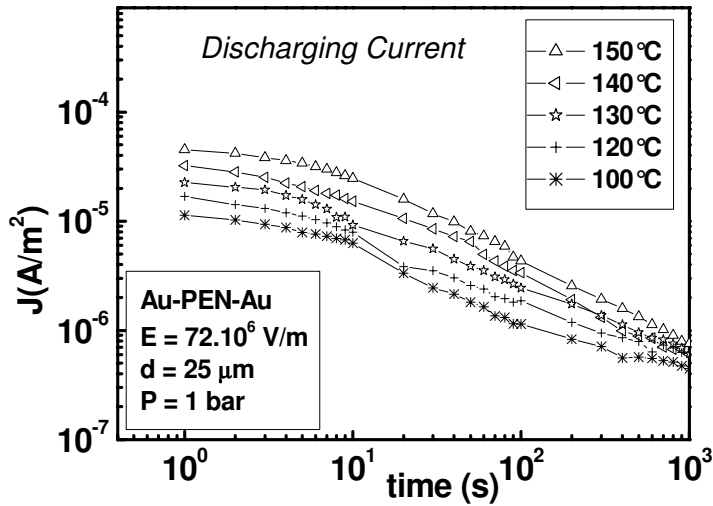Fig 2: Isothermal charging current in PEN at different temperatures.

Fig 3: Isothermal discharging current in PEN at different temperatures.

5. Final non-isothermal discharging currents

The isothermal charging current (Fig.3) becomes low but there is still space charge trapped in the sample. Subsequently the sample is heated up at a constant rate and the released sample is recorded. Figure 4 presents the results obtained for a sample charged at 100°C under an electric field of 72 MV/m for 4000s, and isothermally discharged during 4000s. The non-isothermal discharge current was obtained for a heating rate of 2.4°C/min from 100 to 180°C. The variation of temperature as a function of time was linear. That allow us to represent the variation of current I as a function of temperature T. A high increase of the current was observed. An increase in temperature seems to release some of he trapped charges. Two peaks were obtained: the first one located at 128°C near T_g and the second one, which appears as a shoulder on the first one, at 150°C.

6. Thermal behavior

The DSC thermogram of sample was given in figure 5. As the temperature increases, the glass transition, and the melting of PEN can be observed successively. The DSC thermogram of the semi-crystalline PEN shows a T_g (glass transition temperature) at about 128°C, and T_m (melting temperature) at 267°C. A small endothermic pre-melting peak was also observed at $T_{pm} = 150$°C. The presence of double melting peak was already reported [11] in PEN.

These experiments have allowed us to calculate the degree of crystallinity using the equation:

$$\chi(\%) = 100 \frac{\Delta H_m - \Delta H_c}{\Delta H_f} = 44\% \qquad (3)$$

where ΔH_m and ΔH_c were respectively crystallization and melting enthalpy. $\Delta H_f = 103.4$ Jg^{-1} was a melting enthalpy of 100% crystalline PEN.
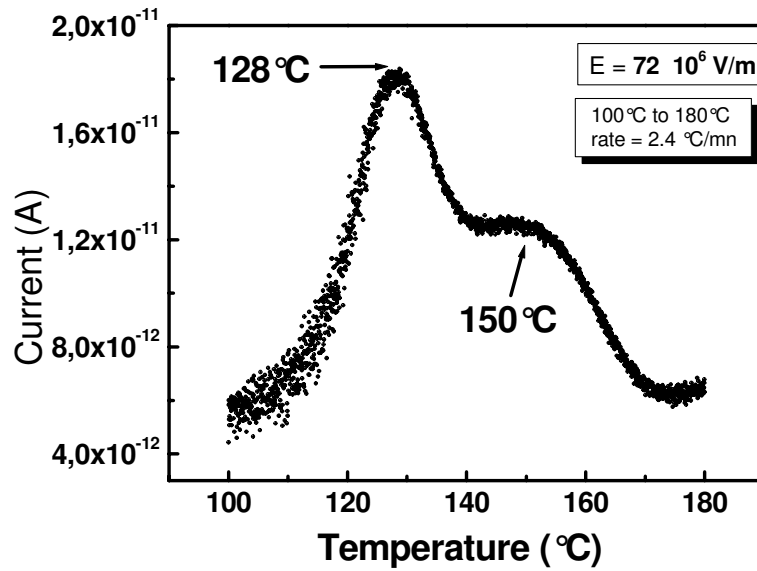
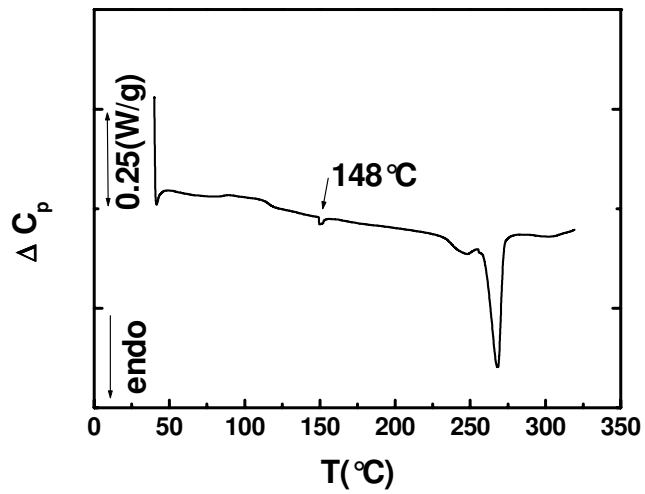Fig 4: Non-isothermal discharging current in PEN

Fig 5: DSC thermogram of semicrystalline PEN.

7. Discussion

The increase of a non-isothermal discharge current is related to detrapping of space charge. The release of charges may be a result of increased motion of the polymer chains at the higher temperature, especially near T_g. Indeed, it is well known that glass transition temperature is associated with chain segment cooperative motion induced by diffusive rearrangement of main chain portions [12].

The peaks are localized at two temperatures which are in good concordance with DSC analyze and current-field J(F) characteristics presented in previous works [13]. It would be noticed that this temperature (150°C) was presented by industrials as the maximal temperature of use. The two peaks suggest that there are, at least, two sorts of traps in which charges may be trapped. The first sort of traps is shallow ones and the second type is deep ones. These traps can be of chemical or physical nature. According to Patsch [14], they may be foreign molecules, carbonyl groups or even polarized states that are able to capture a charge via self-trapping.

On the other hand, the charge concerned may be trapped in different sites associated with amorphous region, amorphous-crystalline boundaries and crystalline region, since the polymer studied is semi-crystalline.

In previous works [1] we have shown that, above 150°C, the shape of isothermal charging current change with temperature reflecting that at low temperature the charge is rapped mainly in the amorphous region but for higher temperature the charge penetrate into and is trapped in the crystalline region [15].

8. Conclusion

These results implies that, after isothermal discharging there was still space charge trapped which cannot release from the sample and require an increasing of temperature. Indeed, the peak in non-isothermal discharge current at T_g, indicate that the change of polymer state, from glassy to rubbery, favor molecular motion and then facilitate the space charge detrapping process. The second peak was associated to premelting of crystallites.

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