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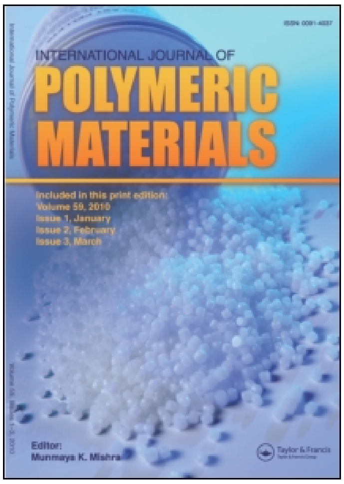

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Salah Mahrous[a]

[a] Physics Department, Faculty of Science, Cairo University at Fayium, Fayium, Egypt

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DIELECTRIC BREAKDOWN OF POLYISOXAZOLINE

Salah Mahrous
Physics Department, Faculty of Science, Cairo University at Fayium,
Fayium, Egypt

Dielectric breakdown of polyisoxazoline was studied over the temperature range from room temperature to 150°C and thickness range 1–10 mm by applying a linearly rising voltage. It was found that at room temperature, the dielectric breakdown strength (E_b) decreased with increasing sample thickness, whereas above 50°C, E_b was almost independent of the thickness. The bond breakdown is most probable in the studied range, and thermal fluctuations at the conducting microcrack tips should result in conducting crack growth, which eventually causes breakdown failure of the polymer.

Keywords: dielectric breakdown, polyisoxazoline, microcracks

INTRODUCTION

The dielectric breakdown strength is interpreted as a measure of the time an insulating material can withstand a certain electrical field until it breaks down. Dielectric breakdown occurs with very little warning by way of an increased current preceding the breakdown, although short pulses of current may be observed. In this sense, the situation is highly nonlinear and critical to the magnitude of the electric field, temperature, and so on but it is not in any evident sense related to direct current transport or to low-field dielectric losses [1].

Breakdwon results in the catastrophic formation of a narrow discrete channel, or a treelike pattern of channels of destruction [2] in what might appear to be initially a homogeneous medium. Quite evidently a massive dissipation of energy is involved in a very confined volume. One may say that the moving particle leaves behind a "tunnel" of residual polarization decaying with time after the passage of the charge.

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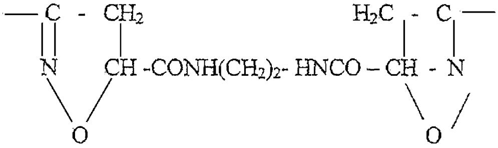
Address correspondence to Dr. Salah Mahrous, Physics Department, Teachers College, Riyadh 4341-11491, Saudi Arabia. E-mail: smahrous@yahoo.com

This is equivalent to the presence of a distribution of virtual charges of the opposite sign to that of the moving charge, giving rise to part of the drag force [3].

In this work, the mechanisms of the dielectric breakdown and molecular orientations of polyisoxazoline are studied over the temperature range from room temperature to 150°C.

EXPERIMENTAL WORK

Polyisoxazoline preparation is described elsewhere [4]. It has the chemical structure shown in Scheme 1.



Polyisoxazoline powder was compressed to form discs with 1–10 mm thickness and 5 mm diameter. The electrodes were deposited by evaporating gold under vacuum on both polymer surfaces in a sandwich geometry. A thin wire was attached to each face of the sample by a small droplet of silver paste. Before measurements, the samples were discharged by heating to 90°C while in a short-circuited state.

Measurements were made on samples immersed in silicone oil (KF-965, IOOcs, Shin-etsu Chemical Industry Co. Ltd), which was kept at a given temperature T_0 between room temperature and 150°C. After the sample was immersed for about 5 min, a linearly rising voltage was applied to the sample at various rates until a breakdown occurred. The electric current accompanied with breakdown was detected with a resistor connected to the sample, and immediately after the current exceeded 10 mA, the applied voltage was removed. About 10 tests were performed and the average of the obtained results was taken as the breakdown voltage.

RESULTS AND DISCUSSIONS

The temperature dependence of the dielectric breakdown strength, E_b, of polyisoxazoline at a rising rate of electric field of 0.03 MV m^{-1} s^{-1} is shown in Figure 1. Each point represents an average of about 10 tests. As can be seen in Figure 1, E_b at room temperature was in excess of 8 MV m^{-1}, but falls rapidly inversely with temperature T_0, above 50°C. This can be explained by considering the presence of impurities

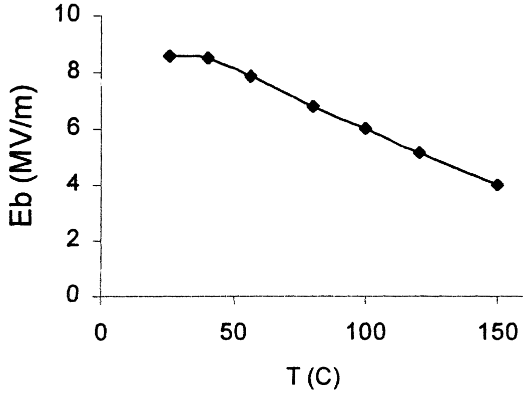


FIGURE 1 Temperature dependence of the dielectric breakdown strength E_b of polyisoxazoline at a rising rate of electric field of 0.03 MV m^{-1} s^{-1}.

in the polymer matrix that could produce an increase in the dc conduction leading to a lowering of the breakdown voltage.

Figure 2 shows the relation between the dielectric breakdown strength and the rate of rise of the applied electric field on the polymer at various temperatures ranging from room temperature to 150°C. At room temperature, E_b was almost independent of the rate of rise of the field within the experimental limit, but increased with the rise in rate at above 50°C. At room temperature the independence of E_b of sample thickness indicates that the breakdown is possibly due to an electronic

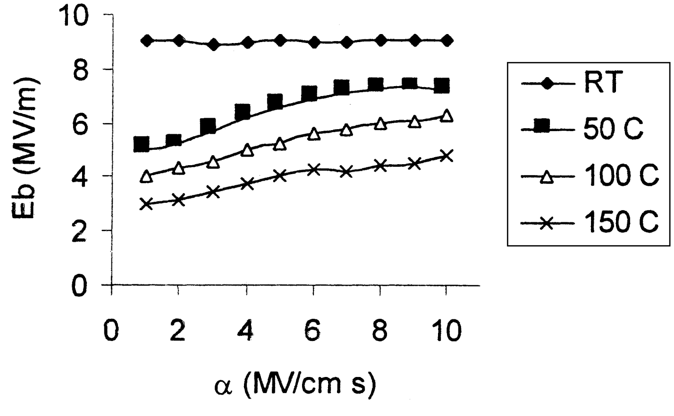


FIGURE 2 Relation between the dielectric breakdown strength (E_b) and the rate of rise of the applied electric field (α) of polyisoxazoline.

process [5]. At higher temperatures, there are two possible mechanisms of the decrease in E_b with T: thermal breakdown and electromechanical breakdown.

The thickness dependence of E_b at a rate of rise of field of about $0.2\,\mathrm{MV\,m^{-1}\,s^{-1}}$ at various temperatures, as measured on the 1–10 mm samples, is shown in Figure 3. It is noticed from this figure that the dielectric breakdown strength decreases with sample thickness, d, at any temperature, but only slightly at 150°C. The variation of E_b with d at high temperature is smaller than that at low temperature.

The mechanical stresses induced by electric field due to the orientation of lamellae structures can lead to weak bonds breaking and, thus, to conducting microcrack formation [6]. In these conducting microcrack regions, the local electric field increases, therefore causing more conduction in these weak regions. The size of the conducting microcrack is of the order of 10–1000 Å [7] and is dependent on the polymer structure. Some of these conducting microcracks coalesce into macroscopic conducting cracks (i.e., electric tree initiation), and produce rapid failure under the applied electric field conditions.

The conducting microcrack growth rate is given by [8]:

$$\frac{dc}{dt} = V_o \exp\left(\frac{\alpha\pi E^2 c - U_o}{kT}\right) \tag{1}$$

where

$$V_o = L_b\left(\frac{\omega}{2\pi}\right)$$

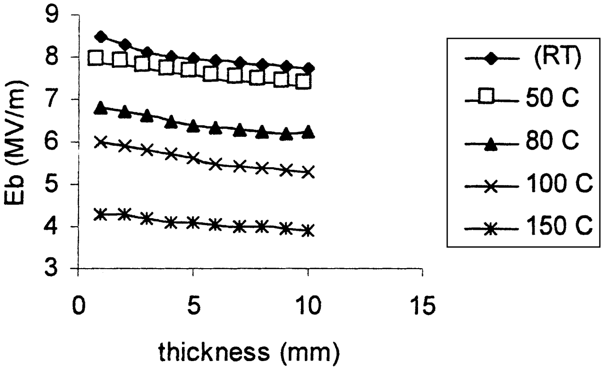


FIGURE 3 The thickness dependence of the dielectric breakdown strengh at various temperatures for polyisoxazoline.

here c is the conducting microcrack length, U_o the activation energy of the breakdown process, U_o and α are adjustable material constants for a particular polymer, k the Boltzmann's constant, T the absolute temperature, E the energy required for microcrack propagation, L_b refers to the scale of the nonlinear region for breakdown, $\omega = 2\pi f$, f is the characteristic frequency of atoms oscillating in a solid. From Eq. 1, it is clear that the growth of a conducting microcrack to the next increment of size by successive polymer bond breakdown at the conducting microcrack tip depends on the local electrical stress at the conducting microcrack tip and thus has a nonlinear dependence on the conducting microcrack size.

The low strains generated by the applied electric fields in the studied range cannot break backbone bonds, whose strength is in the 1–3 eV range. The weaker bonds in a polymer are the van der Waals bonds with strength as low as 0.1 eV [8]. The conducting cracks may be formed in these weak van der Waals regions. The polymer bonds may also be disrupted by the formation of quasi-phonon particles caused by thermal fluctuations [9]. Because electric stresses in polymers are highest at conducting microcrack tips, bond breaking is most probable there, and thermal flucuations at the conducting microcrack tips should result in incremental conducting crack growth, which eventually causes breakdown failure. The growth of a conducting microcrack to the next increment size by successive polymer bond breaking at the conducting microcrack tip depends on local electrical stress at the conducting microcrack tip and hence has a nonlinear dependence on the conducting microcrack size.

The polymer is deformed more or less elastically by the applied field. Polyisoxazoline is a semi-crystalline polymer, so the amorphous regions are easily deformed by stress. When the amorphous phase deformed sufficiently, bond breaking will occur because the crystalline regions are much tougher. The bond breaking may be due to Joule heating, that leads to thermal decomposition, or due to hot electrons that can break polymer bonds. The thermal breakdown is treated according to the equation [10].

$$C_v(dT/dt) = JE \tag{2}$$

where C_v is the specific heat at constant volume, t is the time, J the conduction current density, T the temperature during the breakdown process, and E the electric strength. From Eq. 2 it is noticed that the E_b is independent of the sample thickness.

The conduction current obtained for a 4 mm thick sample obtained under a dc electric field is shown in Figure 3 with heating rate of

2°C/min. At high temperatures, it is noted that there is a tendency for the current to rise steeply with temperature above a critical temperature T_c, although the temperature dependence was much smaller below T_c. The steep rise in current with temperature is probably reasonable for the breakdown process in the high-temperature region. This is due to the enhanced segmental rearrangements in the amorphous region, which enhances electron transport and facilitates the occurrence of electron avalanche breakdown. At low temperatures, the breakdown characteristics are temperature independent and the dependence of E_b on thickness, Figure 2, is negative. This behavior may be due to the space charge effect [11].

On the other hand, the dc conduction (Figure 4) shows higher activation energy than the transient conduction, and the critical temperature T_c above which the current reaches the steady state is increased as the time is shortened. In other words, at a given time, the activation energy for the conduction is higher above T_c than that below T_c. The higher voltage rise rate corresponds to shorter time. Therefore, if the breakdown is thermal even in the low-temperature region, the critical temperature for breakdown is expected to rise with the voltage rise rate (Figure 2).

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