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Anomalous Phenomena in Solid Dielectrics under High Electric Fields

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Abstract: Solid dielectrics have been used extensively in electrical and electronic industries. In the power industry, ultra high voltage transmission and cost saving of power equipment often result in that the dielectrics are operating under high electric fields. On the other hand, reduction in size for modern electronic systems also requires that the dielectrics operate reliably under high electric fields. It is therefore imperative to understand the response of dielectric materials to the high electric fields. In addition to high conduction current, power loss and potential breakdown, it has been observed over the years there are several anomalous phenomena occurred under high electric fields. These are surface potential cross-over phenomenon in corona charged dielectrics, discharging current flowing in the same direction as charging current, space charge fast decaying and transient space charge limited current.

In the present paper all these phenomena have been experimentally demonstrated but most importantly a bipolar charge injection model has been used to understand the observed phenomena under high dc fields. Preliminary simulations based the model reveal that the presence of bipolar charges is the key to explain these anomalous phenomena.

Keywords: high fields, solid dielectrics, anomalous phenomena, bipolar charge injection, simulation.

INTRODUCTION

Over the years, a great deal of work has been done in order to understand the mechanism of electrical conduction and properties of solid polymeric dielectrics. However, due to various limitations such as availability of suitable experimental tools and limiting power in modeling, the charge transportation in dielectric materials is not well understood and this is especially true at high electric fields. On the other hand there is a great urgency in understanding the charge transport mechanisms in polymeric materials under the high electric fields as they are increasingly used in high field conditions in both electronic and electrical industries. While high-density, digital circuits have made extensive use of polymer dielectric packaging materials, there has been increasing effort to incorporate these materials into high power electronic applications. Demand for smaller and lighter power supplies and related electronics has created a need for new innovations in packaging designs and materials. To ensure proper operation of a high power electronic circuit, proper isolation must be ensured between adjacent conductors. The use of polymer materials as interlayer dielectrics in thin film packaging can be attributed to their relatively low dielectric constants and their ease of processing. For power applications, the high currents and voltages associated with power electronic circuits require materials with specific attributes such as a high dielectric strength or breakdown voltage and small values for loss tangent or dissipation factor.

With the increasing demands for electrical energy, the power transmission voltages are increasing. On the other hand, the advance in material processing and equipment manufacturing, the insulation materials used for high voltage insulation get thinner and thinner. This means that polymeric material insulation will experience a very high electric field. It has been noticed that there are several anomalous phenomena observed under the high electric fields. These phenomena were reported in the literature and studied separately. Therefore, the understanding of electric charge transportation under high electric fields is important to ensure the reliability of the insulation system.

In this paper, the several anomalous phenomena under high dc electric fields were briefly reviewed with examples from the results obtained by the author. The unified model based on a bipolar charge injection has been used to simulate the phenomena. Preliminary results showed all the anomalous phenomena observed can be qualitatively explained.

ANOMALOUS PHENOMENA UNDER HIGH DC ELECTRIC FIELDS

The electrical properties of a dielectric are determined by the interaction of charge and electric fields. When the applied field is increased, the phenomenon observed are often surprising and beyond our anticipations. Here are a few typical examples.

Transient Space Charge Limited Current
There will be a current flowing through the circuit following the application of a step voltage to a dielectric material. Typically, the current will decay monotonically over a period of time (polarization current) and finally reach to a steady value (conduction current). However, under a high electric field in the

presence of charge injection and trapping in the bulk of the material, a transient space charge limited current can be observed where a current peak occurs. The transient space charge limited current was a hot topic in 1960s due to its importance to the rapid development in electronics and solid state devices. Theoretical work on the topic was pioneered by Many and Rakavy [1] in 1962. With a number of simplifying assumptions, solutions were derived for the time-dependent current and space-charge distribution following the onset of single type of charge injection. The transient current peak can be used to estimate the mobility of the charge carrier. This transient current peak was observed subsequently in polymeric materials at high fields [2-4]. The mobility of charge carrier obtained from the time when the current peak occurs shows both temperature and field dependent.

Depolarization Current Flowing in the Same Direction as Polarization Current
Depolarization current normally flows in the opposite direction to the polarization current. However under the high electric fields the depolarization current can drop very rapidly and passes through the zero point. After that the current flows in the same direction as the polarization current and decays to zero gradually. The phenomenon has been experimentally observed by several researchers [2-5]. So far no theory has been put forward except an over simplified model based on the bipolar charge injection [5].

Cross-over Surface Potential Decay
Corona charging and surface potential decay were studied extensively in 1970's as it provides a useful tool to monitor charge movement in the insulating materials. In 1967, surface potential cross-over phenomenon was firstly reported by Ieda et al [6], i.e. initially the surface potential of a sample charged to high-potential decays more rapidly than one charged to a lower potential. Most of the theories addressed the time evolution of the surface potential in terms of surface conduction [7], charge injection [8–10], and polarization [11]. The recent literature on potential decay measurement is dominated by the hypothesis of the injection into the bulk of the charge deposited on the surface accompanied by a slow polarization processes within the bulk under the influence of the deposited charge. It is worthy to point out that the fast potential decay occurs only when the electric field across the sample exceeds a certain value. All the theories and models are based on the single charge injection. To explain the crossover phenomenon, various unrealistic assumptions were made, such as partial injection, time dependent injection.

Cross-over in Space Charge Decay
With the recent progress in space charge measurement techniques, it is possible to observe charge evolution within the bulk of the insulting material. After the volts

on measurement, it is also possible to observe charge decay in the sample. Generally, the total charge in the sample decay monotonically with time. It has been recently observed that the charge decay becomes fast in the sample stressed at high electric fields than that stressed at lower stress [12]. This phenomenon is very similar to that described in the above section. The bipolar charge injection has been used to explain the phenomenon. This is supported by the experimental observation.

EXPERIMENTAL RESULTS FROM THE TONY DAVIES HIGH VOLTAGE LABS

One of the core researches in the Tony Davies High Voltage Laboratory at Southampton University is the application of dielectric materials in high voltage engineering. Over the years, we have studied electrical properties of polymeric materials under high dc electric fields. All the anomalous phenomena mentioned above have been observed. Together with the results from space charge measurements, it appears to us that all the anomalous phenomena come from the same origin in terms of physical mechanism. Bipolar charge injection, trapping, recombination and transportation are responsible for the observed phenomena especially at high electric fields.

Multi-current Peaks
Our early work on conduction mechanism in polyethylene [2] revealed so called transient space charge limited current as shown in Figure 1. The peak shifts towards the shorter time when the applied field or temperature increases.
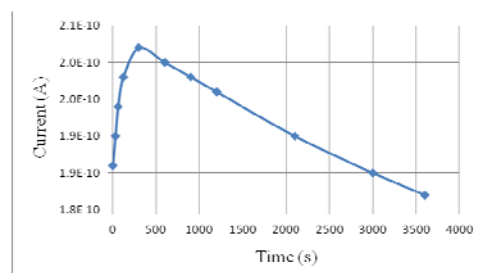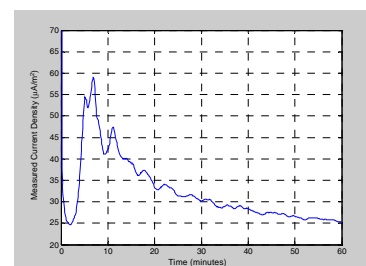

Figure 1 Transient space charge limited current in LDPE (E_{appl}=13.4 kV/mm) at 70°C.

More recently we have measured the current and space charge distribution simultaneously in cross-linked polyethylene sample [13] and multi current peaks have been observed at high fields as shown in Figure 2.
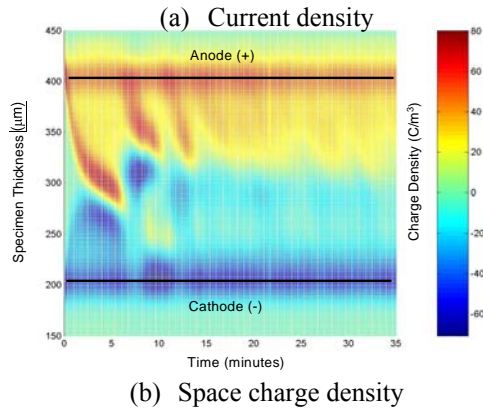
(a) Current density

(b) Space charge density

Figure 2 Current density and space charge density colour contour graph for the first 35 minutes in fresh (E_{appl}=100 kV/mm).

From Figure 2 (b) it is clear that bipolar charge injection has taken place and charge recombination occurs in the bulk of the sample. By comparing the current and space charge dynamics, it is evident that bipolar charge injection and recombination are responsible for the current peaks observed.

Depolarization Current
Anomalous discharge current was observed following the application of high electric field. Figure 3 shows a typical anomalous discharge current flowing in the opposite direction [2].
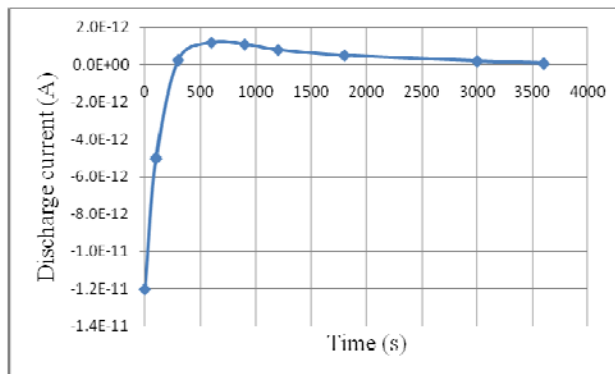

Figure 3 Anomalous discharge current in LDPE (E_{appl}= 26.7 kV/mm) at 70°C.

There were a few reported anomalous discharge currents in the literature. However, there is no proper explanation proposed so far except one simple model based on bipolar charge injection [5].

Cross-over Space Charge Decay
Space charge formation in LDPE sample was measured under dc electric fields [12]. Charge decay after the removal of the applied field was also monitored. The charge decay with time for samples after the removal of different applied fields is illustrated in Figure 4. The space charge distribution clearly shows that both positive and negative charge injections take place. It is

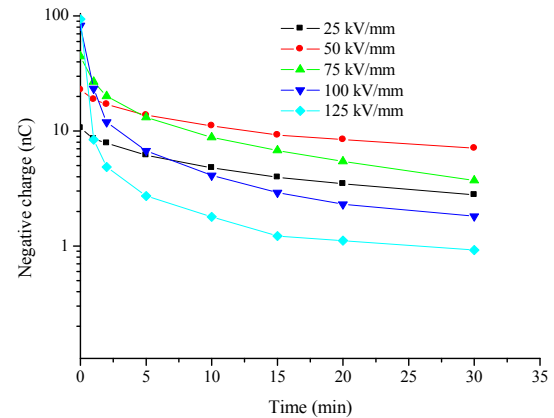believed that the feature has a close link with anomalous discharge current.

Figure **4** Charge decay after the removal of the applied electric fields (t_p=60 min).

Cross-over potential decay
Following the corona charging, the surface potential of a polymer material decays with time. Study of surface potential decay can provide useful information about charge trapping and transport in the material. Figure 5 illustrates a typical surface potential crossover for LDPE sample following corona charged at different voltages.
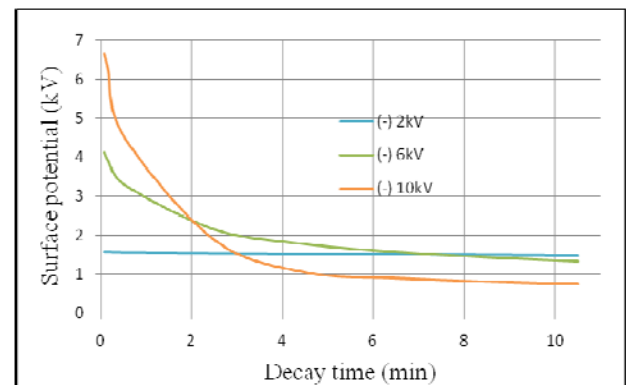

Figure 5 Potential decay for 50μm LDPE sample after different corona voltages.

Author is one of the fewer people who have used the pulsed electroacoustic (PEA) technique to measures space charge in corona charged polymeric materials [14-15]. It has been confirmed without any doubt that bipolar charge injection taking place during charging and decay processes.
Figure 6 shows the space charge measurement in corona charged LDPE film. To minimize the disturbance to the deposited charge, top and bottom surface of the samples were protected by an extra layer 50μm LDPE film. There are four distinctive charge peaks present across the sample from the left to right. The first and the fourth peaks are known as induced charge peaks on the PEA electrodes. The presence of the other two peaks is due

to the existence of charge in the sample. The second peak corresponds to the bottom surface of the 50μm corona-charged film while the third peak to the top surface. The third negative peak is expected as the sample was exposed to negative corona. The first space charge measurement was performed 2 minutes after the sample has completed corona charging as the sample has to be transferred to the PEA setup. The bulk charge is not evident when a short charging time is adopted. However, the bulk charge is clearly seen when the charging time is extended to 10 minutes as shown in Figure 6 (b). It can also be seen that a small amount of negative charge presents across the sample. This may be served as the evidence that charge injection from the top electrode has taken place.

Figure 6 also shows that the charge decay rate is different for different charging times. Charge decay is more rapid when a longer charging time is adopted.
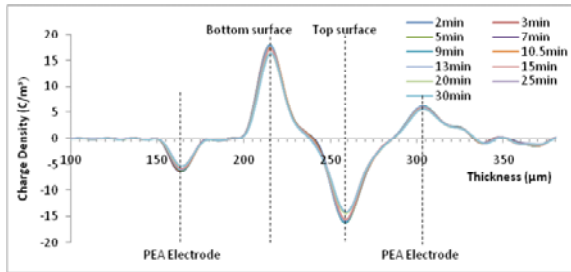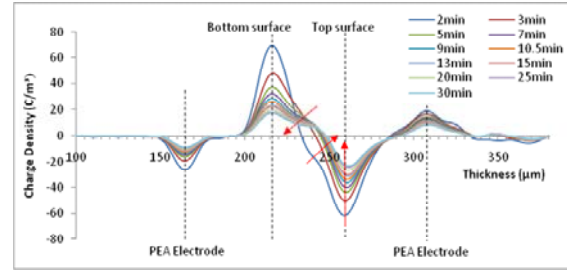

(a) t_c = 2 minutes
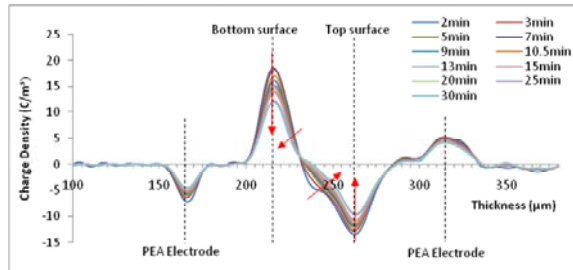
(b) t_c = 10 minutes

Figure 6 Space charge distribution in 50μm sample corona charged at 2 kV for (a) 2 minutes and (b) 10 minutes.

When the corona charging voltage increases, charge injection becomes more obvious as shown in Figure 7. It is clear that both positive and negative charges are present in the bulk of the sample. When the charging time is extended to 10 minutes significant positive charge injection can be observed as shown in Figure 7 (b). Charge decay rate increases significantly compared with the sample charged at 2 kV.

There is clear evidence that bipolar charge injection occurs and the dynamics of these bipolar charges (trapping, transport and recombination) in the material are responsible for the surface potential decay.

NEW MODEL AND SIMULATION RESULTS

To understand these observed anomalous phenomena, it is crucial to remember that almost in all the cases, bipolar charges are involved. By exploring the models in literature, it is noticed that the bipolar charge injection model proposed in [16] has a potential to simulate all these anomalous phenomena. In addition, the bipolar charge injection model has been used extensively to simulate charge transport, trapping and recombination under dc voltage. The simulation results for space charge dynamics in LDPE show a good match with experimental results [17].

(a) t_c = 2 minutes
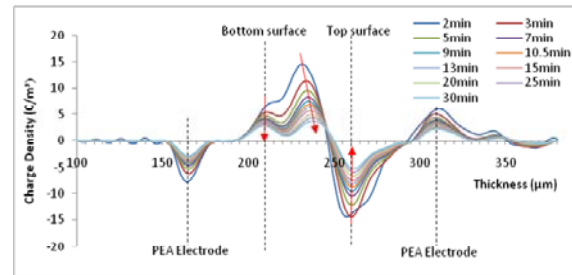
(b) t_c = 10 minutes

Figure 7 Space charge distribution in 50μm sample corona charged at 8 kV for (a) 2 minutes and (b) 10 minutes.

Bipolar Charge Injection Model and Governing Equations

The model [16] aims to effectively describe the bipolar transport and space charge phenomena in solid dielectrics under high dc stress. The bipolar transport is being described as a conduction process governed by an effective mobility. This feature distinguishes the model from the others. In effect, charge carriers are injected from the electrodes, electrons from the cathode and holes from the anode. Injection occurs based on the *Schottky* mechanism whereby overcoming a potential barrier W at the interfaces.

$$J_S = AT^2 \exp\left(-\frac{W - \sqrt{\frac{e^3 E}{4\pi\varepsilon_r \varepsilon_0}}}{kT}\right) \qquad (1)$$

where A is a constant related to the material, T the temperature, k the Boltzmann constant, e the electron charge, E the electric field at the interface, ε_r the relative permittivity of the material and ε_0 the permittivity of

vacuum.

It can be seen that the potential barrier is lowered down because of the application of the electric field. This will lead to an increase in the injected current.

After penetrating into the material, the carriers, under the influence of the applied field, will drift across the material characterized by an effective mobility. Throughout its motions, some carriers are trapped in the localized states i.e. deep trap centres and therefore, the total amount of charges moving across reduces. However, no extraction barrier is introduced in the model and on the other hand; they are prone to recombine with their opposite species (electrons with holes).

Due to the fact that oppositely charged species, electrons and holes are being considered in the numerical computation, charge trapping and mutual annihilation or recombination between these species were introduced into the model. The model contains four *species* of charge particles namely mobile electrons (e_μ) and holes (h_μ), and immobile electrons (e_t) and holes (h_t) (at trap sites) as shown in Figure 8.
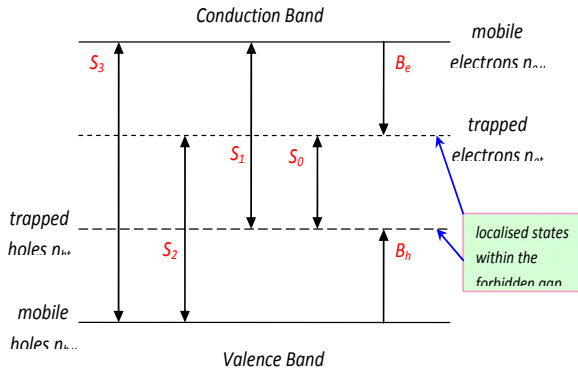

Figure 8 Schematic representations of the conduction and trapping model. S_i are recombination coefficients, $n_{e\mu}$, n_{et} , n_{ht} , $n_{h\mu}$ are mobile and trapped electron and hole densities. B_e and B_h are electron and hole trapping coefficients.

Charge transportation in solid dielectrics is essentially governed by a set of basic equations. They describe the behaviour of charge carriers in the system through a time and space dependent total flux $j(x,t)$ and by neglecting diffusion [16]:

Transport equation:
$$j_C(x,t) = \mu n(x,t) E(x,t) \quad (2)$$

Continuity equation:
$$\frac{\partial n(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x} = s \quad (3)$$

Poisson's equation:
$$\frac{\partial E(x,t)}{\partial x} = \frac{\rho(x,t)}{\varepsilon} \quad (4)$$

where μ is the mobility of carriers, n the density of

mobile species, E the electric field, j the current density, x the spatial coordinate, t the time, s the source term, ε the dielectric permittivity and ρ the net charge density.

4.2 Simulation results

Using the above model space charge dynamics in the bulk of the material under dc voltages can be calculated [17]. In addition, it is also possible to calculate the current density J and its relationship with time. Figure 9 illustrates initial simulation results of current density under various applied electric fields. It is clear that a current peak appears. Similar to the experimental results, the peak shifts towards the shorter time when the applied electric field increases.
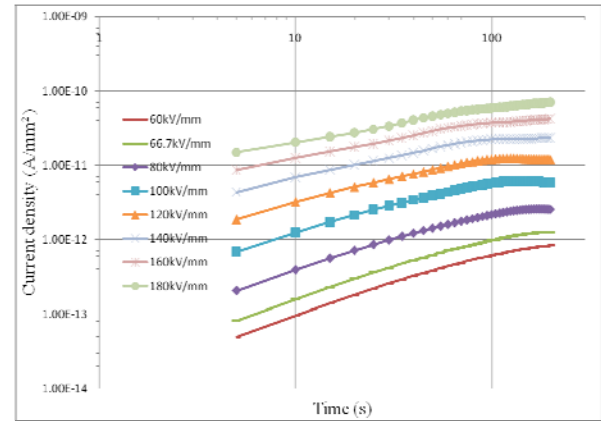

Figure 9 Transient space charge currents obtained from the bipolar charge model.

Once the applied electric field is removed, the discharge current can also be calculated. The discharge current flowing in the opposite direction decays monotonically. However, when the applied electric field increases, the discharge current decreases very rapidly and cross the zero. It flows in the same direction as the charging current before decaying to the zero as shown in Figure 10.
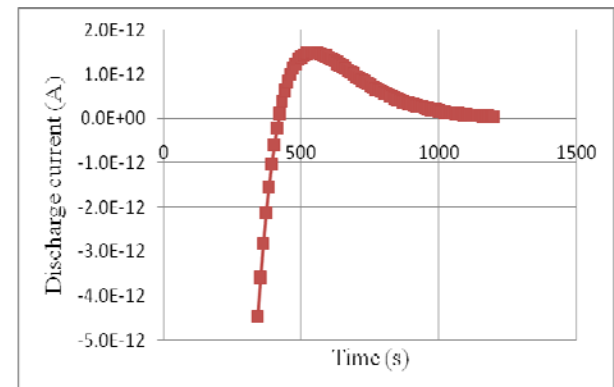

Figure 10 Discharge current obtained from the bipolar charge injection.

The related space charge decay based on the above simulations is shown in Figure 11. It has been found

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that for the same length of voltage application time, the injected charges travel further with increasing applied electric field. The amount of the injected charges also increase rapidly with the applied electric field.
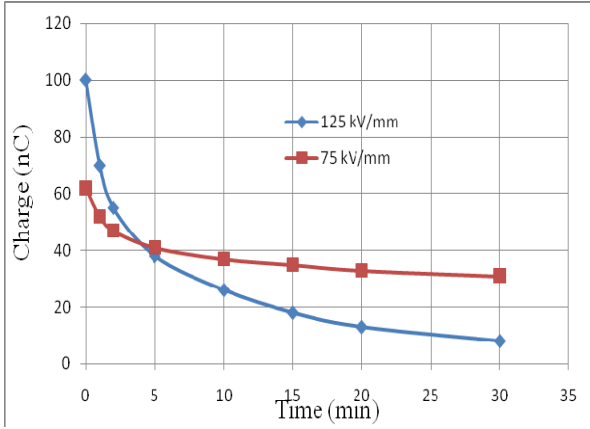

Figure 11 Space charge decay after the removal of the electric fields.

Considering the new evidence of bipolar charge injection during the corona charging and decay processes, the bipolar charge injection model can also be applied to explain the surface potential decay with some modifications. This deviates significantly away from the previous surface potential decay models. Additionally, a tunneling process has been proposed to account for charge injection from the top surface while the conventional Schottky injection is used for the bottom surface.

Based on the space charge measurement from the PEA, the initial charge distribution immediately after the corona charging in a sample can be illustrated in Figure 12.
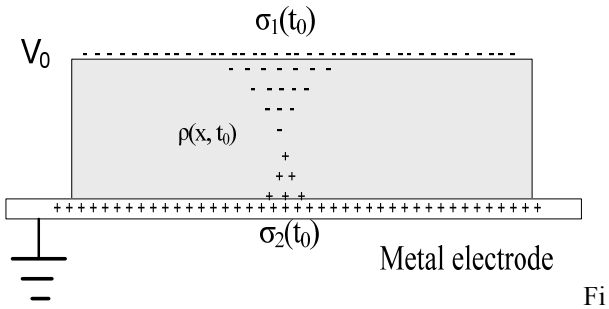

Figure 12 Schematic diagram of surface charge and space charge distribution immediately after corona charging.

Here V_0 is the grid voltage. $\sigma_1(t_0)$, $\rho(x, t_0)$ and $\sigma_2(t_0)$ are surface charge on the top surface, space charge in the sample and induced charge on the metal electrode respectively. The initial values will depend on the grid voltage and charging time. They all change with time leading to a change in surface potential $V(t)$.

Charge injection into the bulk from the top electrode is governed by Fowler-Nordheim tunneling rather than Schottky injection. The FN tunneling current is normally expressed as:

$$I_{FN} = AE^2 \exp\left(-\frac{B}{E}\right) \qquad (5)$$

where E is the electric field at the top surface, A and B are constants related to potential barrier at the top surface and effective charge mass.

It is clear from the above equation that the increase in the electric field will lead to an increase in the FN tunneling current since the square of electric field plays more dominating role that the inverted electric field in the exponential power.

Once charges are injected into the bulk, the rest description of charge trapping, detrapping, recombination and transport can be readily applied using the existing bipolar model.

It has been well accepted that the mobility of charge carriers is a function of the electric field. In fact several models have attempted to use field dependent mobility to account for the cross-over phenomenon without success. The field-dependent mobility can also be easily implemented in the proposed model. A power law proposed by Wintle [18] has been given as below:

$$\mu = cE^n \qquad n \geq 1 \qquad (6)$$

where c is a constant and n is a fixed component.

The charge transport in the bulk of the sample is determined by the electric field. The electric field in the sample at any time consists of contributions from three components, i.e. space charge $\rho(x, t)$, surface charge density at the top $\sigma_1(t)$ and the induced surface charge density at the bottom electrode $\sigma_2(t)$. Let us assume the field components are represented by $E_\rho(t)$, $E_{\sigma 1}(t)$ and $E_{\sigma 2}(t)$ respectively. The surface potential across the sample can be calculated by integrating the total electric field:

$$V_0(t) = \int_0^d [E_\rho(t) + E_{\sigma_1}(t) + E_{\sigma_2}(t)]dx \quad (7)$$

In addition, the total charge in the system at any time must in balance, i.e.

$$\sigma_1(t)S + \sigma_2(t)S + \int_0^d \rho(x,t)Sdx = 0 \qquad (8)$$

where S is the surface area where charges are present.

Clearly, $\sigma_1(t)$, $\sigma_2(t)$ and $\rho(x, t)$ are not independent quantities. Based on the modified model, it is possible to calculate $\rho(x, t)$ during the corona charging until a predefined charging time $t=t_0$. The quantities $V_0(t_0)$ and $\rho(x, t_0)$ are the initial condition for surface potential decay. This allows one to determine $\sigma_1(t_0)$ and $\sigma_2(t_0)$ using the above two equations.

Once these initial four quantities are determined, one can calculate new space charge distribution $\rho(x, t_0+\Delta t)$ based on the proposed model and the two surface density $\sigma_1(t_0+\Delta t)$ and $\sigma_2(t_0+\Delta t)$ using the FN tunneling

and Schottky injection respectively. In addition, as the system is an open circuit, the injected charge must satisfy the following condition:

Maxwell's equation for the total current:

$$J(t) = J_c(x,t) + \varepsilon \frac{\partial E(x,t)}{\partial t} = 0 \qquad (9)$$

The surface potential at time $t=t_0+\Delta t$ can finally be computed using equation (7). Repeating the process the relationship between the surface potential and time can be calculated. Our initial simulation work shows it is possible to achieve surface potential cross-over providing the initial surface potential and space charge distribution are set right as shown in Figure 13.
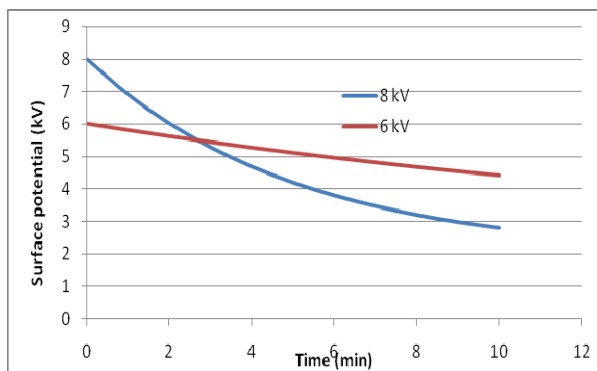

Figure 13 Initial simulation showing cross-over of surface potential decay.

From the above results it can be seen that the bipolar charge injection model shows a good qualitative agreement with all the anomalous phenomena observed at high dc fields. This suggests that they come from the same origin in terms of physical mechanism. Further work is required to quantify the parameters used in the simulation. Once the parameters have been thoroughly tested, they will be useful to characterize the materials.

CONCLUSIONS

This paper reviews several anomalous phenomena occurred in polymeric materials when subject to high dc electric fields. New experimental evidences presented in this paper clearly indicate that bipolar charge injection, trapping, recombination and transport are responsible for the observed phenomena. Our initial simulation results based on bipolar charge model can reveal all features observed in anomalous phenomena. This suggests that all these observed phenomena have the same origin in terms of physical mechanism. Although the new experimental evidences can be explained qualitatively with the bipolar charge injection model, further work is urgently required in terms of detailed theories.

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