AIP | Journal of Applied Physics

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Citation: J. Appl. Phys. **112**, 034116 (2012); doi: 10.1063/1.4745897
View online: http://dx.doi.org/10.1063/1.4745897
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v112/i3
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Investigation into the formation of charge packets in polyethylene: Experiment and simulation

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(Received 7 February 2012; accepted 17 July 2012; published online 10 August 2012)

The phenomenon of charge packet has been reported in polymeric insulation materials under the application of dc electric fields in recent decades. It is noted that such charge packets could lead to substantial modification of local electric stress, which increases the possibility of failure of insulating materials. The physics of charge packets has not yet been revealed clearly. In this paper, the dynamics of positive charge packets in polyethylene is observed using the pulsed electro-acoustic technique. Negative differential mobility of positive charge carrier is found, which is believed to be crucial to the formation of charge packets. This negative differential mobility is introduced into a bipolar charge transport model to simulate the packet-like space charge in polymers. Simulation results show that not only the negative differential mobility but also weaker trapping characteristic are required to generate a positive charge packet in polyethylene under dc stress. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4745897]

I. INTRODUCTION

Due to the demand of electricity energy, the voltage level in power transmission system has been steadily raised. In modern cities, underground cables are preferred for the transmission lines under the environmental considerations. In terms of the insulation, polyethylene has been widely used for high voltage cables due to its excellent electrical performance and low cost. However, the accumulation of space charge has been observed in polyethylene-based insulation materials under dc applications.[1,2] It could create enhanced electric field regions in the cable and lead to partial discharge or even failure of cable insulations.[3] Space charge has been extensively investigated through experiments combined with complicated theory of charge dynamics in polymers. A packet form of space charge, which travels in polyethylene-base materials, has been reported in the recent decade.[4–6] These charge packets cause substantial increase of electric stress and could accelerate the breakdown of polymeric insulations.[6] The exact nature of charge packets has not yet been revealed properly. This complicates the general understanding of charge dynamics and dielectric properties of polymers. Further experimental evidence and supporting theory need to be established to achieve a clear picture.

Pulsed electro-acoustic (PEA) technique, which is widely used to measure space charge in solid dielectrics, is employed to observe the dynamics of positive charge packets in polyethylene under various dc voltages. To understand the formation of positive charge packets, a bipolar charge transport model is developed to simulate this phenomenon by using the velocity values obtained from experiments. A comparison between simulation and experimental data is given in the discussion.

II. EXPERIMENT

A film of commercial additive free low density polyethylene (LDPE) is used for the experiments. The thickness of LDPE film is $100 \pm 5\,\mu$m. The film sample is sandwiched between two flat electrodes. The top electrode is semiconducting carbon-black loaded polymer, while the bottom is aluminium. Constant dc voltage is applied across the sample. Then test is done using a pulsed electro-acoustic (PEA) setup, which has a spatial resolution of $5\,\mu$m. The principle of PEA technique can be found in Ref. 7. Each sample is polarized under dc voltage for several minutes and space charge profiles are recorded at specific time steps. The applied dc voltage ranges from 1 to 12 kV. All the measurements are performed at a room temperature of around 22 °C.

A. Low electric field

1. Pulse excitation method

Under low electric fields, only normal space charge distribution can be observed by the PEA measurement. No charge packets are observed to occur in polyethylene. Therefore, a pulse excitation method, first proposed and reported by Hozumi *et al.*,[8] is employed to initiate a charge packet in polyethylene. The pulse excitation method is illustrated in Fig. 1. A dc bias voltage is applied across the sample from the very beginning; once a quasi-stable distribution of space charge is achieved after several minutes, a pulse voltage with a width of 250 ms and large amplitude of several kilovolts is superimposed onto the dc voltage. The peak amplitude of the overall voltage is up to 15 kV. Due to the excitation of space charge by a large pulse voltage, a packet of positive charge immediately forms at the anode and travels towards the cathode under the dc bias voltage. Hence the dynamics of positive charge packets under the biased electric field can be observed in polyethylene.

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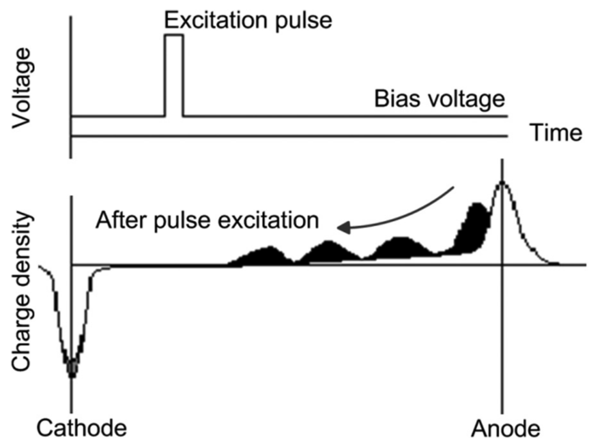

FIG. 1. Principle of pulse excitation method.[8]

2. Excited charge packets

Measured space charge profiles at a bias dc field of 20 kV mm^{-1} after pulsed excitation are shown in Fig. 2(a). This result does not demonstrate clearly the formation and movement of charge packets. Hence the quasi-stable distribution of space charge achieved at 300 s since the application of dc bias voltage but prior to the pulse excitation is subtracted from all of the acquired space charge profiles. Then, the remaining charge profiles clearly show the development of a positive charge packet as shown in Fig. 2(b). A small packet of positive charge carriers is formed at the anode and travels slowly into the bulk of polyethylene. The dynamics of the positive charge packet at a dc field of 50 kV mm^{-1} is shown in Fig. 3.

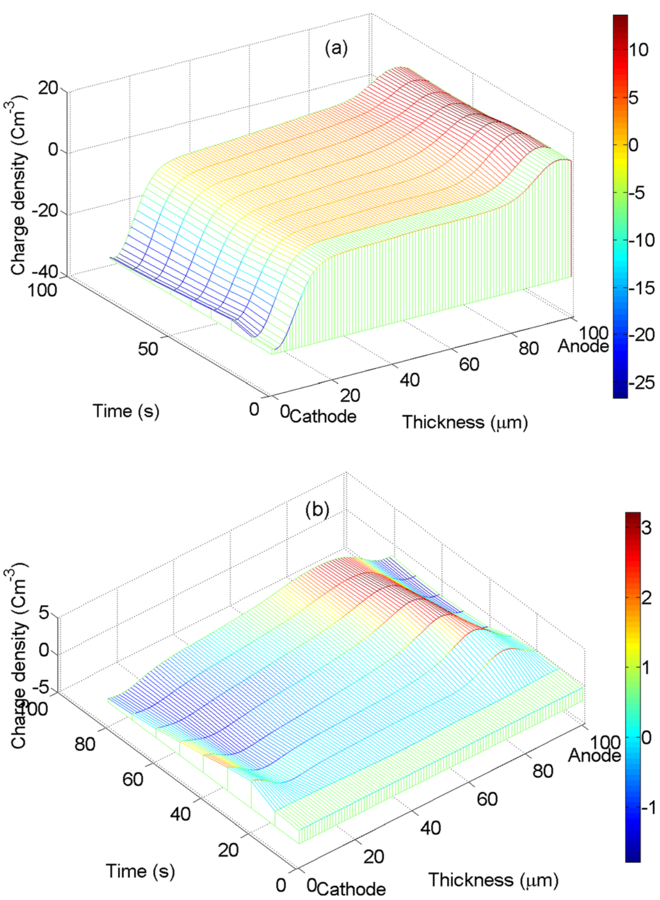

FIG. 2. Space charge profiles at 20 kV mm^{-1} after pulse excitation: (a) unsubtracted; (b) subtracted.

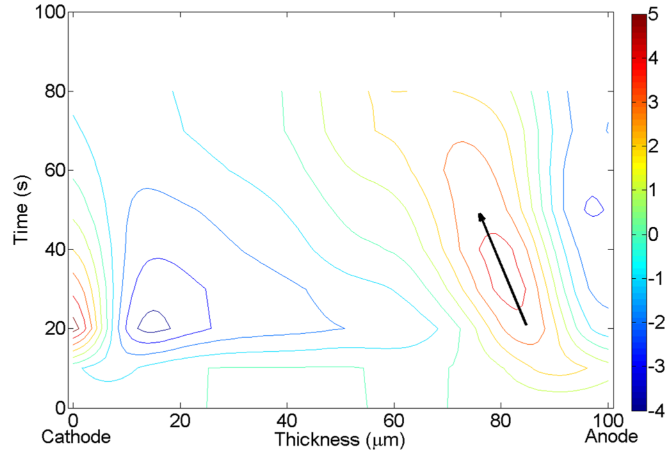

FIG. 3. Contour plot of subtracted space charge profiles at 50 kV mm^{-1} after pulse excitation.

3. V-E curve

To characterize positive charge packets, it is preferable to evaluate the velocity of the packet. The moving packet-like charge may change the electric field distribution with stressing time and hence suggests that the moving distance of charge packets is not proportional to stressing time. However, the experimental observation of the two-dimensional contour plot shows an apparent straight line movement of positive charge packets (indicated by red colour and the arrow) with stressing time as shown in Fig. 3. Therefore, the average velocity of positive charge carriers under the applied dc field can be evaluated. This velocity is averaged from more than three measurements. Moreover, its dependence on the electric field can be determined from repeated tests under dc electric fields ranging from 10 to 70 kV mm^{-1}. The dependence of the velocity of positive charge carriers on the applied electric field is plotted in Fig. 4. It is noticeable that the velocity does not constantly increase with the electric field. The velocity starts to

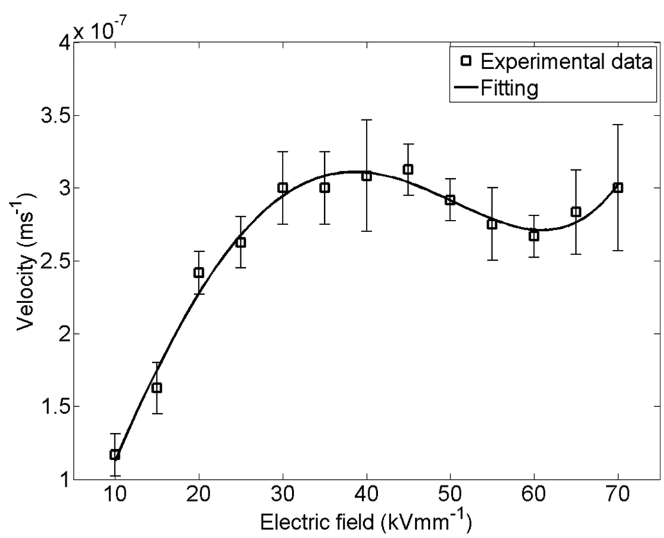

FIG. 4. The dependence of velocity on the low applied electric field.

decrease gradually when the electric field exceeds 40 kV mm^{-1}, followed by a second rise at higher stress above 60 kV mm^{-1}. This type of dependence was first observed in polyethylene.[9] It resembles the "Gunn Effect" seen in semiconducting materials[10] and suggests that a negative differential mobility is involved in the behaviour of positive charge carriers in polyethylene. The apparent mobility of positive charge carriers evaluated from the velocity curve is plotted in Fig. 5. It shows that positive charge carriers in polyethylene have the mobility of the order of 10^{-15}–10^{-14} m^2 V^{-1} s^{-1}. It tends to decrease when the field is increased. On the other hand, as shown in Fig. 3, negative charge carries seem to travel faster and reach the opposite electrode more quickly than positive species even though there is no formation of negative charge packets in polyethylene.

B. High electric field

1. Charge packets

When the applied field is increased above 55 kV mm^{-1}, a positive charge packet can be formed and observed in polyethylene without additional pulse excitation. The charge packet appears as soon as 15 s after the application of the dc voltage. Therefore, a traditional space charge measurement is performed to capture the dynamics of positive charge packets. To clearly present the characteristics of charge packets, subtraction from the original charge distribution at 15 s instance prior to the appearance of the charge packet is performed. The remaining charge profiles in polyethylene at a dc field of 80 kV mm^{-1} and 100 kV mm^{-1} are shown in Fig. 6. It is clearly observed that the positive charge packet formed at 100 kV mm^{-1} travels slower than the one formed under an applied dc field of 80 kV mm^{-1}.

2. V-E curve

The average velocity of positive charge carriers is also evaluated using the same method as described for the low applied fields and its dependence on the applied electric field is shown in Fig. 7. A reduction of velocity with increased field is again observed. The velocity decreases until the breakdown field of polyethylene is achieved as reported in

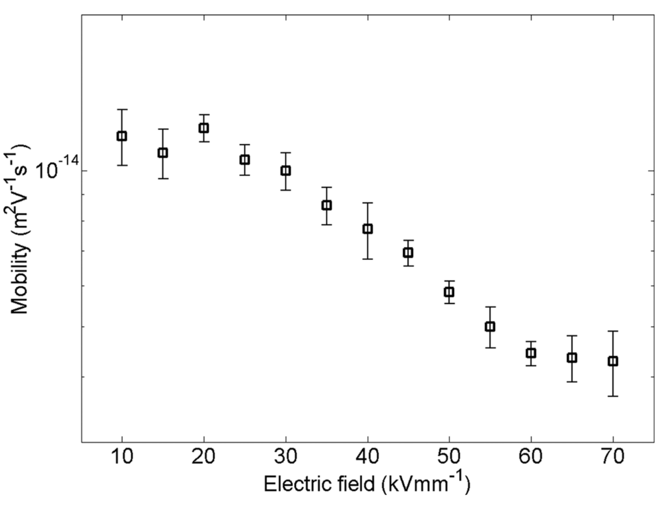

FIG. 5. Mobility of positive charge carriers under low applied electric fields.

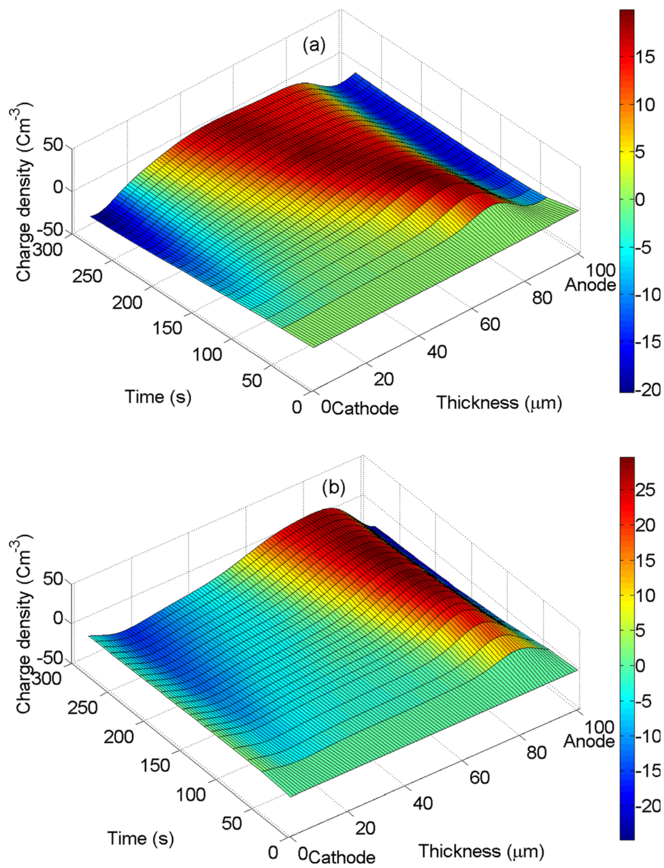

FIG. 6. Subtracted charge profiles at higher electric fields: (a) 80 kV mm^{-1}; (b) 100 kV mm^{-1}.

Ref. 6. But large variations occur at a field of 70 kV mm^{-1}, which is similar to the upturn shown in Fig. 4. The mobility of positive charge carriers under high electric fields is shown in Fig. 8. It shows an overall negative differential dependence on the electric field.

C. Negative differential mobility

In the case of a positive charge packet, the higher the applied electric field is, the slower the charge packet travels

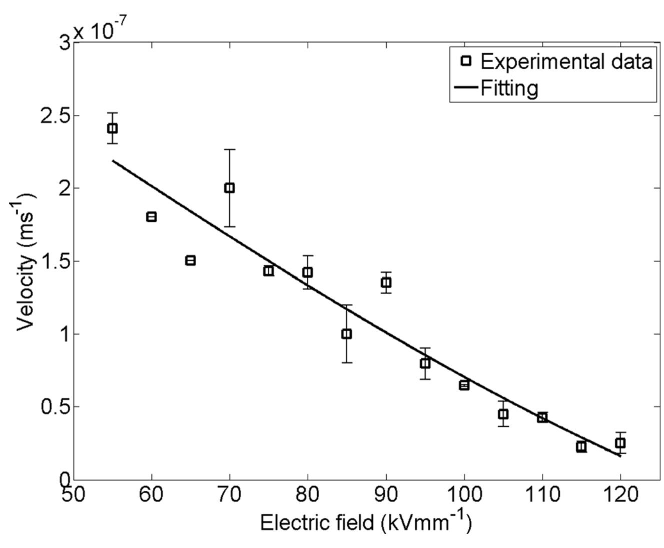

FIG. 7. The dependence of velocity on the high applied electric field.

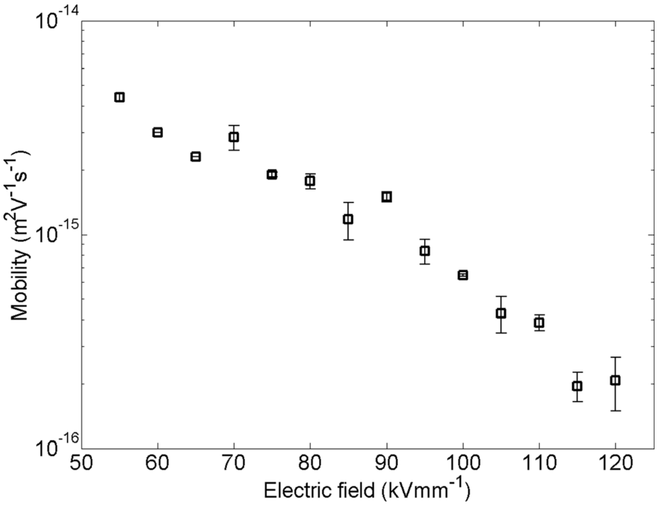

FIG. 8. Mobility of positive charge carriers under high applied electric fields.

in the dielectric. The mobility of positive charge carriers obtained at low applied electric fields and high electric fields both decreases with the electric field. This is referred to as a negative differential mobility. The mobility of positive charge carriers in polyethylene tends to decrease from the order of 10^{-14} to 10^{-16} m^2 V^{-1} s^{-1}.

III. SIMULATION

The reduction of velocity and negative differential mobility inspire the investigation into the formation of charge packets in dielectrics. It is believed that this interesting velocity curve contributes to the charge packet phenomenon. Hence, an idea of developing a numeric model employing the experimental velocity value comes up. It could be an effective approach to build a theoretical understanding of possible physics behind the charge packets.

A. Numeric model

The bipolar charge transport model, which involves bipolar charge injection from the electrodes, charge transport with trapping and recombination processes in the dielectrics, is developed to simulate the dynamics of charge packet in polyethylene under the application of electric field. The numerical treatment of the dielectric and solution of involved equations can be found in Ref. 11. The computation model is based on several essential assumptions. These are as follows:

(1) As additive free low density polyethylene is used in the experiment, it is considered that charge carriers are only generated by charge injection, i.e., electrons and holes from the electrodes without any ionization process in the bulk of dielectric. Electrons and holes are injected from negative and positive electrodes, respectively, controlled by the Schottky law, which involves a barrier height that constrains the injection rate of charge carriers. It is widely used to describe the injection of electronic charge from the metal or semiconducting materials into the dielectric and is accepted for the simulation of space charge formation in polymers.[12,13]

(2) The injected electrons and holes travel in the bulk of the material under the applied electric field. The transportation of charge carriers are characterized by the ohmic conduction law.

(3) Charge carriers can be trapped in the defect centers in the material. Opposite polarity charge carriers can recombine when they encounter each other in the material or at the interfaces.

The point is how charge carriers transport in the dielectric when there is a charge packet formed or how the transportation effects the formation of charge packets. In order to investigate this, the velocity curve obtained from the dynamics of positive charge packets in polyethylene is introduced into the model.

Experiments indicate that negative charge carriers move more quickly than positive charge carriers. Therefore, the transport of electrons is described using a relatively larger constant mobility value in the model. The flow of electrons is expressed by the conduction equation

$$J_e = \mu_e n_e E, \tag{1}$$

where J_e is the electrons flux density; μ_e is the constant mobility; n_e is the density of mobile electrons; and E is the local electric field. On the other hand, positive charge carriers, holes, move slowly in polyethylene and they have a different way of transport, which is described by the experimental velocity value. The velocity used in the simulation is calculated from the polynomial fitting of the experimental *V-E* curve shown in Figs. 4 and 7

$$J_h = v_h E, \tag{2}$$

where J_h is the holes flux density; v_h is the velocity of holes obtained from experiments.

In the simulation, a dc voltage is applied across a polyethylene film with a thickness of 100 μm and a permittivity of 2.3, the dynamics of space charge in polyethylene is then simulated. Numeric computation is performed at an absolute temperature of 295 K. The other parameters in the simulation are given in Table I. A relative low injection barrier height for holes (1.14 eV) than electrons is employed to produce an obvious dominant positive charge distribution in the bulk and the equivalent amount of charge compared with the experimental data. The low trapping coefficient and trapping density for holes are also used to achieve the dominance of mobile positive charge carriers in the system. The relaxation of charge carriers in the system is through extraction from the electrodes and the annihilation process in the bulk due to the recombination of opposite polarity charge carriers.

B. Simulation results

1. Low electric field

The experimental V-E curve obtained in the low field range from 10 to 70 kV mm^{-1} indicates that velocity starts to decrease at the field around 40 kV mm^{-1}. Hence, two typical field values, 20 kV mm^{-1} in the positive differential region and 50 kV mm^{-1} in the negative differential region are

TABLE I. Parameters in the simulation of charge packets.

Parameter	Value	Unit
Barrier height for injection		
w_{ei} (electrons)	1.15	eV
w_{hi} (holes)	1.14	eV
Transport		
μ_0 (for electrons)	2.5×10^{-13}	$m^2V^{-1}s^{-1}$
Velocity of holes	Experimental value	
Trap density		
N_{0et} (electrons)	100	Cm^{-3}
N_{0ht} (holes)	10	Cm^{-3}
Trapping coefficients		
B_e (electrons)	7×10^{-3}	s^{-1}
B_h (holes)	7×10^{-5}	s^{-1}
Recombination coefficients		
S_0 trapped electron-trapped hole	4×10^{-3}	$m^3C^{-1}s^{-1}$
S_1 mobile electron-trapped hole	4×10^{-3}	$m^3C^{-1}s^{-1}$
S_2 trapped electron-mobile hole	4×10^{-3}	$m^3C^{-1}s^{-1}$
S_3 mobile electron-mobile hole	0	$m^3C^{-1}s^{-1}$

applied across the polyethylene film in the simulation. Simulated space charge profiles under a dc field of 20 kV mm^{-1} and 50 kV mm^{-1} are shown in Fig. 9. For a dc field of 20 kV mm^{-1}, holes and electrons are injected into the bulk of polyethylene and move towards the opposite electrodes. No charge packets are formed. In contrast, a positive charge packet forms at the anode and travels to the cathode at an applied field of 50 kV mm^{-1}. Furthermore, a second broad charge packet is generated at the anode once the first packet is absorbed at the cathode, which reproduces the repetition of charge packets. This suggests that the decrease of the velocity with electric fields is crucial to the formation of charge packets.

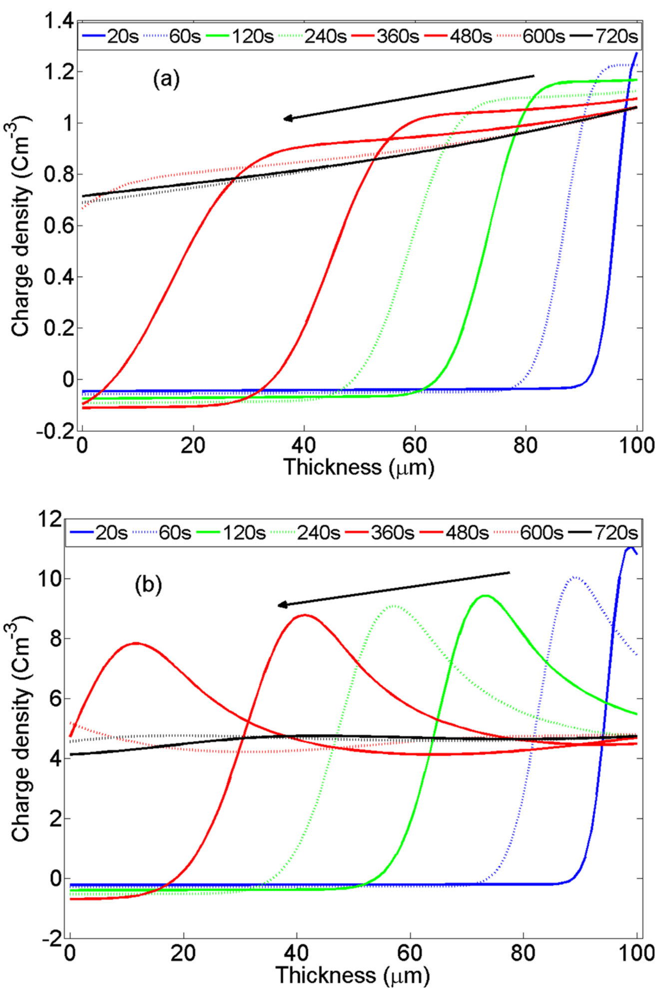

FIG. 9. Simulated space charge profiles at lower dc electric fields: (a) 20 kV mm^{-1}; (b) 50 kV mm^{-1}.

2. High electric field

For a high dc field of 100 kV mm^{-1}, simulated space charge profiles are shown in Fig. 10. It is clearly observed that a large positive charge packet is generated at the anode and it increasingly grows when travelling into the bulk of polyethylene. This agrees with the experimental observation of positive charge packets in low density polyethylene under high electric fields greater than 100 kV mm^{-1}.[6] The increase of the amplitude suggests a lower velocity in front of the charge packet where the field is continuously raised by the charge packet. In other words, the velocity of positive charge carriers continuously decreases under higher electric fields.

3. Trapping coefficient

From the numeric simulation, the decrease of velocity with electric field or the negative differential mobility is confirmed to be crucial to the formation of charge packets in polyethylene. However, this is not the only reason. The travelling charge packet also suggests the dominance of mobile charge carriers over the trapped charge carriers in the system, which indicates a low trapping rate in the region where a charge packet occurs. Hence, the effect of trapping coefficient on the formation of charge packets has been examined using the simulation. The simulated charge profiles in polyethylene at dc field of 50 kV mm^{-1} in the case of various trapping coefficients are shown in Fig. 11. The results show that positive charge packets cannot be formed in the case of a large trapping coefficient greater than 7×10^{-3} for holes. A large trapping coefficient leads to less mobile holes, which cannot raise significantly the electric field in the bulk of polyethylene and consequently ensure a lower velocity in

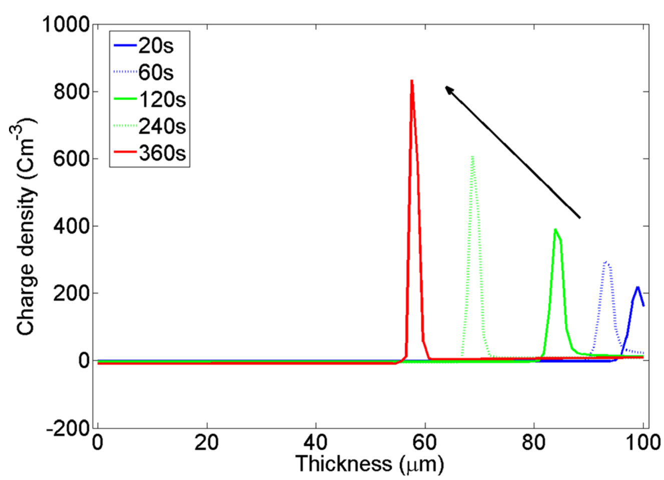

FIG. 10. Simulated space charge profiles at a high dc field of 100 kV mm^{-1}.

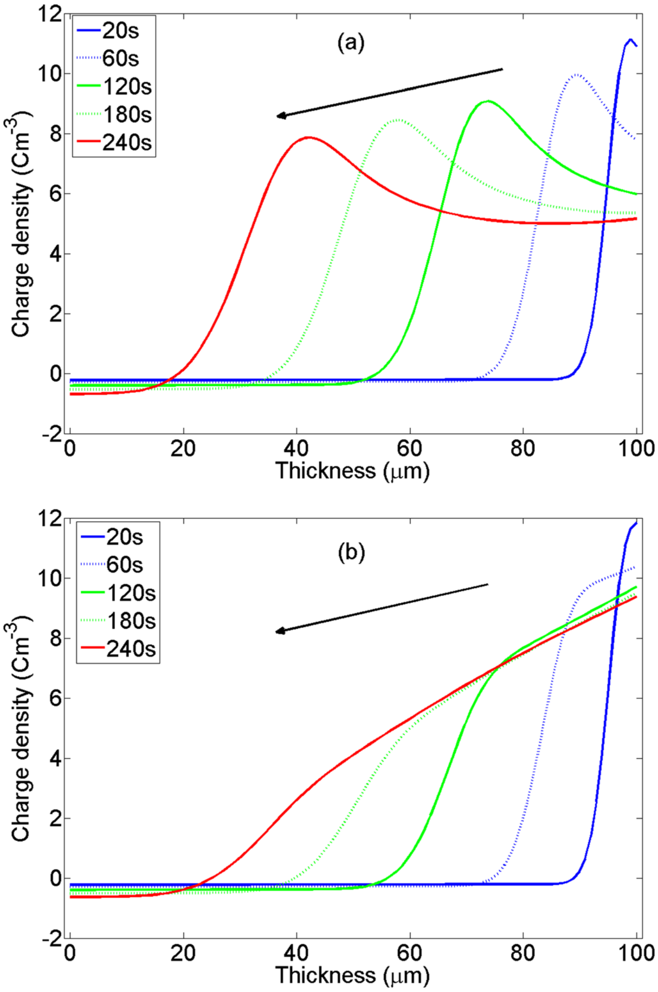

FIG. 11. Simulated charge profiles at a dc field of 50 kV mm^{-1} for different trapping coefficients: (a) 7×10^{-4}; (b) 7×10^{-3}.

front of the charge carriers. Therefore, the charge packet cannot be observed. This suggests a correlation between the trapping characteristics and the formation of charge packets in polyethylene.

IV. DISCUSSION

Positive charge packets can only be observed in polyethylene under low electric field conditions with the additional requirement of excitation by a large pulse voltage. But they can be directly observed at high dc electric fields without additional excitation. This suggests that there is a dependence of the appearance of charge packets on the electric field. When further investigating the behaviours of positive charge packets under various electric fields, the occurrence of the charge packets demonstrates a close correlation with the reduction of velocity at increased electric fields. Therefore, there must be an effect of the electric field on the transport of positive charge carriers (holes). The reason why a higher electric field leads to slower transport of holes in polyethylene is not fully understood. A single crystal of polyethylene of lamella thickness has demonstrated high conductance.[14] The overall low mobility of holes is therefore predominantly constrained by holes transfer in the amorphous inter-lamella regions, which introduces more localized trapping energies.[15] The question would be how the electric field affects hole transfer through inter-lamella space. The low trapping coefficient required for generating the charge packets is also related to the electric field. If conduction due to holes transfer is trap-limited, the unfilled trapping centres region in the front of the charge packets would lead to low carriers mobility and the filled trapping centres region in the rear would lead to high mobility and this would be a reasonable hypothesis for the physics behind. This verifies the explanation and analysis from Matsui *et al.*[6] who considers a low conductivity at high electric field regions and a high conductivity in low field regions. However, the velocity and its dependence on the electric field obtained from experiments reveals the negative differential mobility of positive charge carriers in polyethylene and that the formation of charge packets could only be resulted from the reduction of velocity of charge carriers with electric fields.

V. CONCLUSION

The dynamics of positive charge packets in polyethylene has been investigated through experiment and numeric modeling under various dc electric fields. The decrease of velocity with the electric field or the negative differential mobility is reported and has been confirmed to be crucial to the formation of charge packets. A weak trapping characteristic is equally essential to the charge packets. This indicates the change of dielectric properties of polymers in the presence of charge packets.

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