A model and simulation of fast space charge pulses in polymers

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Abstract
The transport of space charge packets across polyethylene and epoxy resin in high electric fields has been characterized as fast or slow depending on packet mobility. Several explanations for the formation and transport of slow space charge packets have been proposed, but the origins of fast space charge pulses, with mobilities above $10^{-11} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$, are unclear. In one suggested model, it is assumed that the formation of fast charge pulses is due to discontinuous electromechanical compression and charge injection at the electrode-insulation interface, and their transport is related to corresponding relaxation processes. In that model, charges travel as a pulse because of group polarization. This paper provides an alternative model based on the reduction of charge carrier activation energy due to charge density triggered polymer chain movement and subsequent chain relaxation times. The generation and transport of fast charge pulses are readily simulated by a bipolar charge transport model with three additional parameters: reduced activation energy, charge density threshold, and chain relaxation time. Such a model is shown to reproduce key features of fast space charge pulses including speed, duration, repetition rate and pulse size. This model provides the basis for a deep understanding of the physical origins of fast space charge pulses in polymers.

Keywords: space charge packet, electric field assisted chain movement, activation energy, chain relaxation, HVDC cable

(Some figures may appear in colour only in the online journal)

1. Introduction

Space charge accumulation is an important issue for high voltage direct current (HVDC) systems, as it can distort the electric field distribution in polymeric insulation, and thereby reduce the reliability of equipment. Space charge packets have been observed in polyethylene (PE) and epoxy resin. In these cases, space charge moves as a packet with an almost constant shape from an injection electrode to a counter electrode under the influence of a high electric field [1]. In general, space charge packets are divided into two types: fast charge pulses and slow charge packets [1]. Fast space charge packets are usually called fast charge pulses in most of the previous literature. Slow charge packets have mobilities in the range $10^{-16}$ to $10^{-14} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ under fields of $10^{-8} \text{ V m}^{-1}$, which is still in the range of normal space charge mobility under such conditions. The charge density of slow packets is usually several $\text{ C m}^{-3}$ or even higher. By contrast, fast space charge pulses can have mobilities as high as $10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ under fields of $10^{-8} \text{ V m}^{-1}$, and the charge density of a fast charge pulse is much lower than that of a slow charge packet with values between 0.07 and 0.1 $\text{ C m}^{-3}$ [1, 2].

There are several theories that have been used to explain the formation and movement of slow charge packets [3–7]. One is based on pulse-like charge injection from the electrodes due to electric field fluctuations which may cause hysteresis in
injection behavior [3, 4]. Another theory is based on negative differential mobility (NDM) of charge carriers in the bulk of the polymer [3, 5–7]. Chen, et al verified this effect by experiment, and simulated the behavior of slow charge packets with a bipolar charge transport model that takes NDM into account [5]. Baudoin et al compared the theories by simulation using a hysteresis loop in the injection process to verify the first theory and negative differential mobility to verify the other [3]. Nevertheless, the mechanism of fast charge pulse propagation remains unclear, and neither of these theories can explain the very high mobility of fast charge pulses.

In earlier work, Dissado et al studied fast space charge pulses in polyethylene, and concluded that the generation of fast charge pulses is due to discontinuous electromechanical compression and pulse-like charge injection at the electrode-insulation interface. They proposed that transport of charge pulses is related to a corresponding relaxation processes, which may help holes tunnel between nearby chains and electrons penetrate the hindering chain [2]. Their recent work supports the idea that group polarization around the fast charge pulse may produce a potential well that can bind the moving charges together [8]. Even so, there is still no numerical model for fast charge pulse generation.

In this work, an alternative model is proposed. Numerical simulations of fast charge pulses are presented, along with a detailed explanation of their formation, shape, and transport.

2. Physical model

One key characteristic required for charge transport in the form of a packet is that the charge mobility at the back of the packet should be at least as high as that in the front. Otherwise the carriers at the tail cannot follow those at the front, and the packet will disperse. The mobility change in slow packets can be explained by NDM. The region in front of a fast space charge pulse will have a mobility equivalent to the normal charge transport process. Within the fast space charge pulse the mobility is 4–6 orders of magnitude higher than normal. However, the NDM model does not explain such a significant mobility change.

Changing charge mobility by orders of magnitude suggests a physical mechanism that reduces the local activation energy. One proposed mechanism is that electric field assisted chain movement may separate polymer chains to allow electrons to pass between, or may rotate chains bringing them closer allowing holes to tunnel between them [2]. This results because the polymer chains have negative electron affinity [9], so electrons travel in free volume between the polymer chains, and holes transport along the chains, as shown in figure 1. As a result, close adjacent chains block free electron transport, while large distances between chains prevent holes hopping between chains. When the electric field and accumulated charge is high enough, it may effectively promote chain movement. As suggested in [2], β-mode chain movement may open up a blockage in front of pulses of electrons in an amorphous region, allowing electrons to move to next free volume. This enhancement of electron transport is illustrated in figure 1.

Hole transport may be enhanced through the ‘crank-shaft’ displacement of polymer chains (γ-mode chain movement) causing a chain with positive charge to kink towards an adjacent chain, allowing inter-chain transfer via electron tunneling. This effect is also illustrated figure 1. However, there is no direct proof yet to that specific chain movement is responsible for carrier transport.

Two questions arise. The first question is: ‘what triggers chain movement?’ Chain movement is heavily influenced by temperature and high electric fields [9]. A fast charge pulse peak charge density is only around 0.1 C m \(^{-2}\), and the total charge quantity is only 100–130 pC [1]. Such small charge quantities may not be considered large enough to cause sufficient local electric field distortion to trigger the movement of a polymer chain. Here two points should be noted: Firstly, the pulsed electro-acoustic (PEA) space charge measurement system, widely used for space charge mobility characterization, has a resolution limit. Even the surface charge accumulation at the electrode shows as a quasi-normal distribution with several tens of µm width [10]. The observed fast charge pulses always have a similar or smaller width than the charge peak at the electrodes, and the tail of the pulse is always flat (the density is below measurement sensitivity) [1, 2]. So the fast charge pulse is probably relatively narrow (sub µm), and the peak charge density much higher than the apparent value. Secondly, it is noted that carrier mobility in a crystalline region is much higher than that in an amorphous region. Furthermore, the crystalline-amorphous interfaces may also act as barriers blocking charge transport [11]. As a result, carriers may accumulate in amorphous regions, or even be localized at crystalline-amorphous interfaces, and thus the charge density would be even higher in such micro-structures creating more localized areas of carriers at a greater density. This results in relatively small charge quantities within the fast pulse causing much greater local charge densities at the µm level, potentially creating high enough fields to influence local polymer chain movement. The current measurements reported in [12] are supportive of this. It is reported that when the fast charge pulse reached the counter electrode, the out-circuit current sharply increased due to the increase of extraction current. The rising edge of the current is caused by the charge in the fast pulse reaching the electrode/polymer interface, so its high rate of change reflects the narrowness of the fast charge pulse. However, due to the nature of the PEA experimental system and the limited resolution of spatial distribution of space charge measurement, it is difficult presently to robustly model resulting electric field distortion. As a result, electric
3 field thresholds cannot simply be used as trigger conditions to determine conditions for chain movement.

Rather than attempt to model local fields on the micron scale, here we use the observed critical charge quantity \((C_m^{-2})\) or the corresponding charge density of fast pulses as the trigger condition for polymer chain movement. The critical charge quantity \((C_m^{-2})\) is seen to be very stable during individual pulse transport events [1]. We assume that when the charge quantity or the corresponding charge density reaches a threshold value, the local electric field is high enough to assist chain movement, thereby allowing local carriers to move with a reduced activation energy. In this paper, the charge density is thus used as the critical value.

The second question that will now be addressed is 'how does a charge pulse transport with an apparent stable shape?'

Physical defects block carrier movement, so that space charge transport can be understood as charges passing through a series of traps, as shown in figure 2. The difference between typical charge transport and a fast space charge pulse is that the electric field assisted chain movement assists carriers to traverse through those traps with lower activation energy and higher mobility than would otherwise be possible. Figure 2 illustrates this effect; polymer chains C, D and F moving in response to the presence of localized space charge, enabling passage of charges. This process is equivalent to an intelligent traffic light system on a highway. When the space charge density is lower than the threshold value, most of the space charge would be blocked, and has to wait for more space charge to accumulate, if the conditions permit. Once the space charge density reaches a threshold value (enough cars are queued at the lights) the chain moves removing the barrier to allow charge transport with high mobility. If the barrier is re-established immediately the charge density drops below the threshold, then space charge cannot transport in the form of a single pulse. If the barrier is re-established (the lights turn red) only after all the charge passes through with high mobility (as chains A, B and E illustrate in figure 2), the space charge will move collectively until it reaches a new barrier, and then wait until it reaches the threshold density again. If a charge pulse is large enough to overcome all barriers in its path, it will pass through the system as an entity with high mobility. Then the space charge moves in the form of a fast charge pulse.

As a result of its physical nature, the relaxation process of a polymer chain requires a characteristic, temperature dependent time \(t_r\) [9]. Thus, once the chain movement has been triggered by the enhanced local electric field, the following carriers can still pass through that location with high mobility (low activation energy) within \(t_r\) even after the electric field has reduced. From the analogy of the intelligent highway system, it can be seen that the chain relaxation time \(t_r\) is the key parameter that determines the overall sustainability and mobility of such a fast charge pulse.

In summary, three hypothesis are considered in the proposed model of fast charge pulses. These are based on conventional concepts of charge transport:

1. Local high electric fields can assist polymer chain motion which can reduce the overall activation energy of charge mobility in close proximity to the fast charge pulse;
2. The charge pulse magnitude needs to reach a threshold value, above which the local space charge distribution can cause a high enough electric field to assist chain movement;
3. The rate of chain movement is determined by a characteristic, temperature dependent relaxation time, \(t_r\). The reduced activation energy is retained for time \(t_r\) after the electric field is reduced by passage of the space charge pulse.

The three hypotheses define the physical nature, trigger conditions and relaxation conditions of chain movement, and they also correspond to three key parameters: reduced activation energy of mobility, \(W_r\), threshold space charge density, \(C_m^{-2}\).
Charge transport through the bulk insulation can be described with the convection-reaction equation, the Poisson’s equation and the transport equation shown below:

\[
\begin{align*}
\frac{\partial n}{\partial t} + \nabla \cdot \vec{J}_n &= S_a \\
\frac{\partial E}{\partial t} &= \frac{\rho_{em} - \rho_{hm}}{\varepsilon_0 c_n} \\
\vec{f}_a &= \pm \left( \mu_a E - D_t \frac{\partial \vec{E}}{\partial t} \right)
\end{align*}
\]  

where, the subscript a represents the four types of carrier (em, et, hm, ht) with em = mobile electrons, et = trapped electrons, hm = mobile holes, ht = trapped holes; \(n_a\) is the density of the carriers; \(\rho_{all}\) is the total charge density of all the four kinds of carriers at a specified position; \(f_a\) is the carrier flux under the electric field, which is constituted by the drift flux and diffusion flux; \(E\) is the local electric field; \(D_t\) is the diffusion coefficient; \(\mu_a\) is the carrier mobility; \(S_a\) is the reaction term, which includes carriers trapping, de-trapping and recombination:

\[
\begin{align*}
S_{em} &= -e\varepsilon_{em}\mu_{em}\rho_{em} - B_e n_{em} \left(1 - \frac{\mu_{em}}{\mu_{et}}\right) + \exp \left(\frac{\phi_m}{k_B T}\right) n_{em} \\
S_{et} &= -e\varepsilon_{et}\mu_{et}\rho_{et} - e\varepsilon_{et}\mu_{et}\rho_{et} - B_e n_{et} \left(1 - \frac{\mu_{et}}{\mu_{em}}\right) - \exp \left(\frac{\phi_e}{k_B T}\right) n_{et} \\
S_{hm} &= -e\varepsilon_{hm}\mu_{hm}\rho_{hm} - e\varepsilon_{hm}\mu_{hm}\rho_{hm} + B_e n_{hm} \left(1 - \frac{\mu_{hm}}{\mu_{ht}}\right) + \exp \left(\frac{\phi_m}{k_B T}\right) n_{hm} \\
S_{ht} &= -e\varepsilon_{ht}\mu_{ht}\rho_{ht} - e\varepsilon_{ht}\mu_{ht}\rho_{ht} + B_e n_{ht} \left(1 - \frac{\mu_{ht}}{\mu_{hm}}\right) - \exp \left(\frac{\phi_e}{k_B T}\right) n_{ht}
\end{align*}
\]  

Here \(S_{em,et,hm,ht}\) are the recombination coefficients between the charges of opposite signs. We ignore the recombination between the mobile electrons and holes as in [10], \(B_e\) and \(B_h\) are respectively the trapping coefficients of the electrons and holes. \(N_e\) and \(N_h\) are the deep trap density of the electrons and holes. The carriers in deep traps have a possibility to de-trap, \(v\) is the attempt-to-escape frequency, \(W_{et}\) and \(W_{hm}\) are separately the depths of the deep traps for electrons and holes.

The charge mobility \(\mu_a\) in previous simulations is either constant or electric field/temperature dependent [10, 13–16]. However, none of the models can explain the packet-like charge transport. As discussed in section 2, the fast charge pulse is considered to be caused by an increase of charge mobility due to electric field assisted chain movement. To simulate the fast charge pulse, a rapid change of mobility driven by the critical charge density is included in the simulation. This is the key difference between the model in this paper and previous models.

Critical charge densities \(\rho_{c}\) for positive and negative charge pulses are separately set to be 1.2 C m\(^{-3}\) and 0.6 C m\(^{-3}\), which are slightly lower than the values calculated by \(\sigma_c/\Delta d\). It is indicated in [2] that electron transport is due to hopping conduction, as shown in equation (5), and hole transport due to tunneling, as shown in equation (6).

\[
\begin{align*}
\mu_{em} &= \frac{k_b T R_{s1}}{h} \cdot \exp \left( -\frac{\Delta W_{e,em} - eE R_{s1}/2}{k_b T} \right) \\
\mu_{hm} &= k \cdot \frac{e R_s^2}{h} \cdot \exp \left( -\frac{\Delta W_{h,em}}{k_b T} \right)
\end{align*}
\]  

In equation (5), \(h\) is Planck’s constant; \(\Delta W_{e,em}\) is the activation energy of negative charges hopping in a negative fast charge pulse, and a value of 0.52 eV is given by [2]; \(R_{s1}\) is the size
of a free volume region; 5 nm is given by [2]. In equation (6), \( \Delta W_{e,h} \) is the activation energy for positive charges tunnelling in a positive fast charge pulse, 0.24 eV is the value given by [2]; \( R_{c2} \) is the compression produced by the pulse charge in the applied field, and considered to be 0.32 nm at 40 kV mm\(^{-1}\); the dimensionless constant \( k \) equals \( 2.45 \times 10^{-2} \) which is also given by [2]. Equations (5) and (6) yield a good fit to the experimental results of the apparent mobility of the charge pulse. An extra coefficient (2.4 for both \( \mu_{e} \) and \( \mu_{h} \)) is added to the two equations to define the mobility of carriers in fast charge pulses in the simulation to better fit the experimental results, and is discussed later. In the simulation, activation energies for mobilities are firstly set as \( W_{c} = 0.85 \text{ eV} \) for electrons and \( W_{h} = 0.65 \text{ eV} \) for holes to simulate the low charge mobility (\( \sim 10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1} \)); when the charge density of a layer is higher than the critical charge density \( \rho_{c} \), the activation energies of equation (5) are reduced to \( W_{c,e} \) and \( W_{h,h} \); when the charge density decreases to lower than \( \rho_{c} \), the activation energies revert to \( W_{c} \) and \( W_{h} \) after a relaxation time \( t_{r} = 40 \text{ ms} \). The time step of the simulation is 0.1 ms, and the total simulation time is 5 s. Carrier recombination, de-trapping from deep traps and non-blocking carrier extraction (extraction rate is based on Ohmic contacts) are considered. These are conventional settings for space charge simulation in polyethylene. Other details and parameters are the same as the previous simulation described in [10]. The parameters used in the simulation are listed in table 1.

Table 1. Parameters used for the simulation.

| Parameter | Value | Unit |
|-----------|-------|------|
| Thickness (d) | 1.5 | mm |
| Average electric field \( (E_{av}) \) | 40 | kV mm\(^{-1}\) |
| Temperature \( (T) \) | 308 | K |
| Time step \( (\Delta t) \) | 0.1 | ms |
| Simulation time \( (\tau) \) | 5 | s |
| Relative permittivity \( (\varepsilon_r) \) | 2.3 |
| Barrier of Schottky law for electron injection \( (W_{ei}) \) | 1.12 | eV |
| Barrier of Schottky law for hole injection \( (W_{hi}) \) | 1.09 | eV |
| Critical charge density for negative charge pulse \( (\rho_{c,-}) \) | 0.6 | C m\(^{-3}\) |
| Critical charge density for positive charge pulse \( (\rho_{c,+}) \) | 1.2 | C m\(^{-3}\) |
| Reduced activation energy for negative charge pulse \( (\Delta W_{e,h}) \) | 0.52 | eV |
| Reduced activation energy for positive charge pulse \( (\Delta W_{e,h}) \) | 0.24 | eV |
| Activation energy for electrons transport \( (W_{c}) \) | 0.85 | eV |
| Activation energy for holes transport \( (W_{h}) \) | 0.65 | eV |
| Size of free volume region \( (R_{d}) \) | 5 | Nm |
| Compression produced by the pulse charge \( (R_{c2}) \) | 0.32 | Nm |
| Coefficient in equation (6) \( (k) \) | \( 2.45 \times 10^{-2} \) |
| Chain relaxation time \( (t_{c}) \) | 40 | Ms |
| Diffusion rate \( (D_{e}) \) | \( 1 \times 10^{-13} \) | m\(^2\) s\(^{-1}\) |
| Extraction rate \( (C_{e}, C_{h}) \) | 1 |
| Recombination rate \( (S_{e+h,he}, S_{et,h}) \) | \( 1 \times 10^{-4} \) | m\(^3\) C\(^{-1}\) s\(^{-1}\) |
| Deep trapping rate \( (B_{e}, B_{h}) \) | \( 1 \times 10^{-3} \) | s\(^{-1}\) |
| Deep trap density \( (N_{e}, N_{h}) \) | 5 | C m\(^{-3}\) |
| Attempt-to-escape frequency \( (v) \) | \( 6.2 \times 10^{12} \) | Hz |
| Deep trap depth \( (W_{e}, W_{h}) \) | 1 | eV |

4. Simulation results and discussion

4.1. Generation and transport of fast charge pulses

Figure 3 shows the simulation results for a 1.5 mm thick sample. Each line corresponds to the space charge distribution at a specific time. The lines in figure 3(a) are predicted bulk space charge, and each peak is a fast charge pulse. The charge pulses in figure 3(a) show sharp peaks, however as discussed previously, PEA measurements yield charge densities with Gaussian-like peaks, irrespective of the real distribution. This effect lowers the apparent peak charge density and limits the system resolution. To compare the simulated results with experimental data, a Gaussian-like system function can be applied to the simulated data to mimic the loss of resolution associated with experiment. The lines in figure 3(b) show the apparent space charge which is the result of bulk space charge convolved with the Gaussian-like system function evaluated based on the results in [2]. Figure 3(a) omits the electrode surface charge for clarity. Figure 3(b) includes the surface charge at the electrodes where the charge density is much higher (about 8 C m\(^{-3}\)) than in the fast charge pulses. After convolution with the system function, the peaks in figure 3(b) can be favorably compared with the experimental results shown in [2] for both magnitude and shape. Even though the charge density of the apparent charge pulse is much lower than that shown in figure 3(a), the total charge quantity (C m\(^{-3}\)) is similar to the bulk space charge pulse.

It can be seen from figure 3(a) that at 0.4 s, the negative charge pulse has not appeared near the cathode, while a positive charge pulse has traveled one fifth of the insulation thickness from the anode. This is because the negative charge injection rate is lower than the positive, and so the negative charge accumulated near the cathode needs more time to reach the threshold density to generate the charge pulse. Positive charge pulses travel from anode to cathode, and negative charge pulses from cathode to anode. Both of them move with a stable mobility, shape and total charge. At 0.7 s, there are two positive charge pulses in the dielectric at the same time. This is possible because the positive charge pulse needs 0.52 s to pass through the 1.5 mm insulation, and a new positive charge pulse is generated each 0.3 s (consistent with the experimental data). At 0.7 s, the first negative and the first positive charge pulses have passed through each other, and the peak charge densities stay the same as shown in figure 3(a). This is because the period during which the positive and negative charge pulses pass through each other is very short and the recombination between the electrons and holes is limited.
Thus the overlap does not result in a change to the charge quantities within the pulses.

Table 2 compares the fast charge pulse characteristics of the experimental and simulation results. It can be seen that the simulations have successfully reproduced the features of the experimental results from PE under $40 \text{ kV mm}^{-1}$ at 308 K [2]. As the charge extraction is set to be Ohmic, heterocharge readily dissipates before the next fast charge pulse arrives again. In this model then, heterocharge does not accumulate. In order to simulate the heterocharge accumulation, a partial blocked charge extraction should be considered [10].

No extra features or hypotheses, beyond those already discussed, are needed to model charge pulse generation. Figure 4 illustrates the simulated pulse generation process from the anode. Before the generation of a pulse, positive charges are continuously injected from the anode into the polyethylene. However, due to low charge mobility, most of the injected charge accumulates in a layer (labelled 1st in the figure) which is nearest to the anode. At 0.275 s, the charge density in the 1st layer reaches the charge density threshold, and as a consequence, the activation energy of charge mobility decreases in the local polymer due to assisted chain movement. Then the charge in the 1st layer is quickly transported under the high field as the blue and red lines show in figure 4 (to the region labelled as the 2nd layer). At this moment, the mobility in the 1st layer is still in the higher state due to the time delay of the relaxation of chain movement while, if the charge density of second layer has not reached the threshold, the charges in that layer still have relatively limited mobility. The charges at the 2nd layer are effectively slowed enabling those in the 1st layer to catch up. During this period, the charge density of 1st layer decreases, the charge density of 2nd layer increases, and the peak charge density shifts from the 1st layer to the 2nd layer. When the charge density of 2nd layer reaches the threshold, the pulse continues to progress through the polymer through the same process. As shown in figure 4, the peak
shifts from the 2nd layer to the 3rd layer between 0.284 s and 0.292 s after the experiment starts. It can be seen that based on this model, no extra hypothesis is needed for pulse generation. Once the accumulated space charge density reaches the threshold, the pulses can be generated from within the sample: The generation and transport of fast charge pulses in this model are the same process. As a result, the current in the circuit changes continuously and gradually with the field distortion due to slow homocharge accumulation. If, based on the hypothesis that the generation of fast charge pulses is due to discontinuous electromechanical compression as discussed in [2], the charge injection rate should change significantly with electromechanical compression, and would be pulse-like. Also the injection current would act as a pulse which corresponds to the injection of the fast charge pulse. This contradicts the measured circuit currents which show that peaks correspond to extraction of charge pulses when the fast charge pulses reach the counter electrode, rather than charge pulse injection [12].

4.2. Influence of chain relaxation time

In this model, three parameters corresponding to three hypotheses control the generation and transport of fast charge pulses. Here, the influence of chain relaxation, charge density threshold and reduced activation energy are discussed. Firstly, the chain relaxation time is considered, as it is the parameter that determines shape preservation of the fast charge pulse.

In the simulation above, the relaxation time was chosen as $t_r = 40$ ms. Here a shorter relaxation time $t_r = 2$ ms is employed for comparison. The simulation results with the shorter relaxation time are shown in figure 5. In figure 5(a) both positive and negative charge propagate as a pulse with a much lower speed than that in figure 3. The apparent mobility of positive charge pulse is $1.4 \times 10^{-12}$ m$^2$ V$^{-1}$ s$^{-1}$. The apparent mobility of negative charge pulse is $2 \times 10^{-12}$ m$^2$ V$^{-1}$ s$^{-1}$. The speed is much lower than the fast charge pulse, but still much higher than the normal charge transport ($10^{-14}$ m$^2$ V$^{-1}$ s$^{-1} - 10^{-16}$ m$^2$ V$^{-1}$ s$^{-1}$). The charge pulses have a long tail behind the peak with a considerable charge density. After convolution with the system function of the measurement system, the apparent space charge dynamic appears as a propagation front (or accumulated homocharge with a density about 0.6 C m$^{-3}$ for positive and 0.2 C m$^{-3}$ for negative), rather than a fast charge pulse, as shown in figure 5(b).

In summary, by considering the space charge density triggered electric-field-assisted chain movement, and a long enough relaxation time, observed fast charge pulses can be reproduced by simulation; but with a short relaxation time, a fast ‘propagation front’ is predicted to be observed by PEA instead of fast space charge pulses.

As discussed previously, the electric field assisted chain movement helps carrier transport by opening the traps which may block and accumulate carriers. Here we suppose the traps have an average distance $\Delta x$ between each other. When the total charge $\sigma(x, t)$ accumulating at position $x$ reaches the critical value of $\sigma_c$ (or the corresponding charge density reaches $\rho_c = \sigma_c/\Delta x$), the chain is able to move, the trap is opened, and the charge mobility increases greatly with the decrease of activation energy. Here, this moment is set as $t = 0$. Then charges start to travel to the next trap with a speed of $\rho(x, t) \times E \times \mu$. Where $\rho(x, t)$ is the charge density, $E$ is the local electric field, and $\mu$ is the mobility of the charge in the fast charge pulse. The total charge flow can be described as:

$$d\sigma(x, t) = -\rho(x, t) \cdot E \cdot \mu \cdot dt = -\frac{\sigma(x, t)}{\Delta x} \cdot E \cdot \mu \cdot dt. \quad (7)$$

So that the charge left at location $x$, at time $t$ after the chain moves and before the chain relaxes back its origin position is given by:

$$\sigma(x, t) = A \cdot e^{-at} \quad (8)$$

where $a = E \times \mu/\Delta x$. When $t = 0$, the total charge amount is $\sigma_c$, so $A = \sigma_c$. The longer the time $t$, the smaller amount of charge left. So a long relaxation time would ensure most of the charge can follow the pace of the fast charge pulse, as shown in figure 3; a short relaxation time would cause charge to be left behind as a tail, as shown in figure 5.
Equation (8) can also be re-written:

\[ t = -\frac{\Delta x}{E \cdot \mu} \cdot \ln \left( \frac{\sigma(x, t)}{\sigma_c} \right). \]  

(9)

Here three points need to be discussed. Firstly, the first term is the time (set as \( t_0 \)) for one charge in a fast charge pulse to travel \( \Delta x \). If \( \sigma(x, t) \) is 1% of \( \sigma_c \), then the time is \( 4.6 \times t_0 \). In the simulation, the mobility of positive charge with reduced activation energy is \( 1.63 \times 10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1} \), and the mobility of negative charge with reduced activation energy is \( 2.4 \times 10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1} \). The distance between traps is the thickness of a simulation layer 15 \( \mu \)m. The average electric field is 40\( \text{kV mm}^{-1} \). So it is easy to get the \( t_0 \) for positive and negative charge separately as 2.3 ms and 1.56 ms. The chain relaxation time of 40 ms is much longer than these values of \( t_0 \), so that most of the charges are able to follow the pulse. The chain relaxation time 2 ms will allow only 58% of positive charge and 72% of negative charge to follow in the pulse. It cannot allow the charge pulse to move as an entity. Besides, 42% of positive charge (42% \( \times \) 1.2 C m\(^{-3} \) \( \approx \) 0.5 C m\(^{-3} \)), and 28% for negative charge (28% \( \times \) 0.6 C m\(^{-3} \) \( \approx \) 0.17 C m\(^{-3} \)) are left behind and form the ‘tails’ after the propagation front with a charge density of about 0.6 C m\(^{-3} \) for positive and 0.2 C m\(^{-3} \) for negative, as shown in figure 5.

Secondly, the charge mobility with reduced activation energy is higher than the apparent mobility of fast charge pulses. This is because when the charges in a fast charge pulse reach the next trap, they are blocked and have to wait for the chain movement. At that time they have to overcome a high activation energy to get through the trap, so the mobility is very low. This is the reason why an extra coefficient is used to define the mobility in simulation based on the experimental results.

Thirdly, from equation (9) it can be seen that the smaller the distance between traps, the shorter the chain relaxation time required to enable fast charge pulse formation. Presently we have little information concerning the chain relation times and the distance between the traps. However, as discussed above, when the fast charge pulse reaches the counter electrode, the out circuit current has a fast rise time [12]. The increase of the current is due to sudden increase of the extraction current when the fast charge pulse passes through the last trap and reaches the surface of the polymer. If we suppose the extraction current is linear with the amount of charge on the polymer surface, then the rising edge of the current starts at the first moment when the front of fast charge pulse reaches the polymer surface, and ends when most of the charge has reached the polymer surface. So the duration of the rising edge (\( \Delta t \)) can be used to estimate the minimum chain relaxation time, and roughly calculate the distance between traps as \( \Delta x = E \times \mu \times \Delta t \). Here the apparent mobility of a fast charge pulse can be used for \( \mu \). With this information, further observations about the chain movement and corresponding trap level can be made.

### 4.3. Influence of charge density threshold

According to the discussion above, the electric field has a threshold for electrical assisted chain movement. Because of

Figure 6. The first fast charge pulses in simulations with different threshold charge densities. Each is shown at different time so that they are spatially separated in the illustration.

the difficulty in modelling the values of local electric fields on appropriate micron scales, the charge density has been used in this model as an alternative threshold.

To explain the effect of charge density threshold, simulations with different threshold charge densities (\( A = 0.6, B = 1.2, C = 1.8, D = 2.4, E = 3 \text{ C m}^{-3} \)) are employed with a fixed chain relaxation time of 40 ms. All parameters for positive and negative charges (injection, extraction, charge mobility, et al) are set to be the same in this simulation, so that symmetrical results can be obtained and are readily analyzed. Other parameters are the same as in the previous simulation above. Figure 6 shows the simulated first charge pulses in the simulations with different threshold charge densities. For better illustration, pulses at different times are shown so that the pulses appear at different positions. It can be seen that the higher the set threshold value, the higher the peak value of the fast charge pulse. Table 3 lists the total charge in a pulse, the start time (for the first charge pulse), the duration of each pulse and the repetition rate.

From table 3, it can be seen that the higher the threshold charge density, the higher the total charge quantity, and a longer time is required to generate a fast charge pulse. A higher threshold charge density results in a lower pulse repetition rate. This is because more time is required for charge injection to accumulate enough charge to reach the threshold. The total charge in the pulse and the time of the first charge pulse have a linear relationship: the ratio being the injection current density.

Moreover, table 3 shows that the charge pulse transit time (duration) decreases with the increase of threshold value. At higher threshold values, more charge would transport deeper into the sample before the charge pulse is generated. Thus when the pulse moves deeper to the sample, the pre-existing charge at that position would reduce the time for the charge density to reach the threshold value. This would provide a higher current flux, and so less time is needed for the forward position to reach the threshold value and less time is needed for the fast charge pulse to traverse the sample.
In summary, the conditions for polymer chain movement determine the conditions for fast charge pulse formation, and the associated relaxation time determines the shape preservation of the fast charge pulse. These in turn depend on the microstructure of the polymer chains and polymer morphology [17–19].

In PE there are three main characteristic relaxation processes (chain movement) which are designated as α, β and γ relaxations [17–19]. α relaxation corresponds to the motion of chain units in crystalline region. β relaxation corresponds to the motions of smaller groups or segments, such as chain-end motion, or branch-point motion in amorphous or interphase regions. γ relaxation corresponds to the rotation of C–C bonds in amorphous regions [17, 18]. Different chain motions thus require different activation energies to trigger movement: α - 1.2 eV, β - 0.6 eV and γ - 0.2 eV [17]. They also need different times to move or return to original positions (relaxation times) [18, 19]. Direct measurements and simulations show that polyethylene has multi-levels of traps (identified as chemical and physical in their nature) which limit the charge transport [20, 21]. The physical traps are formed by the disorder of the chain structure and local morphology [21, 22]. Those traps together control the apparent charge mobility [20–22]. The electric field assisted chain movement is regarded as opening traps down to a certain depth. If those trap levels dominate the normal charge transport process, the resulting chain movement will greatly increase the apparent mobility. The study of the chain movement should then provide information on the trap level.

5. Conclusions

The model proposed here for the generation and transport of fast charge pulses in polymer insulation is based on a specific physical mechanism: electric field assisted chain movement. There are three key assumptions:

1. Local high electric fields can assist polymer chain movement, thereby reducing the overall activation energy of charge mobility around fast charge pulses.

2. The charge pulse density can reach a threshold value above which the local space charge distribution generates a high enough electric field to assist chain movement.

3. After field assisted chain movement, the polymer chains need a characteristic, temperature dependent time \( t_r \) to relax. As a result, the reduced activation energy is retained for a time \( t_r \), after the charge pulse has passed, and the local electric field is reduced.

As shown by the simulations, fast charge pulse behavior is revealed in a bipolar charge transport model with just three parameters: charge density threshold, activation energy, and chain relaxation time. These three parameters determine the possibility and consequent nature of fast space charge pulse transport. The success of this model in reproducing experimental behaviour provides the basis for an understanding of the origins of fast space charge pulses observed in polymers.

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