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Observation of negative differential mobility and charge packet in polyethylene

George Chen and Junwei Zhao

Faculty of Physical and Applied Science
University of Southampton
Southampton SO17 1BJ United Kingdom

Abstract

Charge packet has been observed on many occasions but its physical mechanisms have never been properly understood. One of the models proposed by Lewis et al shows the presence of negative differential mobility with an electric field in semi crystalline polyethylene. In this letter we have observed the negative differential mobility using the transient space charge profile measured by the pulsed electroacoustic technique. By superimposing a short pulse voltage to a dc voltage, it is possible to obtain the velocity of holes at different applied fields. To our knowledge we have for the first time observed negative differential mobility in polyethylene. This observation provides crucial evidence to support Lewis's model and allows one to simulate charge packet and its behaviours.

Polymeric materials have been widely used as insulation in power engineering due to their excellent dielectric properties. With the advances in manufacturing and processing technology, the defects in these materials have been reduced significantly over the years. Accordingly, the operating electric field of the materials has been increased steadily to minimise the cost of power equipment and devices. On the other hand, electrical phenomena under high electric field become of significant importance as they are not only scientifically interesting but also related to the reliable operation of many devices. It has been recognized that the reliability issue of the insulation materials operating at high electric field is closely related to space charge formation and its dynamics.

The formation of space charge in dielectric materials at high electric field is a well-known phenomenon and has been a hot topic of scientific research in the last few years thanks to the significant developments in space charge mapping techniques. The presence of space charge will distort the applied electric field and may lead to a more severe electric field enhancement resulting in material degradation/ageing [1]. On the other hand, space charge dynamics in the material have been used as an ageing marker to assess the status of the material [2-3]. One of the observed and well-published phenomena in space charge research is charge packet and its dynamics [4-10]. A charge packet can be loosely defined as a pulse of net charge that propagates across the material under the influence of electric field while maintaining its shape. It has been termed as an unexpected phenomenon and usually formed under high electric field and propagates through the material from one electrode to the other. In many cases, positive charge packet was reported although negative charge packet was also observed. Different mechanisms have been put forward to explain various features of charge packet. In recent paper by Lewis et al [11], they proposed the velocity-field characteristic for holes based on their charge transport model in polyethylene. The velocity of holes increases initially with the field and reaches its maximum at the threshold field E_T . After E_T the velocity of holes decreases with the increasing electric field as shown in Figure 1. When the applied field is lower than E_T , any injected holes from the anode will modify the applied field. The embryo of charge packet is characterised by the field E_A (rear) and E_B (front) with $E_B > E_A$. The corresponding velocities are V_A and V_B . From Figure 1, it can be seen that $V_A < V_B$, meaning the charge packet will disperse during its propagation. However, when the applied field is above E_T the velocities of rear and front charge packet change to V'_A and V'_B . As $V'_A > V'_B$ the charge packet will increase. The growing packet reduces E'_A and increases E'_B until the corresponding velocity becomes the same, i.e. $V'_A = V'_B = V_P$.

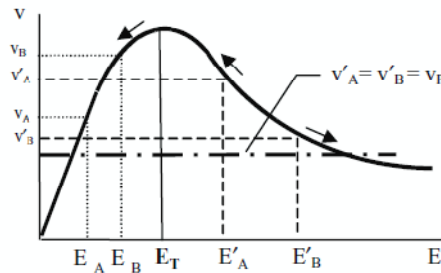


Figure 1 Velocity – field relationship for holes in polyethylene [11].

The characteristic described above is similar to part of the Gunn Effect [12] observed in semiconductors but the mechanisms are probably very different. For the Gunn Effect, it is believed that electrons can exist in a high-mass low velocity state as well as their normal low-mass high velocity state in semiconductors. The normal states can be forced into the high-mass state by an electric field of sufficient strength. In this state electrons form clusters or domains which cross the region at a constant rate causing current to flow as a pulse. The detailed mechanism can be traced back to two conduction band energy levels, Γ (normal lower valley) and L (satellite valley). In the lower Γ valley, electrons exhibit a small effective mass and very high mobility while in the satellite valley electrons possess a large effective mass and very low mobility. The two valleys are separated by a small energy gap. Initially, most electrons reside near the bottom of the lower Γ valley and they can readily be accelerated in a strong electric field to the energy in the order of the Γ -L inter-valley separation. Electrons are then able to scatter into the satellite valley, resulting in a decrease in the average electron mobility. Above the threshold field E_T , most electrons reside in the L valley. In polyethylene, the energy landscape for semiconductors may not apply. According to the model proposed by Lewis et al [11], hole transport in polyethylene can only occur via electron vacancies in or closely associated with the valence band and is consequently confined to polymer chains. Continuous movement of holes requires inter-chain hole transfer via tunnelling between closely adjacent polymer chains including chains in amorphous regions. Therefore, hole conduction process is more sensitive to morphology of polyethylene. When an electric field is applied to the material reorganisation of molecular chains in amorphous regions will take place due to induced mechanical stress. They have concluded that the above process would have adversely influence on hole tunnelling,

consequently reducing hole mobility and encouraging hole trapping, leading to the proposed velocity-field diagram shown in Figure 1.

The above velocity and field characteristic has been used in simulation of charge packet behaviour in polyethylene [13]. However, there is no convincing experimental evidence so far to verify the model. In this letter, we report the velocity-field characteristic in polyethylene for holes based on transient space charge profiles. The pulsed electroacoustic (PEA) technique was utilised for space charge measurement and the applied field ranges from 10 kVmm^{-1} to 70 kVmm^{-1} . Additive free low-density polyethylene (LDPE) films with a thickness of $100 \mu\text{m}$ were used in the present study. All the measurements were carried out at room temperature.

Different methods have been used to measure charge mobility in solid dielectrics such as time-flight method, transient space charge limited current method and surface potential decay method. Recently, Hozumi et al [14] have proposed a new method to estimate mobility based on transient space charge measurement. By superimposing a pulse voltage to the applied dc voltage, it is possible to generate a charge packet that moves under the influence of the electric field. The schematic principle of the method is shown in Figure 2.

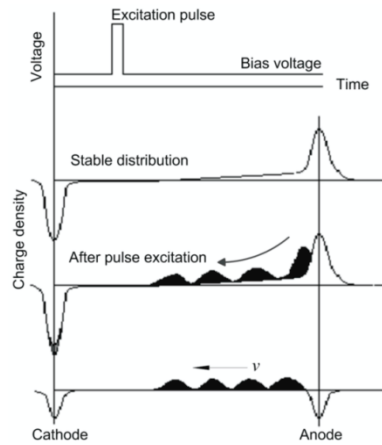


Figure 2 Transient space charge profile and velocity estimation for holes.

When the external applied field is high enough charge injection takes place due to either Schottky injection or tunnelling. The injected charge will move under the influence of the electric field and may reach a quasi-equilibrium after a period of time. In addition to the material itself, this time also depends on the applied field strength and temperature. An electric pulse is then applied and an extra charge packet will be initiated and move across the material. By subtracting the steady-state charge profile the small charge packet due to the pulse voltage becomes clearly visible and its movement can be used to estimate the velocity of the charge as shown in Figure 2. It has been reported [4] that the semicon electrode (polyethylene loaded with carbon black) injects more easily than the electrodes such as aluminium or gold. So in the present case the semicon was used as the anode and aluminium as the cathode to promote hole injection, and therefore, positive charge packet.

Figure 3 shows the transient charge distribution at an applied field of 20 kVmm^{-1} and subtracted plots. The magnitude of the pulse voltage is about 10 kV with a pulse width of 0.25 s . The pulse magnitude is decreased when the applied voltage is increased while the pulse width remains the same. The packet charge is not obvious without the subtraction, but it can be clearly seen that the movement of charge packet goes towards the cathode after the subtraction as shown in Figure 3 (b). The velocity of holes depends on the applied field as shown in Figure 4 where the applied field is 50 kVmm^{-1} . It can be seen that the velocity of the charge packet is faster compared with that at 20 kVmm^{-1} . On the other hand, the injected negative charge seems to move very fast.

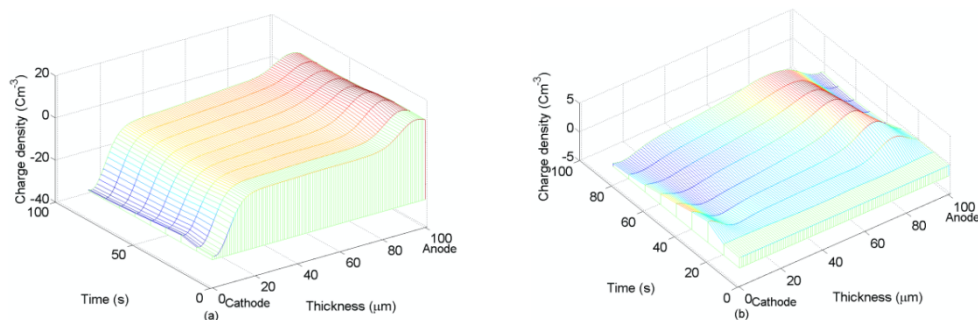


Figure 3 Transient space charge profiles at 20 kVmm^{-1} after pulse excitation: (a) before subtraction; (b) after subtraction.

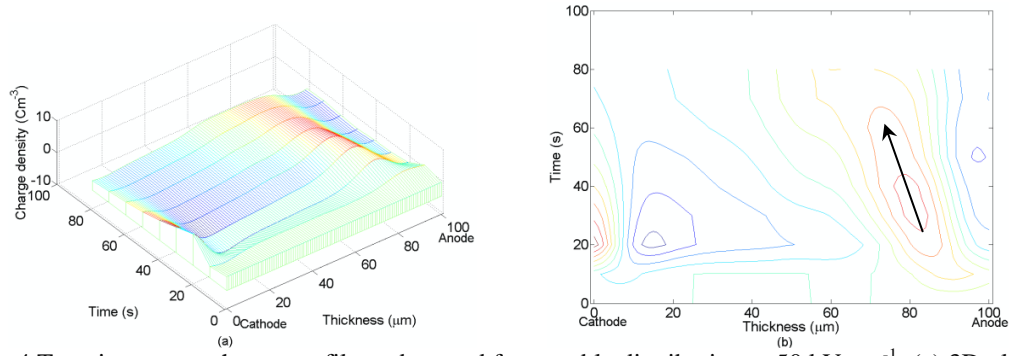


Figure 4 Transient space charge profiles subtracted from stable distribution at 50 kVmm^{-1} : (a) 3D plot; (b) contour plot.

A range of applied electric fields from 10 kVmm^{-1} to 70 kVmm^{-1} were investigated and the velocity obtained is illustrated in Figure 5. Each data point is the average of at least three measurements. When the applied field exceeds 70 kVmm^{-1} it takes a longer time to achieve the steady state. Therefore, the present results are limited to 70 kVmm^{-1} . However, it becomes very clear that the proposed characteristic of the velocity-field by Lewis et al [11] has been, to our knowledge, for the first time experimentally observed. The velocity and electric field relationship in Figure 5 deviates slightly from the one where the applied field is exceeding 60 kVmm^{-1} . Instead of decreasing continuously the velocity of holes increases. The shape is strikingly similar to the Gunn effect occurred in semiconductors.

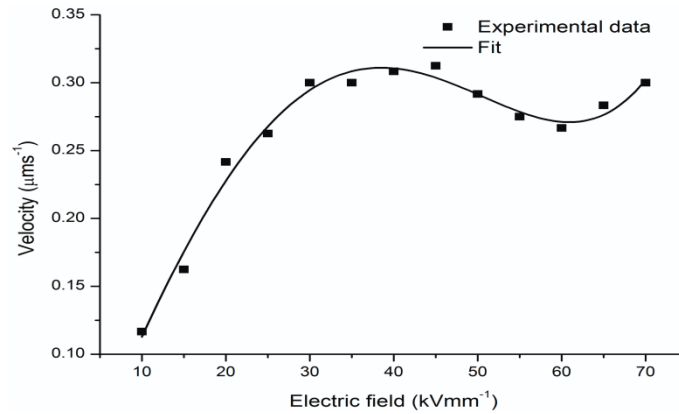


Figure 5 Dependence of hole velocity on the applied electric field.

The Gunn Effect is responsible for the negative differential resistance observed in semiconductor materials. The above relationship may be directly related to the negative differential resistance observed in polyethylene reported many years ago [15-16] where the current and voltage characteristics of very thin polyethylene film with varying thickness from 10 to 50 nm were investigated. Negative differential resistance was observed and the onset of the negative differential resistance was found as low as 27 kVmm^{-1} . Crine [17] has termed the negative differential resistance in polyethylene as one of the unexplained disturbing phenomena in the electrical properties of dielectric polymers. Based on the result obtained in this letter, the negative differential resistance observed in polyethylene can be attributed to the change in charge carrier mobility. As mentioned earlier, the charge packet has been observed under a wide range of conditions, the negative differential mobility obtained in the present work may be responsible for those observed in the electric field range below 80 kVmm^{-1} . It is believed that electron injection takes place as well especially when the positive charge packet moves close to the cathode. However, injected electrons seem to move very fast which may contribute to the formation of charge packet. On the other hand, the real charge packet always changes its shape during the propagation, i.e. diffused. This may be related to recombination of holes and electrons. The charge packet observed above 100 kVmm^{-1} may take place based on the different mechanisms such as a consequence of charge injection, ionisation and recombination. Currently, simulations using the velocity-field characteristic are underway to study charge packet behaviours in polyethylene. More importantly, the mobility versus electric field in aged polyethylene will also be investigated to unravel the relationship between ageing and charge mobility.

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