Combined Electronic and Thermal Breakdown Models for Polyethylene and Polymer Laminates

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Abstract: A comprehensive DC dielectric breakdown model has been extended for polyethylene in which specific time and voltage regimes were identified for thermal and electrical failure modes. In addition to electron injection processes, space-charge formation and the associated electric field distribution are critical in determining the conditions for insulator failure. Space charge formation and distribution are governed by electron and hole injection from the electrode as well as charge mobility and trapping. Breakdown fields of 5 MV/cm are predicted through a computational model for polyethylene films in the 5 to 200 μm thickness range and there is weak dependence of breakdown field on thickness. The model is applied to predict the effect of interfaces for multilayer polymer structures that act as barriers.

Introduction

The high voltage reliability of polyethylene is important for future DC cable applications. Past studies have explored dielectric breakdown mechanisms including electronic, thermal, electromechanical and partial discharge assuming no space charge [1-5]. Space-charge-limited current arises from local electric field distortion which affects current density[6-8]. Space charge mediated dielectric breakdown has been initiated through pre-stress experiments[9, 10]. Moreover, advances in nondestructive measurement techniques such as laser-induced pressure pulse (LIPP), pulsed electro-acoustic (PEA) and thermal pulse methods have made the direct measurement of space charge induced from high electric field possible[11, 12]. Space charge from charge injection and transport in low-density polyethylene(LDPE) was observed through the PEA method and analyzed with a trapping/detrapping behaviors of charge carriers[13]. Therefore, the breakdown models of polyethylene should be modified to accommodate space charge effects.

A theoretical breakdown model that predicts not only electronic breakdown but also thermal breakdown was first proposed by Fukuma et al. for polypropylene[14]. This model assumes electron injection at the cathode through Schottky mechanism and charge transport by hopping conduction mechanism through the bulk of the polymer film. Space charge effects on electric field distribution and charge transport are governed by Poisson's equation. Thermal breakdown was expected when elevated local temperature from Joule heating exceeded melting temperature of polypropylene before electric field reached intrinsic breakdown strength. A similar model was applied to LDPE where pressure effects on breakdown strength were also investigated[15]. Although these models have provided general agreement with experiments, the Richardson constant among model parameters showed unrealistically low values of $0.1 \text{ Am}^{-2}\text{K}^{-2}$. Recently, bipolar charge transport model was proposed for LDPE and their modeling of charge profile and electric field distribution showed good agreement with experiments[16-18]. Bipolar charge transport model assuming constant mobility was also applied to electronic breakdown of LDPE and used to explain thickness dependence of breakdown strength at room temperature[19].

The aim of this work is to develop combined thermal and electronic breakdown models based on bipolar charge transport and charge injection. For the mobility of charge carriers, two kinds of mobility will be addressed including constant mobility and hopping mobility. This model provides not only a prediction of the breakdown strength but also field, time and temperature effects on the space charge dynamics and the resultant electric field evolution during breakdown process. Recently, polymer laminates with high breakdown strength have been demonstrated where the dielectric breakdown fields have doubled over neat polymers[20]. The model is extended to predict the role of added interfaces on the dielectric breakdown strength of polyethylene.

Breakdown Model and Numerical Techniques

Electronic breakdown is initiated by the local electric field enhancement from space charge formation and thermal breakdown occurs from Joule heating by a transient conduction current. Considering that the thickness is much smaller than the width and breadth of the sample, a one-dimensional problem is assumed for thermal breakdown.

$$C_v \frac{\partial T}{\partial t} - \kappa \frac{\partial^2 T}{\partial x^2} = J(x, t)E(x, t) \quad (1)$$

where x is the distance from the cathode in the direction of thickness, T is the local temperature, t the time, C_v the volumetric heat capacity, κ the thermal conductivity, J the transient current density, and E the local electric field. At the breakdown criterion, thermal breakdown occurs when a local temperature in the dielectric attains the melting temperature of LDPE. For impulse thermal breakdown, thermal conduction is ignored, and this assumption has often been used to model breakdown process in polymers [14, 15]. However, impulse thermal breakdown is only valid if the time to breakdown is smaller than thermal relaxation time [21]. For a LDPE with thickness of 25 μm the time to breakdown is about 30 seconds at 25 $^{\circ}\text{C}$ assuming 300 V/s of ramp rate. Meanwhile, the thermal relaxation time (τ_d) governed by

$$\tau_d = \frac{d^2 C_v}{4\kappa} \quad (2)$$

where d is the sample thickness, which yields a thermal relaxation time in the millisecond range. The time to breakdown is much longer than the thermal relaxation time and thermal conduction cannot be ignored and assumption of impulse thermal breakdown is invalid in our case.

Charge injection and space charge formation in dielectric materials are closely related with the electronic breakdown. Space charge dynamics plays an important part in breakdown process because it governs the local electric field. It has been experimentally demonstrated that under high DC electric field the charge injection and space charge formation occurs in polyethylene[13, 22-24]. In the breakdown model, it is assumed that injected charges from

electrodes produce space charge within bulk and its electric field enhancement determines the breakdown. Breakdown occurs when local electric field within the bulk reaches intrinsic breakdown strength of polyethylene. The intrinsic breakdown strength of polyethylene is determined based on the literature and set to 680 MV/m [25].

The theoretical breakdown field based on electronic and thermal models can be determined if the electric field and the temperature in the insulator is numerically calculated for each time and each position. For every time step, the local temperature is compared with the melting point and the local electric field is compared with intrinsic breakdown strength. Dielectric breakdown occurs when either of the two criteria is reached. Both electric field and temperature are evaluated from charge injection and charge transport through Poisson's equation and Joule heating.

Charge generation and transport models are described by injection, transport, trapping and recombination of bipolar charges to predict space charge evolution in polyethylene [17, 18]. Charge carriers are injected at the interface of electrode/dielectric according to the Schottky process. The two kinds of carriers can be either trapped or mobile within the dielectric. Under an externally applied electric field, mobile electrons in the conduction band (or holes in the valence band) drift with an effective mobility that is governed by trapping and detrapping in shallow traps. Deep trapping is described as a single traps level in which electrons or holes have insufficient energy for detrapping. The recombination process between the electron-hole pairs are also taken into account. After recombination, the carrier in the trap is released and thus trapping is feasible again. Initially no mobile charges are assumed within the dielectric. Dipolar polarization has been ignored considering the non-polar nature of polyethylene.

Assuming one-dimensional problem along the thickness, the current densities at the cathode ($x=0$) and the anode ($x=d$) from the Schottky mechanism is governed by

$$J_e(0, t) = AT^2 \exp\left(\frac{-w_{ei}}{k_B T}\right) \exp\left(\frac{e}{k_B T} \sqrt{\frac{eE(0, t)}{4\pi\epsilon}}\right),$$

$$J_h(d, t) = AT^2 \exp\left(\frac{-w_{hi}}{k_B T}\right) \exp\left(\frac{e}{k_B T} \sqrt{\frac{eE(d, t)}{4\pi\epsilon}}\right), \quad (3)$$

where $J_e(0, t)$ and $J_h(d, t)$ are the fluxes of electrons and holes at cathode and anode respectively, A is the Richardson constant, T the temperature, k_B the Boltzmann constant, $E(0, t)$ and $E(d, t)$ the electric fields at the cathode and the anode, ϵ the permittivity of the dielectric, e the elementary electron charge, w_{ei} and w_{hi} the potential barriers for electrons and holes. Assuming no extraction barriers at electrodes, extraction current densities for electrons at the anode and holes at the cathode are described by

$$J_e(d, t) = \mu_e n_{em}(d, t) E(d, t),$$

$$J_h(0, t) = \mu_h n_{hm}(0, t) E(0, t), \quad (4)$$

where n_{em} and n_{hm} are charge densities for mobile electrons and holes, and μ_e and μ_h are the effective mobility for the electron and the hole.

The behavior of charge carriers within dielectrics as a function of time and distance are governed by three fundamental equations:

Poisson's equation

$$\frac{\partial E(x, t)}{\partial x} = \frac{\rho(x, t)}{\epsilon}, \quad (5)$$

Transport equation

$$J(x, t) = \mu n(x, t) E(x, t), \quad (6)$$

Continuity equation

$$\frac{\partial n(x, t)}{\partial t} + \frac{\partial J(x, t)}{\partial x} = s, \quad (7)$$

where E is the electric field, ρ the net charge density, n the charge density for each carrier, and s is the source term. The source term defines variations in local charge density due to trapping and recombination processes. For each charge type, mobile or trapped, the source term is described by

$$\begin{aligned}
s_1 &= -S_1 n_{ht} n_{em} - S_3 n_{hm} n_{em} - B_e n_{em} \left(1 - \frac{n_{et}}{N_{0et}}\right) \\
s_2 &= -S_2 n_{et} n_{hm} - S_3 n_{hm} n_{em} - B_h n_{hm} \left(1 - \frac{n_{ht}}{N_{0ht}}\right) \\
s_3 &= -S_2 n_{hm} n_{et} - S_0 n_{ht} n_{et} + B_e n_{em} \left(1 - \frac{n_{et}}{N_{0et}}\right) \\
s_4 &= -S_1 n_{ht} n_{em} - S_0 n_{ht} n_{et} + B_h n_{hm} \left(1 - \frac{n_{ht}}{N_{0ht}}\right)
\end{aligned} \tag{8}$$

where s_1 , s_2 , s_3 , and s_4 are the respective source terms for mobile electrons, mobile holes, trapped electrons, and trapped holes; the charge density terms n_{em} , n_{et} , n_{hm} , and n_{ht} are for mobile electrons, trapped electrons, mobile holes, and trapped holes; S_0 , S_1 , S_2 , and S_3 are the recombination coefficients between four different electron-hole pairs; B_e and B_h are the trapping coefficients for electrons and holes; N_{0et} and N_{0ht} are the trap densities for electrons and holes.

To solve the coupled problem of Eqns. (1), (5), (6), and (7) for the breakdown model, several numerical techniques are utilized. The electric field and potential in the dielectric are evaluated using finite difference method (FDM) through direct discretization of Eqn. (5). The modeled LDPE sample is divided into m slices and the electric field difference at each cell associated with the net charge density can be calculated with the boundary condition. The continuity equation, Eqn. (7), can be evaluated using a partitioning technique such that in a first step the continuity equation without the source term is calculated using upwind scheme and then in a second step the charge density is updated with the source term. The time step in the calculation is selected based on the condition of Courant-Friedrichs-Lewy which is necessary for the stability of the numerical technique. The power balance equation, Eqn. (1), can be calculated using the FDM with the backward Euler method in time and the forward Euler method in spatial coordinate. The backward Euler method in time is applied due to its superior numerical stability regardless of the time step although the computation is much rigorous.

Bipolar transport model with the constant mobility was used to predict space charge dynamics, external current density, and electroluminescence in LDPE material has been supported by experimental measurements under various DC electric field conditions[17, 26, 27]. Constant mobility for electrons and holes is assumed and the mobility of electrons is higher than that of holes. These parameters as shown in Table 1 will be employed to describe electronic breakdown and thermal breakdown process for LDPE samples with thickness between 5 and 200 μm under dc stress from 100 to 500 V/s voltage ramp rate.

Table 1 Parameters in the simulation of breakdown strength using constant mobility.

Parameters	Value	Unit	Reference
Barrier height for injection			
w_{ei}	1.15	eV	[27]
w_{hi}	1.14	eV	[27]
Mobility			
μ_e	9.0×10^{-15}	$\text{m}^2 \text{V}^{-1} \text{s}^{-1}$	[28]
μ_h	1.5×10^{-16}	$\text{m}^2 \text{V}^{-1} \text{s}^{-1}$	[28]
Trap density			
N_{0et}	100	Cm^{-3}	[27]
N_{0ht}	10	Cm^{-3}	[27]
Trapping coefficients			
B_e	7×10^{-3}	s^{-1}	[27]
B_h	7×10^{-5}	s^{-1}	[27]
Recombination coefficients			
S_0	4×10^{-3}	$\text{m}^3 \text{C}^{-1} \text{s}^{-1}$	[27]
S_1	4×10^{-3}	$\text{m}^3 \text{C}^{-1} \text{s}^{-1}$	[27]
S_2	4×10^{-3}	$\text{m}^3 \text{C}^{-1} \text{s}^{-1}$	[27]
S_3	0	$\text{m}^3 \text{C}^{-1} \text{s}^{-1}$	[27]

Simulation results

Space charge dynamics and electric field evolution are estimated assuming constant mobility. Figures 1(a) and 1(b) show local electric field distribution and net charge distribution at four different times during the voltage ramp (ramp rate of 300 V/s) for 25 μm thick LDPE sample. The electric field distribution increases with time until the sample reaches the intrinsic breakdown strength of 680 MV/m at 40 s. Figure 1(b) shows that negative charge has developed near the cathode and net positive charge has developed near the anode during the linear voltage rise. The magnitude of both charges increases with time, although the negative charge has much smaller magnitude than that of the positive charge and the negative is distributed over wider thickness region due to the higher electron mobility. The higher electron mobility also results in the arrival of electrons at the anode for times as short as 20 s although holes do not transport to the cathode even as the breakdown event is reached as shown in Figures 1(c) and 1(e). Figure 1(d) and 1(f) show that trapped electrons are present over the entire region except next to the anode and trapped holes are present only near the anode.

Space charge also contributes to the local electric field in addition to the applied electric field as shown in Figure 1(a). At about 10 s, near the initial stage of the voltage rise, the applied field is not high enough to promote charge injection from the electrodes. Therefore, the electric field contribution from space charge is very low and the resulting local electric field is nearly uniform and given by the applied electric field. At 20 s, the charge injection contributes to the formation of space charge within the dielectric and the local electric field is altered. The maximum local electric field occurs near the anode. Higher electric field with increasing time results in more space charge and the position of maximum local electric field moves towards the cathode. This result suggests that the electronic breakdown takes place within the dielectric and not at the interface between dielectrics and electrodes.

Figure 1 Modeling computational outputs from the constant mobility values for holes and electrons for 25 μm LDPE sample under 300 V/s until breakdown.

Breakdown strength of LDPE samples with thickness from 5 μm to 200 μm under 300 V/s voltage ramp was modeled using the constant mobility. For samples with this thickness range the local electric field near the anode reached the intrinsic breakdown strength as described previously and dielectric breakdown takes place. Breakdown strength of LDPE samples decreases as thickness increases and the rate of decreasing in breakdown strength is reduced with the thickness as shown in Figure 2. This trend coincides with the modeling result of LDPE by Chen et al[19]. The difference of about 20 % in the magnitude of the breakdown strength comes from different assumption of hole mobility[19]. Compared to constant hole mobility Chen et al. used field dependent hole mobility which decreases with electric field. Then in their case there is more space charge buildup near anode which decreases breakdown strength.

The thickness dependence of the breakdown strength is often plotted in the logarithmic scale. This plot shows that breakdown strengths in the model for LDPE are weakly thickness dependent. This agrees with Chen's results which also show weakly thickness dependent breakdown strengths (Figure 2). In low-alkali BAS glasses Lee and Murata also showed weakly thickness dependent breakdown strengths[29, 30]. The temperature rise from Joule heating until the breakdown is also tested. For all of the different samples the temperature increased by less than 1 $^{\circ}\text{C}$. Therefore at 25 $^{\circ}\text{C}$ the effect of temperature on the breakdown strength was negligible.

Figure 2 Breakdown strength of LDPE with thickness from 5 μm to 200 μm on the log-log plot compared with Chen et al.'s modeling data[19]

This model has been applied to predict the effect of interfaces on breakdown strength for multilayer polymer structures that act as barriers. It is assumed that a 200 μm LDPE sample consists of 4, 8, 16, and 32 layers respectively. At periodic boundary within the LDPE sample current density for electrons and holes can be described by

$$J_{pb} = f_b J_{w/o pb} \quad (9)$$

where J_{pb} is current density at periodic boundaries, f_b transfer factor at periodic boundaries, $J_{w/o pb}$ current density without periodic boundaries. The transfer factor ranges from 1 to 0.001. These values are consistent with values given by Waser for boundaries in ceramic materials[31]. Figure 3 shows modeling outputs assuming transfer factor of 0.01 for 32 multilayers. At the periodic boundaries, electrons and holes are partially blocked and both electron and hole density increase. The modified charge distribution results in an altered electric field distribution near periodic boundaries.

Figure 4 shows computational breakdown strength with periodic boundary distance for a 200 μm thick LDPE sample. For transfer factor(f_{pb}) of 0.01 breakdown strength increased from 490 MV/m to 540 MV/m as periodic boundary distances decreased from 50 μm to 6.25 μm . The breakdown strength with periodic boundary distance of 6.25 μm was 12 % higher compared to breakdown strength without periodic boundaries. For transfer factor(f_{pb}) of 0.001 breakdown strength increased from 500 MV/m to 640 MV/m as periodic boundary distance decreased from 50 μm to 6.25 μm . Then there was 32 % increase in breakdown strength compared to that without periodic boundaries. The modeling results can explain experimental breakdown strength increase for multilayer polymer structures reported by Mackey et al[20] where the breakdown strength increased from 500 MV/m for the individual polymer phase to 1000 MV/m for nanolaminate structures with additional electronic transport barriers.

Figure 3 Modeling computational outputs from the constant mobility values for holes and electrons for 200 μm LDPE sample under 300 V/s until breakdown assuming transfer factor of 0.01 at boundaries of 32 layers.

Figure 4 Breakdown strength of 200 μm LDPE sample with periodic boundary distance for two different transfer factors (f_b). Dotted line represents breakdown strength without periodic boundary layer.

Conclusions

A model for electrical breakdown under DC conditions has been extended for low-density polyethylene and polymer laminates with electronic transport barriers. The model is based on bipolar charge injection and the space charge formation under higher electric fields which is experimentally verified. From the modeling it is shown that the space charge dynamics lead to a weak thickness dependent breakdown strength at room temperature which is consistent with other researchers. Temperature dependence of electron mobility and hole mobility are other factors for applying this numerical model to temperature dependence of breakdown strength. Furthermore, the model also predicts enhanced breakdown strength as a result of partially blocking interface for multilayer polymer structures.

One goal of this study is to promote further experimental work for breakdown strength in polyethylene with control variables such as thickness, ramp rate. The role of addition transport boundaries through laminate composite synthesis should also be explored.

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