Numerical Simulation on DC Breakdown of Polyimide Based on Charge Transport and Molecular Chain Displacement

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Abstract. A DC breakdown model combining charge transport and molecular chain displacement is utilized to simulate the thickness-dependent DC electrical breakdown of polyimide and reveal the physical mechanism of DC breakdown. The free volume existing in dielectric materials provide electrons with free path to be accelerated and gain energy under the electric field. Molecular chains with occupied deep traps can be displaced by Coulomb force under electric field, furthermore, the displacement will enlarge the local free volume. The energy of electron w is determined by the local electric field F and the length of free volume λ_L, which can be expressed as $w = eF\lambda_L$. When the maximum energy of electrons exceed the deep trap energy level, the local current and temperature will rise in a surge, triggering breakdown eventually. The simulation results reveal the dynamics of space charge and electric field inside polyimide material before the DC electrical breakdown occurs. The breakdown strength F_b of polyimide films obtained from the DC breakdown model decrease with an increase in sample thickness d, which satisfies an inverse power law $F_b = kd^{-n}$ with $n = 0.30$. A strong dependence can be found between breakdown field and sample thickness when the influence from molecular chain displacement on free volume is taken into consideration. The simulation results indicate that the DC electrical breakdown may be the result of the interaction of space charge accumulation effect and molecular chain displacement.

Keywords: DC breakdown model · Polyimide · Molecular chain displacement · Free volume · Thickness

1 Introduction

It is known from the literature review that the electrical breakdown strength F_b of dielectric materials are decreased with an increase in sample thickness d, following the inverse power law $F_b = kd^{-n}$ [1–3]. The relationship above is only an empirical equation

B. Németh (Ed.): ISH 2019, LNEE 598, pp. 108–117, 2020.
https://doi.org/10.1007/978-3-030-31676-1_11

obtained from experimental results without reasonable theories for interpretation. With the fast-growing development of electrical power systems, thinner dielectric films with higher breakdown strength are increasingly required. Therefore, in order to promote the DC breakdown strength of dielectric materials more effectively, it is extremely necessary to understand the physical mechanism of DC electrical breakdown in dielectric materials.

The formation and dynamic of space charges inside dielectric materials under applied high voltage will determine the distribution of internal electric field and have a strong influence on the electrical breakdown strength [4–6]. Matsui et al. have found that the maximum electric field in LDPE before the insulation breakdown are almost the same value of about 520 $V\mu m^{-1}$, which may be an intrinsic breakdown filed existing in the polymeric materials [5]. Chen et al. proposed a model based on the bipolar charge injection model and the formation of charge packet under high electric filed to explain the thickness-dependent DC electrical breakdown in LDPE [7]. The simulation results demonstrated that the electric breakdown was consequent upon the charge dynamics. In addition, free volume exist in polymers, whose scale are very small below the nm level, usually referring to the vacancy without molecular chains or the unoccupied volume at the end of molecular chains [8, 9]. According to the free volume breakdown theory proposed by Artbauer [10], free volume provide electrons with free path to be accelerated and gain energy under the electric field. The molecular chain with occupied deep traps will move a distance driven by the Coulomb force under electric field, which will cause the free volume around enlarged. As a result, the expansion of free volume enable electrons to gain higher energy. When the energy of electrons are high enough to overcome the potential barrier of deep traps, the local current and temperature will rise in a surge, which will trigger breakdown eventually. In the previous work, the relationship between sample thickness and electrical breakdown field was simulated successfully in LDPE based on the effect of enlarged free volume caused by molecular chain displacement [11].

Polyimide (PI) is a macromolecule polymeric material with excellent performances in thermal, electrical, mechanical and radiation resistant properties and is widely used for power equipment insulation [2, 12]. In this paper, a DC electrical breakdown model combining space charge dynamics and molecular chain displacement effect is proposed to simulate the thickness-dependent DC electrical breakdown of polyimide and research the physical mechanism of DC electrical breakdown.

2 DC Breakdown Model

2.1 Charge Transport

Figure 1 shows the schematic diagram of bipolar charge transport model. In bipolar charge transport model [7, 13, 14], electrons and holes are injected into the dielectric material from cathode and anode respectively by Schottky thermionic emission. The contact potential barriers exist between both metal electrodes and dielectric material, expressed as $E_{in(e)}$ and $E_{in(h)}$ in eV. Electrons and holes will migrate toward anode and cathode under electric filed after injected into the dielectric material, which may be captured by the deep traps during migration. The accumulation phenomenon of space

charges will form gradually at the interfaces near both metal electrodes with more and more electrons and holes injected and trapped. The deep traps are only with a single energy level, $E_{T(e)}$ and $E_{T(h)}$ for electrons and holes, respectively. Recombination occur when electrons and holes encounter each other.

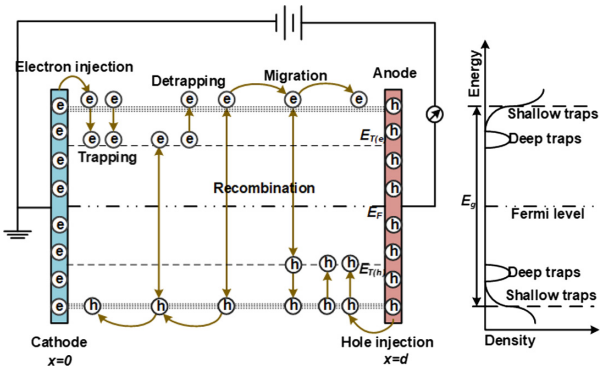

Fig. 1. Schematic diagram of bipolar charge transport model.

The injection currents densities at cathode and anode are determined by the contact potential barriers between the material and its electrodes, electric fields at the interfaces, and temperature [7, 13–16].

$$j_{in(e)}(0,t) = AT^2 \exp\left[-\frac{E_{in(e)}}{k_B T}\right] \exp\left[\frac{\sqrt{eF(0,t)/4\pi\varepsilon_0\varepsilon_r}}{k_B T}\right] \tag{1}$$

$$j_{in(h)}(d,t) = AT^2 \exp\left[-\frac{E_{in(h)}}{k_B T}\right] \exp\left[\frac{\sqrt{eF(d,t)/4\pi\varepsilon_0\varepsilon_r}}{k_B T}\right] \tag{2}$$

Here, $j_{in(e)}$ and $j_{in(h)}$ represent the injection current densities in Am^{-2} for electrons and holes respectively; A is the Richardson constant (=1.20 × $10^6 Am^{-2}K^{-2}$); k_B is the Boltzmann constant; T is the temperature in K; e is the elementary charge; ε_0 is the permittivity of free space; ε_r is the relative permittivity of the insulating material; $F(0, t)$ and $F(d, t)$ are electric fields in $V\mu m^{-1}$ at the interfaces of cathode and anode, respectively.

The space charges injected into the dielectric material will migrate with assistance of shallow traps, and the conduction current densities of electrons and holes can be expressed as:

$$j_{c(e)}(x,t) = q_{free(e)}(x,t)\mu_{0(e)}F(x,t) \tag{3}$$

$$j_{c(h)}(x,t) = q_{free(h)}(x,t)\mu_{0(h)}F(x,t) \tag{4}$$

Here, $j_{c(e)}$ and $j_{c(h)}$ are the conduction current densities for electrons and holes respectively in Am^{-2}; $q_{free(e)}$ and $q_{free(h)}$ are the density of free electrons and holes in Cm^{-3}; $\mu_{0(e)}$ and $\mu_{0(h)}$ are the mobilities for electrons and holes in $m^2V^{-1}s^{-1}$, controlled by shallow traps [7, 13–16].

The probability of charges trapped in deep traps $P_{tr(e,h)}$ is proportional to the carrier mobility and deep trap density $N_{T(e,h)}$ in m^{-3}, but inversely proportional to the dielectric constant [17]. which can be expressed as,

$$P_{tr(e,h)} = eN_{T(e,h)}\mu_0/\varepsilon_0\varepsilon_r \tag{5}$$

The detrapping probability $P_{de(e,h)}$ of trapped charges released from deep traps can be expressed as,

$$P_{de(e,h)} = v_{ATE}\exp(-\frac{E_{T(e,h)}}{k_BT}) \tag{6}$$

Here, $E_{T(e,h)}$ are the deep trap energy level for electrons and holes respectively in eV, and v_{ATE} is the attempt-to-escape frequency in s^{-1}.

The recombination coefficient between free electrons and holes $R_{e\mu,h\mu}$ and trap-assisted recombination coefficient between free electrons and trapped holes $R_{e\mu,ht}$, and that between trapped electrons and free holes $R_{et,h\mu}$ can be described as [18, 19]:

$$R_{e\mu,h\mu} = \left(\mu_{0(e)} + \mu_{0(h)}\right)/\varepsilon_0\varepsilon_r \tag{7}$$

$$R_{e\mu,ht} = \mu_{0(e)}/\varepsilon_0\varepsilon_r \tag{8}$$

$$R_{et,h\mu} = \mu_{0(h)}/\varepsilon_0\varepsilon_r \tag{9}$$

The unit of recombination coefficient is $m^3C^{-1}s^{-1}$.

Charges in dielectric materials satisfy a set of charge continuity equations, which can describe the trapping, detrapping, and recombination processes of electrons and holes [7, 13–16].

$$\frac{\partial q_{free(e)}(x,t)}{\partial t} + \frac{\partial j_{c(e)}(x,t)}{\partial x} = -P_{tr(e)}q_{free(e)}\left(1 - \frac{q_{trap(e)}}{eN_{T(e)}}\right) + P_{de(e)}q_{trap(e)} - R_{e\mu,h\mu}q_{free(h)}q_{free(e)} - R_{e\mu,ht}q_{free(e)}q_{trap(h)} \tag{10}$$

$$\frac{\partial q_{trap(e)}(x,t)}{\partial t} = P_{tr(e)}q_{free(e)}\left(1 - \frac{q_{trap(e)}}{eN_{T(e)}}\right) - P_{de(e)}q_{trap(e)} - R_{et,h\mu}q_{trap(e)}q_{free(h)} \tag{11}$$

$$\frac{\partial q_{free(h)}(x,t)}{\partial t} + \frac{\partial j_{c(h)}(x,t)}{\partial x} = -P_{tr(h)}q_{free(h)}\left(1 - \frac{q_{trap(h)}}{eN_{T(h)}}\right) + P_{de(h)}q_{trap(h)} - R_{e\mu,h\mu}q_{free(e)}q_{free(h)} - R_{et,h\mu}q_{trap(e)}q_{free(h)} \tag{12}$$

$$\frac{\partial q_{trap(h)}(x,t)}{\partial t} = P_{tr(h)} q_{free(h)} \left(1 - \frac{q_{trap(h)}}{eN_{T(h)}}\right) - P_{de(h)} q_{trap(h)} - R_{e\mu,ht} q_{free(e)} q_{trap(h)} \tag{13}$$

The subscripts μ and t represent the mobile and the trapped charges, respectively.

The electric field can be calculated from the electric potential, $F = -\nabla\phi$. The electric potential distribution inside dielectric materials can be obtained by the Poisson equation [7, 13–16].

$$\frac{\partial^2 \phi(x,t)}{\partial^2 x^2} = -\frac{q_{net}(x,t)}{\varepsilon_0 \varepsilon_r} \tag{14}$$

Here, q_{net} is the net charge density inside the dielectric material.

A boundary condition is required in order to solve the Poisson equation. In the following simulation, the applied voltage is set as a positive ramp voltage with a rising rate of 1 kVs^{-1}. The electric potential at anode is equal to the value of applied voltage and the potential at cathode is zero. Thus the boundary conditions can be expressed as $\phi(d, t) = V_{\text{appl(t)}} = k_{\text{ramp}} t_{\text{ramp}}$. Here, V_{appl} is the applied voltage in V, k_{ramp} is the rising rate of the applied voltage in kVs^{-1}, and t_{ramp} is the elapsed time after applying a DC voltage.

2.2 Molecular Chain Displacement

The molecular chain with deep traps occupied by charges will move away from the initial location driven by the Coulomb force under the electric field, as shown in Fig. 2. The velocity equation for the motion can be expressed as: [11, 20]

$$\frac{d\lambda_D}{dt} = \mu_{mol} F - \frac{\lambda_D}{\tau_{mol}} \tag{15}$$

Here, λ_D is the displacement of molecular chains in m; μ_{mol} is the mobility of molecular chains in $\text{m}^2\text{V}^{-1}\text{s}^{-1}$; and τ_{mol} is the relaxation time of molecular chains in s. The mobility of molecular chains μ_{mol} is determined by the carrier mobility controlled by shallow trap and the probability of trapping and detrapping, namely $\mu_{mol} = \mu_0 P_{de}/(P_{tr} + P_{de})$. The relaxation time constant of molecular chains τ_{mol} is equal to the retention time of charges in deep traps, $\tau_{mol} = \tau_0 \exp(E_T/k_B T)$.

The displacement of molecular chains with deep traps occupied by charges under electric field will enlarge the free volume existing in the polymer. Electrons will be accelerated to obtain energy in the free volume under the influence of electric field. The energy w of electrons depends on the local electric field F and the length λ_L of the free volume, defined as $w = \lambda_L eF$. Therefore, the energy w will be increasing with the increase of the length of free volume and the local electric filed. When the maximum energy of electrons $w_{\max} = (\lambda_L eF)_{\max}$ exceed the deep traps energy level E_T, insulation breakdown will be initiated.

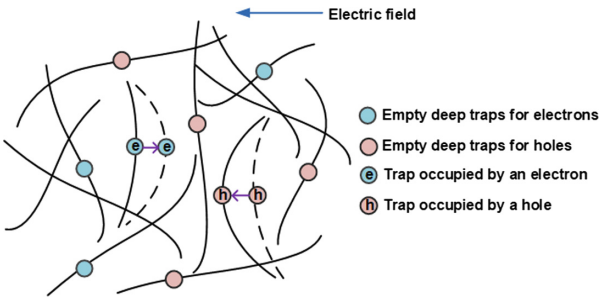

Fig. 2. Schematic diagram of the displacement of molecular chains with trapped charges.

In order to calculate the displacement of molecular chains, the distribution of electric field inside the PI films need to be determined firstly. Then, we can obtain the velocity and displacement of molecular chains with trapped charges by solving Eq. (15). Since the initial length of free volume is negligible, it can be assumed that the length of free volume is approximately equal to the displacement of molecular chains with occupied deep traps. Hence the maximum energy of electrons can be expressed as $w_{\max} = (\lambda_D eF)_{\max}$. When the maximum energy of electrons is higher than the deep trap level, a large number of electrons will jump across the deep trap barriers without hindrance to form a large current, which will eventually lead to the insulation breakdown. Therefore, a DC electrical breakdown model combining the space charge effect and molecular chain displacement is proposed.

2.3 Parameters

In the following simulation, the thickness of PI films ranged from 30 to 230 μm. The temperature was set as 303 K. The energy of deep traps was extracted from the TSDC experimental results. The density and energy of deep traps for electrons and holes were assumed to be the same value, which were 6.25×10^{20} m^{-3} and 0.82 eV. The effective contact barrier between the metal electrode and its dielectric material was 1.0 eV. The carrier mobility of electrons and holes controlled by shallow traps in PI samples were 4.1×10^{-14} $m^2V^{-1}s^{-1}$ and 2.0×10^{-14} $m^2V^{-1}s^{-1}$, respectively, which were obtained from the surface potential decay (SPD) test. The relative permittivity of PI was 3.4, and the applied DC voltage increased at a rate of 1 kVs^{-1}.

3 Simulation Results

Figure 3(a) and (b) indicate the distribution of space charge and internal electric field inside a PI film with a thickness of 150 μm as a function of position at different times calculated from the DC breakdown model. It can be seen from Fig. 3(a) that the charge density due to space charge accumulation is small at the initial time, since the applied voltage is low and the amount of charge injection from metal electrodes into the PI film is little at the beginning. The accumulation of space charges increase gradually with the

increase of applied voltage. Positive and negative charges accumulate near the anode and cathode respectively, and then migrate toward cathode and anode under the electric field. The local electric field is determined by the applied voltage and space charge distribution. As shown in Fig. 3(b), at the beginning, the internal electric field has a uniform distribution with no significant distortion. The space charge density increases with the increasing applied voltage, leading to a more and more serious distortion of the local electric field. For instance, at $x = 15$ μm near the cathode, the space charge density $q = -8.99$ cm^{-3} at $t = 10$ s, $q = -142.89$ cm^{-3} at $t = 30$ s, and $q = -251.78$ cm^{-3} at $t_b = 52.4$ s. If α is defined as the ratio of the local maximum electric field to the applied electric field, we can get $\alpha = 1.06$ at $t = 10$ s, $\alpha = 1.29$ at $t = 30$ s, and $\alpha = 1.33$ at $t_b = 52.4$ s. The maximum value of internal electric field appears at the middle of the PI film, which increases non-linearly with time.

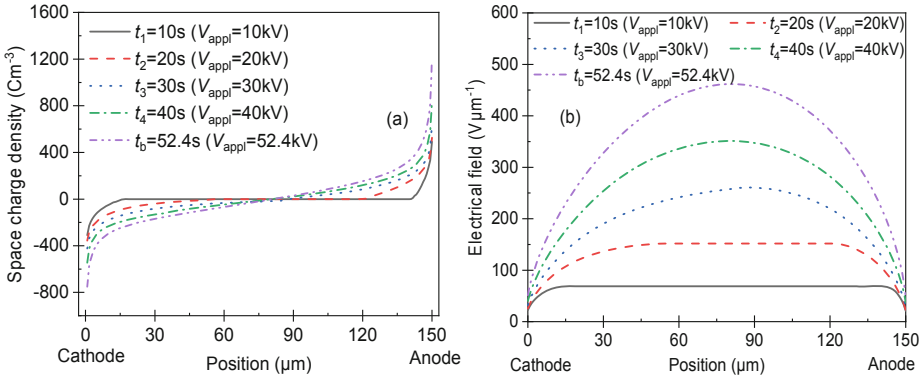

Fig. 3. Numerical results of the space charge density (a) and internal electric field (b) as a function of position at various times. t_b corresponds to the moment of electrical breakdown, and V_{appl} represents the applied voltage at different times.

Figure 4(a) demonstrates the displacement of molecular chains with occupied deep traps inside the PI film with a thickness of 150 μm as a function of position at various times. According to the equation of molecular chain displacement, molecular chains with positive and negative charges will move toward cathode and anode respectively under the internal electric field, and the velocity of molecular chain displacement is proportional to the electrical field. Therefore, the curves of Fig. 3(b) and (a) have the same trend. It can be seen that the maximum value of molecular chain displacement λ_{max} increases as the applied voltage increases. For example, $\lambda_{max} = 0.06$ nm at $t = 15$ s, $\lambda_{max} = 0.53$ nm at $t = 30$ s, and $\lambda_{max} = 1.80$ nm at $t = 52.4$ s. The maximum value of molecular chain displacement also appears at the middle position of the PI film, and the value of the displacement is relatively small near both metal electrodes.

Figure 4(b) shows the maximum energy of electrons obtained from the local electric field. The molecular chain with occupied deep trap will move a distance driven by Coulomb force under the electric field. The displacement of molecular chain could

cause the free volume around enlarged. In the initial state, since the length of free volume inside PI is negligible at room temperature, it can be assumed that the length of free volume λ_L is equal to the value of molecular chain displacement λ_D. Electron can be accelerated to get a certain energy in free volume under electric field. The energy of electron w obtained from electric field in free volume depends on the length of free volume and the local electric field strength, which can be described as $w = \lambda_D Fe$. When the maximum energy of electrons $w_{max} = (\lambda_D Fe)_{max}$ are higher than the deep trap energy level E_T, local current and temperature will rise in a surge, hence triggering electric breakdown eventually. As shown in Fig. 4(b), breakdown occurs when the maximum energy of electrons exceed the deep trap energy. The breakdown time t_b of PI films are increased non-linearly with the increase of films thickness d, so the breakdown field F_b of PI films also increase with the thickness d due to $F_b = k_{ramp} t_b / d$.

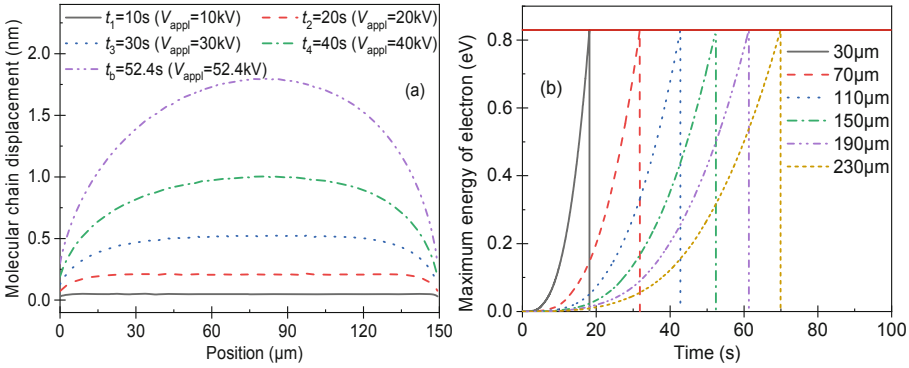

Fig. 4. Numerical results of molecular chain displacement as a function of position at various times (a) and numerical results of the maximum energy of electrons as a function of time of polyimide films with various thickness (b).

Figure 5 shows the DC breakdown strength of PI films as a function of thickness calculated by the DC breakdown model in double logarithmic coordinates. It is obvious that the DC breakdown strength of PI decreases with an increase in films thickness. There is a linear relationship between electric breakdown strength F_b and thickness d in the double logarithmic coordinates, which can be expressed as an inverse power function $F_b = kd^{-n}$ with an inverse power index $n = 0.30$. A strong dependence can be seen between breakdown field and sample thickness when considering the influence from the molecular chain displacement on free volume.

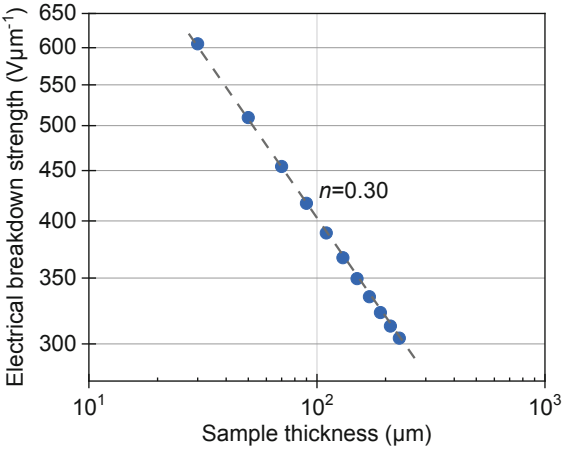

Fig. 5. Numerical results of the DC electrical breakdown field as a function of thickness of polyimide films in the double logarithmic coordinates.

4 Conclusions

A DC electrical breakdown model modulated by charge transport and molecular chain displacement are introduced. The molecular chain with trapped charges can move a distance driven by the Coulomb force under local electric field, which will lead to the free volume enlarged. The length of free volume is approximately equal to the displacement of molecular chain. Electron can obtain a certain amount of energy by being accelerated in free volume under local electric field, and the obtained energy is determined by the length of free volume and the local electric field strength, namely $w = \lambda_L Fe$. When the maximum energy of electrons are large enough to jump across the deep trap barriers, local current and temperature will increase in a surge, eventually triggering electrical breakdown. The dynamics of space charge and the distribution of internal electric field before insulation breakdown are calculated by the charge transport model.

The simulated results of electrical breakdown field F_b of PI films decrease with the increase of sample thickness d, following an inverse power law with power index $n = 0.30$. A strong dependence between electric breakdown field and sample thickness can be seen from the simulation results when considering the influence from molecular chain displacement on the free volume and taking the maximum electron energy exceeding the deep trap barrier as the criteria for the occurrence of insulation breakdown. The thickness-dependent DC electrical breakdown may be the result of the interaction of space charge accumulation effect and molecular chain displacement. Further experiments are needed to verify the simulation results.

Acknowledgments. This work was supported by State Key Laboratory of Advanced Power Transmission Technology (Grant No. GEIRI-SKL-2018-010), the National Basic Research Program of China (grant No. 2015CB251003), and the National Natural Science Foundation of China (grant No. 51507124).

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