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Charging and Discharging Current Characteristics of Polypropylene Film under Varied Electric Fields

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Abstract: Charging and discharging current behavior under high DC electric field in polypropylene (PP) film is closely related to the charge transport and accumulation process, which has an important effect on the electrical insulating properties of PP. In this paper, the dependence of the charging and discharging current of polypropylene films on time and electric field has been comprehensively studied. The results showed that the transient and steady current values of the charging and discharging process increase with the increase of electric field. Dependence of the charging current on the electric field conformed well to the space charge limited current (SCLC) theory with a transition electric field of 270 kV/mm, at which the charge transport changed from ohmic conduction to SCLC conduction. The carrier mobility derived from the discharging current became significantly smaller with increase of the charging electric field. The charge accumulation after discharging was derived from the integration of the difference of the charging and discharging current and it showed an increase with the electric field and increased sharply above a certain threshold electric field (the same as the transition electric field in SCLC theory). It was proved that the conduction current and charge accumulation evolution and dependence on the electric field were mainly determined by the balance between the electrode charge injection process and the bulk conduction process.

Keywords: charge transport; DC breakdown; conduction current; polypropylene film

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1. Introduction

As energy storage elements, direct current (DC) support capacitors are applied to the DC side rectifier bridge arm modules in flexible DC power transmission, which mainly plays the roles of voltage support and harmonic filter [1]. Polypropylene is widely used as a dielectric material in support capacitors, due to its low loss and high breakdown strength [2]. There are many factors that affect the breakdown of metallized polypropylene (PP) capacitor films, including unfavorable working conditions, such as high electric field, current, frequency, temperature and geometry, microstructure morphology, mechanical properties and electrical properties, defects and other material internal factors [3–5]. Space charge accumulation at high electric field and its effect on the distortion of the local electric field is the main reason for the breakdown of metallized capacitor film [6,7].

Charging–discharging current measurement is an effective method for characterizing the process of charge injection, transportation and space charge accumulation in dielectric materials. There is much research on the charging and discharging current of polypropylene film. A. Mittal et al. studied the charging–discharging characteristics of stretched and unstretched polypropylene films at low electric field [8,9]. Liu et al. studied trap characteristics and breakdown characteristics of PP films at different polarization times [10]. Gao et al. studied the effect of charging–discharging cycles on the breakdown characteristics of polypropylene [11]. M. Moudoud et al. experimentally investigated the charging and discharging currents of polypropylene and polystyrene materials under a DC stress of 1000 V. The results showed that the charging and discharging currents were obviously different, and space charges were formed in the samples [12]. However, few researchers have studied the charging–discharging characteristics and charge transport mechanism of polypropylene film for DC supporting capacitors at high DC electric field (≥230 kV/mm). The normal operating electric field of polypropylene (PP) capacitor films is about 230–270 kV/mm, but the highest breakdown electric field is 710 kV/mm at 20 °C. The specific reason why the films operate at such a low electric field is yet to be proven. Examining the accumulation mechanism of space charges under different electric fields is helpful to analyze the causes behind why the films operate as they do. Factors such as the threshold electric field and the amount of charge accumulation have a significant impact on the breakdown of polypropylene films for capacitors. This helps to increase the operating voltage of the capacitor.

In this study, we investigated the charging and discharging current characteristics of polypropylene film at high electric field, exhibiting their dependence on time and electric field. Further, the charge transport mechanism during charging and discharging is elaborated. It provides the basis of space charge generation and accumulation.

2. Materials and Methods

2.1. Materials

Biaxial-orientated polypropylene (BOPP) films with thicknesses of 9 µm and crystallinity of 52% were prepared by Anhui Tongfeng Electronics Co., Ltd. located in Tongling, China. Aluminum electrodes with a diameter of 30 mm were steamed on both sides of the films to make specimens. All the specimens were short-circuited for 24 h at 60 °C in a vacuum oven to remove stray charges before experiments. The experiment was carried out in dried silicone oil.

2.2. Charging and Discharging Current Test

The charging current (I_c)of PP films were measured by applying a DC voltage for 2 h. The discharging current I_d of the PP films were obtained by removing the applied voltage after 2 h. Both are functions of time. The difference between I_c and I_d gave the conductivity contribution [13]. The I_c and I_d of the specimens were measured by increasing voltage step by step until the sample broke down. The principle of voltage rise was the electric field went by 40 kV/mm each time. The initial electrical field was 200 kV/mm. The next electrical field was from 230 kV/mm to the breakdown electric field strength, increasing by 40 kV/mm each time. All measurements were performed at 20 °C. The I_c and I_d testing system is shown in Figure 1.

The charging current was monitored for 2 h after the application of the electric field, after which the applied field was removed and the sample was allowed to relax for 2 h at test temperature. The sample was used for the next higher field until it broke down. To confirm that memory effect did not play a role, fresh samples, subjected to only the preconditioning step were measured once, and the results were compared to those from the sample that had undergone a series of fields at the same temperature. All measurements were conducted in dried Silicone oil.

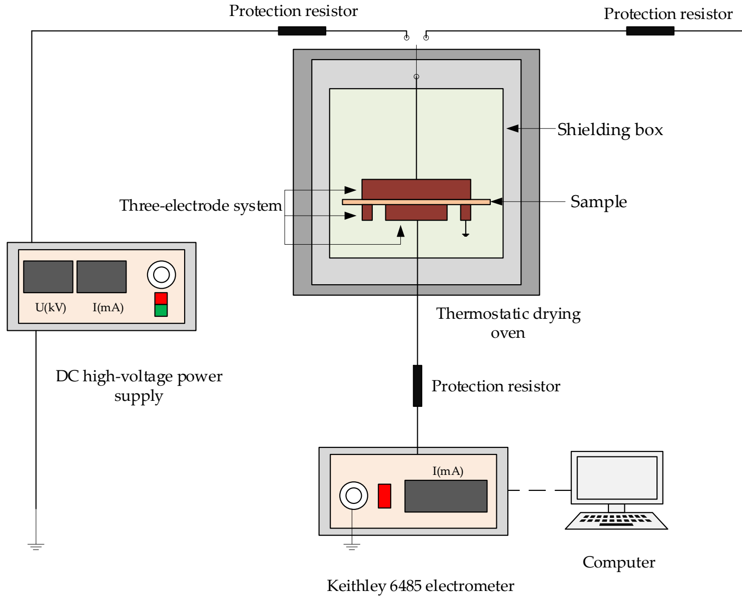


Figure 1. Charging and discharging current testing system.

3. Results and Discussion

3.1. Time and Electric Field Dependence of Charging Current and Discharging Current

Figure 2 shows the charging and discharging currents at different electric fields. As shown, both I_c and I_d decayed with time and I_c nearly reached a steady state at 7200 s.

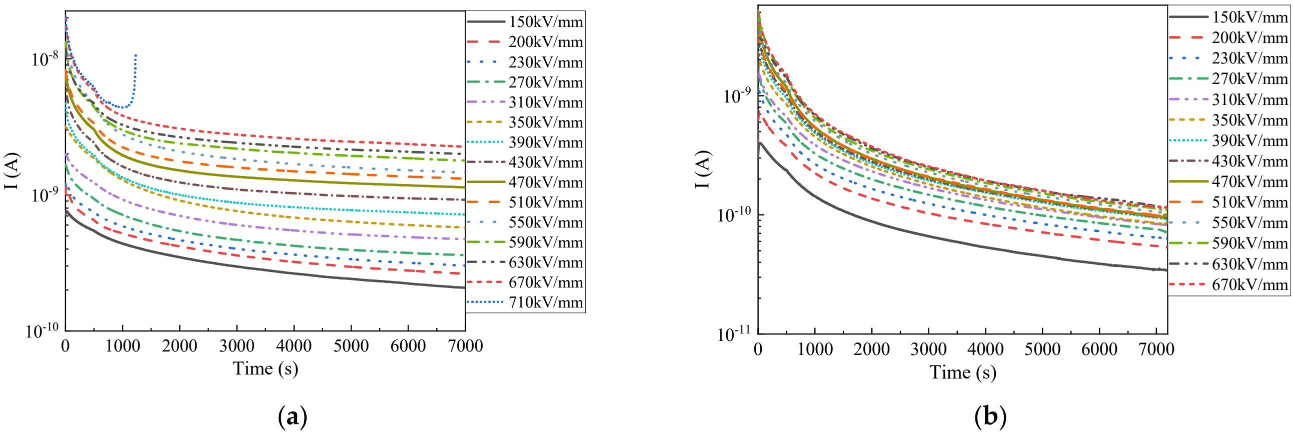


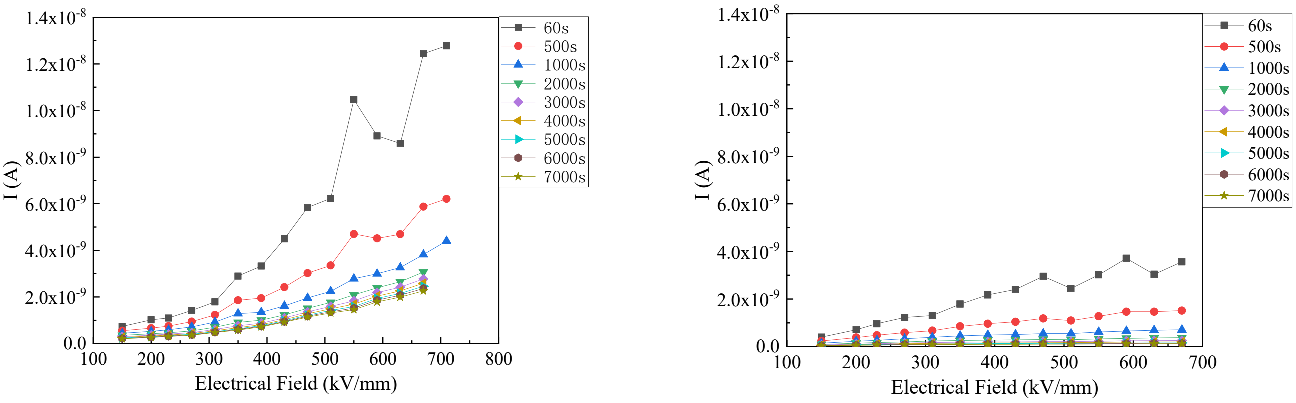
Figure 2. Charging and discharging currents of PP films. ((**a**): The charging current versus time; (**b**): The discharging current versus time).

At 150 kV/mm, the initial I_c was about 7.35×10^{-10} A. Ass time increased, I_c decreased, and the steady-state I_c was about 2.07×10^{-10} A. As the electric field increased, the initial I_c and steady-state I_c increased slightly and the charging current reached a steady state faster. Under 670 kV/mm, the initial I_c and steady-state I_c were 2.01×10^{-8} A and 2.26×10^{-9} A, respectively. When the electric field was 710 kV/mm and applied for 1500 s, the current rose sharply and breakdown occurred. Under 150 kV/mm, the initial value of the discharging current was about 3.95×10^{-10} A, then I_d decreased rapidly, and eventually stabilized at around 3.45×10^{-11} A. Obviously, the initial I_d was less than the I_c, but they were in the same order of magnitude. As time increased, the steady-state I_d was one order of magnitude smaller than the steady-state I_c. This phenomenon continued until the electric field was greater than 510 kV/mm. Under 670 kV/mm, the initial I_d and steady-state I_d, respectively, were to 4.92×10^{-9} A and 1.15×10^{-10} A. The I_d was always one order of magnitude smaller than the I_c. With increase of the electric field, the I_d increased gradually. It was obvious that I_d was much less than I_c.

We found that the dependence of the charging current characteristics and its evolution with time on the electric field could be divided into two stages. In the first stage, the I_c rapidly declined, then declined slowly and gradually reached a quasi-steady state at the second stage (I_c did not reach the quasi-steady state until the electric field was larger than 430 kV/mm). The I_c at 60 s was much larger than that of 500–7000 s. The rapid decrease of the conduction current (Stage 1) was attributed to the ohmic contact formation process between the electrode and PP, which resulted in the fermi level balance and a charge injection barrier around the electrode-PP interface area. The I_c behavior at Stage 2 was ascribed to the charge transport process, which was closely related to the processes of charge injection across the interface barrier and passed through the bulk of PP by means of a hopping mechanism. The process of Stage 1 was much faster than Stage 2 considering the slow charge transport process through the bulk material, leading to the slower decay of the I_c in Stage 2. A higher electric field promoted the charge transport process and shortened the time it took to reach a quasi-steady state in Stage 2.

Under high electric field, due to the steady-state leakage of current and space charge accumulation, the charging current and discharging current had no mirror symmetry. The I_c was much larger than the I_d, which implied remarkable space charge accumulation in the bulk materials [14,15]. Carriers hopping between traps and forming conduction current was the dominant mechanism under high electric field, so the current was trap-controlled. Therefore, it could be assumed that the variation trend and difference of I_c and I_d were related to charge tapping and de-trapping processes. This will be described in more detail in a later section.

Figure 3 shows the variation trend of charging and discharging current versus electric fields at different times. It can be clearly understood from Figure 3 that the I_c increased with the increase of the electric field at any time. However, the I_d only went up with increasing electric field before the 2000 s point and then kept a low value close to zero after 2000 s. The increase of the I_c with increasing electric field was owing to the decreased effective charge injection barrier and the high charge injection rate, along with enhancement of bulk conduction for accelerating the de-trapping of the trap charges [16]. The above two aspects contributed to the current increase by increasing the carrier density and the carrier mobility, respectively. The rapid decay of the I_d originated from the rapid decay of the free charges and the charges in the shallow traps during the short circuit. The current stabilization value tended to zero because the process of the trapped charge de-trapping in deep traps was much more difficult and slower than in shallow traps.



(**a**) (**b**)

Figure 3. The variation trend of charging and discharging current versus electric fields. ((**a**): The charging current versus electric field; (**b**): The discharging current versus electric field).

In order to further explore the dependence characteristics of charging current on the electric field, the quasi-state current value versus electric field was presented by ln*I*-ln*E*, as shown in Figure 4. The curve clearly shows two obvious sections with different slopes. The variation trend of ln*I*-ln*E* was essentially the same between 60 s and 7000 s. The slope of the curve was 0.93 and 2.04 below and above 270 kV/mm, separately. This behavior coincided well with the space charge limited current (SCLC) theory. According to SCLC theory, the charge injection and bulk conduction process reach a balance at a low level with less space charge accumulation and the current is dominated by ohmic conduction, due to the thermally-activated intrinsic charge at low electric field, corresponding to Section 1 in the curve with the slope close to 1. Charge injection was extremely enhanced with further increase in the electric field and a large number of injected charges were trapped to form space charges. Thus, the conduction current was dominated by space charges with a slope close to 2 in the ln*I*-ln*E* curve [13,17]. The charge injection was enhanced at high electric field because both the height and width of the potential barrier at the electrode-dielectrics interface were reduced.

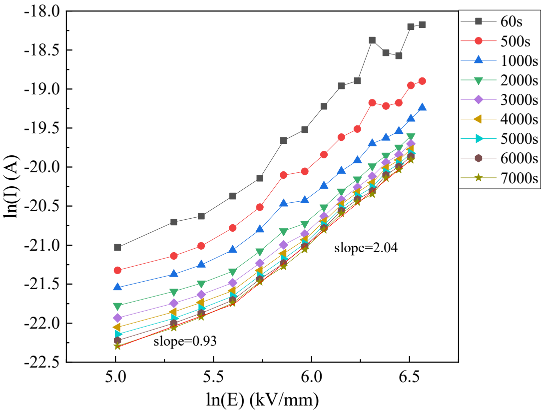


Figure 4. Electric field dependence of Ic in PP films.

3.2. Time and Electric Field Dependence of Carrier Mobility

Carrier mobility can be calculated by the isothermal short-circuit discharge current [18]. During the isothermal discharge current test, the injected charge returns to the injected electrode under the application of the internal electric field and generates the induced current in the external circuit. The correlation between carrier mobility and time can be obtained from the changes of external circuit current and injected charge. In the short-circuit process, the relationship formula between the residual injected charge and time can be expressed as

$$q(t) = \int_0^t I(t)dt \tag{1}$$

where *I(t)* refers to the short-circuit I_d, which can be expressed as

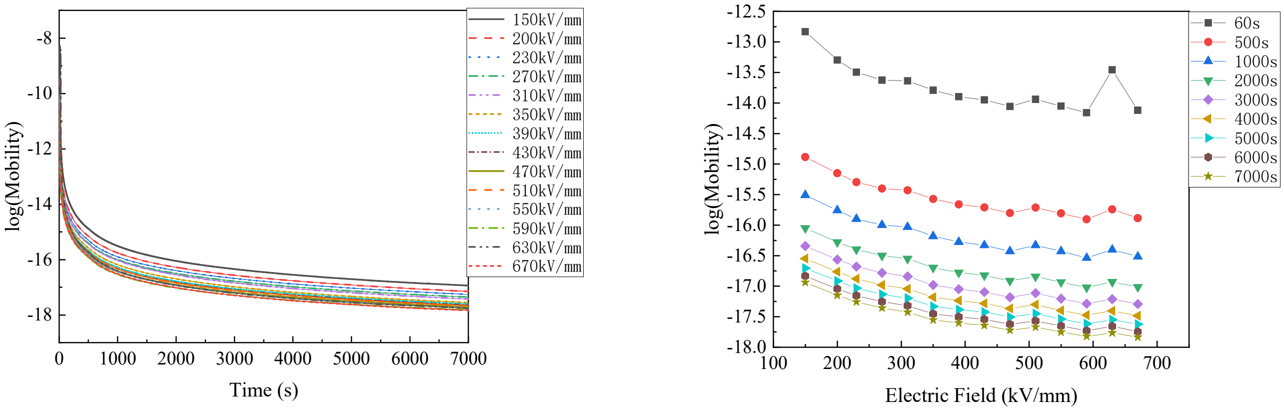
$$I(t) = \mu \rho E S = \mu E l q(t) \tag{2}$$

where, μ means carrier mobility, ρ means injected charge density, E means the electric field, and l means charge injection depth. The electric field can be expressed by the Poisson equation as $E = \rho l / \varepsilon$. It can be obtained by substituting the expressions of electric field and injected charge into Equation (2)

$$\mu(t) = \frac{I(t)\varepsilon l S}{q^2(t)} \tag{3}$$

The change of carrier mobility versus time can be obtained based on Equation (3) and the isothermal discharge current (IDC).

Figure 5 shows the carrier mobility of biaxial polypropylene films under different electric fields. Mobility decreased with time and electric field. It is clear from Figure 5a that the various carrier mobilities of polypropylene films can be generally divided into two stages. In stage 1, the initial mobility is high and sharply decreases over time. In stage 2, the mobility decreases slowly over time, then it tends to flatten out. The two stages correspond to the fast decay of free charge and de-trapping of space charge in shallow traps in short circuits and the de-trapping of space charge in deep traps, respectively. In stage 2 of a short circuit the free charge and the space charge in shallow traps basically become de-trapped, and only the space charge in the deep traps remain de-trapped. From Figure 5b it can be observed that the mobility decreased slightly with increase of the electric field. The mobility was very dependent on the electric field and all the mobility varied from $10^{-12.5}$ to 10^{-18} $m^2/V{\cdot}s$. Space charges were mostly trapped in deep traps under the high electric field because the space charges easily de-trapped in shallow traps. The higher the electric field, the greater the percentage of deep traps, and the slower the de-trap of space charge, which had less and less mobility. The above-mentioned trend and mechanism of carrier mobility are consistent to that of discharging current.

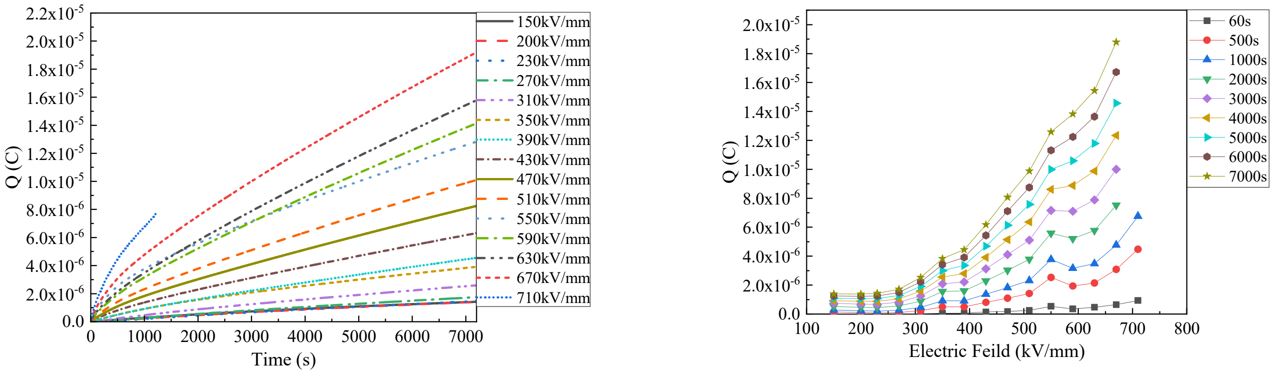


(**a**) (**b**)

Figure 5. The carrier mobility of biaxial polypropylene films at 20 °C. ((**a**): The carrier mobility versus time; (**b**): The carrier mobility versus electric field).

3.3. Time and Electric Field Dependence of Charge Accumulation

Figure 6 shows space charge accumulation in the sample during continuous pressurized charging and discharging at 20 °C. Under each electric field, it is obvious that the space charge accumulation (Q) increased with time. When the electric field was less than 270 kV/mm, Q accumulated slowly with time. When the electric field was greater than 270 kV/mm, the accumulation rate increased with time, that is, the larger the electric field, the faster the charge accumulation. At 150 kV/mm, the total space charge accumulation (Q_t) was about 1.40 × 10^6 C, which was similar to the Q_t at 200 kV/mm and 230 kV/mm. After that, the Q_t increased slightly with the increase of electric field, but there was no significant change, and at 270 kV/mm, the Q_t was about 1.42 × 10^6 C. When the applied electric field exceeded 270 kV/mm, the average charge density increased significantly with the increase of electric field strength. At 310 kV/mm, Q_t was 2.57 × 10^6 C, much larger that at 270 kV/mm. At 670 kV/mm, Q_t was 1.91 × 10^5 C. At 710 kV/mm, the electric field before breakdown, Q was also increasing. In general, with the increase of electric field, the charge accumulation started slowly, and when a threshold electric field was reached, the charge accumulation increased significantly [17,19]. The electric field threshold for rapid increase of charge accumulation was about 270 kV/mm at 20 °C, which is the lowest field in which the sign of aging occurs. The threshold is related to the lower limit value, at which space charges are steadily trapped in PPs, corresponding to the transition of conduction current from ohmic conduction to SCLC conduction [20]. As the conduction current density began to increase beyond this electric field threshold, the space charge accumulation in PPs also increased, as shown in Figure 6, since it is usually proportional to the current density [21]. This indicated that there was a lot of space charge available, and enough space in the films to store space charge. Failure of PP electrical properties is accelerated due to the space charge accumulation at high DC electric fields at 20 °C.



(**a**) (**b**)

Figure 6. The space charge accumulation of PP films. ((**a**): Q versus time; (**b**): Q versus electric field).

The relationship between the space charge accumulation and transport can be described as follows. Below the threshold electric field of charge accumulation, the injected charge is all extracted from the other electrode. Almost no charge is present in the polypropylene film. De-trapping of carriers in deeper traps results in reduced mobility. At electric fields above the threshold, the space charge accumulation is significant, due to the carriers being trapped. The electric field becomes higher, and the space charges accumulate in the deeper traps. The space charges that accumulate under electrical field are far greater than the space charges that migrate away during short-circuit measurements. A large amount of space charge exists inside the sample at the end of short-circuit measurement. In conclusion, due to the limitation of the traps, the carriers migrate freely in the beginning, then hop between the shallow traps, and are finally restricted in the deep traps.

4. Conclusions

We investigated the charge transport and space charge accumulation behavior in PP film by comprehensively studying the dependence of charging and discharging current on time and electric field. The following conclusions can be drawn from the experimental results and theoretical analysis.

(1) The transient and steady current values of the charging and discharging process increase with increase of electric field.
(2) Dependence of the charging current on the electric field conforms well to the space charge limited current (SCLC) theory with a transition electric field of 270 kV/mm, at which the charge transport changes from ohmic conduction to SCLC conduction.
(3) The carrier mobility becomes significantly smaller with increase of the charging electric field, due to the fact that many more charges are restricted in the deep traps.
(4) The charge accumulation increases with the electric field and increases sharply above the transition electric field, which is in good agreement with SCLC theory.
(5) The conduction current and charge accumulation evolution and dependence on the electric field are mainly determined by the balance between the electrode charge injection process and the bulk conduction process.

The charge accumulation of polypropylene films was calculated based on charging and discharging currents for 2 h. However, during the operation of the capacitors, the polypropylene films need to be in the electric field for a long time, and the short-time charging and discharging behaviors provide some insights into the study of charge accumulation during aging. However, there are more factors affecting charge accumulation during aging (e.g., edge discharging), so the failure mechanism of polypropylene films during aging needs to be further investigated.

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