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Influence of Polarization and Non-Uniform Electric Field on Failure Characteristics of Polypropylene Film

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ABSTRACT Polypropylene (PP) film is widely used in the high energy density capacitors due to its excellent electrical properties. However, polarization and non-uniform electric field have great impact on the dielectric reliability of PP films, which were investigated in this paper focusing on the failure characteristics. By using the polarization and depolarization current method, the polarization tests of PP films with different thickness were conducted to reveal the variation of the capacitance, dielectric loss and volume resistivity. The accumulation of space charge was measured based on PEA method, and then the trap density and trap depth of PP films at different polarization levels were calculated. Finally, the DC breakdown properties at non-uniform electric field were analyzed based on the breakdown voltage and the quantitative indication of breakdown craters. Obtained results reveal that the increasing polarization degree causes the decrease in the volume resistivity, the increase in the polarization current, dielectric constant and dielectric loss. In addition, the trap density and trap depth both increase that promote the accumulation of space charge. The breakdown voltage, failure area and angular second moment of breakdown craters show the increasing tendency with the increase of the electric-field uniformity, but the decreasing tendency with increasing polarization degree.

INDEX TERMS Polypropylene film, polarization degree, non-uniform electric field, failure characteristics, quantitative indication of breakdown craters.

I. INTRODUCTION

With the rapid development of smart grid and micro grid in China, high energy density capacitors (HEDC) are widely applied in power system as one of the significant storage components. In HEDC, polypropylene (PP) film is the main insulation due to its excellent dielectric properties [1], [2]. However, PP film is usually subjected to the electrical problems due to effects of physical installation, high ambient temperature and electric field during the actual operation, which can accelerate the dielectric degradation, even the insulation breakdown, to hazard the operating stability and reliability of HDEC [3], [4].

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Multiple factors that can result in the insulation degradation of PP films have been studied. Considering the relation between the electrical-thermal aging and the physicochemical properties of PP film, a wide variety of aging properties, from the electrical, thermal and chemical aging to the physical aging have been identified [5]. Ritamäki and Kurimský have analyzed the dielectric properties of bi-axially oriented polypropylene (BOPP) film under accelerated electrical-thermal aging [6], [7]. Rytöluoto analyzed the insulation properties of PP films under multi-electrical stresses [8]. Wu et al have studied that partial discharge, space charge, and thermal factors can be failure factors of composite insulation (polypropylene film and paper impregnated with oil) capacitors [9]. Moreover, dielectric properties of PP films such as dielectric response, breakdown phenomenon et have been widely investigated under the factors mentioned above.

Li et al have analyzed the polarization characteristics of metallized PP film capacitors under different temperatures and the insulation resistance under different electric fields [10], [11]. Laihonen and Fujii have investigated the DC and AC breakdown strength properties of PP films while exposing to electrical stresses, respectively [12], [13]. And the breakdown characteristics of PP films under nanosecond voltage pulses have been obtained through experimental study by Chen et al [14]. Ho et al have studied the effect of UV treatment on the dielectric strength of BOPP capacitor films [15].

Furthermore, other properties of PP films have also been considered. The negative corona charging and surface potential decay (SPD) characteristics of nonwoven PP fabrics have been investigated to analyze dielectric properties of the film capacitors [16]. Du et al have studied the effects of direct fluorination on surface charge accumulation and decay of PP film [17]. The effects of different temperature, voltage waveforms and nanoparticles on surface charge and trap characteristics of PP film have been researched by other scholars respectively [18]–[20].

In this paper, effects of polarization and non-uniform electric field on failure characteristics of PP film were investigated. The variation of polarization degree, film thickness, and electric field with different distance and uniformity was considered to measure the capacitance, dielectric loss, volume resistivity, accumulation of space charge, trap density, trap depth, breakdown voltage and quantitative indication of breakdown craters.

II. EXPERIMENTAL SETUP

PP films with different thicknesses (20, 60 and 100 μm) were prepared by controlling the tableting machine [6], [17]. All the specimens were wiped by using the pure ethanol and kept in the dust free environment at room temperature for more than 24 h before the experiments.

A. POLARIZATION TEST

Based on the polarization and depolarization current method [21], the specimens were charged for different polarization time of 0, 2, 6, 10 h, which could simulate different operating time of the film capacitors. Figure 1 shows that the stable power source could supply positive direct-current (DC) voltage, which could make the specimens conduct polarization tests under electric field of 20 kV/mm. When the switch was placed at K_1 , the sample was charged by the power source, and the current flowing through the sample was called the polarization current. The time relay was used to control polarization time. After completing the charging, the switch was placed at K_2 and the sample was directly short-circuited to the ground. At this time, the current flowing through the sample was called the depolarization current. The polarization and depolarization currents could be measured by the ammeter (KEYTHLEY6485) for 1000s test time. The capacitance and dielectric loss could be tested by LCR meter (TH2826A) and volume resistance could be tested by the digital high resistance meter (PC68).

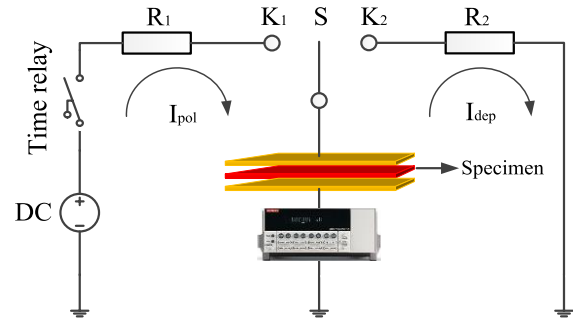


FIGURE 1. Polarization and depolarization setup.

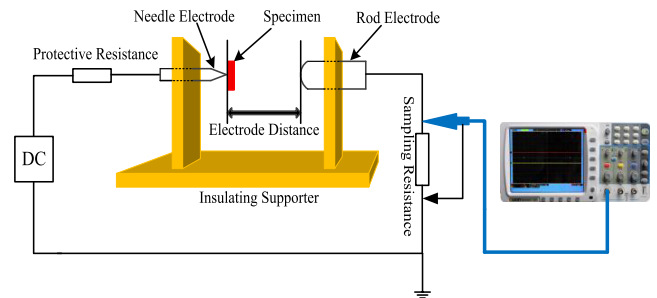


FIGURE 2. Breakdown experimental setup.

B. BREAKDOWN EXPERIMENT

The breakdown experiments were constructed with different electric-field structures and electrode spaces at room temperature in accordance with IEC 60243-2:2013. The two electrode structures were the needle-rod electrode and needle-needle electrode, and the electrode spaces were 2, 4, 6 mm respectively. The breakdown experimental setup is shown in Figure 2, where the curvature radius of needle electrode was 0.5 mm and the diameter of rod electrode was 10 mm. The specimen was firstly cut into the dimension of 20 mm \times 20 mm and then set at the high voltage electrode (needle electrode). The high voltage DC transformer was controlled to boost at the rate of 500 V/s until the breakdown occurred through the specimen and PP films of each thickness were conducted 20 breakdown experiments. The breakdown voltage could be recorded and the waveform of breakdown current could be obtained by the sampling resistance (1.2 Ω) and oscilloscope (EDS102C). The images of breakdown craters were captured through applying a monocular-video-zoom microscope and breakdown craters was statistically analyzed by the image processing technology.

C. PEA TEST

The accumulation of space charge in polypropylene under different electric fields was measured by space charge measuring device based on the pulsed electro-acoustic method [21]. The polarized electric field intensity was +45 kV/mm and the thickness of the sample was 300 μm . The schematic diagram of the measuring device is shown in Fig. 3. The temperature of the experiment was 25°C. Before the beginning of the experiment, the thickness of the

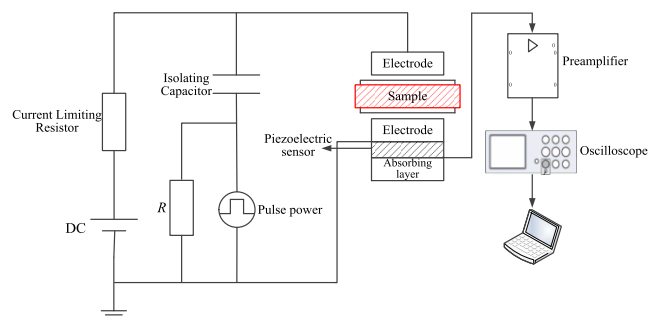


FIGURE 3. Pulsed electro-acoustic method.

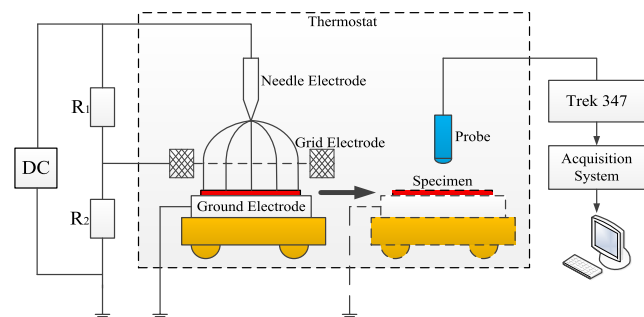
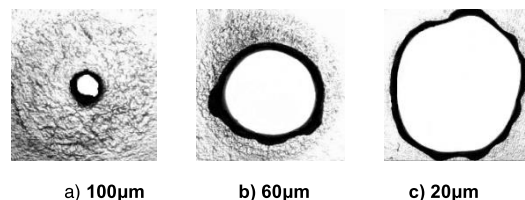


FIGURE 4. SPD experimental setup.

sample was measured and recorded by the thickness gauge. A little silicone oil was dripped into the center of the surface of the aluminum electrode, and the sample was placed in close contact with the electrode. Then semi-conductive electrode and high voltage electrode were placed on the sample and pressed equally. During the experiment, oscilloscope, data acquisition system, space charge measurement platform, pulse power supply and high voltage DC power supply were turned on in turn. The parameters of data acquisition interface were set to measure and record 30-minute voltage waveform. After the experiment, the space charge signal was recovered and processed by signal processing software. The space charge density of polypropylene film at different voltage time was obtained by frequency domain deconvolution algorithm.

D. SPD TEST

Figure 4 shows the schematic diagram of the isothermal surface potential decay experimental device, which could be divided into the charging process and the test process [22]. During the charging process, the specimen was corona charged by the needle-grid-plane electrode for 5 min. The charging voltage was 5 kV and the grid voltage was 2 kV. After charging, the sample was quickly moved to the bottom of the surface potential measuring probe, and the distance between them was 3 mm. Surface potential of the specimen was measured for 60 min by the kelvin type probe (Trek347) connected with the electrostatic voltmeter. The SPD experimental setup was set in the thermostat of 25°C and 21% RH.



a) 100µm b) 60µm c) 20µm

FIGURE 5. Breakdown morphology of PP films.

III. DATA PROCESSING

A. PROCESSING OF BREAKDOWN CRATERS

For convenient analysis, the breakdown craters are defined as three parts from the inside to the outside, which are breakdown area, carbonized area and outer area respectively. This morphology is formed due to the heating effect caused by the large jump current while breaking down. In Figure 5, breakdown area is in the middle of the breakdown craters, and its shape is round or oval. Carbonized area is close to the breakdown area, where deposition of large amount of carbide causes significant changes in morphology. Outer area is the outermost layer of the breakdown craters, where very small change happens in morphology.

The breakdown area is analyzed by the image processing method with the help of MATLAB software. Gray level co-occurrence matrix is used to analyze the breakdown area and carbonized area of the PP films after breaking down. As shown in Figure 5, with the increase of the PP film thickness, the percentage of breakdown area and carbonized area tends to be equal in the image, which leads to greater uniformity of gray distribution and smaller Angular second moment(ASM). And ASM approaching higher value indicates the proportion of breakdown area is much larger than that of carbonized area. As a result, it can establish the relationship between pattern discrimination and breakdown characteristics.

IV. ANALYSIS AND DISCUSSION

A. DIELECTRIC PROPERTIES

As shown in Figure 6(a), as the polarization time increases, volume resistivity of PP film shows the decreasing trend. And the rate of declination becomes slower with the increase of thickness. The non-crystalline region of PP films is more likely to be influenced by polarization than the crystal region, which makes the free volume smaller, leading to the decrease of volume resistivity and the increase of conductivity. Therefore, in Figure 6(b), polarization current increases and the current curve tends to move up with the increase of polarization time. In addition, as shown in Figure 6(c), dielectric constant and dielectric loss both increase with the increase of polarization time for 60 and 100µm PP films. Polypropylene material undergoes degradation and break of chemical bonds on the effect of high-energy electric field, which makes some large molecular chains destroy and produces a large number of small molecules. They can be rapidly turning-direction polarized under the applied electric field, causing the dielectric

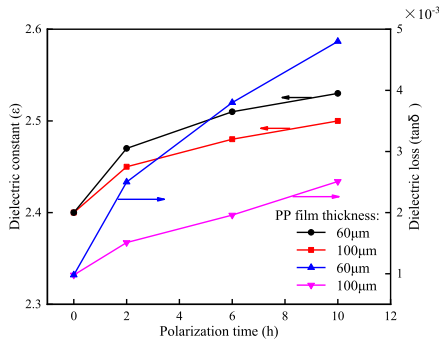
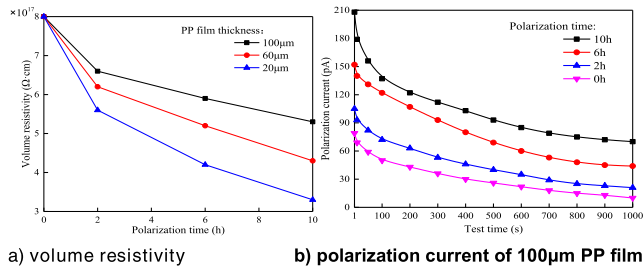


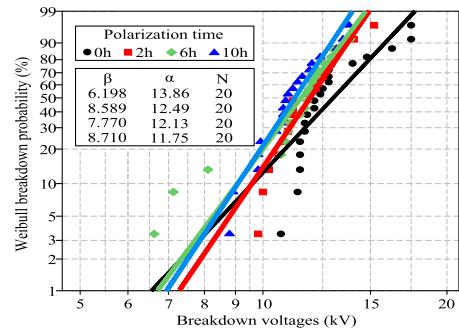
FIGURE 6. Dielectric properties of PP films at different polarization time.

constant increasing. At the same time, the turning-direction polarization is greatly hindered by friction, resulting in larger dielectric loss [23].

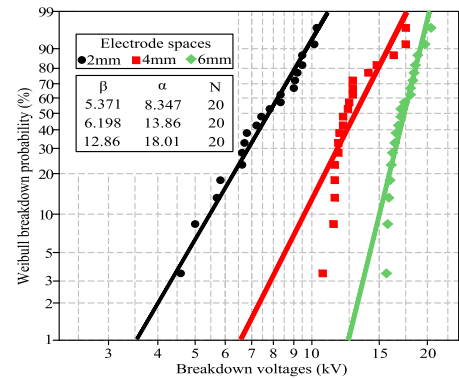
B. BREAKDOWN VOLTAGES

For analyzing the influence of polarization on breakdown characteristics of PP films, specimens are carried out breakdown tests under the needle-rod electrode at different polarization time, the distance of which is 4 mm. As shown in Figure 7(a) and Figure 8(a), the breakdown voltages decrease with the increase of the polarized duration for PP films of different thicknesses. Due to the charge injection during the polarization process, the crystal structure of PP films is destroyed and reduced, and the effect of polarization on the non-crystalline region is more significant, which reduces the electric field strength. Besides, the charges produced on the polymer surface improve the electric field at electrode junctions, as well as resulting in breakdown voltage declining.

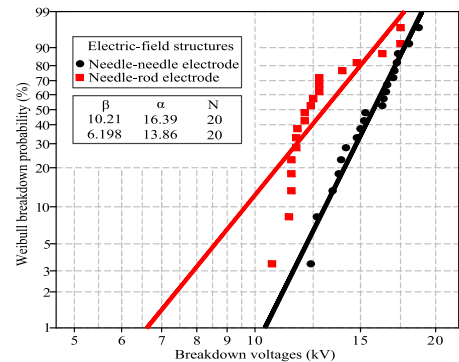
For analyzing the influence of the non-uniform electric field on breakdown characteristics of PP films, unpolarized samples are carried out breakdown tests under the needle-rod electrode at different electrode spaces [7]. As shown in Figure 7(b), Figure 8(b), the breakdown voltages increase considerably with increasing the electrode spaces. The electric field uneven coefficient becomes larger as the electrode space increases. More energy is needed to make the film breakdown. Moreover, unpolarized samples are also carried out breakdown tests at different electrode structures, the distance of which is set to 4 mm. In Figure 7(c) and Figure 8(c), the breakdown voltage under the needle-needle electrode is higher than that under the needle-rod electrode. The main



a) Weibull distribution of breakdown voltages at different polarization time



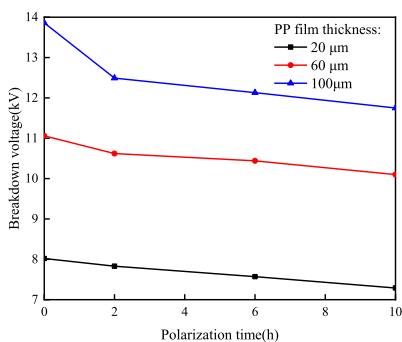
b) Weibull distribution of breakdown voltages at different electrode spaces



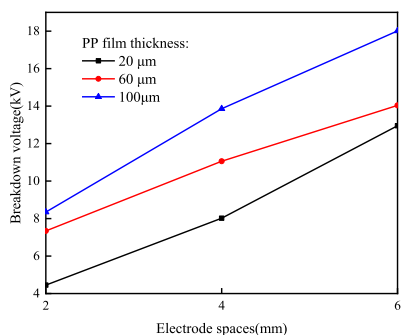
c) Weibull distribution of breakdown voltages at different electric-field structures

FIGURE 7. Weibull distribution of breakdown voltages of 100µm PP film.

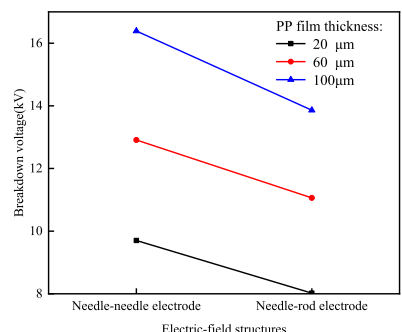
reason is that the electric field distribution under the needle-needle electrode is symmetrical, which results in the electric field non-uniformity less than that under the needle-rod electrode [24]. Similar conclusions can be verified for 20 and 60µm PP films. It can be found that the non-uniform electric field has a greater impact on breakdown characteristics than polarization. Furthermore, with increasing the thickness, the breakdown voltages also increase. Due to the volume effect, the electric field stress at the contact point decreases with the increase of the barrier thickness. It means that the higher applied voltage is needed to increase the ionization process for the thicker solid barrier, for the purpose of raising the electric field stress at the contact point to reach the breakdown condition.



a) Breakdown voltages at different polarization time



b) Breakdown voltages at different electrode spaces

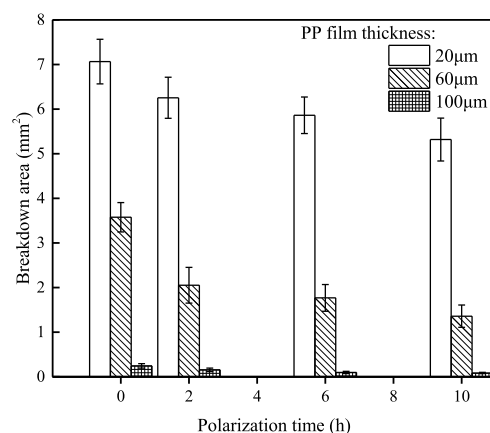


c) Breakdown voltages at different electric-field structures

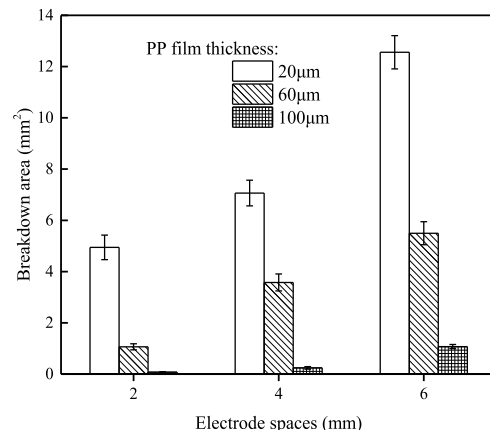
FIGURE 8. Breakdown voltages of 20,60,100 μm PP films.

C. BREAKDOWN CRATERS

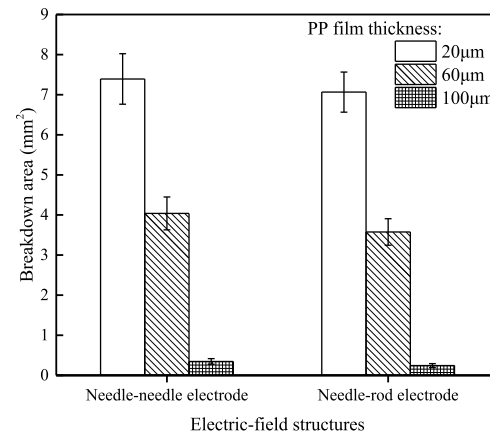
In Figure 9(a), with the polarization time increases, breakdown area shows the obvious decreasing trend for the PP films of different thicknesses. As shown in Figure 10(a), the parameter of ASM also decreases, which falls faster for the thinner film. The PP films are damaged by the polarization causing chemical reactions and structural changes, which can form low density region and small molecules. Therefore, the conductive electrons and ions are obviously increased in conductive energy levels of PP film. Under the influence of DC electric field, the conductive electrons can obtain enough energy to damage the crystal structure, leading to the easy occurrence of breakdown. Because the dielectric breakdown and dielectric properties are based on the film structure, with increasing the polarized duration, the broken degree of crystal structure is enhanced and the needed energy of breakdown is reduced, resulting in the decrease of breakdown area and ASM.



a) breakdown area at different polarization time



b) breakdown area at different electrode spaces



c) breakdown area at different electric-field structures

FIGURE 9. Breakdown area of 20, 60 and 100 μm PP films.

In Figure 9(b) and Figure 10(b), with increasing the electrode space, the breakdown area and ASM both increase. The electrode spaces have significant effect on the breakdown characteristics of PP films, especially for the thinner films. The breakdown voltage and breakdown current increase rapidly, which can generate more energy to destroy the film and form a larger breakdown crater. Moreover, as shown in Figure 9(c), the breakdown area under the needle-needle electrode is larger than that under needle-rod electrode.

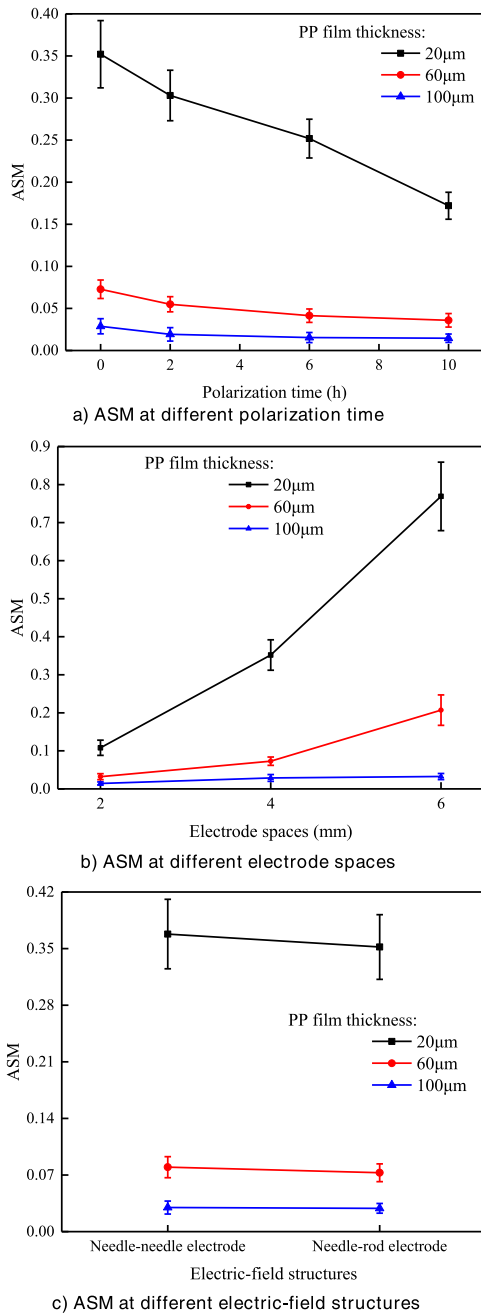


FIGURE 10. ASM of breakdown area and carbonized area of 20, 60 and 100 μm PP films.

Due to a slight increase of the breakdown voltage and breakdown current, there is more serious damage happening on the film, which is reflected in the increase of ASM, as shown in Figure 10(c). It also can be found that breakdown area and ASM decrease with increasing the thickness. Although more energy can be produced by the thicker film owing to the larger breakdown voltage and breakdown current, the larger volume can prevent heat from transferring and protect the film structure. As each breakdown image shows the unique ASM, the feature for discriminating and quantifying the patterns is consistent with the breakdown characteristics. As a result, the

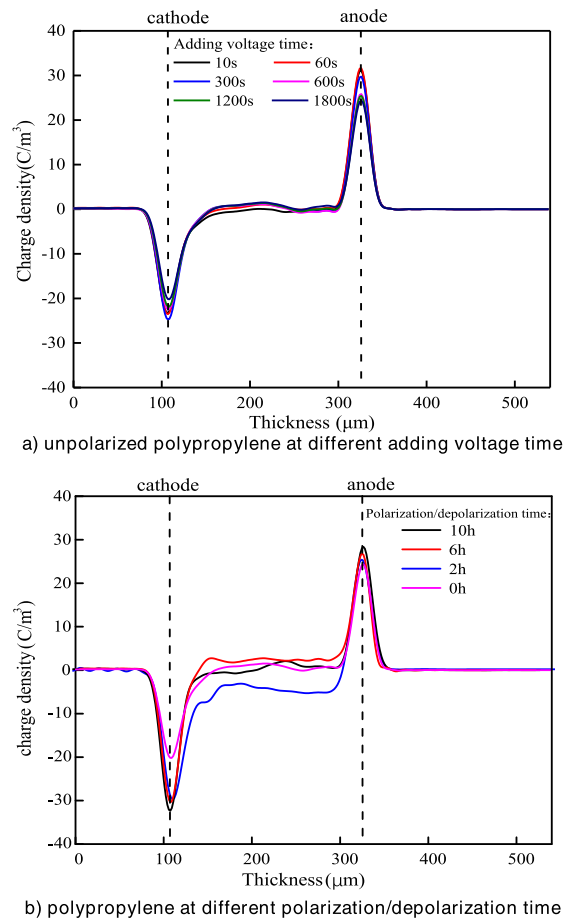


FIGURE 11. Space charge accumulation waveform of polypropylene.

gray level co-occurrence matrix can be used to quantify the breakdown phenomena of PP films.

D. SPACE CHARGE ACCUMULATION

The space charge accumulation waveform of polypropylene under different polarization/depolarization time can be obtained by deconvolution algorithm with signal processing software. Figure 11(a) shows that under the +45 kV/mm DC electric field, there is obvious homopolar charge injection near the cathode in unpolarized polypropylene. The charge injection amount and injection depth also increase with the increase of adding voltage time. From Figure 11(b), it can be concluded that the charge density of pure polypropylene increases with the increase of polarization/depolarization time after 30-minute adding voltage time, and the charge accumulation becomes more serious. The further increase of homopolar charge injection near the cathode will have a negative impact on the insulation performance and long-term operation performance of the material.

E. TRAP DISTRIBUTION

From Figure 12(a), the typical surface potential decay behavior of 100 μm pure polypropylene film at different

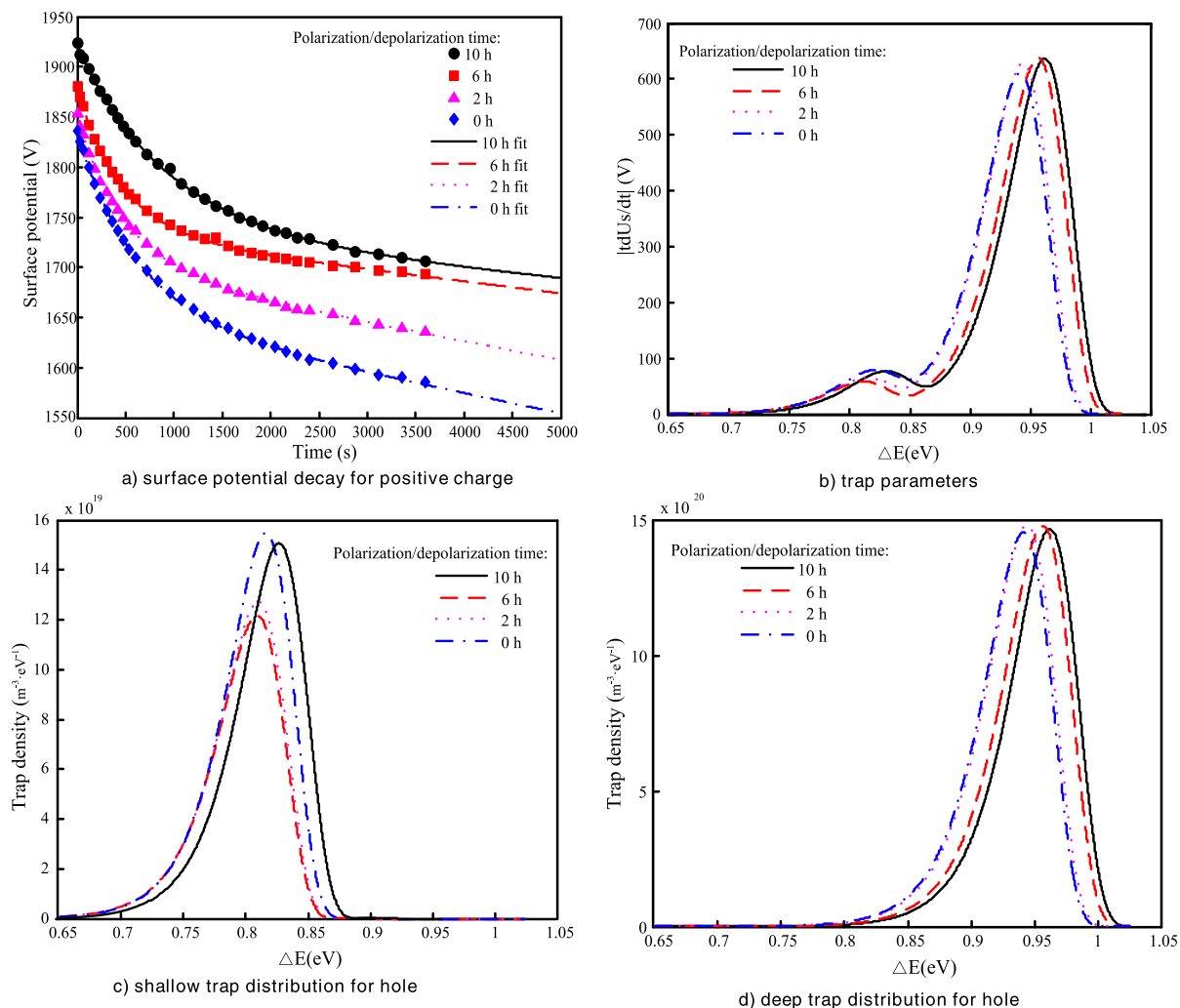


FIGURE 12. Trap distribution of 100um PP film at different polarization time.

polarization/depolarization time (0 h, 2 h, 6 h and 10 h) is measured under positive polarization voltage (under electric field of 20 kV/mm). With the polarization time increases, the initial surface potential of the sample becomes higher and the rate of surface potential decay becomes slower. As shown in Figure 12(b), the trap energy density is characterized by $|tdUs/dt|$ which is proportional to the trap energy density and it can be seen that the polypropylene film has shallow trap and deep trap energy centers, in which the hole trap energy level is from 0.7 eV to 1.02 eV. In Figure 12(c), the shallow trap density and trap depth both show a trend of decreasing first and then increasing. And under the influence of charging time and polarization time, the variation of density and depth of shallow traps results in the fluctuation of energy range 0.8-0.9 eV. However, in Figure 12(d), the deep trap distribution curve appears to move right with increasing the polarization time. The deep trap density shows a small increasing trend, while the deep trap depth shows the increasing tendency obviously. It includes that the trap density and depth are larger than those of the unpolarized one and the trap depth ranges

from 0.9384 eV to 0.9586 eV, which is the domain of deep traps. The molecular chain of PP film is destroyed by the continuous DC electric field. The free radical is formed and the chain reaction of the free radical is triggered, which can generate large number of small molecules and form more local states, resulting in the increase of the deep trap density and energy level. The process of charge detrapping becomes more difficult and the accumulation of charge in the polarization process leads to obvious electric field distortion, which causes low voltage and easily leads to breakdown [25].

The surface potential attenuation of 100 μm pure polypropylene film at different polarization/depolarization time (0 h, 2 h, 6 h and 10 h) is measured under negative polarization voltage. From Figure 13(a), the experimental results show that during corona charging, charged particles migrate to the surface of polypropylene material continuously and are trapped. Then charge transfer occurs between charged particles and samples, which makes charged particles change from ionic state to electronic state. Because the negative corona is more likely to occur than the positive corona under

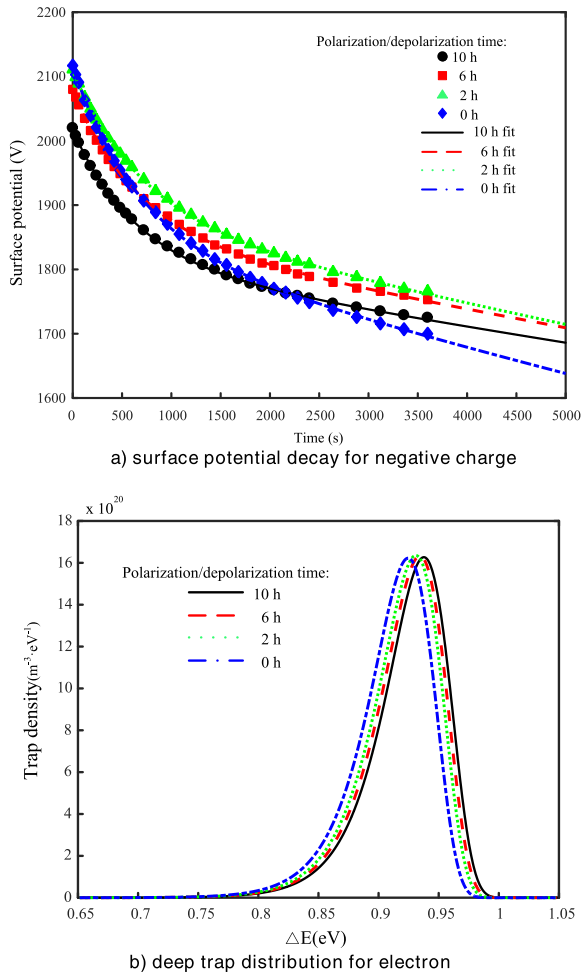


FIGURE 13. Trap distribution of 100um PP film at different polarization time.

the same applied voltage. Therefore, the number of negative charges is larger than that of positive charges, which makes the absolute value of negative potential higher than that of positive potential at the initial time of attenuation. With the increase of polarization/depolarization time, the decay of surface potential becomes slower. Moreover, the surface potential of unpolarized PP film decays faster compared to other samples due to the lower trap density and depth. From Figure 13(b), it can be seen that the deep trap under negative voltage shows the same trend as that under positive voltage. The distribution curve of deep trap moves to the right with the increase of polarization/depolarization degree and the depth of deep trap increases significantly. Compared with unpolarized samples, the density and depth of electron traps of polarized samples increase, and the depth of deep trap increases from 0.9243 eV to 0.9375 eV.

V. CONCLUSION

This paper investigated the trap characteristics and the breakdown characteristics of PP films at different polarization time. The effects of electrode spaces and electric-field structures

were also taken into consideration. The main conclusions are as follows:

(1) With increasing the thickness of PP films, breakdown voltages increase as well, but breakdown area and ASM decrease.

(2) As the polarization time increases, dielectric constant, dielectric loss and polarization current all increase, but volume resistivity decrease. And the trap density and trap depth both increase, which can promote the accumulation of space charge trapped by traps. Moreover, breakdown voltages, breakdown area and ASM all decrease for PP films.

(3) With the increase of electrode spaces, breakdown voltages, breakdown area and ASM all increase.

(4) With increasing the uniformity of electric field, the breakdown voltages, breakdown area and ASM all show the increasing tendency.

The obtained results reveal the polarization and non-uniform electric field and their effects on the breakdown characteristics of PP films, which is beneficial to improving the dielectric reliability of PP films during the operating lifetime of HEDC.

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