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Influence of Space Charge on Dielectric Property and Breakdown Strength of Polypropylene Dielectrics under Strong Electric Field

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Abstract: Space charge accumulation in polypropylene (PP) affect the dielectric properties and breakdown strength of the material. The pre-injected charge in PP under the action of different polarity voltage is quantitatively characterized, and the effects of the pre-injected charge inside the dielectric on the dielectric properties and breakdown strength are measured and analyzed. Based on the molecular simulations, the influence mechanism of the temperature on dielectric properties and breakdown are discussed. The experimental results show that the injected charges in PP under the negative polarity voltage is significantly larger than that of the positive polarity. These charges have a great influence on the dielectric constant and breakdown performance of PP, and the effect is different for different charge polarity. The effect of negative polarity pre-voltage conditions on the dielectric constant is much greater than that of positive polarity, and the dielectric constant of PP decreases from 2.2 to 1.3, decreasing about 41% under the negative polarity pre-voltage. By contrast, the dielectric constant slightly increases under the effect of the homopolar preload. Furthermore, the breakdown strength of the dielectric after the heteropolar preload is 249 kV/mm, which is 36% lower than that of PP without pre-voltage, and it slightly increases after the positive polarity pre-voltage. As the temperature increases, the increase in free volume favors the development of electron collision ionization and electron collapse processes, leading to a decrease in breakdown voltage at high temperatures. This work has a good guiding significance for the comprehensive evaluation of energy storage parameters.

Keywords: polypropylene; space charge; dielectric properties; breakdown strength

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Citation: Xing, Z.; Gu, Z.; Zhang, C.; Guo, S.; Cui, H.; Lei, Q.; Li, G. Influence of Space Charge on Dielectric Property and Breakdown Strength of Polypropylene Dielectrics under Strong Electric Field. *Energies* **2022**, *15*, 4412. https://doi.org/10.3390/en15124412

Academic Editor: Issouf Fofana

Received: 29 April 2022
Accepted: 11 June 2022
Published: 17 June 2022

1. Introduction

Polypropylene (PP) material has been widely used in high power pulse power supplies, power capacitors, and other energy storage devices with high voltage withstand high insulation impedance, and low loss [1–4]. In engineering applications, the subjected electric field of polypropylene is high, and the metal electrode injects charge into the thin film, space charge accumulation is an important factor affecting the properties of the material [5–8].

At present, the research on energy storage dielectrics represented by PP mainly focused on enhancing the energy storage density of composites including nanoparticle surface modification [9–12], multiphase filler blending [13,14], and multilayer structure design [15]. M. Takala et al. [16] filled POSS nanoparticles into PP, which can effectively inhibit the charge transport and increase the breakdown strength of nanocomposite. Cho et al. [17] prepared the ternary nanocomposites of PP/r-PANI/RGO, and they exhibited a high dielectric constant and high energy density. Hari et al. [18] enhanced the interfacial polarization of the material using strontium titanate and flax fibers, raising the dielectric constant of PP from 0.866 to 2.62. Wang et al. [19] grafted styrene onto a PP matrix to increase the crystallization temperature and accelerate the crystallization rate of PP. Zhou et al. [20] studied the modification of PP with maleic anhydride, which can improve the compatibility between the matrix and the nanoparticles, and significantly enhance the capacitance performance at high temperatures.

However, the current evaluation of PP performance mainly concerns the dielectric constant at a low electric field and the short-time breakdown strength. There are few reports about the effect of space charge accumulation inside the dielectric on the dielectric properties and breakdown strength during long-term operation. Under the action of DC voltage, the metal electrode injects charge into the insulation material; these charges in PP cannot be released in time during the discharge process under the long-term operation, resulting in the accumulation of space charge [21–23]. Wei et al. [24] studied the ageing characteristics of thermally stressed oil paper and found that the space charge increased with ageing time, while raising the corresponding trap energy level. Zhou et al. [25] studied the space charge evolution of SIR at different temperatures with polarity reversal voltages and found that the presence of space charge led to a larger electric field. Park et al. [26] investigated the effect of SiC nanoparticles on the space charge behavior of silicon nanocomposites and found that the breakdown strength was greatest under positive polarity conditions. Li et al. studied space charge distributions in PP and found that the maximum charge density reaches 60 C/m^3 [27]. It has been found that the amount of space charge in PP after charging and discharging 3000 times significantly increases, which has an impact on the life of the capacitor. However, the effect law of space charge on the dielectric and breakdown properties is not fully understood.

In this paper, the effect of space charge on the dielectric and breakdown properties of PP under a strong electric field was studied. Firstly, the quantitative characterization of pre-injection space charge in PP under the effect of different polar electric fields was performed using the thermally stimulated depolarizing current method. Then, the laws and mechanisms of the influence of the polarity of the pre-injection space charge on the dielectric properties are investigated. Finally, the effect law of the polarity of the pre-injection space charge on the breakdown characteristics was investigated, and the effect mechanism of the temperature on breakdown characteristics was discussed.

2. Materials and Methods

2.1. Materials

In the experiments, PP film samples were prepared by the melt blending method. Firstly, the PP raw material was dried in the drying oven at 60 °C for 12 h. Secondly, the appropriate amount of PP particles was placed in the metal mold and heated in the plate vulcanizing machine for a certain time. Then, it was treated at 10 MPa and 195 °C for 15 min. Finally, the mold was cooled at the plate vulcanizing machine, and the film samples of PP with a thickness of 100 µm were obtained.

2.2. Methods

In the experiment, a space charge injection system was designed under a strong electric field. Before the performance testing, space charge was injected into PP sample by applying voltage. The sample is placed in a temperature-controlled chamber to ensure experimental temperature stability while reducing the interference from external conditions. Space charge in PP is pre-injected by the DC high-voltage power supply, as shown in Figure 1. The experimental temperature is 30 °C, the applied field strengths are +50 kV/mm and −50 kV/mm, respectively, and the applied time is 30 min. After the space charge was pre-injected, the thermally stimulated depolarization current test system was used to measure and analyze the amount of the injected charges in PP.

A broadband dielectric spectrometer was used to test the dielectric spectrum of the specimens. The upper and lower surfaces of the specimens were coated separately before the test and dried in an oven at 60 °C for 6 h to avoid the effect of moisture. The test frequency range is 10^{-1}~10^6 Hz, and the temperature changes from room temperature to 100 °C. The thickness of the sample is 0.1 mm.

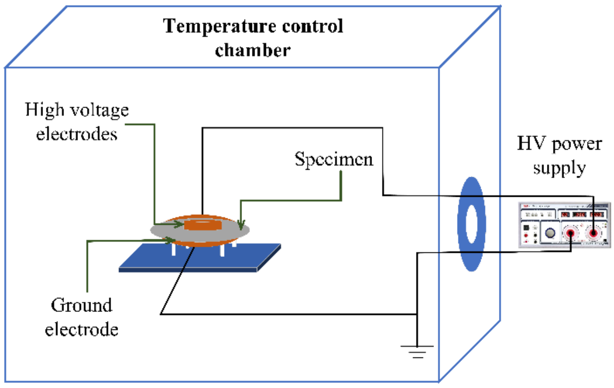

Figure 1. Schematic diagram of the pre-injected space charge in PP.

The voltage breakdown tester was used for breakdown testing. Figure 2 shows a schematic diagram of the breakdown experimental setup, which is composed of high voltage electrode, DC power supply, and a data acquisition system. The sample is placed between the cylindrical electrode with a diameter of 25 mm, and the thickness of the sample is 0.1 mm. According to the IEC 60243-1, the rate of voltage is 500 V/s until breakdown, the test was repeated 5 times. Experimental data were analyzed by Weibull breakdown probability distribution.

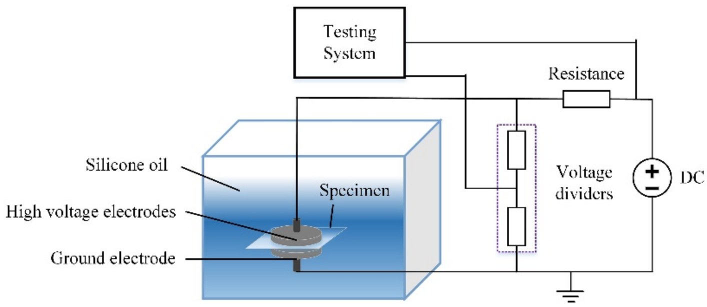

Figure 2. Schematic diagram of the breakdown device.

3. Results

3.1. Quantitative Characterization of the Pre-Injected Space Charge

Firstly, the space charge was pre-injected by the space charge injection system under the electric field of +50 kV/mm and −50 kV/mm, and the applied time was set to 30 min. Secondly, the thermally stimulated depolarization current system was used to measure the thermally stimulated depolarization current (TSDC) from −50 °C to 100 °C, and the amount of pre-injected space charge was extracted. The results are shown in Figure 3.

Under the action of DC strong electric field, the electrons in the metal electrode can cross the interface potential barrier and inject into the PP, and gradually migrate into the interior along the thickness direction. The charge migration process in the material can be affected by the impurity defects of the material, resulting in the accumulation of space charge. Normally, this part of the charge is harder to break free from the trap center at room temperature, and as the temperature increases, the charges trapped gradually gain energy and become free again. Therefore, the TSDC of the specimen after the pre-injected space charge was measured to calculate the amount of the injected space charge.

It can be seen from Figure 3 that the depolarization current of PP is small and almost unchanged before 40 °C, indicating that when the temperature is lower than 40 °C, the trapped charges cannot get enough energy to get out of the trap center. As the temperature increases, the depolarization current significantly increases and reaches its maximum value at around 60 °C, with values of 3.30×10^{-11} A and -1.35×10^{-12} A under 2 different polarization electric fields of −50 kV/mm and +50 kV/mm, respectively. It can be seen that the pre-injected space charge in PP polarized by the negative voltage is significantly larger than that of the positive voltage. Further, the amounts of injected charges in PP under different polarization conditions are calculated, as shown in Figure 4.

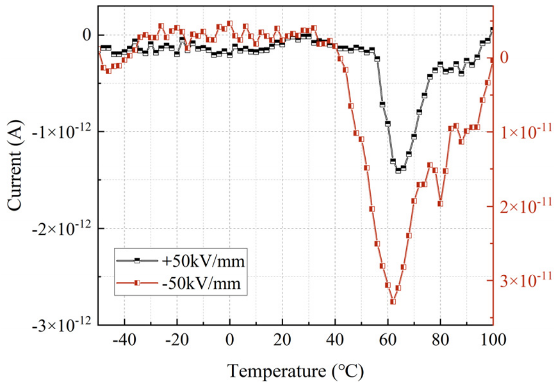

Figure 3. TSDC curves of PP at different polarization electric fields.

The trapped charges inside the dielectric can be calculated from the *TSDC* curve:

$$Q_{TSDC} = \frac{60}{\beta} \int_{T_0}^{T_1} I(T)dT \quad (1)$$

where, β is the heating rate. T_0 and T_1 are the starting and ending temperatures, respectively. $I(T)$ is the *TSDC*.

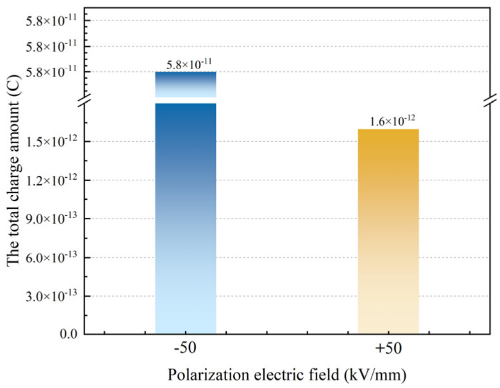

Figure 4. The total charge in PP under different polarization conditions.

Figure 4 shows the total charge in PP under different polarization conditions. When the sample was polarized by the negative electric field of 50 kV/mm, the total charge is 5.8×10^{-11} C. In comparison, it is 1.6×10^{-12} C in PP under the case of the positive electric field. It can be seen that more charges are injected in PP under the negative voltage, which is because the charges are more easily injected across the interfacial potential barrier to the interior of the material under the negative voltage. The changes in space charge in the dielectric under different polarization conditions can affect the dielectric properties and breakdown strength of the material.

3.2. Effect of Space Charge on Dielectric Properties

Firstly, the permittivity and dielectric loss of PP with temperature were tested, the applied voltage is 1 V, and the temperature changed from room temperature to 100 °C.

Secondly, the samples were pre-injected charges under a different applied electric field of +50 kV/mm and −50 kV/mm for 30 min. Then, the permittivity of PP after pre-injected charges was tested by the broadband dielectric spectrometer to study the effect of space charge on dielectric properties. The test results are shown in Figure 5.

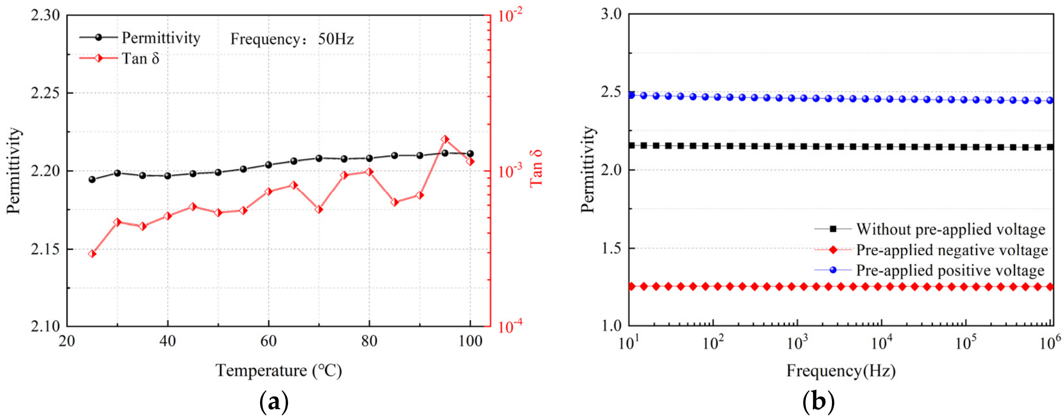

Figure 5. The change in permittivity of PP before and after pre-injected space charge: (**a**) the change in permittivity and dielectric loss of PP with temperature before pre-injected charge; (**b**) the change in permittivity of PP with frequency after pre-injected charge.

As can be seen from Figure 5a, the permittivity of PP slightly increases with the increase in temperature. At the low temperature, the molecular chain segments are closely aligned with each other and the rotation is difficult. As the increase in temperature, the small molecular chain segments and groups gradually separate and turn, resulting in a slight increase in permittivity. The permittivity of PP is less affected by the temperature because PP belongs to a non-polar dielectric with the dominant polarization types of electron displacement.

From Figure 5b, it can be seen that the permittivity of PP varies little with the frequency, which is 2.16 at the high frequency and 2.2 at the low frequency. In addition to the electron displacement polarization, the thermionic polarization also occurs at the low frequency, owing to the effect of some impurity ions, and the thermionic polarization cannot be established at the high frequency, which needs about 10^{-2} s.

To better analyze the effect of space charge on the dielectric properties of PP, the infrared absorption spectra of samples before and after pre-injected space charge were tested, and the wavenumber changes from 400 cm^{-1} to 4000 cm^{-1}. The infrared test results are shown in Figure 6.

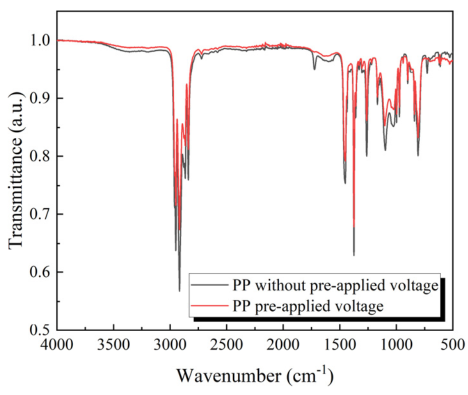

Figure 6. Infrared spectra of PP before and after pre-applied voltage.

As can be seen in Figure 6, the IR spectra of PP after pre-injected space charge are the same as the sample without space charge, and no new absorption peaks appear. It indicates that space charge in PP does not affect the internal functional groups, and the effect of space charge on the dielectric properties is mainly influenced by the physical processes.

It can be seen from Figure 5b that the permittivity of the PP with the homopolar charges increases from 2.2 to 2.4, enhancing by about 9%. It is analyzed that some part of pre-injected positive charges can affect the polarization process. The local electric field generated by this part of the charge is consistent with the original electric field, which enhances the internal electric field and promotes the separation of the positive and negative charge centers, thus improving the permittivity. Meanwhile, the local electric field is beneficial to the thermionic polarization process the ion displacement polarization. The weakly bound ions caused by impurity defects in the material can migrate to the local region, resulting in thermionic polarization. Figure 7 shows a schematic diagram of the effect of space charges on the polarization processes. In the polarization properties testing, the applied voltage is AC voltage, and here shows the influence of space charge on the polarization process during the applied voltage is positive half period.

The accumulation of heteropolar space charge has a great influence on permittivity. It decreases from 2.2 to 1.3, decreasing by approximately 41%. When the negative voltage is applied, a large number of heteropolar charges are injected near the PP surface, which can generate a local electric field in the opposite direction of the original electric field. It can weaken the displacement polarization process and thus reduces the internal dielectric capacity. Comparing the effect of the space charge polarity in PP on the dielectric constant, the effect of heteropolar charges on the dielectric constant is much greater than that of the homopolar charge. This is due to the fact that more space charges are injected in PP under the negative voltage, which is about 5.8×10^{-11} C. In comparison, the amount of injected space charge under the positive voltage is 1.6×10^{-12} C.

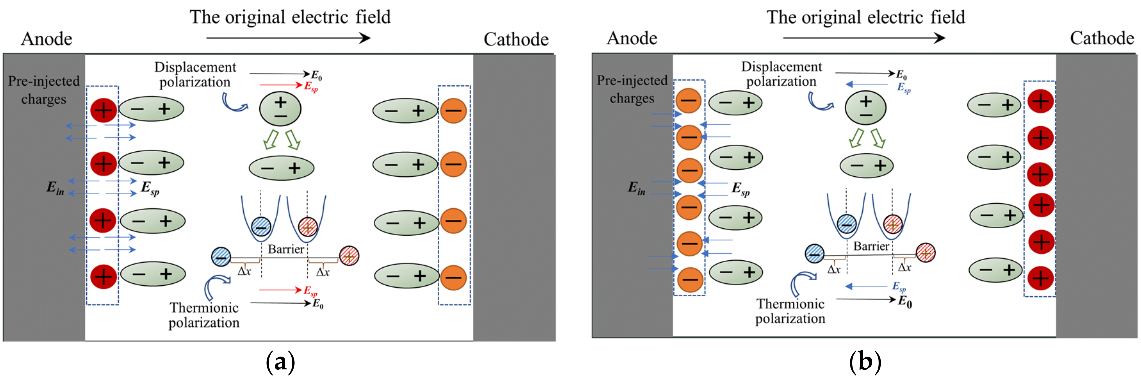

Figure 7. Schematic diagram of the effect of space charge on polarization processes. (**a**) the positive polarity pre-voltage; (**b**) the negative polarity pre-voltage.

3.3. Effect of Space Charge on Breakdown Strength

Firstly, the samples were pre-injected charges under a different applied electric field of +50 kV/mm and −50 kV/mm for 30 min, respectively. Then, the breakdown strength was tested by the breakdown testing system. The results are shown in Figure 8, and the Weibull parameters are shown in Table 1.

As can be seen from Figure 8, the DC breakdown field strength of the PP without pre-voltage is about 388 kV/mm. The breakdown strength of PP after pre-applied by the positive voltage increases slightly. In comparison, the effect of negative pre-applied voltage on the breakdown strength is relatively large, decreasing about 36% than that of without pre-applied.

The analysis shows that this is related to the internal space charge of PP under different pre-voltage conditions, as shown in Figure 9. When the positive voltage is pre-applied, the electrode interface can inject the same polarity charge and migrate into the interior of the material. The local electric field generated by this part of the charge near the interface is opposite to the original electric field, which can weaken the interface electric field.

Meanwhile, the electric field generated inside the material is consistent with the original electric field, which can enhance the internal electric field, as shown in Figure 9a. Usually, compared with the dense structure inside the material, the interface between the metal electrode and the dielectric is more prone to breakdown due to defects such as air gap or electrode burr. When the applied voltage is high, the breakdown discharge initials from the interface and extend to the interior and eventually form a penetrating discharge channel.

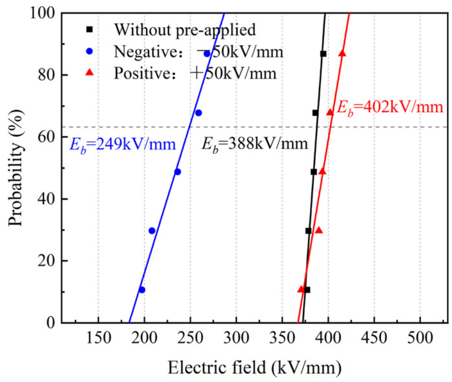

Figure 8. Weibull distribution of breakdown strength of PP.

Table 1. Parameters for breakdown behavior.

	Scale Parameter α (kV/mm)	Shape Parameter β
Without pre-applied	388	70
Negative: −50 kV/mm	249	9
Positive: +50 kV/mm	402	29

In contrast, the pre-injected space charge inside PP under the negative pre-voltage can enhance the interfacial electric field, resulting in the degradation of the breakdown strength, as shown in Figure 9b. Comparing the breakdown voltage of PP under different pre-voltage conditions, the effect of the heteropolar preload on the breakdown strength is greater than that of the homopolar preload, owing to the injection of more space charge in PP under the heteropolar preload conditions.

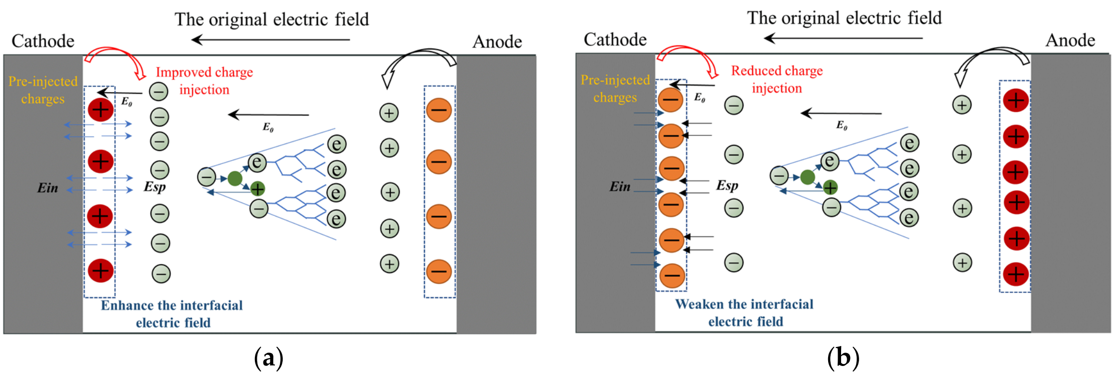

Figure 9. Schematic diagram of the effect of pre-applied space charge on breakdown strength. (**a**) heterocharges; (**b**) homocharges.

3.4. Effect of Temperature on Breakdown Strength

Temperature is an important factor affecting the breakdown characteristics of PP dielectric. In this section, the breakdown characteristics of samples at different temperatures from 25 °C to 90 °C were tested, and the result is shown in Figure 10, and the Weibull parameters are shown in Table 2. Further, the changes in microscopic parameters such as the free volume and the mean square displacement of PP at different temperatures were analyzed by molecular simulation, and the breakdown mechanism of PP dielectric was discussed.

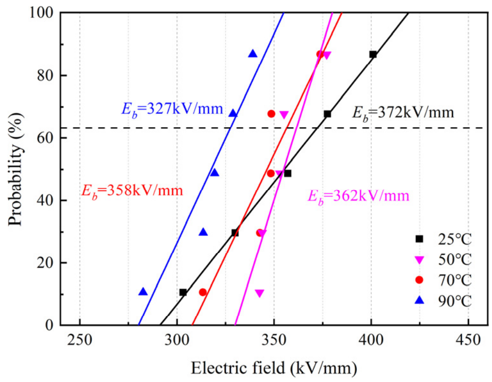

Figure 10. Weibull distribution of breakdown strength of PP at different temperatures.

Table 2. Parameters for breakdown strength.

	Scale Parameter α (kV/mm)	Shape Parameter β
25 °C	372	11
50 °C	362	20
70 °C	358	17
90 °C	327	33

The Weibull distribution of breakdown strength of PP is shown in Figure 10. As can be seen, the breakdown strength of PP has a slight decrease as the temperature increases. When the temperature is 90 °C, the maximum breakdown strength is 327 kV/mm, descending about 12%. This is related to the changes in the carrier migration and the free volume caused by the temperature. The increase in temperature can lead to an increase in carrier migration speed and higher accumulated energy in the material, which is more conducive to the development of electron collision ionization and electron collapse processes. Besides, the development process of breakdown relates to the free volume in the material. The rising temperature increases the free volume in the dielectric, and the free stroke of electrons inside the dielectric becomes larger, accumulating more energy and making breakdown more likely to occur. Moreover, the heat generated inside the material at high temperatures cannot be diffused in time, resulting in the accumulation of internal heat, which can increase the probability of thermal breakdown.

To analyze the effect of temperature on the breakdown characteristics, the changes in microscopic parameters such as the free volume and the mean square displacement of PP dielectric at different temperatures were analyzed by the molecular simulation. Firstly, the molecular model of PP is constructed as Figure 11, and the molecular structure is optimized to obtain a stable molecular space structure. Then the optimized model was subjected to molecular simulations to analyze the effect of temperature on the microstructure of PP.

The volume of the molecule is distinguished into occupied volume and free volume, which in turn contains the free volume hole and the closed space [28]. Figure 11b shows the free volume of the PP dielectric, where the blue part is the free volume hole and the gray part is the closed space. Figure 12 displays the variation of the free volume of PP dielectric with temperature, in which the free volume increases with the increase in temperature. The minimum value is 1.23 nm^3 at 25 °C, and it rises to 1.34 nm^3 when the temperature rises to 70 °C and dramatically increases to 1.56 nm^3 when the temperature rises to 90 °C. The increase in free volume would enlarge the free stroke of electrons inside the dielectric, which is conducive to the development of electron collision ionization and electron collapse processes.

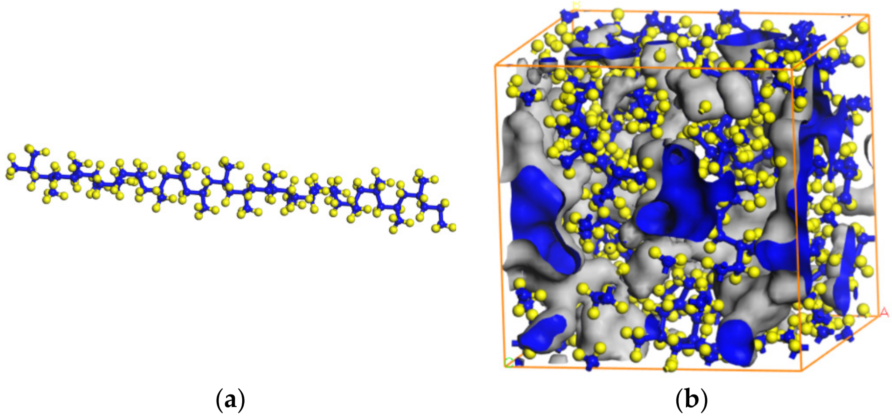

Figure 11. Molecular model. (**a**) molecular chain structure of PP; (**b**) the free volume of PP.

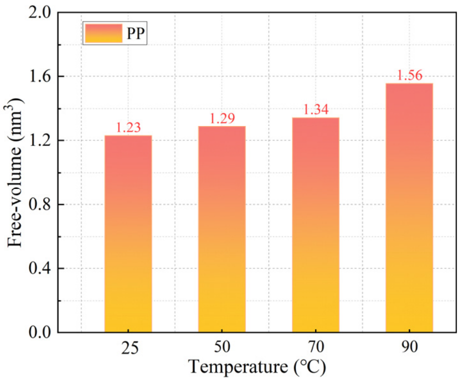

Figure 12. Variation of the free volume of PP with temperature.

The molecular chain motion characteristics of PP at different temperatures were further analyzed, and the mean square displacement variation curves of PP dielectric at different temperatures were shown in Figure 13. As can be seen, the molecular chain movement intensifies with the increase in temperature, and the mean square displacement gradually increases from about 33 Å^2 at 25 °C to 52 Å^2 at 90 °C. The change in means square displacement in the PP is expressed macroscopically as an increase in the dielectric constant of the dielectric with increasing temperature.

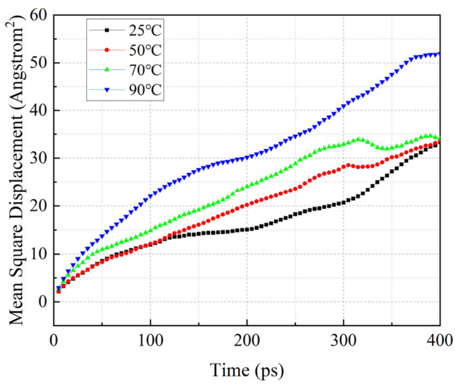

Figure 13. Variation of mean square displacement of PP at different temperatures.

4. Conclusions

The amount of space charge in PP under the action of different polarity voltage is quantitatively characterized, and then the influence law and mechanism of space charge pre-injected in PP on dielectric properties and breakdown strength were investigated. The main conclusions are as follows.

(1) The injected charges in PP under the negative polarity voltage was significantly larger than that in the positive polarity case. The total charges are 5.8×10^{-11} C and 1.6×10^{-12} C under conditions of -50 kV/mm and +50 kV/mm, respectively. This is because the charges are more easily injected across the interfacial potential barrier to the bulk of the material under the negative polarity voltage;
(2) Space charge in PP affect its dielectric properties, and the effect of negative polarity pre-voltage on the dielectric constant is much greater than that of positive polarity. The dielectric constant of PP under hetero-polar pre-voltage conditions decreases from 2.2 to 1.3, decreasing by approximately 41%. In comparison, it slightly increases under homo-polarity pre-voltage. This is mainly related to the polarity and amount of pre-injected space charges in PP. It is necessary to pay more attention to the effect of space charge accumulation on the dielectric constant of energy storage material in long-term operation;
(3) The breakdown strength of PP has a slight increase after homo-polarity pre-voltage, and the breakdown strength of hetero-polarity decreases by approximately 36% than that of PP without pre-voltage. This is mainly related to the change in the interfacial electric field caused by the space charge near the electrode. In addition, the breakdown strength decreases with the increasing temperature, which is related to the carrier migration and free volume change caused by the temperature. Compared with the room temperature and 90 °C, the free volume increases by 26.8% and the increase in electron free travel is beneficial to the development of electron collision ionization and electron collapse.

Author Contributions: G.L. put forward the idea and analyzed the data. Z.X., Z.G. and G.L. carried out the experiments and wrote the paper, C.Z., S.G. and H.C. assisted in completing the experiment. Q.L. analyzed the mechanism. All authors have read and agreed to the published version of the manuscript.

Funding: This study is supported by the State Key Laboratory of Advanced Power Transmission Technology, grant No. GEIRI-SKL-2020-014.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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