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Polycyclic Compounds Affecting Electrical Tree Growth in Polypropylene Under Ambient Temperature

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ABSTRACT Polypropylene (PP) has great potential to be used as recyclable high-voltage direct current (HVDC) cable insulation material. Electrical tree is the key factor that leads to cable insulation failure and limits the increase of HVDC transmission voltage level. In this study, three different polycyclic compounds, namely 4,4'-bis(dimethylamino)phenylazo, 2-hydroxy-2-phenylacetophenone and 4-phenylbenzophenone, are added into PP matrix with the addition content of 0.1 wt%. The electrical treeing experiments are conducted under impulse superimposed DC voltage at 30, 70 and 90 ◦C. Then the polycyclic compound with the best effect on inhibiting electrical tree is selected, and its filling ratio is further changed from 0.1 wt% to 0.3 wt% to get the optimal inhibition effect. It is found that the addition of 2-hydroxy-2-phenylacetophenone can inhibit the electrical tree degradation. However, the addition of 4-phenylbenzophenone and 4,4'-bis(dimethylamino)benzene will increase the electrical tree length and accumulated damage. As the content of 2-hydroxy-2-phenylacetophenone increases, the electrical tree length and accumulated damage decrease firstly, and then increase. The effect of polycyclic compounds on the electrical tree degradation is affected by the relative polarity of impulse superimposed DC voltage and temperature. The experimental results reveal that adding $0.1 \text{ wt\% } 2$ -hydroxy-2-phenylacetophenone polycyclic compound to PP can effectively inhibit the electrical tree degradation, which has great potential for application in recycle HVDC cables.

INDEX TERMS HVDC cable, polypropylene, electrical tree, polycyclic compounds, ambient temperature.

I. INTRODUCTION

The transmission of HVDC cable through its narrow corridors, large transmission capacity, low line loss and high efficiency develops rapidly, and plays an important role in the integration of renewable energy generation, offshore platforms and island power transmission [1], [2]. At present, the most commonly used HVDC cable insulation material is cross-linked polyethylene (XLPE) [3]. However, there are still some disadvantages that cannot be avoided although its insulation level has reached 500 kV [4], [5]. The main problem is that XLPE is difficult to recycle after reaching the end

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of its service life due to thermosetting properties, resulting in environmental pollution [6]. Therefore, the development of new recyclable cable main insulation materials is very important to us in the near future. PP has excellent electrothermal chemistry, and has the advantages of no cross-linking, recyclability, high production speed and high efficiency in the production process [7], [8]. Japanese scholar Kurahashi et al. reported a 22 kV AC recyclable cable demonstration circuit using PP material as the main insulation in 2002 [9]. It was found that the breakdown strength and dielectric loss of PP were completely meet engineering requirements. In 2016, HV Cable Solutions Company manufactured the 35 kV PP DC cable. Prysmian developed a PP-based P-Laser cable sample with a voltage rating of $\pm 640 \text{ kV}$ rencently [10]. PP is

a crucial direction in the future development for recycable HVDC cable insulation [11]–[13].

The electrical tree degradation is an important indicator for evaluating cable insulation properties [14]. Once the electrical tree is generated, it will develop at an extremely fast speed, resulting in insulation breakdown and power failure [15]. Electrical tree degradation restricts the development of cable insulation and limits the increase of transmission voltage level [16], [17]. At present, two ways to inhibit the growth of electrical tree are generally used, one is to add nanoparticles to the polymer matrix, and the other is to fill the polycyclic compounds [18], [19]. As an organic compound, the polycyclic compounds can be directly added into the polymer during the production process by mechanical blending, which has the advantages of low cost and simple production process. It was found that polycyclic compounds had excellent effect on inhibiting electrical tree growth [18]–[21]. Simplex Wire and Cable reported a series of polycyclic compounds based on nitrotoluene structure that can significantly improve the electrical tree resistance in PE [20]. The inhibition efficiency of various polycyclic compounds with an added amount of 0.5 wt% on electrical trees were studied, and it was found that the highest efficiency of bismuth increased the electrical tree initiation voltage in LDPE by 1.6 times [21]. The addition of polycyclic compounds such as benzil, acetophenone derivatives and benzophenone derivatives to XLPE could also increase the DC breakdown field strength and inhibit electrical tree growth [22], [23]. However, there are few studies on the effect of polycyclic compounds on electrical trees in PP.

Power electronic devices in the HVDC transmission system, circuit breaker operation and lightning intrusion may cause voltage waveform distortion to form repetitive impulse voltages in the fluctuating state [24], [25]. Direct current (DC) cable insulation is affected by the DC rated voltage and these impulse overvoltages, which can easily lead to the initiation and breakdown of electrical tree in a short period of time, affecting the safe operation of power equipment [26], [27]. At the same time, during the operation of the HVDC cable, the wire core will generate more heat due to the high current, which will cause the cable to work in a high temperature environment [28], [29]. In our previous works, we studied the effects of impulse superimposed DC voltage and temperature on the growth and breakdown characteristics of electrical tree in PP respectively [30], [31]. It was found that the impulse superimposed DC voltage and high temperature accelerate the electrical tree degradation. However, there are few reports on influence of polycyclic compounds on electrical tree degradation inhibition under impulse superimposed DC voltage, especially under different temperatures.

In this study, three different polycyclic compounds are added into PP matrix with the addition content of 0.1 wt%. The electrical tree degradations in PP and PP/polycyclic compounds composites are studied under impulse superimposed DC voltage at 30, 70 and 90 ◦C. Then the polycyclic compound with the best effect on inhibiting electrical tree is selected, and its filling ratio is further changed from 0.1 wt%

(a) 4,4' bis(dimethylamino)benzil

(b) 2-hydroxy-2-phenylacetophenone

(c) 4-phenylbenzophenone

to 0.3 wt% to get the influence of filling ratio on the growth rate and accumulated damage of electrical tree. The influence mechanism of polycyclic compounds and the effect of impulse superimposed DC voltage and temperature on electrical tree growth characteristics are discussed based on the experimental results.

II. MATERIALS AND METHODS

A. PREPARATION OF PP AND PP/POLYCYCLIC COMPOUNDS COMPOSITES

It was reported that adding benzil, acetophenone derivatives and benzophenone derivatives to XLPE could inhibit electrical tree growth under AC voltage. It was also found in our previous works that 4,4'-bis(dimethyl amino)benzil and benzophenone derivatives showed an excellent ability to suppress space charge in XLPE [32]. Therefore, three different polycyclic compounds are selected to be added to the PP matrix in this study. They are: A. 4,4'-bis(dimethylamino)benzil, a kind of benzil. B. 2-Hydroxy-2-phenylacetophenone, a kind of acetophenone derivatives. C. 4-phenylbenzophenone, a kind of benzophenone derivatives. Fig. 1 shows the chemical structural formula of them. An internal mixer and a thermoforming machine were used to prepare PP and PP/polycyclic compounds composites. The specific preparation process of PP/polycyclic compounds composites was as follows: First of all, the experimental materials were placed in an oven at 60 ◦C for 24 h. Secondly, the appropriate content of PP pellets was put into the internal mixer at 150 ◦C with the rotation speed of 50 r/min for 3 minutes, and then a certain amount of polycyclic compound was added to mixer and was mixed for 10 minutes. Next, put the PP/polycyclic compound composite into a special mold, and then placed the mold between the thermoforming machine. Next, melted the composite at 190 \degree C for 15 min, and then pressed the sample at 190 °C and 15 MPa. Finally, reduced the pressure to 0 MPa and turned off the heating power. During the cooling process, when the temperature dropped between $120-140$ °C, the needle electrode was inserted into the special mold. The mold size was 150 mm \times 15 mm \times 3 mm. The distance between the needle tip and the edge of the sample was set to 2 mm. The curvature radius was 3 μ m.

TABLE 1. Specific samples kinds.

For the convenience of distinction, as shown in Table 1, PP was used to represent neat PP without polycyclic compound. The PP/4,4'-bis(dimethylamino)benzil composite with the polycyclic compound content of 0.1 wt% was represented by PP-AN0.1. The PP/2-hydroxy-2- phenylacetophenone composite with the polycyclic compound content of 0.1 wt% was represented by PP-BN0.1. The PP/4-phenylbenzophenone composite with the polycyclic compound content of 0.1 wt% was represented by PP-CN0.1. In order to compare the effect of polycyclic compound content on the growth characteristics of electrical tree, 0.2 wt% and 0.3 wt% PP/2-hydroxy-2-phenylacetophenone composites were prepared, represented by PP-BN0.2 and PP-BN0.3 respectively.

B. ELECTRICAL TREEING EXPERIMENTS

The experiments were carried out on the variable temperature environment experimental platform described in [19]. The experimental temperature was set at 30, 70, 90 ◦C. Because the maximum operating temperature of HVDC XLPE cable that is also the most commonly used HVDC cable at present is generally designed to be 70 °C [33]. Therefore, we set 70 °C to study the effect of polycyclic compounds on electrical trees in PP. At the same time, because PP has a higher melting point of about 150 ◦C, it can increase the operating temperature to about 90 °C [30]. Therefore, a temperature of 90 °C was set to investigate whether the polycyclic compounds can still inhibit the electrical tree at this temperature. The main purpose of setting 30 $°C$ as the room temperature is to form a temperature gradient to compare the difference of influence of polycyclic compounds on electrical tree degradation under different temperatures. The impulse superimposed DC voltage power supply was composed of the HVDC power supply and an impulse generator. The experimental voltage was \pm 35 kV impulse superimposed -25 kV DC voltage. The frequency was 400 Hz. The treeing process was recorded by a digital microscope. All experiments were recorded from the electrical tree initiation starting from the needle tip to the electrical tree breakdown with the longest electrical tree

FIGURE 2. Schematic diagram of the accumulated damage calculation method.

channel reaching the ground electrode. Tree length and accumulated damage were used to investigate the electrical tree degradation. Accumulated damage is a parameter introduced to characterize the deterioration area of the electrical tree, describing the degree of polymer damage [30]. Fig. 2 shows the process of calculating accumulated damage. A. The photo containing the electrical tree is taken. B. The image is filtered and binarized. Then calculating the number of pixels in the black area, which is the accumulated damage of the electrical tree. Specific schematic diagram of experimental setup and detailed descriptions could refer to reference [19, 31].

The trap distribution was measured by surface potential decay (SPD) method, and the detailed descriptions of the surface charge test system can be found in [19]. The experiment was conducted at 30 ◦C with a humidity of 20 %. The sample was charged by DC voltage for 10 min. The specific calculation method of trap level distributions can be found in our previous work [19].

III. EXPERIMENT RESULTS

A. EFFECT OF POLYCYCLIC COMPOUND KIND ON ELECTRICAL TREE DEGRADATION

1) ELECTRICAL TREE DEGRADATION UNDER OPPOSITE POLARITY IMPULSE SUPERIMPOSED DC VOLTAGE

Fig. 3 shows the electrical tree structures in PP and PP-BN0.1 under different temperatures. The impulse voltage is $+35$ kV and the DC voltage is -25 kV. Fig. 3(a) is tree structures in PP samples, and Fig. 3(b) is tree structures in PP/polycyclic compounds composites to which the 0.1 wt% 2-hydroxy-2-phenylacetophenone polycyclic compound is added. The treeing time is 2 min. When the temperature is the same, the electrical tree length in PP-BN0.1 is shorter compared with that in PP. The electrical tree channel is thinner. As the temperature increases, the electrical tree channels become thicker, blacker, and the number of branches becomes more.

Fig. 4 and Fig. 5 show the electrical tree growth characteristics with different kinds of polycyclic compounds under opposite polarity impulse superimposed DC voltage. The impulse voltage is +35 kV and the DC voltage is −25 kV. Fig. 4 shows the relationship between the tree length and the treeing time under different temperatures. The electrical tree length in PP-BN0.1 decreases by 19 % compared with PP with the treeing time of 35 min at 30 $°C$. At 70 $°C$, the electrical tree length in PP-BN0.1 decreases by 7.8 % with

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(b) Tree structures in PP-BN0.1

FIGURE 3. Tree structures in PP and PP-BN0.1 under different temperatures.

the treeing time of 7 min. The electrical tree length in PP-BN0.1 decreases by 6.6 % with the treeing time of 4 min at 90 °C. Fig. 5 shows the relationship between the accumulated damage and the treeing time under different temperatures. The accumulated damage decreases by 19.6 % with the treeing time of 35 min at 30 $^{\circ}$ C. At 70 $^{\circ}$ C, the accumulated damage decreases by 10.9 %. The accumulated damage decreases by 7.7 % at 90 ◦C. It can be concluded that the 2-hydroxy-2-phenylacetophenone polycyclic compound has a certain inhibition effect on the electrical tree length and accumulated damage under opposite polarity impulse superimposed DC voltage at different temperatures. It has the best inhibition effect at 30 ◦C. With the increase of temperature, the effect of inhibiting electrical tree degradation gradually decreases. However, when another two kinds of polycyclic compounds, 4,4'-bis(dimethylamino)benzil and 4-phenylbenzophenone, are added, the electrical tree length and accumulated damage increase. The electrical tree length in PP-AN0.1 increases by 3 % compared with PP with the treeing time of 35 min at 30 ◦C, and it increases by 4.2 % and 4.5 % with the treeing time of 7 min at 70 \degree C and 4 min at 90 \degree C, respectively. The accumulated damage increases by 3.9 %, 8.9 % and 14.1 % at 30, 70 and 90 \degree C, respectively. The effect of 4-phenylbenzophenone is the worst. At 30 $°C$, the electrical tree length in PP-CN0.1 increases by 7.8 % and the accumulated damage increases by 7.7 % with the treeing time of 30 min. With the increase of temperature, the effect is even worse. At 70 \degree C, the electrical tree length in PP-CN0.1 increases by 15.8% and the accumulated damage increases by 18.9 % when the treeing time is 7 min. At 90 $^{\circ}$ C, the electrical tree length in PP-CN0.1 increases by 17.6 % and the accumulated damage increases by 25.6 % when the treeing time is 4 min.

2) ELECTRICAL TREE DEGRADATION UNDER THE SAME POLARITY IMPULSE SUPERIMPOSED DC VOLTAGE

Fig. 6 shows the electrical tree structures in PP and PP-BN0.1 under the same polarity impulse superimposed

FIGURE 4. The relationship between the tree length and treeing time with different kinds of polycyclic compounds under different temperatures under +35 kV impulse superimposed −25 kV DC voltage.

DC voltage. The impulse voltage is −35 kV and the DC voltage is -25 kV. Fig. 6(a) is tree structures in PP samples, and Fig. 6(b) is tree structures in PP/2-hydroxy-2-phenylacetophenone polycyclic compounds composites. Because the electrical tree growth rate under the same polarity impulse superimposed DC voltage is different from that under the opposite polarity impulse superimposed DC voltage, the selected treeing time of Fig. 6 which is 5 min is different from Fig. 3 in order to better show the effect of polycyclic compounds to the tree structure under different temperatures. As shown in Fig. 6, the electrical tree length in PP-BN0.1 sample is shorter compared with PP and the number of electrical tree channels is less at the same temperature. As shown in Fig. 6(a), with the temperature increasing, the electrical tree channels become thicker, and the main

FIGURE 5. The relationship between the accumulated damage and treeing time with different kinds of polycyclic compounds under different temperatures under +35 kV impulse superimposed −25 kV DC voltage.

branches and the side branches overlap, leading to the denser electrical tree structures. As shown in Fig. 6(b), the color of the electrical tree channels doesn't change significantly as the temperature increases from 30 \degree C to 70 \degree C. As the temperature increases from 70 $\mathrm{^{\circ}C}$ to 90 $\mathrm{^{\circ}C}$, the electrical tree channels become thicker.

Fig. 7 and Fig. 8 show the electrical tree growth characteristics with different kinds of polycyclic compounds under the same polarity impulse superimposed DC voltage. The impulse voltage is −35 kV and the DC voltage is −25 kV. Fig. 7 shows the relationship between the tree length and the treeing time under different temperatures. Fig. 8 shows the

(b) Tree structures in PP-BN0.1

FIGURE 6. Tree structures in PP and PP-BN0.1 under different temperatures.

relationship between the accumulated damage and the treeing time under different temperatures. It can be seen that the 2-hydroxy-2-phenylacetophenone polycyclic compound has a certain inhibition effect on the electrical tree degradation under the same polarity impulse superimposed DC voltage under different temperatures. Its inhibition effect is best at $30 °C$, and its effect of inhibiting electrical tree growth gradually decreases with increasing temperature. When two kinds of polycyclic compound, 4,4'-bis(dimethylamino)benzil and 4-phenylbenzophenone, are added, the electrical tree length and accumulated damage increase instead. The electrical tree length and accumulated damage in PP-AN0.1 are not much different from those in PP. However, the electrical tree length and accumulated damage in PP-CN0.1 are significantly larger than those in PP. The electrical tree length in PP-CN0.1 increases by 13.2% compared with PP with the treeing time of 15 min at 30 \degree C, and the accumulated damage increases by 17.1%. The same as the experimental results under the opposite polarity impulse superimposed DC voltage, the effect deteriorates as the temperature increases. At 90 ◦C for 15 min, the electrical tree length in PP-CN0.1 increases by 19.1% compared with PP, and the accumulated damage increases by 17.7%.

B. EFFECT OF POLYCYCLIC COMPOUND CONTENT ON ELECTRICAL TREE DEGRADATION

1) ELECTRICAL TREE DEGRADATION UNDER OPPOSITE POLARITY IMPULSE SUPERIMPOSED DC VOLTAGE

Fig. 9 shows the electrical tree structures with different contents of polycyclic compounds under opposite polarity impulse superimposed DC voltage. The impulse voltage is $+35$ kV and the DC voltage is -25 kV. Fig. 9(a) is tree structures in PP samples, Fig. 9(b) is tree structures in PP-BN0.1, Fig. 9(c) is tree structures in PP-BN0.2 and Fig. 9(d) is tree structures in PP-BN0.3. The treeing time is 2 min. The electrical tree structures are not affected by the content of polycyclic compound and temperature, which are branch

FIGURE 7. The relationship between the tree length and treeing time with different kinds of polycyclic compounds under different temperatures under −35 kV impulse superimposed −25 kV DC voltage.

trees under different conditions. However, the appearance is connected with the polycyclic compound content. As the content increases, the number of electrical tree channels decreases first and then increases. The length of the electrical tree first reduces and then increases.

Fig. 10 and Fig. 11 show the electrical tree growth characteristics with different contents of polycyclic compounds. The impulse voltage is $+35$ kV and the DC voltage is −25 kV. Fig. 10 shows the relationship between the tree length and the treeing time under different temperatures. Fig. 11 shows the relationship between the accumulated damage and the treeing time under different temperatures. It can be seen that the tree length and accumulated damage in

FIGURE 8. The relationship between the accumulated damage and treeing time with different kinds of polycyclic compounds under different temperatures under −35 kV impulse superimposed −25 kV DC voltage.

PP-BN0.1 are the least compared with PP, PP-BN0.2 and PP-BN0.3, which means that the addition of 0.1 wt% 2-hydroxy-2-phenylacetophenone has the best inhibition effect on the electrical tree degradation. However, with the increase of the polycyclic compound addition filling ratio, polycyclic compound can no longer inhibit the growth of electrical tree, but promote the growth of electrical tree. At 30 \degree C and 70 \degree C, PP-BN0.2 still has a certain inhibitory effect on the accumulated damage. With the treeing time of 30 min at 30 \degree C, the accumulated damage decreases by 7.5 %. With the treeing time of 4 min at 70° C, the accumulated damage decreases by 7.1 %. However, at 90 \degree C, the accumulated damage in PP-BN0.2 is larger than that in PP, which

FIGURE 9. Tree structures with different contents of polycyclic compounds under different temperatures.

increases by 25.1 %. PP-BN0.2 doesn't have the inhibitory effect on the electrical tree lengths, but increases the electrical tree lengths, which are larger in PP-BN0.2 than those in PP at different temperatures. The electrical tree lengths in PP-BN0.2 increase by 1.6 %, 7.9 % and 66.5 % at 30, 70 and 90 [°]C with the treeing time of 35, 4 and 2.5 min, respectively. The electrical tree length and accumulated damage in PP-BN0.3 are the largest, especially at high temperatures. The electrical tree length increases by 8.9 % compared with PP, and the accumulated damage increases by 11.1 % at 30 ◦C and 30 min. The electrical tree length increases by 52.9 %, and the accumulated damage increases by 50.3 % at 70 ◦C and 4 min. The electrical tree length increases by 80.2%, and the accumulated damage increases by 53.1 % at 90 °C and 2.5 min.

2) ELECTRICAL TREE DEGRADATION UNDER THE SAME POLARITY IMPULSE SUPERIMPOSED DC VOLTAGE

Fig. 12 shows the electrical tree structures with different contents of polycyclic compounds. The impulse voltage is -35 kV and the DC voltage is -25 kV. Fig. 12(a) is tree structures in PP samples, Fig. 12(b) is tree structures in PP-BN0.1, Fig. 12(c) is tree structures in PP-BN0.2 and Fig. 12(d) is tree structures in PP-BN0.3. The treeing time is 5 min. The tree structures under the same polarity impulse superimposed DC voltage are the same like those under the opposite polarity impulse superimposed DC voltage, which the tree structures are branch trees. With the polycyclic compound addition content increases from 0.0 $%$ to 0.1 wt $%$, the number of main branches and channels of the electrical trees reduces obviously. With the addition content increases from 0.1 wt % to 0.2 wt%, the number of main branches and channels of the electrical trees increases gradually. With the addition content increases from 0.2 wt% to 0.3 wt%, the number of main branches and channels increases obviously.

FIGURE 10. The relationship between the tree length and treeing time with different contents of polycyclic compounds under different temperatures under +35 kV impulse superimposed −25 kV DC voltage.

Fig. 13 and Fig. 14 show the electrical tree growth characteristics with different contents of polycyclic compounds. The impulse voltage is -35 kV and the DC voltage is -25 kV. Fig. 13 shows the relationship between the tree length and the treeing time under different temperatures. At 30 \degree C, the electrical tree length in PP-BN0.1 decreases by 11 % compared with PP. The electrical tree length decreases by 10 % with the treeing time of 12.5 min at 90 °C. Fig. 14 shows the relationship between the accumulated damage and the treeing time under different temperatures. At 30 ◦C, the accumulated damage decreases by 15.5 % with the treeing time of 15 min. The accumulated damage decreases by 4.7 % at 90 $^{\circ}$ C. It can be concluded that with the polycyclic compound addition content of 0.1 wt%, the electrical tree growth can be inhibited

FIGURE 11. The relationship between the accumulated damage and treeing time with different contents of polycyclic compounds under different temperatures under +35 kV impulse superimposed −25 kV DC voltage.

and the inhibiting effect gradually decreases with increasing temperature. The electrical tree length in PP-BN0.2 is slightly larger than that in PP under the same polarity impulse superimposed DC voltage, but it still has a certain inhibitory effect on the accumulated damage, which is the similar condition as that under the opposite polarity impulse superimposed DC voltage. Same as experimental results under the opposite polarity impulse superimposed DC voltage, tree length and accumulated damage are the largest with the addition content of 0.3 wt% under the same polarity impulse superimposed DC voltage, which promotes the growth of electrical trees. Comparing the effect of polycyclic compounds under different relative polarities of impulse superimposed DC voltage,

FIGURE 12. Tree structures with different contents of polycyclic compounds under different temperatures.

although adding 0.1 wt% 2-hydroxy-2-phenylacetophenone can inhibit the growth of electrical tree, the suppression effect is better under the same polarity impulse superimposed DC voltage than that under opposite polarity impulse superimposed DC voltage, especially at high temperatures. When the treeing time is 90 min at 90 ◦C, the length of the electrical tree under the same polarity impulse superimposed DC voltage decreases by 29.9 %, and the accumulated damage decreases by 16.7 %. The length of the electrical tree under the opposite impulse superimposed DC voltage decreases by 12.5 %, and the accumulated damage decreases by 11.5%.

IV. DISCUSSION

A. EFFECT OF POLYCYCLIC COMPOUNDS ON ELECTRICAL TREE DEGRADATION

The electrical tree degradation under impulse superimposed DC voltage is closely connected with the thermal electron motion and trap distributions [31]. Fig. 15 shows the trap distributions of different kinds of polycyclic compounds modified PPs. The neat PP has a deep trap depth of 0.9 eV. After adding different kinds of polycyclic compounds, the changes of trap distributions are different. The addition of the polycyclic compound B increases the trap depth and trap density. However, the addition of the A and C decrease the trap depth and trap density.

In our previous works, we discussed the charge motion process under impulse superimposed DC voltage and the relationship between charge motion and electrical tree growth mechanism. When the applied voltage is −35 kV impulse and −25 kV DC, the amplitude change of the impulse voltage causes the charge to trapping and detrapping to generate energy, and the energy is transferred to other charges to form thermal electrons [34]. Thermal electrons are accelerated in free volume and then impinge on molecular chains leading to the initiation and growth of electrical tree [35]. When the applied voltage is $+35$ kV impulse and -25 kV DC, after

FIGURE 13. The relationship between the tree length and treeing time with different contents of polycyclic compounds under different temperatures under −35 kV impulse superimposed −25 kV DC voltage.

the application of the impulse voltage, energy is produced by charges neutralization, exciting the thermal electrons to strike the molecular chain [36]. Since the energy required for excitation or ionization of most aromatic compounds is lower than that of PP, the aromatics will be excited or ionized by thermal electrons under the action of strong electric field, which makes it possible for polycyclic compound B to show the increased charge trap depth [32], [37]. During the process of injecting the charges, charges are easier to be trapped by the deep trap in PP-BN0.1, so the amount of the injected charges is reduced. At the same time, due to the effect of the

FIGURE 14. The relationship between the accumulated damage and treeing time with different contents of polycyclic compounds under different temperatures under −35 kV impulse superimposed −25 kV DC voltage.

polycyclic compound, the ability of samples to capture internal charges is enhanced with the increases of trap density and depth, resulting in a reduction in the amount of free charges. Therefore, the molecular chain will not easily be destroyed and the electrical tree resistance of PP-BN0.1 is improved. However, it was found from the experiments in this study that polycyclic compounds A and C could not inhibit the electrical tree degradation. In our previous research, it was found that the addition of 4-phenylbenzophenone could inhibit the XLPE electrical tree degradation [19], but it was found in

FIGURE 15. The trap distributions of different kinds of polycyclic compounds modified PPs.

this study that the addition of 4-phenylbenzophenone to PP accelerates the growth of electrical tree, indicating that the same polycyclic compound may have different effects on the electrical tree growth characteristics in different polymers. The literature [32], [38] pointed out that since the polycyclic compounds contained the group that could participate in the crosslinking reaction, they could be bonded to the XLPE molecular chain during the crosslinking process, thereby ensuring that the polycyclic compounds were not easily precipitated from the polymer matrix. However, there is no cross-linking reaction in the production process of PP. The polycyclic compounds are only added to PP by physical blending, and the compatibility of polycyclic compounds with PP molecules has a great influence on the electrical tree growth characteristics. Adding 4-phenylbenzophenone and 4,4'-bis(dimethylamino)benzil to PP accelerates the electrical tree growth rate, possibly due to the compatibility of these two polycyclic compounds with PP molecules is not high enough, and some additives migrated and precipitated, leading to the decreased charge trap depth and the degradation of the electrical tree resistance of the PP/polycyclic compounds composites. As the addition content of 2-hydroxy-2-phenylacetophenone increases from 0.1 wt% to 0.3 wt%, the electrical tree growth rate increases. With the increase in the addition content of polycyclic compound, additives will be concentrated in one place, and it is easy to introduce physical defects, resulting in deep trap levels, reduced trap density, and increased carrier mobility, which in turn is equivalent to the introduction of impurities, resulting in reduced resistance to electrical tree.

B. EFFECT OF IMPULSE SUPERIMPOSED DC VOLTAGE AND TEMPERATURE ON ELECTRICAL TREE DEGRADATION

It can be found from the experimental results that the growth trends of electrical trees under the same polarity and opposite polarity impulse superimposed DC voltage are similar, but the effect of polycyclic compounds is worse under opposite

polarity than that under the same polarity. On the one hand, this is related to the transport properties of holes and electrons inside the insulating medium [39]. In the electrical tree growth stage, the polycyclic compounds can capture high energy charge and reduce its average free path. Compared with holes, electrons injected into the needle tip are easily trapped by traps. The electron average free path is shorter, and the electrons are more likely to form the same-polar space charge accumulation [40], [41]. The accumulation of space charge weakens the external electric field, so the polycyclic compounds have a better effect under the same polarity than that under the opposite polarity. On the other hand, after the addition of the polycyclic compounds to PP, the polar groups in the composite increase. Under impulse superimposed DC voltage, polar groups are susceptible to polarization and ionization, producing positive and negative charges [42] . Under opposite polarity impulse superimposed DC voltage, the positive and negative charges injected before and after the polarity change are more easily neutralized by the negative and positive charges generated by the ionization of the polar group, generating energy and accelerating the growth of electrical tree.

The growth of electrical tree to breakdown is a complex electro-thermal cumulative breakdown process. Taking the electrical tree growth process under the same polarity impulse superimposed DC voltage as an example, charges accumulate after −15 kV DC voltage applying. With the impulse voltage applying, since the rising edge is short, a large amount of charges is injected to push the injected charge moving. Thermal electrons are produced by the transferred excess energy produced by charge trapping [31]. These thermal electrons impact and destroy the polymer molecular chain to form micropores. Then partial discharges occur, which will produce local temperature rise on short notice. When $T_{ambient \ temperature} + \Delta T_{discharge} > T_{softening}$ (T_{ambient temperature}: the ambient temperature, ΔT discharge: the local temperature rise caused by partial discharges, $T_{\text{softening}}$: local softening temperature of PP), local thermal breakdown is caused. Under the combined action of charged particle impact, the micropores expand, leading to the electrical tree growth [43]. With the ambient temperature increasing, for one thing, the injected charge density increases, and the charge trapping probability reduces due to the higher charge energy. Therefore, the internal carrier density of the sample increases. The trap charge retention time is relatively shorter, the charge detrapping probability is higher, and the polycyclic compounds' capability to acquire thermal electrons is relatively decreased. For another, the thermal cumulative breakdown is enhanced at high temperatures, the effect of thermal electron on electrical tree degradation is decreased, resulting in weakening effects of the polycyclic compounds on electrical tree degradation at high temperatures.

V. CONCLUSION

In this study, three different polycyclic compounds are added into PP. The electrical tree growth characteristics in PP and

FIGURE 16. The electrical tree breakdown time of different kinds of polycyclic compounds with an addition content of 0.2 wt% under ± 35 kV impulse superimposed −25 kV DC voltage at 30 ◦C.

these PP/ polycyclic compounds composites under opposite and the same polarity impulse superimposed DC voltage are studied at 30, 70 and 90 °C. Then the filling ratio of the polycyclic compound with the best effect on inhibiting electrical tree is further changed, and the influence of filling ratio on the growth rate and accumulated damage are studied. The results show that adding 0.1 wt% 2-hydroxy-2-phenylacetophenone polycyclic compound can effectively inhibit the electrical tree degradation, which has a great potential for application in recycle HVDC cables. The conclusions could be summarized:

1) The addition of 0.1 wt% 2-hydroxy-2 phenylacetophenone polycyclic compound can inhibit the electrical tree degradation in PP under impulse superimposed DC voltage. However, the addition of 0.1 wt% 4-phenylbenzophenone and 0.1 wt% 4,4'-bis (dimethylamino) benzene will increase the electrical tree length and accumulated damage.

2) As the 2-hydroxy-2-phenylacetophenone content increases from 0.0 wt% to 0.1 wt%, the electrical tree length and accumulated damage decrease. As the content increases from 0.1 wt% to 0.3 wt%, the electrical tree length and accumulated damage increase. The addition of 0.1 wt% 2-hydroxy-2-phenylacetophenone has the best inhibitory effect on the electrical tree degradation.

3) The effect of polycyclic compounds on the electrical tree growth characteristics is affected by the relative polarity of impulse superimposed DC voltage and temperature. Although the growth trend of electrical trees under the same polarity and opposite polarity impulse superimposed DC voltage is similar, but the effect of polycyclic compounds is worse under opposite polarity than that under the same polarity. The effect of the polycyclic compounds at high temperatures is weaken.

APPENDIX

Fig. 16 shows the electrical tree breakdown time of three kinds of polycyclic compounds with an addition content

FIGURE 17. The electrical tree breakdown time of different kinds of polycyclic compounds with an addition content of 0.3 wt% under ±35 kV impulse superimposed −25 kV DC voltage at 30 ◦C.

of 0.2 wt% under ±35 kV impulse superimposed −25 kV DC voltage. Fig. 17 shows the electrical tree breakdown time of three kinds of polycyclic compounds with an addition content of 0.3 wt% under ± 35 kV impulse superimposed −25 kV DC voltage. The experiment temperatures of Fig.16 and Fig. 17 are both 30 ◦C. The breakdown times of PP-CN0.2 and PP-CN0.3 are shorter than breakdown time of PP, indicated that C still could not inhibit the electrical tree growth with the addition content of 0.2 % and 0.3 %. With the addition content of 0.2 % and 0.3 %, none of these polycyclic compounds can inhibit the electrical tree growth. The effect of C is the worst and the effect of B is better.

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