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Effect of Crystalline Morphology on Electrical Tree Growth Characteristics of High-Density and Low-Density Polyethylene Blend Insulation

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ABSTRACT The electrical tree initiation and propagation behaviors are a key issue for the polyethylene-based cable insulation. This paper focuses on the effect of crystalline morphology on electrical tree growth characteristics of high-density and low-density polyethylene (HDPE/LDPE) blend insulation. In this paper, the electrical tree growth characteristics of HDPE/LDPE blends with HDPE mass fractions of 0, 10, 15, 20 wt% are investigated under repetitive impulse voltage at 40, 60, 80 °C. It is found that with the rise of HDPE content from 0 to 15 wt%, the growth rate and accumulated damage of electrical tree decreases, with the morphology of electrical tree tending to bush tree. The increasing of voltage amplitude improves the energy of injected charge while the temperature rise leads to the relaxation of molecular chain, which both result in the promotion of collision ionization, thus increasing the density of electrical tree. Compared with branch tree, bush tree illustrates better inhibition of discharge tracks due to uniform electric field at the end of branches. The crystalline characteristics of HDPE/LDPE blends indicate that the crystallinity increases with the addition of HDPE, and the blend comprising 15 wt% HDPE in an LDPE matrix apparently reduces the average size of spherulites and improves the distribution of spherulites evenly. The upsurge of crystal-amorphous interface leads to the increase of deep trap energy level density and the decrease of carrier mobility. Carrier injection and migration at the interface between crystalline and amorphous regions are restrained, thus inhibiting the growth of electrical tree. It is concluded that the crystalline morphology modified by polymer blending has a significant effect on the electrical tree growth characteristics.

INDEX TERMS HVDC cable, polyethylene insulation, electrical tree, crystalline morphology, conductivity, breakdown, trap level.

I. INTRODUCTION

High-voltage direct-current (HVDC) power cables play a significant role in the power transmission today, particularly for high-capacity, long-distance, and regional power grid interconnections [1]–[3]. Thermoplastic polymeric materials are extremely desirable for power cable insulation application because of its recyclability, non-crosslinking and the absence of byproducts during cable production, which results in the reduction of undesirable space charge accumulation and the degassing cost [4], [5].

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Polyethylene thermoplastic cable insulation has been used for HVDC cable insulation, such as 500 kV PE insulated DC cable developed by Nexans [3], [4], [6], indicating that PE based insulation is a promising research direction for HVDC insulation in the future. The insulation materials used for high voltage DC cables shall possess the characteristics of small temperature coefficient of resistance, less space charge injection and accumulation, high dielectric strength and fine thermal conductivity [7], [8]. With the application of electrical stress, the material may deteriorate and eventually leads to the failure of the insulation. Restraining electrical treeing of PE based insulation is a significant modification process for

large-scale reliable application of LDPE insulation in high voltage DC cables [9].

In the past few decades, new catalyst, copolymerization, blending, nanotechnology and additives have been used to improve the electrical property of PE [10], [11]. It is found that the blending of polyethylene with EVA in appropriate proportion will significantly reduce the injected charge and eliminate the triboelectrification phenomenon [12]. A small amount of polyolefin elastomer (POE) dispersed into LDPE can meaningfully reduce the amount of space charge and improve the breakdown strength of LDPE. The reduction of spherulite size and the increase of crystallinity are the main reasons for this productive result [11]. The mixture of HDPE and LDPE is supportive to nucleation of matrix crystal, which reduces the average size of spherulites. Smaller spherulites increase the interface between crystalline and amorphous phases, thus suppressing the charge migration process and reducing the accumulation of space charge [13]. It is also reported that adding a small amount of HDPE to LDPE can reduce DC conductivity and improve mechanical properties in high temperature field [14].

The electrical tree growth characteristics are influenced by the complex non-uniform state of aggregation structure in LDPE. It has been proved that the difference of dielectric constant and density between crystal and amorphous region makes carriers migrate mainly in amorphous region, and easy to be trapped by interface defects [13]. The breakdown strength is higher in the case of a small difference in density between crystalline lamellae and amorphous parts [15]. Improving the crystal structure of LDPE and the stacking density of lamellae in spherulites is an effective method to inhibit the growth of electrical tree [16].

Compared with LDPE, HDPE exhibits higher dielectric strength and volume conductivity. However, HDPE is not suitable for cable insulation due to its high hardness and bending strength. Considering with the perfect compatibility of PE based materials, comprising HDPE in an LDPE matrix will remain the excellent mechanical properties of LDPE and further improve the dielectric strength. This paper aims at improving the electrical tree resistance characteristics of low density polyethylene insulation by blending high density polyethylene. In this paper, the effects of HDPE content of blended insulation on the growth of electrical tree, dc conductivity and dc breakdown strength are investigated. DSC and SEM tests are employed to characterize the crystallinity morphology of blend insulation. The trap level and charge transport characteristics of the blend insulation are analyzed by isothermal surface potential decay (ISPD) experiments. The effects of HDPE content on the growth characteristics of electrical tree are discussed.

II. EXPERIMENTAL ARRANGEMENT

A. SAMPLE PREPARATION

The raw materials of LDPE (LD100BW, Beijing Yanshan Petrochemical Company, with a density of 0.92 g/cm^3) and HDPE (L501, Beijing Yanshan Petrochemical Company,

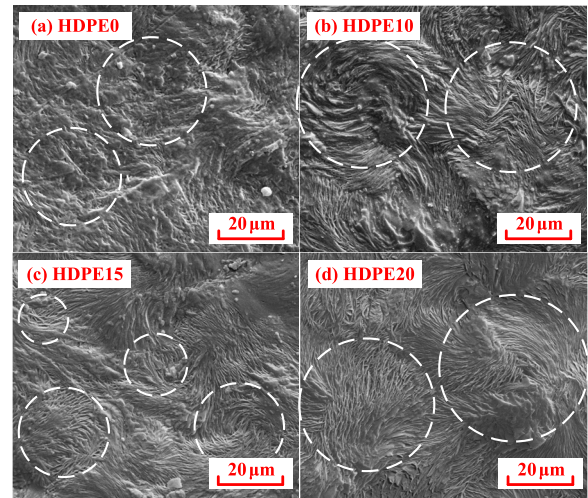


FIGURE 1. SEM images of HDPE/LDPE blends.

with the density of 0.95 g/cm^3) are employed to manufacture the test samples. The raw materials are melted and blended on a twin roll mill at $180 \text{ }^\circ\text{C}$ for 15 min in order to make them disperse and mix uniformly under shear stress. The flat plate specimen is obtained by hot pressing at $160 \text{ }^\circ\text{C}$ with the pressure of 15 MPa for 10 min in the vulcanizer with the size of $80 \text{ mm} \times 30 \text{ mm} \times 1.2 \text{ mm}$. The HDPE/LDPE blends with HDPE content of 0, 10, 15, 20 wt% are obtained, which are designated as HDPE0, HDPE10, HDPE15 and HDPE20, respectively. The made sample is heated to the melting state and the needle electrode is inserted to ensure that the distance between the needle tip and the bottom plate is 2 mm. The diameter of the needle body is $300 \text{ } \mu\text{m}$ and the radius of curvature is $3 \text{ } \mu\text{m}$. When the sample is cooled, the lower surface of the sample is pasted with copper foil [17].

B. CHARACTERIZATION OF CRYSTALLINE MORPHOLOGY

The morphology of the blend insulation is characterized by scanning electron microscope (SEM). Before the characterization, the blended sample is brittle broken in liquid nitrogen, and is immersed in a mixed solution containing 65.7 wt% concentrated sulfuric acid, 33 wt% concentrated phosphoric acid and 1.3 wt% potassium permanganate for 6 hours at room temperature. The sample is repeatedly cleaned by ultrasonic vibration with deionized water to remove the residual acid reagent on the surface.

The SEM morphology of HDPE/LDPE blend insulation is shown in Figure 1. Excellent compatibility of the two materials can be concluded. When the mass fraction of HDPE increases from 0 to 15 wt%, the volume of spherulites atrophies gradually and has uniformly distributed. When the mass fraction of HDPE increases to 20 wt%, the volume of spherulites increases with sparse distribution of spherulites.

The crystallinity and melting point of the blends are measured by differential scanning calorimetry (DSC). The melting curve of HDPE/LDPE measured by DSC is shown in Figure 2. It can be concluded that the crystallinity of samples with HDPE mass fraction of 0, 10, 15, 20 wt% is 36.5%, 37.2%, 41.2% and 47.4% respectively, indicating that

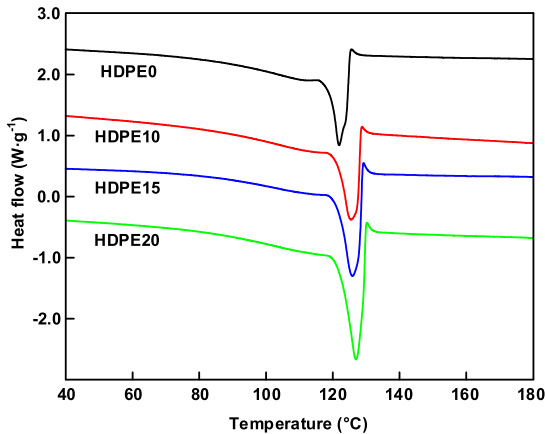


FIGURE 2. DSC curve of HDPE/LDPE blends.

with the increasing addition of HDPE, the crystallinity of the blend sample shows monotonous growth.

C. EXPERIMENTAL METHOD

The electrical tree tests of HDPE/LDPE blend insulation are carried out under repetitive impulse voltage with voltage amplitude (V_p) of 18 kV and 22 kV at 40, 60 and 80 °C. Impulse voltage produced by the high voltage impulse generator is a positive impulse of 200 Hz. The time of rising edge wave front is 50 μ s, and the time of falling edge wave tail is 60 μ s. The experimental process of electrical tree is shown in reference [18]. The optical photographs of electrical trees are recorded by a CCD camera. Each experiment is repeatedly performed for 5 times.

DC conductivity of samples is measured at 40, 60, 80 °C to compare the electrical conductivity and activation energy of HDPE/LDPE blends. The breakdown strength at 40, 60, 80 °C is measured by DC voltage breakdown system, and the DC breakdown strength based on Weibull distribution of HDPE/LDPE blend insulation are compared.

The isothermal surface potential decay test is employed to characterize the trap level distribution of HDPE/LDPE blends [19], [20]. The distance between the needle tip and sample is 5 mm and the tip voltage is set at -6 kV with the corona time of 5min. The experimental humidity is controlled at 20% and the temperature is kept at 60 °C. The double exponential function can be used to accurately fit the surface potential attenuation curve and the general expression of the double exponential function is

$$V_s(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} \quad (1)$$

where $V_s(t)$ is the potential attenuation value of the sample surface at the time t . A_1 , A_2 , τ_1 , τ_2 are the fitting parameters.

The trap energy level E_t , energy level density $N_t(E_t)$ and carrier mobility μ can be calculated by the following formulas [21]:

$$E_t = k_B T \ln(v_{ate} t) \quad (2)$$

$$N_t(E_t) = \frac{4\epsilon_0 \epsilon_r}{q L^2 k_B T} \left| t \frac{dV_s(t)}{dt} \right| \quad (3)$$

$$\mu = L^2 t_T^{-1} U_{s0}^{-1} \quad (4)$$

where k_B is Boltzmann constant; T is the absolute temperature; v_{ate} is the escape frequency of electron; q is the electronic charge quantity; L is the sample thickness; ϵ_0 is the vacuum dielectric constant; ϵ_r is the relative dielectric constant of sample; t_T is inflection point time of surface potential decay rate; U_{s0} is initial potential.

III. EXPERIMENTAL RESULT

A. EFFECTS OF HDPE CONTENT ON ELECTRICAL TREE GROWTH

Figure 3 presents the electrical tree morphology of HDPE0, HDPE15 and HDPE20 samples at 60 °C under repetitive impulse voltage of 18 kV. In terms of tree morphology, HDPE0 and HDPE20 are manifested as branch tree, while HDPE15 is manifested as bush tree. Bush tree propagates toward broad direction and forms crowded discharge channels, indicating higher dimensions of electrical tree growth and shorter electrical tree length. Branch tree is characterized by rapid sparse discharge tracks with vertical deterioration in dominance, resulting in the gathering of discharge channels. With the increase of treeing time, bush tree specifies intensive growth in wide directions while branch tree prompts the development of pointed end for longitudinal extension.

In order to describe the growth process of electrical tree quantitatively, the variation of electrical tree length and accumulated damage percentage [17], [18] of tested samples are employed in this study, as shown in Figure 4. The electrical tree length is measured as the longitudinal growth of electrical tree, defined by the distance from the pointed end of electrical tree and the needle tip. It is found that HDPE15 inhibits electrical tree propagation in electrical tree length and accumulated damage obviously. Compared with the deterioration of HDPE0 and HDPE20, the growth rate of HDPE15 is faster in the first 2 min of treeing time. After 5 min, the electrical tree length of HDPE15 is relatively shorter than that of HDPE0 and HDPE20. With the treeing time of 40 min, the electrical tree length of HDPE15 is 60% of the tree length of HDPE0 and 54% of the tree length of HDPE20 respectively. The electrical tree length of HDPE15 tends to stagnate after 10 min, while the accumulated damage of HDPE15 tends to stagnate with the treeing time of 30 min, indicating the lateral growth with stationary electrical tree length.

Different from the characteristics of bush tree in HDPE15, branch trees in HDPE0 and HDPE20 tends to grow all the time with decreasing growth rate. Electrical tree length tends to stagnate earlier than accumulated damage, reflecting that the longitudinal length increases insignificantly after 10 min of treeing time, but the transverse length of electrical tree remains increasing for all the samples. Due to the disperse growth of bush tree, HDPE15 performs better effects in electrical treeing inhibition.

B. EFFECTS OF VOLTAGE AMPLITUDE ON ELECTRICAL TREE GROWTH

Figure 5 displays the growth of electrical tree of HDPE0, HDPE15 and HDPE20 under repetitive impulse voltage

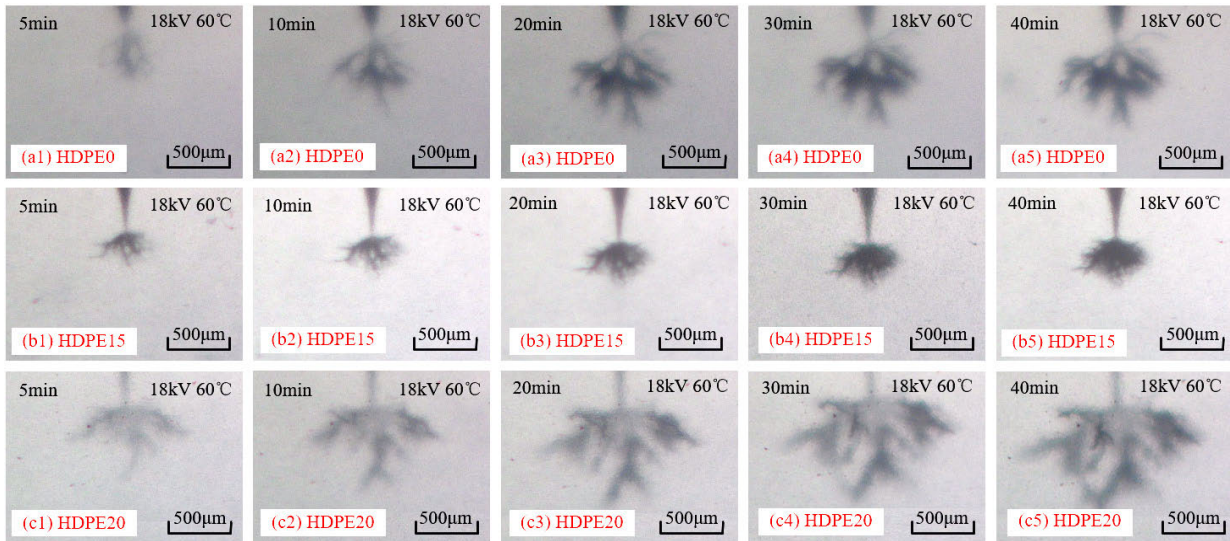


FIGURE 3. Time-varying electrical tree growth of HDPE/LDPE blends at 60 °C under repetitive impulse voltage of 18kV.

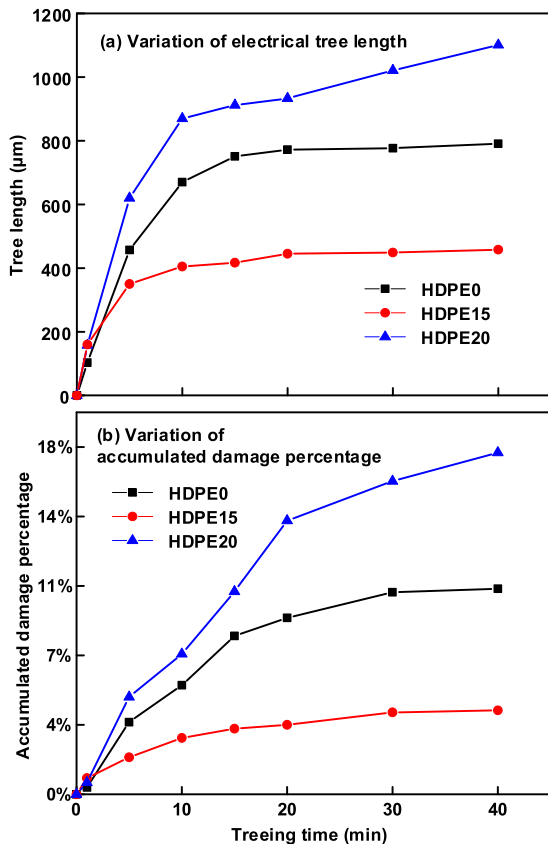


FIGURE 4. Variation of electrical tree length and accumulated damage percentage of different HDPE contents at 60 °C under repetitive impulse voltage of 18kV.

of 18 and 22 kV. The experimental temperature is controlled at 60 °C. Under repetitive impulse voltage of 18 kV, HDPE0 and HDPE20 are termed dense branch tree, while HDPE15 shows bush tree. Under repetitive impulse voltage of 22 kV, bush tree distinguishes among all the samples. When the impulse voltage changes from 18 kV to 22 kV, electrical trees of HDPE0 and HDPE20 indicates morphology

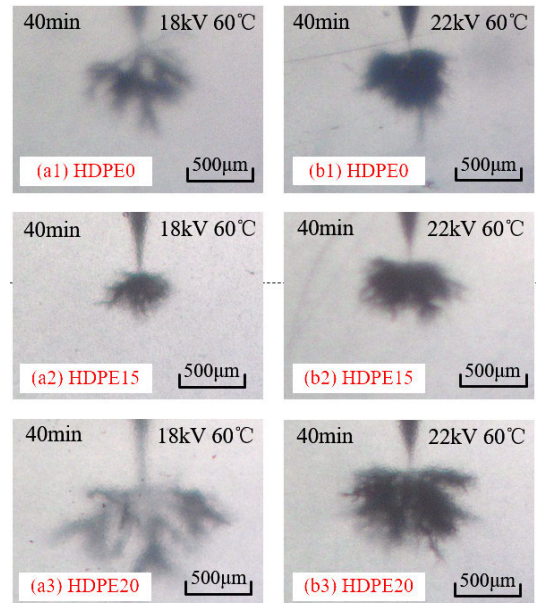


FIGURE 5. Electrical tree morphology of HDPE/LDPE blends at 60 °C under repetitive impulse voltage of 18kV and 22kV for 40 min.

convert from branch to bush, and electrical tree length with treeing time of 40 min significantly reduced. Electrical tree of HDPE15 remains bush with denser discharge channels.

Quantitative electrical tree analysis is available in Figure 6. It can be found that the electrical tree of HDPE20 shows a growth process in 20 min to 40 min of treeing time, while HDPE15 and HDPE0 samples tend to stagnate in the same time period. The electric field intensity of the discharge channel tip can be calculated by the following formula [22]:

$$E = \frac{2U}{r \ln(1 + 4d/r)} \quad (5)$$

where E represents electric field intensity of tip; U is the amplitude of impulse voltage; r is the radius of curvature of tip; d is the distance between tip and grounding electrode.

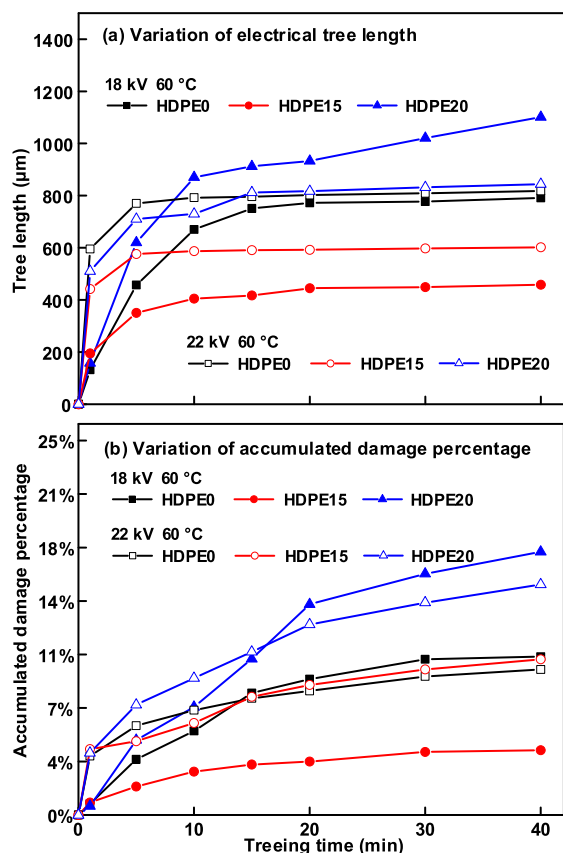


FIGURE 6. Variation of electrical tree length and cumulative damage percentage of HDPE/LDPE blends at 60 °C under repetitive impulse voltage of 18kV and 22kV.

Compared with the bush electrical tree, the curvature radius of the tip of branch channels is small, and the electric field intensity of the branch tip is more concentrated under the same voltage, leading to the promotion of vertical growth of electrical tree. The larger curvature radius at the end of the bush tree disperses the energy injected into the polymer insulation, inhibiting the longitudinal growth of bush tree.

Under impulse voltage of 18 kV, electrical tree lengths of three samples display gradually growth for 20 min and tend to stagnate. Under impulse voltage of 22 kV, electrical tree lengths of three samples grow rapidly in 10 min at the outset and then stagnate earlier than those under 18 kV. Although the length of electrical tree branches tends to be stagnant, the accumulated damage keeps increasing with decreasing gathering speed. Higher voltage amplitude irritates the energy of injected charge, stimulating spread of discharge track and forming a wide range of low density areas at higher voltage level. Meanwhile, higher voltage level impels denser electrical tree, leading to more high energy charges injected into the envelope end of the channels. Due to the electric field shielding formed by the injected charges, the electrical tree tends to stagnate earlier.

C. EFFECTS OF TEMPERATURE ON ELECTRICAL TREE GROWTH

Figure 7 illustrates the electrical tree growth of HDPE15 at 40, 60 and 80 °C under repetitive impulse voltage of 18kV.

In terms of electrical tree morphology, HDPE15 presents branch like electrical tree at 40 °C and bush like electrical tree at 60 and 80 °C. When the temperature increases from 40 to 60 °C, the morphology changes from branch to bush, and the length of tree decreases. When the temperature rises further to 80 °C, bush tree tends to be denser and longer compared with the electrical tree at 60 °C.

The increase of temperature densifies discharge channels of electrical tree. From the standpoint of electrical tree length, the growth rate of electrical tree decreases with the morphology of electrical tree transferring from branch to bush, as shown in Figure 8a. The upsurge of temperature accelerates the growth speed of bush like tree. When the temperature rises from 40 to 60 °C, the shape of electrical tree transfers from branch to bush. Therefore, the shielding effect of electric field formed by injecting electric charge highlights, which makes the growth rate of electrical tree decrease [23]. When the temperature rises from 60 to 80 °C, the bush electrical tree is termed more dense, and the increase of temperature makes the polymer molecular chain segment relax, which makes it easier to form a wide range of low-density areas. Molecular chain relaxation enlarges the free volume of charge motion. As hot electrons are accelerated in the free volume to obtain energy, the energy generated by electron impact ionization increases with the rise of free volume, which accelerates the fracture of the molecular chain, and leads to an enlarged accumulated damage area as shown in Figure 8b.

D. BREAKDOWN PROPERTIES OF HDPE/LDPE BLENDS

DC breakdown strength is an important parameter to estimate the insulation performance of polymers. The Weibull distribution of DC breakdown field strength of HDPE/LDPE blend insulation is shown in Figure 9. At 40 °C, the breakdown field strength of 0, 10, 15, 20 wt% HDPE blend insulation samples with Weibull distribution cumulative failure probability of 63.2% is 265, 281, 295 and 282 kV/mm respectively. When the content of HDPE blends increases from 0 to 15 wt%, the breakdown field strength augments with the rise of the content of HDPE blends. When the content of HDPE blends further increases to 20 wt%, the breakdown field strength declines. Identical trend of breakdown field strength is achieved at 60 and 80 °C. The increase of temperature leads to the relaxation of molecular chain, leading to the decrease of breakdown field strength.

As a typical semi crystalline polymer, the micro crystalline characteristics of HDPE/LDPE blend insulation will inevitably affect its internal trap energy level distribution and charge transport process, and then affect its macro electrical tree growth characteristics, DC conductivity features and breakdown strength.

IV. DISCUSSION

A. EFFECTS OF HDPE ADDITION ON TRAP ENERGY LEVELS AND CRYSTALLINE MORPHOLOGY

Polyethylene-based insulation has a large amount of localized states distributing in amorphous regions as well as the interfaces between crystalline and amorphous regions,

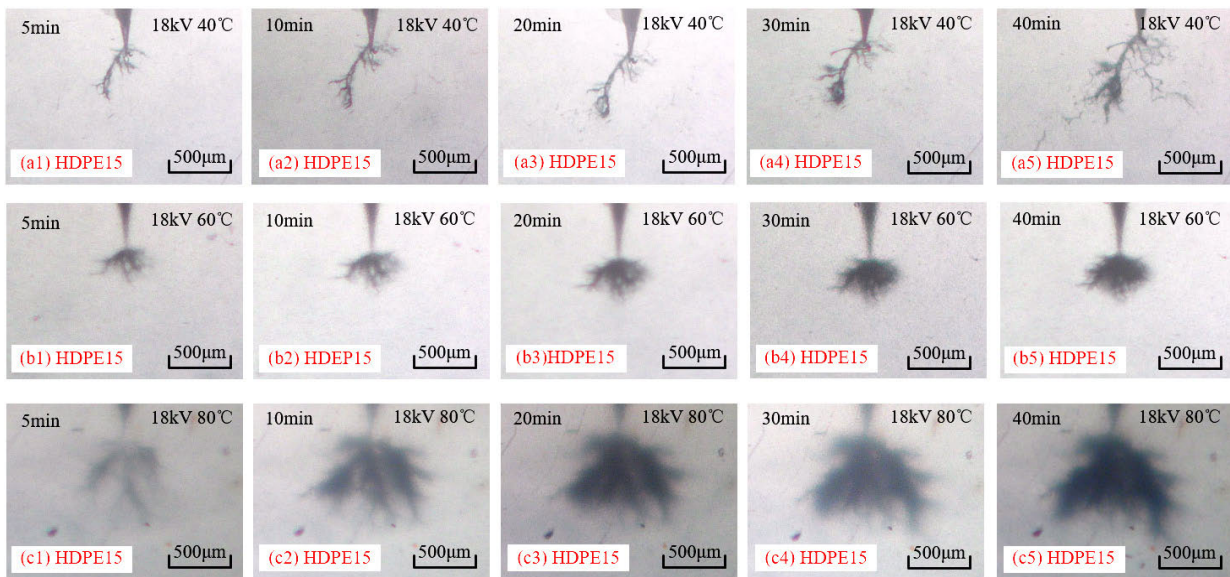


FIGURE 7. Time-varying electrical tree growth of HDPE15 at 40, 60, 80 °C under repetitive impulse voltage of 18kV.

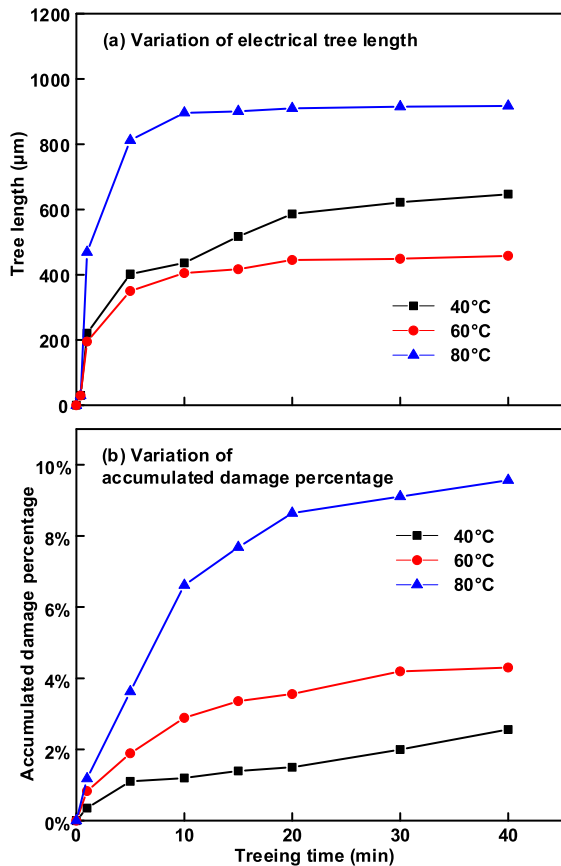


FIGURE 8. Variation of electrical tree length and cumulative damage percentage of HDPE15 at 40, 60, 80 °C under repetitive impulse voltage of 18kV.

which are accessible states for electrons and holes. The localized states are closely related to the process of charge trapping and detrapping, which will directly affect the progression of charge injection, transport and extraction in the sample. The trap distribution characteristics of HDPE/LDPE

based on the isothermal surface potential decay (ISPD) method are shown in Figure 10. The results present that HDPE with a mass fraction of 15 wt% owns the highest deep trap energy density and the lowest shallow trap energy density. The incorporation of HDPE with 15 wt% content decreases the average size of spherulites and improves distribution of crystalline region. The interface between the crystalline region and the amorphous region is raised, which makes it easier to form deep traps that hinder the charge carrier transportation. The upsurge of crystallinity diminishes the amorphous region and consequently weakens the shallow trap density formed by amorphous region.

Figure 11 illustrates the DC conductivity of HDPE/LDPE blend insulation varying with temperature. It can be inferred that with the increase of temperature, more carriers are stimulated to take part in the conductivity, leading to the increasing of volume conductivity. With the surge of HDPE blending content, the DC conductivity of HDPE/LDPE blend sample shows a significant downward trend due to the perfect dielectric properties of HDPE. The DC conductivity is a function of temperature and electric field according to the reference [24]. The activation energy can be calculated by fitting the DC conductivity curve of different samples, and the results are given in Table 1. The deep trap introduced by the addition of HDPE indicates a higher potential barrier for migration process, thus, more activation energy is required for carrier hopping and transportation. The insulations with higher activation energy have a greater temperature dependence of conductivity, which further aggravates the nonuniform distribution of the electric field across the cable insulation under a temperature gradient. The insulation strength and conductivity temperature dependence of cable need to be coordinated through further research in order to achieve the standard of ensuring normal operation of the cable.

It has been proved that the HDPE15 blended sample displays smaller spherulite size and higher crystallinity

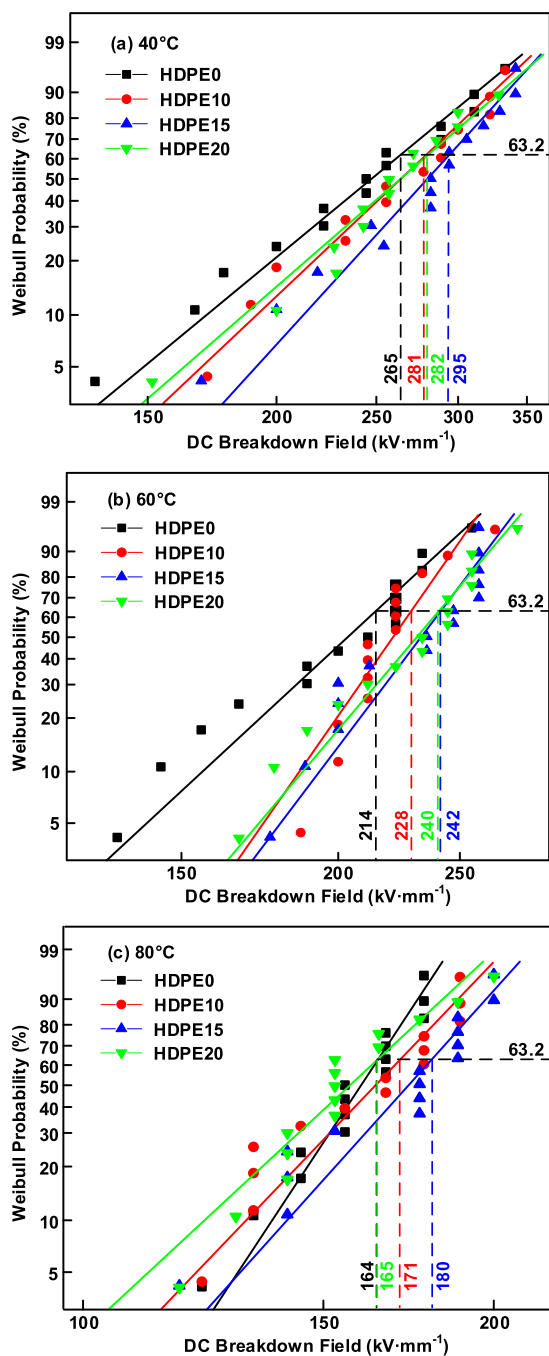


FIGURE 9. Weibull distribution of DC breakdown in HDPE/LDPE blends.

than HDPE0. Due to the different melting points of the components, there will be differences in crystallization sequence when HDPE and LDPE are blended in varying proportions. The effect of this difference on crystallization is shown in Figure 12. When incorporation of HDPE is less than 15 wt%, the HDPE molecules disperse in the LDPE matrix and crystallize earlier at higher temperature, and will become the nucleus of the mixture crystal. These nuclei crystallize out of phase with the surrounding amorphous LDPE, resulting in the decrease of the average size of spherulites. The increase of crystallinity can reduce the shallow traps in the disordered

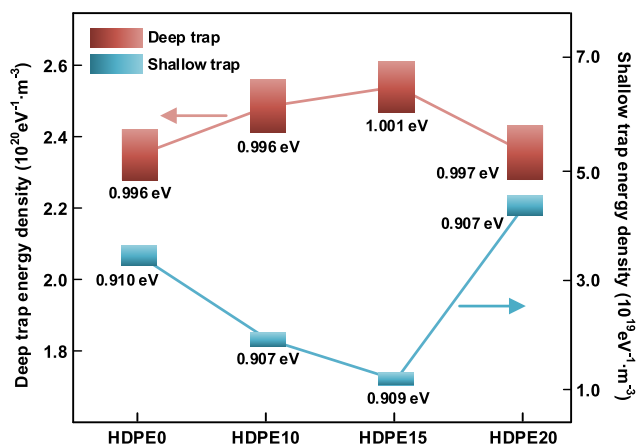


FIGURE 10. Trap level characteristics of HDPE/LDPE blends.

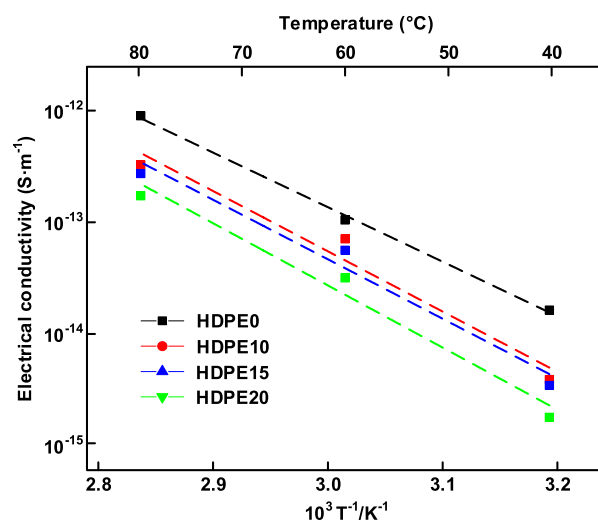


FIGURE 11. DC conductivity of HDPE/LDPE blends under 10 kV/mm at 40, 60, 80 °C.

TABLE 1. The activation energy of HDPE/LDPE blend insulation.

Sample	φ (eV)
HDPE0	0.96
HDPE10	1.07
HDPE15	1.06
HDPE20	1.10

molecular chain in the amorphous region. The decrease of spherulite size and the increase of spherulite interface increase the deep traps at the interface between the crystal region and the amorphous region, resulting in the increase of the deep trap density and the decrease of the shallow trap density in Figure 10 and Figure 11.

When the content of HDPE reaches 20 wt%, HDPE agglomerates in LDPE matrix and extrudes the crystallization space of LDPE in the amorphous region during the formation of spherulites, which is not conducive to the refinement of crystalline spherulites, leading to the enhancement of the average size of spherulites.

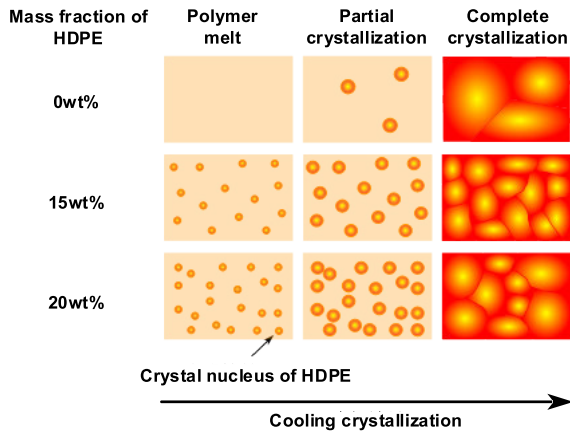


FIGURE 12. Schematic diagram of crystallization of HDPE/LDPE.

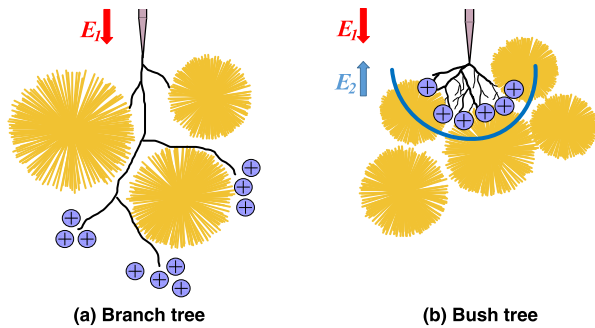


FIGURE 13. Effect of crystalline morphology on electrical tree growth of HDPE/LDPE.

B. EFFECTS OF CRYSTALLINE MORPHOLOGY ON ELECTRICAL TREE GROWTH

The crystalline morphology modified by polymer blending has a significant effect on the electrical tree growth characteristics. The electrical tree growth and propagation mechanism of HDPE/LDPE blend insulation are illustrated in Figure 13. The charge injected from the tip of needle is more likely to migrate in the amorphous region between the spherulites. When the charge migrates to the boundary of the spherulites, it is more likely to be trapped by the deep trap at the interface between the crystalline region and the amorphous region, which leads to the stagnation of the electrical tree growth at the edge of the spherulites or leads to the growth along the edge of the spherulites. When the content of HDPE is 15 wt%, the smaller size and the denser distribution of the spherulites contribute to the inhibition obviously. At the same time, the increase of crystallinity reduces the channels in the amorphous area of the longitudinal growth of electrical tree. Therefore, the electrical tree end encounters resistance when it develops in the longitudinal direction, which is more likely to form partial self-affine fractal or the turning of the discharge path, resulting in the more chance of performing dense bush electrical tree on the macro level. The crystalline morphology influences the trap level distribution, and further makes a significant effect on the electrical tree growth. The bush electrical tree tends to display a lower growth speed,

mainly due to the following reasons [25]–[27]: the bush electrical tree stretches more branch ends, which disperse the injection charge of the needle tip, reducing the local injection charge energy in longitudinal growth. Compared with branch tree, the spherical branch ends of the bush tree make the injection charge easier to form a uniform electrical field shielding, as shown in Figure 13. It is vividly revealed that the electrical field intensity at the end of the bush tree is reduced, so as to inhibit the further charge injection and the growth of electrical branches.

V. CONCLUSION

1) With the rise of HDPE content from 0 to 15 wt%, the growth rate and accumulated damage of electrical tree decreases, with the morphology of electrical tree tending to bush tree. Compared with branch like tree, bush like tree illustrates better inhibition of discharge tracks due to the uniform electric field at the end of branches.

2) The increasing of voltage amplitude improves the energy of injected charge while the temperature rise leads to the relaxation of molecular chain. They both result in the promotion of collision ionization and partial discharge, thus increasing the density of electrical tree.

3) With the rise of HDPE content from 0 to 15 wt%, the crystallinity of the sample increases and the spherulite size diminishes due to heterogeneous nucleation. The deep trap energy level and density of energy level upsurge, hindering the mobility of carrier, thus inhibiting the growth of electrical tree. It is concluded that the crystalline morphology modified by polymer blending has a significant effect on the electrical tree growth characteristics.

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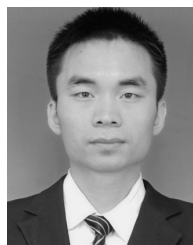
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