Influence of morphological variations on the AC breakdown of XLPE insulation in submarine cable factory joints

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Abstract: Extruded crosslinked polyethylene (XLPE) submarine cables with factory joints play a significant role in cross-sea transmission interconnection projects. Morphological variations in the insulation of submarine cables and factory joints are caused by differences in process parameters. The dielectric properties of XLPE are affected by its morphology. In this study, samples of XLPE insulating materials were taken from the cable bulk, cable–joint transition, and factory joint, respectively, of a 500 kV high-voltage AC submarine cable. The crosslinked morphology was obtained by extraction and infrared spectrometer. The crystal morphology was obtained by the way of etchant, differential scanning calorimetry, and X-ray diffractionmetry. The Weibull distribution was used to gather statistics of AC breakdown strength. The results show that in comparison with the cable bulk, the contents of crosslinked byproducts are higher in the cable–joint transition and factory joint. At the same time, there is a sparse distribution in terms of crystal morphology, with the appearance of smaller grains and lower crystallinity, in the cable–joint transition and factory joint. These morphological variations would generate more heat during electron transport and lengthen the free paths of electrons, then lead to the occurrence of AC breakdown. The results might provide insights into the optimisation of the morphology of XLPE, in particular submarine cable factory joints.

1 Introduction

The production and application of submarine cables have ushered in significant developments in offshore wind power and island interconnection, as well as investment in a series of key construction projects [1–3]. According to the statistics, the total length of submarine cables exceeds 2000 km which carries the voltage of at least 110 kV in China. In addition, the production and operation of 500 kV crosslinked polyethylene (XLPE) submarine cable with a length of 18.15 km have been achieved for the first time worldwide [4, 5]. However, there is a limit on the tonnage of insulating materials for submarine cable production equipment [6, 7]. The factory joints are required to connect with submarine cable lines of great length. Therefore, the research on submarine cable factory joints has become the key issue when the submarine cable projects are undergoing rapid development. At present, the fabrication of factory joints for 220 kV submarine cables has been achieved in China [8–10]. Factory joints for 500 kV submarine cables [11] have passed type tests and pre-qualification tests and they have been commercially used in Zhoushan, Zhejiang province, China.

Submarine cable factory joints are fabricated by insulation recovery on the cable bulk, accompanying with heating and crosslinked processes. There are three forms in insulation, expressed as cable bulk insulation, cable–joint transition insulation, and factory joint recovery insulation. In actual cable fabrication, the process parameters used between the insulation recovery of factory joints and cable bulk insulation are diverse. It reflects the aspects of temperature, heating-up period, and pressure control. These diversities can lead to variations in crosslinked and crystal morphology between the cable bulk insulation and the factory joints.

Some research findings have revealed that the dielectric properties of insulating materials are affected by the morphological variation. Jin found that the grain size was larger, the gap between grains was larger, and the dielectric loss tangent and permittivity were larger in outer bending of cable than that of inner. The reason was crystal morphology of XLPE would change under the action of stress [12]. Harlin found that the crystal morphology of XLPE had a great impact on the electrical tree initiation [13]. Zhong found that the space charge behaviour and DC conductivity of XLPE would be affected by the small size of the grains formed in a factory degassing process [14, 15]. Fothergill found that crystalline material grown from recrystallisation processes that take place in the polarisation stage attains a particularly stable polarisation [16]. However, most of the research objects are thin film samples prepared in laboratory which is quite different from the cable insulation produced actually. Further research into the finished cables and the actual fabrication process is still needed. In addition, there have been no studies about the effect of factory joint insulation recovery on cable–joint transition. This paper provides a reference for factory joint insulation recovery, especially with large insulation thickness.

In this paper, XLPE insulation from the cable bulk, cable–joint transition, and factory joint from a 500 kV submarine cable are studied. Just to be practical, the cable bulk and cable–joint transition have been degassed in factory, and the factory joint is used immediately after fabrication. The morphology and AC breakdown strength are analysed [17], with the aim of examining the influence of morphological variations in XLPE on AC breakdown performance. At the same time, with the purpose of the factory joints fabrication optimisation, the different properties comparing among the cable bulk, cable–joint transition and factory joint are represented.

2 Experimental

2.1 Sample preparation

Samples were obtained from the same 500 kV XLPE submarine cable with joint fabricated in factory. The cable bulk was extruded by vertical continuous vulcanisation. The cable–joint transition...
fulfilled with the test standard ASTM D2765-01. The crosslinked outer insulation. The submarine cable with factory joint and thickness. Therefore, in this paper, the insulation is separated into three parts, in other words, inner insulation, middle insulation, and joint insulation. They were cut by circumferential lathe. The specimens were sealed and kept at room temperature. Considering the thermal conductivity of XLPE, the temperature drop rate at different radial positions would be inconsistent. And this phenomenon is more obvious in cable with larger insulation thickness. The samples used in this paper are originated from the cable bulk but subsequently experienced the joint insulation recovery process. The factory joint underwent injection moulding and vulcanisation. The three kinds samples were segmented into 20 cm sections with an electric saw, and then they were cut by circumferential lathe. The specimens were sealed and kept at room temperature. Considering the thermal conductivity of XLPE, the temperature drop rate at different insulation radial positions would be inconsistent. And this phenomenon is more obvious in cable with larger insulation thickness. Therefore, in this paper, the insulation is separated into three parts, in other words, inner insulation, middle insulation, and outer insulation. The submarine cable with factory joint and samples shown in Fig. 1. The samples used in this paper are based on the actual cable fabrication situation. The cable bulk and cable–joint transition have been degassed in factory, but the factory joint is used immediately after fabrication.

2.2 Measurement methods

2.2.1 Crosslinked morphology: The test of crosslinked degree is fulfilled with the test standard ASTM D2765-01. The crosslinked degree \( C_{gel} \) could be calculated as follows:

\[
C_{gel} = \left( m_1 / m_2 \right) \times 100\%
\]  

where \( m_1 \) is remaining mass (g) and \( m_2 \) is sample mass. Measurement needs repetition for three times and takes the average.

A Nicolet 6700 advanced Fourier transform infrared spectrometer is used to measure the relative contents of crosslinked byproduct. The concentrations are calculated based on the Beer–Lambert law

\[
A = \lg(I_0/I) = C \varepsilon \rho
\]

where \( A \) is absorbivity, \( I_0 \) and \( I \) are the intensity of incident and transmitted (cd), \( C \) is concentration (mol/l), \( L \) is thickness (cm), and \( \varepsilon \) is absorption factor. The ratio of the characteristic peak absorption intensity of byproduct and methylene –CH₂– is taken as the relative content. It is important to note that the relative content of byproduct is a ratio, without units. Ten samples are measured, respectively, and taken the average.

2.2.2 Crystal morphology: To observe the grain size of XLPE, a specimen is immersed in an etchant mixture solution, which composition are \( \text{H}_2\text{SO}_4 \) (18.4 mol/l) and \( \text{KMnO}_4 \), at room temperature for 4 h. Then it is cleaned with dilute \( \text{H}_2\text{SO}_4 \), \( \text{H}_2\text{O}_2 \), \( \text{H}_2\text{O} \), and acetone under ultrasonic oscillation in sequence.

Differential scanning calorimetry (DSC) is used to calculate the crystallinity and crystal surface energy. Each kind of sample is tested for three times and then the average value is used. The crystallinity of high-molecular-weight polymer can be determined as follows:

\[
X_c = (\Delta H / \Delta H_{100}) \times 100\%
\]

where \( \Delta H \) and \( \Delta H_{100} \) are melting enthalpy of sample and fully crystallised (J/g). In this paper, \( \Delta H_{100} = 287.9 \) J/g.

The crystal surface energy can be calculated with Thompson–Gibbs formula [18, 19]

\[
\sigma = \frac{2 \varepsilon \rho}{\Delta H_{100} L_B}
\]

where \( T_m \) and \( T_{m0} \) are the melting temperature of crystal and infinite crystal (K), \( \sigma \) is crystal surface energy (J/m²), \( \Delta H_{100} \) is crystal melting enthalpy (J/m³), and \( L_B \) is thickness of plate crystal (nm). In this paper, \( T_{m0} = 414.6 \) K, \( \Delta H_{100} = 2.88 \times 10^3 \) J/m³, and \( L_B = 10 \) nm.

X-ray diffractometry is used and the interplanar distance is calculated with Bragg diffraction equation

\[
2 \varepsilon \sin \theta = n \lambda
\]

where \( d \) is interplanar distance (nm), \( \theta \) is diffraction angle (°), \( n \) is the diffraction order, and \( \lambda \) is wavelength (nm). Similar to DSC, this measurement also repeats for three times to obtain the average value. In this paper, \( \lambda = 0.15418 \) nm.

2.2.3 AC breakdown strength: The sample is 100 mm × 100 mm × 0.5 mm and put into a cylindrical electrode system with 25 mm diameter, shown in Fig. 2. Then it is immersed into the 25# transformer oil at room temperature. Take 20 samples for AC breakdown test with the rising voltage rate of 1 kV/s. The breakdown voltage is recorded. The sample is wiped clean and the thickness of the breakdown point is measured for three times to take the average value. The AC breakdown strength is calculated according to the Weibull distribution [20]:

\[
F(y) = 1 - \exp \left( - \left( \frac{y}{\alpha} \right)^\beta \right)
\]

where \( y \) is the breakdown strength, \( \alpha \) indicates the breakdown strength when the failure probability is 63.2%, and \( \beta \) represents the dispersion of the test data. The confidence bound is 95% in this paper.
3 Results and discussion

3.1 Radial distribution of crosslinked morphology

During the polyethylene crosslinked, dicumyl peroxide (DCP), which is used as the crosslinked agent, breaks down into two active free radicals. The free radicals capture hydrogen atoms from the polyethylene molecules then activate the chains. It makes chains binding to each other to form carbon-carbon crosslinked bonds, resulting in forming XLPE with three-dimensional structure. The crosslinked process is as follows. In general, the crosslinked degree and crosslinked byproducts can characterise the crosslinked morphology.

3.1.1 Crosslinked degree: The radial distribution of crosslinked degree is shown in Fig. 3. The crosslinked degrees of the cable bulk, cable–joint transition, and factory joint samples are all revealed to be lower in the inner insulation but higher in the outer insulation. It might be that the outer insulation is heated at a higher temperature and for a longer duration than the inner insulation during vulcanisation. These factors make DCP fully decompose to crosslink the polyethylene. This phenomenon is more pronounced in cables with greater insulation thicknesses because temperature difference at inner insulation and outer insulation is larger.

Compared with the cable bulk, there is relatively high in cable–joint transition in terms of crosslinked degree, especially in the inner and middle insulation. However, it is not obvious. In comparison with the cable bulk and cable–joint transition, the crosslinked degree of the factory joint varies markedly in radial direction. The crosslinked degree of outer insulation is about 2.3% higher than that of inner insulation. The cause of this phenomenon may have been the contrast between the vertical continuous vulcanisation and injection moulding vulcanisation.

The higher the crosslinked degree is, the more tertiary hydrogen is contained in the molecular structure. Tertiary hydrogen is more reactive than parahydrogen, which exists in PE. So, it leads to the self-accelerating oxidation and chains break at the crosslinked point more likely. In view of the long-term operation condition of the cable line, the section with prominent crosslinked degree would undergo thermal and oxygen ageing earlier, which leads to degradation in the insulation. This is regarded as a defect and becomes the initiation of insulation discharge, thus causing irreversible damage to cable insulation. Therefore, a uniform distribution of the crosslinked degree, rather than prominent variations, is preferred in cable insulating materials.

3.1.2 Relative contents of crosslinked byproducts: The relative contents of three crosslinked byproducts are shown in Fig. 4. The relative content of α-methyl styrene, acetophenone, and cumyl alcohol do not vary greatly along the radial direction. The reason may be that the byproducts mainly diffuse outward through the insulation shield. It may also be the case that the contents of α-methyl styrene and cumyl alcohol are lower or the mass diffusion coefficient of acetophenone is higher in XLPE.

In comparison with the cable bulk, there are slight increases in the contents of all three byproducts in the cable–joint transition. It indicates that a small amount of DCP in the cable–joint transition decomposes after undergoing the factory joint insulation recovery. There may be re-vulcanisation occurs in cable–joint transition insulation. Owing to the difference between the vulcanisation...
3.2 Radial distribution of crystal morphology

The XLPE molecular chains constitute a three-dimensional network, which is disordered at high temperatures. As it cools, some chains remain disordered and become amorphous, while others form partially ordered regions and are arranged into plates. The chains in an ordered region are neither completely crystalline nor completely amorphous, and it can adopt a semi-crystalline state because of entanglement. Differences in temperature and cooling rate will affect the morphology of amorphous and crystalline regions. The grain size, crystallinity, crystal integrity, and crystal interplanar distance can be used to characterise the crystal morphology.

3.2.1 Grain size distribution: The spherulite surface of XLPE sample is treated with the etchant mixture solution. In order to determine the grain size, images are taken randomly. The number of valid unit cells is at least 100 to ensure the statistical accuracy. The results are shown in Fig. 5.

In the samples, the grains at inner insulation are larger and their distribution is scattered, but the grains at outer insulation are smaller and their distribution is clustered. The reason for this may be that the temperature decreasing rate at inner insulation is tardiness, and hence there is plenty of time for macromolecule chains to be arranged in order to form plate crystals. It can result in a larger spherulite. At the same time, short molecular chains that do not participate in crosslinked will also have formed into small spherulites if the cooling duration is long enough. The fastest temperature drop occurs at outer insulation, and hence most of the grains are small and the short chains are not involved in spherulite formation because of lack of time.

Compared with the cable bulk insulation, the grain size of cable–joint transition is reduced, no matter at inner, middle, and outer insulation. It can be obtained from the curves in Fig. 5b which shift to small size. Spherulites in factory joints are generally small, particularly at outer insulation.

3.2.2 Crystallinity distribution: The crystallinity distribution is shown in Fig. 6 and the standard deviation is listed in parentheses. In cable bulk, there is 0.22% discrepancy between inner and outer insulation. The crystallinity of cable–joint transition decreased after factory joint insulation recovery. The crystallinity of cable–joint transition insulation at inner, middle, and outer insulation decreases by 4.69, 2.80, and 4.03%, respectively, in comparison with the cable bulk. This may be that not only the primary crystalline grains, but also small grains and imperfections would melt, in the process of factory joint vulcanisation. When it cools, partial recrystallisation occurs, but the small grains formed in degassing no longer participate in crystallisation. For another, the network structure of XLPE does not facilitate the ordered folding.
Table 1 Melting temperature $T_m$ (K) at axial insulation positions

| Position | Cable bulk | Cable–joint transition | Factory joint |
|----------|------------|------------------------|---------------|
| inner    | 377.98     | 378.57                 | 378.56        |
| middle   | 379.13     | 379.22                 | 378.91        |
| outer    | 379.22     | 379.55                 | 379.10        |

Table 2 Values of crystal surface energy $\alpha_e$ ($10^{-3}$ J/m$^2$) at axial insulation positions

| Position | Cable bulk | Cable–joint transition | Factory joint |
|----------|------------|------------------------|---------------|
| inner    | 127.19     | 125.14                 | 125.18        |
| middle   | 123.20     | 122.88                 | 123.96        |
| outer    | 122.88     | 121.74                 | 123.30        |

Table 3 Interplanar distance (nm) between {110} plane in level 1 diffraction

| Position | Cable bulk | Cable–joint transition | Factory joint |
|----------|------------|------------------------|---------------|
| inner    | 0.418      | 0.420                  | 0.421         |
| middle   | 0.419      | 0.420                  | 0.421         |
| outer    | 0.420      | 0.420                  | 0.420         |

Table 4 Interplanar distance (nm) between {200} plane in level 1 diffraction

| Position | Sample | Cable–joint transition | Factory joint |
|----------|--------|------------------------|---------------|
| inner    | 0.380  | 0.381                  | 0.381         |
| middle   | 0.381  | 0.381                  | 0.383         |
| outer    | 0.380  | 0.381                  | 0.381         |

of molecular chains, when crosslinked degree of the cable–joint transition increases after insulation recovery, as shown in Fig. 3.

The crystallinity of factory joint is quite different from that of cable bulk. In comparison with the cable bulk, the crystallinity of factory joint reduces, with droop rate manifesting 5.63, 8.43, and 5.02% at inner, middle, and outer insulation, respectively. This is caused by the difference in cooling process and non-uniform crosslinked degree.

3.2.3 Crystal surface energy: The crystal surface energy ($\sigma_e$) signifies free energy per unit area. During the polymer crystallisation, the regular arrangement of macromolecular chains tends to reduce the crystal surface energy [16]. The magnitude of $\sigma_e$ reflects the crystal perfection. In theory, the lower the value of $\sigma_e$ is, the higher the crystal perfection, stability, and melting temperature are; the higher the value of $\sigma_e$ is, the lower the crystal perfection, stability, and melting temperature are.

The melting temperature can be determined from DSC curve, as shown in Table 1. The crystal surface energy can be calculated by (4), as shown in Table 2. The crystal surface energy is higher at inner insulation than outer insulation, in other word, the crystal integrity of inner insulation is inferior to that of outer insulation. This may occur because the grains are smaller and more numerous at outer insulation but more tightly bound at inner insulation. In comparison with cable bulk, the interplanar distances between {110} plane and {200} plane are reduced by 1.61, 0.26, and 0.93%, respectively. That is, the {110} plane are more loosely bound at outer insulation but more tightly bound at inner insulation. In comparison with cable bulk, the interplanar distances between {110} plane and {200} plane in the cable–joint transition are greater. The reason may be the melting of whiskers or defects in the factory joint insulation recovery. On the other hand, there are no significant variations in the interplanar distances in cable–joint transition. The interplanar distances in factory joint are greater than those in cable bulk. Considering the units used (nm), the effect of factory joint insulation recovery on the interplanar distances in cable–joint transition still needs further study.

3.3 AC breakdown strength

The AC breakdown strength is shown in Fig. 7. The AC breakdown strength of factory joint is slightly lower than that of cable bulk.

For convenience of comparison, the average values of inner, middle, and outer insulation results are taken, as shown in Table 5. There are obvious differences among cable bulk, cable–joint transition, and factory joint, in terms of crosslinked morphology and crystal morphology. In comparison with cable bulk, there are greater contents of $\alpha$-methylstyrene, acetophenone, and cumyl alcohol. It may make the permittivity increase. With regard to the cable–joint transition, re-vulcanisation may create interfaces between crosslinked networks formed in different processes. These would increase the dielectric loss and heating during the electron transport process and further accelerate the occurrence of AC breakdown.

Moreover, in cable–joint transition and factory joint, the grain size, crystallinity, and crystal surface energy are reduced and the interplanar distance increases. In other words, the crystal morphology of cable–joint transition and factory joint change to a sparse distribution, manifesting the arrangement of molecular chains in plates, and imperfect, defective, and small grains melted. All these result in wider grain boundaries among crystalline grains. Usually, the free path of electrons is longer, and the energy produced is greater, in an amorphous region than in a crystalline region. In the case of electron collisions, it is easy to break molecular chains, destroy the insulating structure, and form conductive channels, which causes breakdown. Hence, AC breakdown preferentially occurs in amorphous or imperfectly crystalline regions [21, 22]. All these results would lead to a reduction in the AC breakdown field strength.

In factory joints with large insulation thickness (15–30 mm), cable manufacturers usually adopt the following two methods:

(i) Layered extruded vulcanisation: The production period is prolonged and the thermal history of XLPE is inconsistent, due to multiple heating. It would affect the uniformity of insulating material performance.

(ii) One-step extruded vulcanisation: During the pressurisation and vulcanisation, the crosslinked byproduct methane is not easy to dissolve out, and it is easy to cause holes in insulation.

Therefore, it is key to control the vulcanisation conditions in factory joint insulation recovery.
On the other hand, in the fabrication process of factory joint insulation recovery, the temperature is usually about 300°C, and the extrusion vulcanisation rate is much lower than that of vertical continuous vulcanisation. All these lead to the change of cable–joint transition performance, which can be seen from the results.

To ensure the vulcanisation quality of factory joint insulation recovery and the uniformity of thermal history at inner, middle, and outer insulation, it is suggested to reduce the vulcanisation temperature, increase the vulcanisation pressure, and extend the vulcanisation reaction time. The changes of technological conditions can make it consistent basically in radial distribution in terms of crosslinked degree, crystallinity, and grain size, so as to ensure that the dielectric properties would not significantly vary.

4 Conclusions

In comparison with the cable bulk, the difference of morphology in cable–joint transition insulation is obvious. It appears as an increase in the content of crosslinked byproducts and a reduction in grain size, crystallinity, and crystal surface energy. Moreover, the AC breakdown strength of the cable–joint transition is lower. The reason for this might be that re-vulcanisation occurred in the cable–joint transition during the factory joint insulation curing process. The relative contents of crosslinked byproducts in factory joint insulation are notably higher than those in cable bulk, and the crosslinked degree is more variable. Owing to the differences in heating and cooling processes, the grain size, crystallinity, and crystal surface energy are lower. The AC breakdown strength in factory joint insulation is lower than that in cable bulk.

For factory joint with large insulation thickness, it is recommended to adopt low temperature and extended vulcanisation time to ensure the consistency with dielectric performance of the cable bulk. In addition, more attention should be paid to the temperature control of cable–joint transition.

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