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Influence on Aggregation Structure Changes of Retired Cross-linked Polyethylene (XLPE) Cable Insulation under Pre-qualification Test

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Influence on Aggregation Structure Changes of Retired Cross-linked Polyethylene (XLPE) Cable Insulation under Pre-qualification Test

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Abstract. In order to investigate the cross-linked polyethylene (XLPE) insulation characteristics of a retired high-voltage cable that has been in operation for 16 years and to evaluate the reliability of reusing it for practical operation, a 180-day pre-qualification test method was applied to a section of this cable to inspect the changes of aggregation structure. In this paper, the electric field and the thermal field of the inner, medium and outer positions of the insulation under the accelerated aging test were analyzed and converted to the corresponding equivalent condition at the surface of the conductor core. It can be found that equivalent testing conditions of the medium and outer positions are close to the cable practical operation condition. The diagnostic measurements of fourier transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC) and X-ray diffraction (X-ray) were conducted to analyse the aggregation structure of the samples. The results showed that the molecular chain of the inner position of the cable insulation was damaged and the crystalline structure was slightly degraded under high-strength test condition. On the contrary, the crystalline structure of medium and outer positions were both improved after the accelerated aging test because of the reduction of the impurities and the annealing effect. So it can be expected that the rest of this retired cables still have the potential to reuse in long-term actual operation condition.

1. Introduction

Cross-linked polyethylene (XLPE) has been widely used for cable insulation since the 1980s due to its excellent performance in electrical and thermal properties [1~3]. In recent years, a growing number of of high-voltage cross-linked cables are decommissioned because of transmission line relocation or design service life limitation. In the face of the situation that lacks extensive experimental research on reusing retired cables for practical operation, They are dismissed to be a waste. In fact, a large quantity of experimental studies on retired cables indicate that relevant parameters still comply with quality standards and still meet the requirements for continuous operation [4~6].

It is well evidenced that long-term electric field and thermal field can change the morphology of XLPE insulation deeply and lead to the degradation of the insulation [1]. In the actual operation of the cable, the insulation is influenced by many factors. So the aging process of the insulation is complicated and the insulation properties can be enhanced in some certain situations. The reasons can be simply summarized as two points: the thermal effect and electric field effects [7~8]. The thermal

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effect is mainly reflected in the secondary reaction of cross-linking and the re-growth of the crystalline structure [9]. The electric field effect is mainly reflected in the mobility of impurities under the electric field. Therefore, analysis on electric and thermal field plays a decisive role in the aging process of high-voltage cross-linked cables.

The purpose of pre-qualification test is to investigate the long-term safety and reliability of AC cables. It is widely used in extrusion insulation and accessories with rated voltage of 150-500 kV [10]. In this paper, the pre-qualification test method is applied to a 110 kV high-voltage cross-linked retired cable that has been in operation for 16 years, and a 180-day electrothermal cyclic test is carried out under high-strength conditions. This paper aims to explore the XLPE insulation characteristics changes of the retired cable and to evaluate the reliability of reusing it for practical operation.

2. Experimental

2.1. Preparation of XLPE Samples

A section of 110 kV high-voltage cross-linked polyethylene cable that had been in operation for 16 years was selected as the testing sample. It can be observed that the cable sheath and the insulating layer were not damaged, and there was no water permeation before the accelerated aging test. The configuration and critical parameters of the cable are shown in Figure 1 and Table 2, where the terms of Inner, Medium and Outer are defined as the inner, medium and outer position of the cable insulation, respectively.

Table 1. Critical parameters of the cables.

Sample	**Voltage Level (kV)**	**Conductor Area (mm^2)**	**Conductor Diameter (mm)**	**Insulation Diameter (mm)**	**Manufacturer**	**Operation Years**
Parameter	110/63.5	630	32	68	Showa	1999-2015

2.2. Pre-Qualification Test Method Setup

A 180-day pre-qualification test was performed on a test cable with the length of 15 m. A high voltage of 1.7 U_0 (108 kV, U_0= 63.5 kV) was applied to the cable, and the current was applied to the cable metal conductor. The insulation was heated for at least 8 h to the preset temperature of 90 ~ 95 °C and kept for 2 h, then cooled to the room temperature at least for 16 h during the test. No insulation breakdown phenomenon occurred during the accelerated aging test. So we can get the untreated samples and accelerated aging samples in different positions.

2.3. Analysis of Electric Field and Thermal Field under the Pre-Qualification Test

The calculation of the electric field distribution in cable insulation can be deducted by Gauss's theorem. The specific Formula is as follows:

$$E = U \cdot [r \cdot Ln\left({R_O}/{R_I}\right)]^{-1} \tag{1}$$

Where U is the conductor voltage, E is the electric field strength from the core of radius of r, R_I and R_O are the outer diameters of the inner and outer boundaries of the closed cylinder. The calculated values are as follows: U=1.7 U_0=108 kV, R_I=16 mm, R_O=34 mm.

The electric field distribution in the inner, medium and outer positions of the cable insulation can be obtained by the parameters of Table 1 and Formula (1). The results of each insulation layer are as follows: E_I=8.95 kV/mm, E_M=5.73 kV/mm and E_O=4.21 kV/mm.

Through parallel cable in the simulated circuit, the temperature curves of the inner and outer position of the insulation during the accelerated aging test were obtained by the thermo-couple sensors. The thermal circuit model was used to calculate the thermal field distribution of the medium position of the insulation. The temperature curve in each position was profiled in Figure 2.

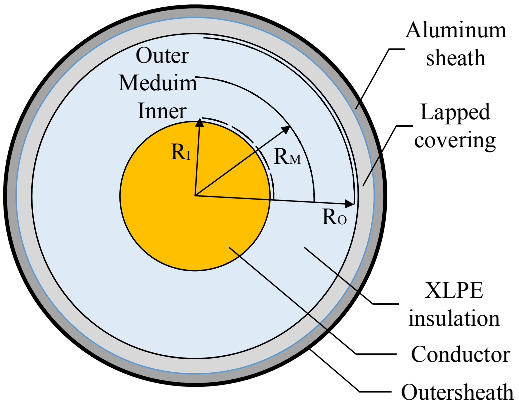

Figure 1. Configuration of the testing cable. R_I, R_M and R_O are the corresponding radius of the inner, medium and outer insulation layer.

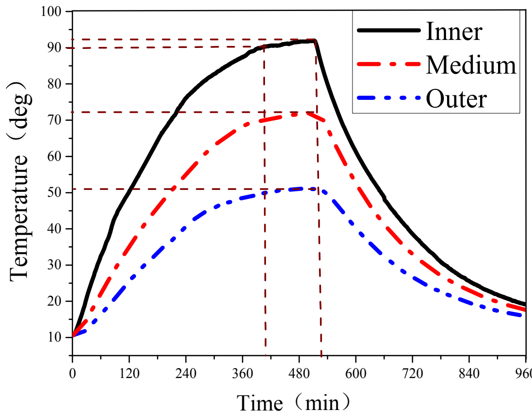

Figure 2. Temperature curve in each position of testing cable(one cycle).

Based on the results of distribution in electric field strength and the thermal field in different positions, a reverse derivation can be proposed to convert the accelerated aging testing condition of the inner, medium and outer insulation layer into corresponding equivalent condition at the surface of the conductor core. Formula (1) is exerted reversely to obtain the equivalent voltage level in each position, which means that E_x $(x = I, M \text{ or } O)$ and R_I are substituted into E and r to obtain the corresponding voltage level of each insulation layer. In addition, each insulation layer can be equivalent to a thermal cyclic aging test of 90~95 ° C, 70~75 ° C and 50~55 ° C from Figure 2. The equivalent accelerated aging test conditions of each position of the insulation are shown in Table 2. We can notice that the equivalent accelerated aging test conditions in medium and outer positions are close to the cable practical operation condition.

Table 2. Equivalent accelerated aging test condition of each position of the insulation.

Sample	**Voltage level (kV)**	**Temperature (°C)**
Inner	1.70 U_0	90~95
Medium	1.08 U_0	70~75
Outer	0.80 U_0	50~55

2.4. Diagnostic Measurements
Fourier transform infrared spectroscop (FTIR), X-ray diffraction experiment and differential scanning calorimetry (DSC) measurement were adopted to analyse the aggregation structure changes of each sample after accelerated aging test.

Micro-structure changes in each sample were performed by VERTEX 70 infrared spectrometer manufactured by German. Each sample was tested at 32 scans in the range of 600~3600 cm^{-1}.

Thermal properties of each sample were analysed by DSC NETZSCH-DSC 214 instrument manufactured by German. 5 mg samples were prepared for the test with the program of a heating phase of 30~140 °C and a cooling phase of 140~30 °C at a constant rate of 10 °C/min under N_2 atmosphere.

Aggregation structure changes of each sample were analysed by Bruker D8 ADVANCE X-ray diffractometer manufactured by German. The working interval of Bragg angle was 2θ=5~90° by step of 0.131°.

3. Result and Discussion
Samples were sliced radially from untreated and accelerating aged cable. We defined the samples of inner, medium and outer position of the untreated cable insulation as Inner-1, Medium-1 and Outer-1,

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while samples of the inner, medium and outer position of the accelerating aged cable insulation as Inner-2, Medium-2 and Outer-2.

3.1. Result and Discussion of FTIR Spectroscopy Measurement

XLPE is a high molecular polymer whose methylene groups(-CH_2) are the most characteristic group of XLPE. This absorption peak appears at a wavelength of 720 cm^{-1}, and it is confirmed by the peaks that locate at 1471 cm^{-1}, 2856 cm^{-1} and 2937 cm^{-1}. Dicumyl peroxide (DCP) is commonly used as cross-linking agent and its by-product mainly consists of acetophenone, cumyl alcohol and α-methylstyrene. The corresponding characteristic groups of these compounds are phenylenyl, carbonyl (=C=O) and hydroxyl (-OH), and the infrared absorption peaks appear at 1600 cm^{-1}, 1680 cm^{-1} and 3371 cm^{-1}, respectively. In addition, absorption peaks ranging from 1700 cm^{-1} to 1800 cm^{-1} can be considered as the thermo-oxidative products. Carbonyl index and double band index were chosen for research. The indexes are shown in Table 3.

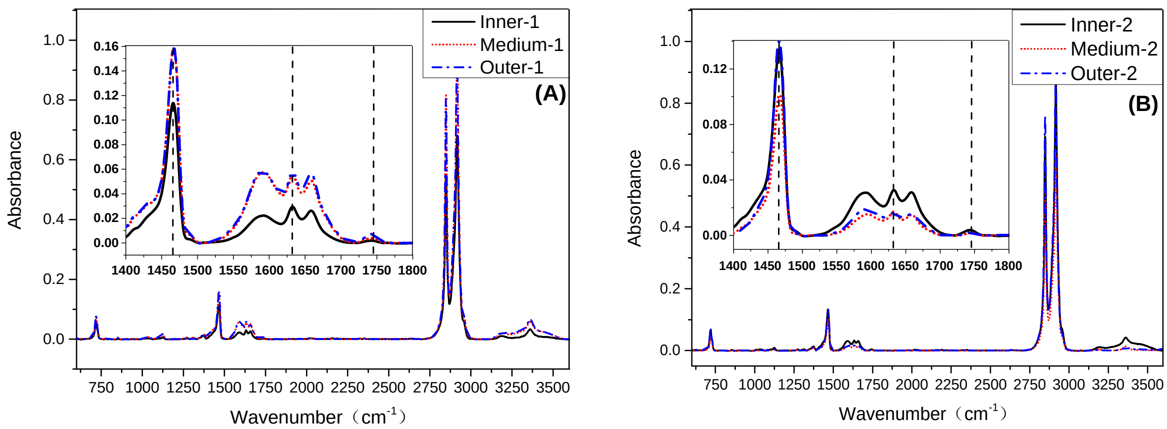

Figure 3. FTIR spectrum of each layer before and after the accelerated aging test.

In Figure 3 (A), it is clearly observed that obvious absorption peaks appear at 1600 cm^{-1}, 1680 cm^{-1} and 3371 cm^{-1} in Medium-1 and Outer-1. It indicates that a large quantity of cross-linking by-products remain in the medium and the outer insulation layer. As to the inner position, thr absorption intensity of these three peaks are the lowest among the samples. It can be inferred that the insulation layer near the conductor core of the cable is subjected to a stronger thermal and electric field effect, so that the inside impurities can be reduced by evaporation or migration in the actual operation of a cable.

From Figure 3 (B) and Table 4, it can be found that the index of the two groups in each position changed greatly after the accelerated aging test. The decrease in absorbance of phenylenyl, carbonyl and hydroxyl group in the medium and the outer layer insulation signifies the decreases in the cross-linking by-products after the accelerating aging test. We can also observe that both carbonyl index and unsaturated band index in the medium and outer positions decrease after accelerated aging test, which means the process of oxidation degradation did not happened basically under the corresponding aging test condition. On the contrary, groups of thermo-oxidative products in the inner position increase slightly after the accelerating aging test, which shows that a certain oxidation degradation happened and leads to the damage in molecular chain and the formation of more oxidation polar groups and broken chains.

3.2. Result and Discussion of DSC Measurement

With the results of DSC measurement, two phases were analyzed, including the heating and cooling phase, which can be used to analyze crystalline structure and the ability of recrystallization of the samples under different equivalent accelerated aging test. Figure 4 (A) and (B) show the heating and cooling phase thermograms of each sample. The specific parameters are obtained or calculated and listed in Table 3, where T_m is melting temperature, ΔH_f is enthalpy of fusion, T_c is the crystallizing temperature and ΔH_c is the enthalpy of crystallization.

After the accelerated aging test, the aggregation structure of each insulation layer has changed greatly. As is shown in Figure 4 (A), a sharp endothermic peak that symbolizes the process of crystalline structure melting, has emerged in all samples. The heat flow curve of the inner position drifts slightly to lower temperature and the melting range of the main DSC peak becomes wider. This phenomenon is ascribed to the generation of short chain segment, resulting from the broken of XLPE macromolecules under the severe testing condition. As to the medium position, it can be figured out the melting range of the main melting peak is greatly narrowed and the melting enthalpy is obviously increased, even though the main melting peak moves towards lower temperature. Therefore, the crystalline structure becomes uniform and the crystalline area is enlarged because of the adequate chain's mobility and impurities' reduction under the equivalent testing condition. Similarly, the crystalline structure of the outer position becomes more uniform, reflecting as the narrowing of the melting range and higher melting peak in DSC thermogram.

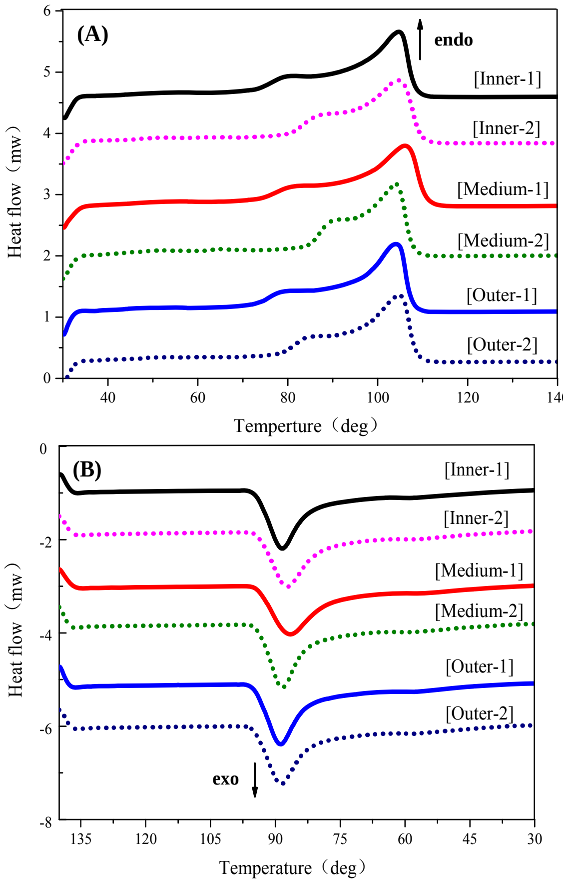

Figure 4. DSC thermogram of each layer before and after the accelerated aging test.

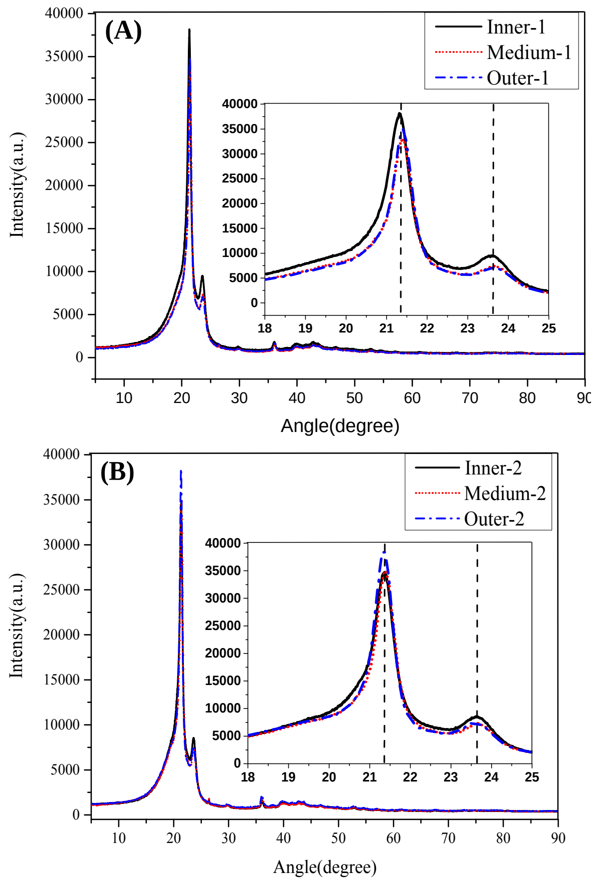

Figure 5. X-ray spectrum of each layer before and after the accelerated aging test.

In Figure 4 (B), an exothermic peak which characterizes the process of recrystallization of the polymer was observed in all the samples. As to the inner position, a slight displacement of the exothermic peak towards lower temperatures and the lifting of supercooling degree ΔT indicate that certain damage in the XLPE macromolecules leading to decline of the ability to recrystallization and degradation in the crystalline structure under the severe equivalent testing condition. As to the medium position, the heat flow curve shifts to higher temperature, exothermic peak area is enlarged and the crystalline rate is quicken. The phenomenon is attributed to integrity and regularity of the XLPE macromolecules and decreasing of impurity molecules. Therefore, the equivalent testing condition promotes the re-growth of the crystal in a period of time. As to the outer position, the recrystalline strength and the form of crystal remain as the same comparing to the untreated one, which demonstrates that the equivalent testing condition hardly causes damage to the aggregation structure in the process of accelerated aging test.

3.3. Result and Discussion of X-ray Measurement

The change of the aggregation structure of each sample before and after the accelerated aging test was analyzed by X-ray diffractometer, to observe the changes in the crystal phase of each layer under the equivalent testing condition.

Figure 5 displays the X-ray spectrum of each position in cable insulation before and after the accelerated aging test. It can be observed that two main crystalline peaks of each sample appear at 2θ=21.22° and 2θ=23.63°, which correspond to the (110) and (200) lattice planes. There is no excessive displacement in the position of the peaks or in their splitting among the samples, but the intensity and the shape of the peaks are different. It is indicated that accelerated aging hardly produces any new crystalline phase in the aggregation structure, but results in the changes in the crystallinity and the grain size of the samples.

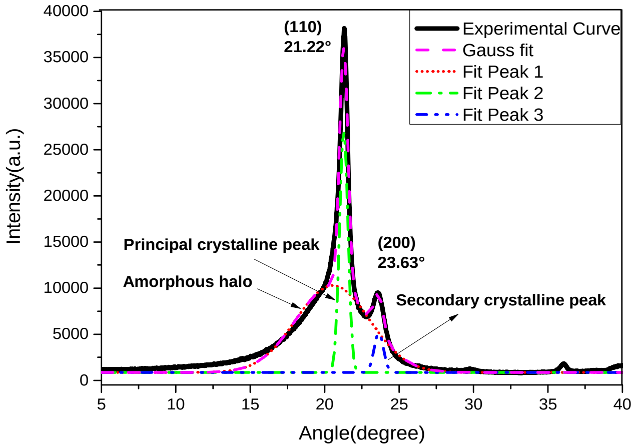

Figure 6. Gaussian fitting of the crystalline peaks and the amorphous halo by Hinrichsen method.

The crystallinity percentage can be calculated by Hinrichsen method. The X-ray spectrum of each sample is fitted by Gaussian functions. Figure 6 displays the corresponding three Gauss fit peaks obtained by using the original 9.1. The calculating process is given as follows:

$$\chi(\%) = [(S_2 + S_3)/(S_1 + S_2 + S_3)] \times 100 \quad (2)$$

Where χ (%) is crystallinity percentage, S_1 is the area of the amorphous halo, S_2 is the area of the main crystallization peak at 2θ=21.22° and S_3 is the area of the secondary crystallization peak at 2θ=23.63°.

Table 3. Parameters obtained from FTIR, DSC and X-ray spectrum of each layer.

Sample	Carbonyl index	Unsaturated band index	T_m (°C)	ΔH_f (J/g)	T_c (°C)	ΔH_c (J/g)	χ (%)
Inner-1	0.017	0.26	104.7	110.4	88.4	-101.2	27.42
Medium-1	0.025	0.35	106.2	114.6	86.5	-103.4	25.75
Outer-1	0.049	0.36	104.1	114.6	88.8	-106.0	27.48
Inner-2	0.030	0.25	104.6	111.9	87.2	-103.3	26.04
Medium-2	0.019	0.18	104.0	116.5	88.5	-104.2	26.19
Outer-2	-105.0	16.2	104.8	114.2	13.6	88.6	28.84

Basing on the formula (2), we present the crystallinity percentage in Table 3. Before the accelerated aging test, the damage to molecular chain and the deterioration of each position are slight in the actual operation condition. The crystallinity and the grain sizes are mainly affected by the content of impurities. For the inner position, the crystallinity is high, indicating that the spherulites are tightly distributed in the insulation and the crystalline structure is relatively perfect due to the suitable cable operation condition. With regard to the medium position of the insulation, the grain sizes of the main crystal and the secondary crystal are larger but the crystallinity is relatively low. That means that spaces among the crystalline structures are expanded and the integral crystalline structure is not closely arranged. This can be attributed to the large dispersion of impurities and crystalline structure. As to the outer position of the insulation, the crystalline structure is compact with the fact of high crystallinity because the influence of the electric field and the thermal field are weak during the actual operation.

The variation of crystalline structure among the samples is enlarged after the accelerated aging test. The inner position of the insulation is subjected to higher electric field and thermal field, and the crystallinity is decreased. The backbone of molecular has been damaged to some extent, leading to appearance of a certain of small molecular chains and free groups. It can also account for the increasing of secondary crystals. On the other hand, the increasing of the crystallinity in the medium and outer positions of the insulation reflect the homogenization and tightness of crystalline structure. It can be inferred the phenomenon is caused by reduction of impurities and annealing effect [6] under the corresponding equivalent testing condition.

4. Conclusion

Relevant diagnostic measurements were adopted to analyse the changing trend of the aggregate structure of the samples after the accelerated aging test, so as to evaluate the cable insulation condition effectively. Following conclusions can be made:

1) Under the severe condition of accelerated aging of 1.70 U_0 and 90~95 °C, a slight of degradation happens in the crystalline structure of the inner position of the insulation due to the increasing of broken molecular chains and free polar groups;

2) Under the condition of accelerated aging of 1.08 U_0 and 70~75 °C, the crystalline structure of the medium position of the insulation becomes more compact and uniform with the fact of high crystallinity and decreasing of secondary crystals in the insulation;

3) Under the condition of accelerated aging of 0.80 U_0 and 50~55 °C , annealing effect promotes the movement of the chains and reduces the volatile impurities in the outer position of the insulation to generate a better crystalline structure.

After the comprehensive evaluation on the aggregation structure changes of each insulation layer, it can be expected that the rest of the retired cables still have the potential to reuse in long-term actual operation condition.

5. Acknowledgments

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