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Morphological, Structural, and Dielectric Properties of Thermally Aged AC 500 kV XLPE Submarine Cable Insulation Material and Its Deterioration Condition Assessment

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ABSTRACT Cross-linked polyethylene (XLPE) has been widely used as insulation material for cables. In 2019, the highest voltage level AC 500 kV XLPE submarine cable has been operated in Zhoushan, China. The irreversible degradation poses a safety hazard to the operation of submarine cable. Therefore, it is necessary to investigate the physical, chemical and electrical properties of XLPE submarine cable insulation under aging condition and explore the method for its deterioration degree assessment. In this paper, the AC 500 kV XLPE submarine cable insulation material was thermally aged at 130 °C. The deterioration characteristics of materials were analyzed based on morphology, chemical structure, mechanical property, thermal property and dielectric properties. Multiple characteristic parameters were extracted and new deterioration condition assessment model was established. Results show that the physicochemical characteristic parameters including retention rate of elongation at break, carbonyl index (*CI*) based on FTIR, full-width at half-maximum (FWHM) of the diffraction peak based on XRD, melting enthalpy based on DSC analysis, and the dielectric characteristic parameters including modified Cole-Cole model parameter $\chi_{s\alpha}$ and AC breakdown strength are sensitive to deterioration condition of XLPE material. According to the change of elongation retention at break, the deterioration condition of XLPE sample was divided into enhanced stage, wear-out stage and disposal stage. The new multi-factor deterioration condition assessment model including six characteristic parameters (retention rate of elongation at break, *CI*, FWHM, melting enthalpy, modified Cole-Cole model parameter $\chi_{s\alpha}$, AC breakdown strength) was successfully established by grey correlation analysis. The new model can reflect the property of mechanical, chemical, thermal and dielectric property and has prominent effect in differentiating deterioration degree of the AC 500 kV XLPE submarine cable insulation material.

INDEX TERMS Cross-linked polyethylene (XLPE), thermal aging, physicochemical properties, dielectric properties, condition assessment.

I. INTRODUCTION

Cross-linked polyethylene (XLPE) has outstanding physical and chemical properties [1]. The AC 500 kV power interconnection project in Zhoushan, Zhejiang province of China is currently the highest voltage and longest routing XLPE cable project throughout the world. During the service of cable, XLPE material will suffer from various stresses (electrical, thermal, mechanical, etc.), which result in the changes of its chemical composition, physical morphology and electrical properties [2]. The irreversible degradation poses a safety hazard to the operation of submarine cables. Therefore, it is

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necessary to investigate the physical, chemical and electrical properties of XLPE submarine cable insulation under aging condition and explore the method for its deterioration degree assessment [3].

Studies about physical and chemical properties of XLPE material under thermal stress reveal its degradation process. With the development of aging, the retention rate of elongation at break and the tensile strength showed a trend of decline after a slight increase [4], [5]. In terms of thermal performance analysis, differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA) X-ray diffractometry (XRD) and fourier-transform infra-red (FTIR) spectroscopy has been used [6], [7]. It was found that the crystallinity of the XLPE material decreases significantly and a large amount of carbonyl groups (-C = O-) were produced [1], [8]–[10]. The breakdown strength and endurance life under multi-stress also showed a downward trend with the development of thermal aging [11]–[13].

In addition, the dielectric properties of XLPE have received extensive attention in recent years [14], [15]. M. Nedjar compared the changes of dielectric constant and dielectric loss angle of XLPE under high frequency and it was found that the dielectric permittivity was less affected by the aging [16]. Chonung Kim et al studied the dielectric behavior of thermally aged XLPE cable in the high frequency range and suggested that dielectric loss tangent and ac resistivity can be considered as useful diagnostic parameters for estimating the deterioration degree of the thermally aged XLPE cable [17]. Mengyan Hu et al found that permittivity decreased firstly and then increased with the elongation of aging time and the dissipation factor increased [10]. Dongxin He et al found that the dielectric constant decreases with aging time in high frequency areas (10^3-10^6 Hz), while the dielectric loss factor increases with aging time at low frequency (10^{-2}-0 Hz) [5]. Pulong Geng et al measured the characteristic parameters (complex permittivity, leakage current, complex dielectric modulus) and found that the test temperature had an influence on AC leakage current, which mainly reflected in the low frequency region [18].

In recent years, scholars have proposed a variety of aging state criteria from the perspectives of mechanical, thermal, chemical structure and dielectric properties. The elongation at break retention rate of 50% is often used to indicate the XLPE material reaches its end life [19]. J. C. Fothergill et al suggested that the most likely candidates for aging markers, may include the change in inter-phase fraction using FTIR, changes in micro-void size distribution, quantitative chemical characteristics extracted from FTIR spectra [20]. However, these criteria are often based on a single performance change that is difficult to reflect the aging state of the overall performance. To analyze the aging degree of material from different physicochemical and dielectric properties, some researchers have reported the gray correlation, neural network methods [21]–[23]. These methods mainly used in the oil-paper insulation condition assessment, few researchers reported the XLPE multi-factor deterioration degree judgment method.

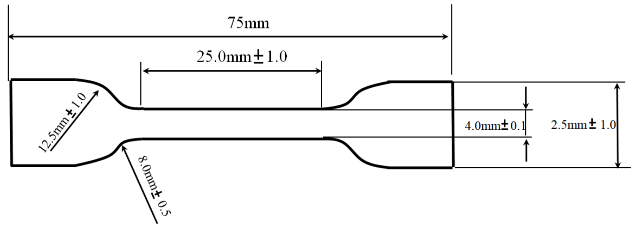


FIGURE 1. Dumbbell XLPE piece for tensile test.

For 500 kV submarine cable, although the insulation material is XLPE, the difference in raw material, manufacturing process will result in difference in the physical, chemical and electrical properties of the insulation material during the aging process. At present, few researchers reported the thermal aging of 500 kV XLPE submarine cable. It is necessary to perform the aging experiment on 500 kV XLPE submarine cable insulation material and explore its aging properties. In this paper, the physical, chemical and dielectric properties of the AC 500 kV XLPE submarine cable insulation material before and after the thermal aging was investigated. A new multi-factor deterioration degree judgment method based on the grey correlation analysis was introduced for the deterioration condition assessment of the AC 500 kV XLPE submarine cable insulation material. Firstly, the XLPE samples were aged at 130 °C and samples were taken for the mechanical, thermal, chemical structure and dielectric properties measurement. A modified Cole-Cole model was used to analyze the dielectric characteristic parameter. Secondly, changes of characteristic parameters of mechanical, thermal, chemical structure and dielectric properties were extracted and a multi-factor deterioration degree judgment method based on the grey correlation analysis was provided. Finally, the healthy threshold of the AC 500 kV XLPE submarine cable insulation material based on the thermal aging was proposed.

II. EXPERIMENTS

A. SAMPLE PREPARATION AND THERMAL AGING

The AC 500 kV XLPE submarine cable insulation material was provided by the Zhoushan Power Supply Company. 2 mm thick dumbbell XLPE specimens (FIGURE 1) were prepared for the test on retention rate of elongation at break and 1 mm thick rectangular piece was prepared for thermal, chemical structure and dielectric property tests. The 401-C air heat aging chamber was used for accelerated thermal aging and the aging temperature was controlled at 130 °C. Samples were taken out at 0-day, 40-day, 95-day, 125-day, 135-day and 160-day.

B. CHEMICAL STRUCTURE CHARACTERIZATION

The Nicolet IS5 FT-IR Infrared Spectrometer manufactured by Nicolet Inc. was used to measure the infrared spectrum of the samples with different thermal aging conditions. The spectrum range was 4000-500 cm^{-1} and the resolution was 4 cm^{-1}. The XRD patterns was obtained by an X-ray

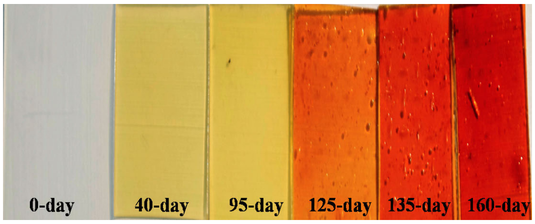


FIGURE 2. Surface morphology changes (color).

diffractometer (Panalytical Empyrea, Almelo, Netherlands),whose scan rate was 10°/min and a scan angle 2θ in the range of 5° to 90°.

C. TENSILE TEST

The Computer-controlled electronic universal testing machine (ZY-13) was used for the tensile test. The dumbbell shaped XLPE sample was vertically sandwiched between upper and lower fixture to ensure the specimen in the tensile process was under vertical tensile stress. The tensile speed was 25 mm/min. The experimental temperature was room temperature. The test was repeated three times on samples of each aging stage and the average value was taken as the final result.

D. THERMAL PERFORMANCE TEST

The United States TA Instruments Q200 DSC was used to obtain the recrystallization and melting curves of the aged samples. It was chosen as a heating-cooling-heating process. Firstly, the temperature was increased from room temperature to 120 °C to eliminate the effect of the thermal history of the sample. Then temperature was decreased to −40 °C and then increased to 120 °C. The increasing and cooling rate of temperature were 10°C/min under nitrogen atmosphere.

E. DIELECTRIC PROPERTY TEST

The XLPE samples were measured by the Concept80 broadband dielectric spectrometer of NOVOCONTROL Company. Samples were sandwiched tightly between the upper and lower electrodes with diameter of 40 mm. The measuring frequency ranges from 10^{-2}Hz to 10^{5}Hz under 70°C.The AC breakdown test was carried out by the Huace Insulation Breakdown Tester (HCDJC-100kV). The electrode used was a plate-plate electrode. The diameter of the electrode is 25 mm and the voltage boost rate is 1 kV/s. The samples of each aging stage were tested five times and the average value was analyzed.

III. EXPERIMETAL RESULTS AND DISCUSSIONS

A. SURFACE MORPHOLOGY CHANGES

As shown in FIGURE 2, three different changes can be observed. Firstly, the surface color changed from transparent white (0-day) to yellow (40-day/95-day). It shows dark brown with the development of aging (125-day/135-day/160/day). Secondly, the deterioration of the AC 500 kV XLPE submarine cable insulation material tended to spread from local part of samples to global parts. Material deterioration started from the edge of the sample and gradually developed into the interior (95-day). With the deepening of the overall color, the aging was obvious after thermally aged 125 days. As shown in FIGURE 2 and FIGURE 3, bubbles can be found in the severely deteriorated samples (135-day/160-day). The length of these bubbles is approximately ten micrometers, which are round, elliptical and irregular. More micro-bubbles and some big bubbles (>30μm) were observed in the severely aged samples. The samples before and after aging were broken in liquid nitrogen and observed by scanning electron microscope (SEM), as shown in FIGURE 4. It can be found that fresh samples have uneven cross sections and suspected crystal stacking can be observed, while the section of the sample aged 160 days is smooth and the crystal structure cannot be observed.

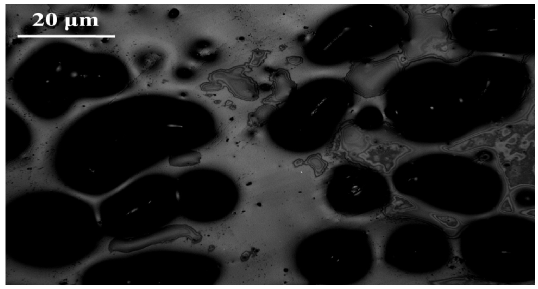
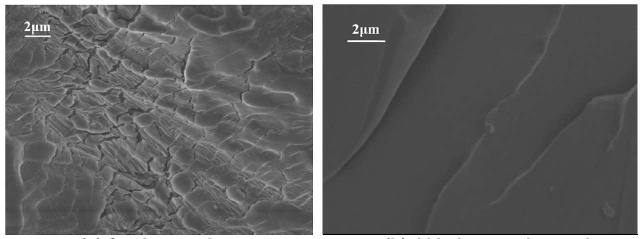


FIGURE 3. Bubbles (black shadows) in the severely deteriorated samples (135-day/160-day).



(a) fresh sample (b) 160-day aged sample

FIGURE 4. SEM before and after aging.

B. CHEMICAL STRUCTURE ANALYSIS

FTIR results are shown in FIGURE 5. The significant absorption peaks at wavelength of 2915 cm^{-1}, 2847 cm^{-1}, 1470 cm^{-1}, and 718 cm^{-1} are characteristic absorption peaks for -CH_2- in the polyethylene carbon chain [24]. The absorption peaks at wavelength of 1170 cm^{-1} and 1710 cm^{-1} are characteristic absorption peaks for -C = O-. The carbonyl peak of 0-day and 40-day is not obvious. However, it can be found obviously in the FTIR results of 95-day, 125-day, 135-day and 160-day samples.

Thermal oxidation of XLPE cable insulation samples can be evaluated by the carbonyl index (*CI*) [25]:

$$CI = I_{1720}/I_{2915} \tag{1}$$

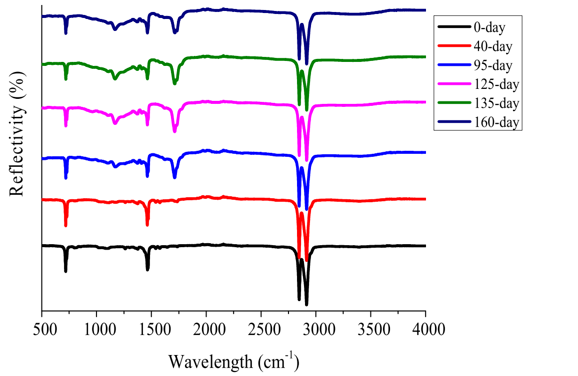


FIGURE 5. FTIR spectra of XLPE samples with different aging conditions.

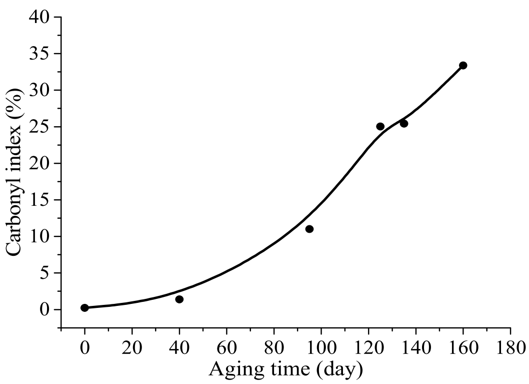


FIGURE 6. Diagram of carbonyl index of XLPE samples with different aging conditions.

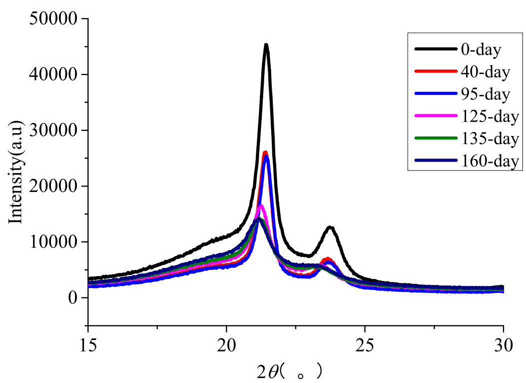


FIGURE 7. XRD spectra of XLPE samples with different aging conditions.

where I_{1720} is the intensity of the absorption peak at 1710 cm^{-1}, I_{2915} is the intensity of the absorption peak at 2915 cm^{-1}. FIGURE 6 shows the changes of *CI* with aging progress. It can be found that *CI* increases slowly from less than 5% of 40-day sample to more than 30% of 160-day sample. The *CI* results present that the thermo-oxidative aging process accelerated from the 95th day because the *CI* values increases rapidly after being aged 95 days.

The XRD results shown in FIGURE 7 present an amorphous hallo and two major crystalline peaks. With the development of aging process, the position of peaks moves to left slightly and no new peaks appears. This means that no new crystalline phase in the material structure produced. The intensity of the peaks decreases in the aging process, which means that the percentage of crystallinity decreases for the aged XLPE samples. The full-width at half-maximum (FWHM) of the diffraction peak was calculated from the XRD curves to characterize the inter-planar spacing and changes in grain size [26]. It can be found in FIGURE 8 that the FWHM decreases slightly in the beginning period and increases rapidly from less than 0.55 to more than 0.7 with the development of aging process, which means that the grain size increases slightly at the beginning of the aging, then decreases significantly with more small crystals produced. FIGURE 8 also shows that the crystal structure of the samples begins to change significantly after being aged 95 days.

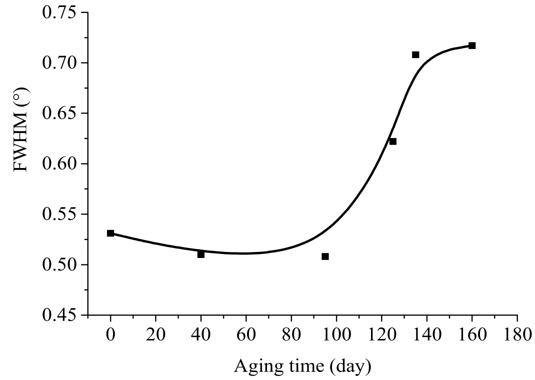


FIGURE 8. FWHM of XLPE samples with different aging conditions.

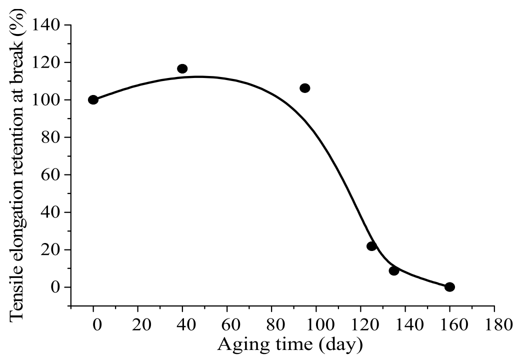


FIGURE 9. Changes in elongation retention at break of XLPE samples with different aging conditions.

C. TENSILE TEST ANALYSIS

It can be found in FIGURE 9 that the tensile property decreases approximately abruptly from 95-day samples to 125-day samples. At the beginning 40 days of aging, the tensile elongation retention at break increases gradually to about 120%, reaching its peak. After being aged for 95 days, the tensile elongation retention at break drops back to about 100%. Then the value decreases to about 20% at 125th day rapidly. From 135th day to 160th day, the value decreases gradually to about 0%. FIGURE 9 also shows the mechanical properties of the samples decreases seriously after being aged 95 days.

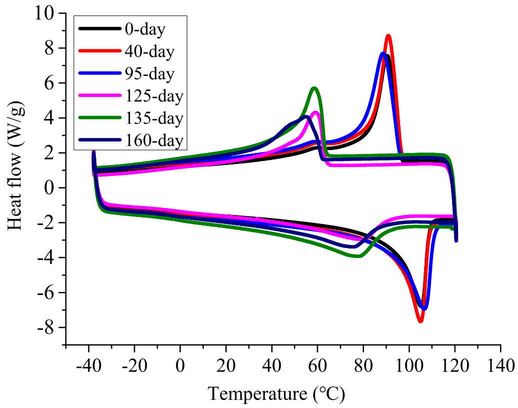


FIGURE 10. Changes in DSC of XLPE samples with different aging conditions.

TABLE 1. Results of DSC for different aged samlpes.

Sample	Glass transition temperature /℃	Melting enthalpy /J/g	Melting peak temperature /℃	Crystallinity /%
0-day	82.35	86.77	105.15	30.20
40-day	82.21	91.55	104.98	31.87
95-day	79.57	89.45	106.62	31.13
125-day	49.51	29.47	79.55	10.26
135-day	46.30	29.31	78.08	10.20
160-day	33.26	29.24	75.87	10.18

D. THERMAL PERFORMANCE ANALYSIS

DSC results for samples with different aging condition are shown in FIGURE 10. An endothermic peak and an exothermic can be observed. They correspond to the melting and crystallization temperature, respectively. TABLE 1 lists the glass transition temperature, melting enthalpy, melting peak temperature and crystallinity. The expression for the crystallinity calculation in Table 1 is:

$$Xc = {}^{\Delta H}/_{\Delta H_0} \tag{2}$$

where Xc is the degree of crystallinity, ΔH is a melting enthalpy, ΔH_0 is a melting enthalpy with a crystallinity of 100%, $\Delta H_0 = 287.3$J/g [27]. FIGURE 10 and TABLE 1 show that the melting and crystallization peak of XLPE changes obviously before and after aging. The peak shape broadened and the melting enthalpy decreases, indicating that the molecular structure of XLPE has been seriously deteriorated. On the other hand, the glass transition temperature of XLPE shifts toward the lower temperature, indicating that XLPE will be less heat-resistant after thermal aging. The results shown in FIGURE 10 and TABLE 1 present that the AC 500 kV XLPE submarine cable insulation material is more easily change from a glassy state to a highly elastic state in the thermal aging process.

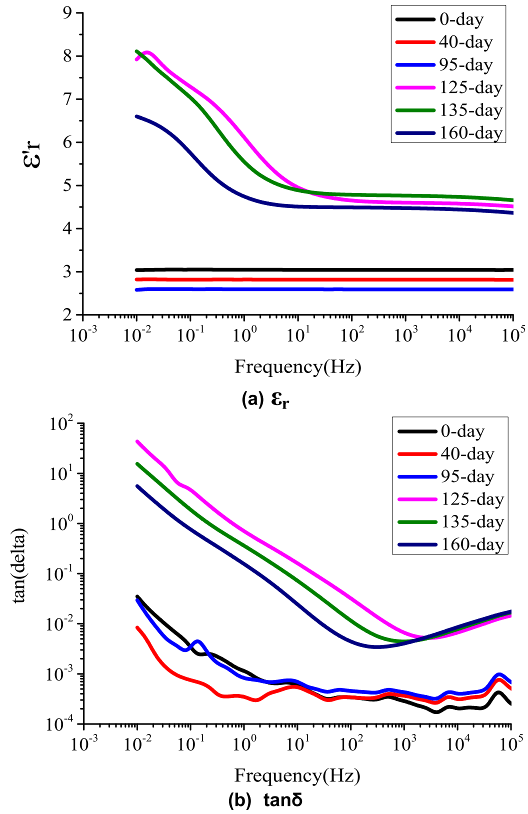


FIGURE 11. Changes in ε_r and tan δ at different aging conditions at 70 °C (a) ε_r (b) tanδ.

E. DIELECTRIC PROPERTY ANALYSIS

FIGURE 11 illustrates the changes in the $\varepsilon' r$ and tanδ of samples with different aging conditions under 70 °C. Firstly, Values of $\varepsilon' r$ and tanδ of severely aged samples are much higher than fresh samples or samples with slightly aging condition. Secondly, $\varepsilon' r$ and tanδ show the same changes at the same frequency. $\varepsilon' r$ and tanδ decrease slightly at the beginning of the aging progress (0-day, 40-day, 95-day). These characteristic parameters increases significantly when samples aged for 125 days. However, these parameters decreases slightly for sample aged 160 days. Combining with the phenomenon of bubbles in the sample observed under the microscope (FIGURE 2, 3), this phenomenon may be the result of complex polarization process between the bubbles and the XLPE material. Finally, the characteristic parameters of severely aged samples increase significantly with the decrease of frequency. The tanδ of severely aged samples firstly decreases and then increases with the decrease of frequency, and a significant inflection point appears in the mid-frequency band.

Studies of dielectric spectrum characteristics of materials mainly focus on the qualitative characterization [28]. Dielectric response function based on the classical "single relaxation time" Debye model has been proposed by various researchers to model the experimental data [29]. D. K. Das-Gupta et al discussed different relaxation models (Debye model, Cole-Cole model, Davidson-Cole model, Havriliak-Negami two-parameter model) and their possible implication in the interpretation of the relaxation spectra of polymers [30]. S. K. Ojha et al estimated function representing distribution of relaxation frequencies [29]. Cole-Cole model and modified Cole-Cole model have been widely used to extract characteristics of frequency dielectric spectroscopy of oil-paper insulation [31]–[34]. However, few researchers reported the Cole-Cole model and the new parameters to diagnose the XLPE insulation material property. The modified Cole-Cole model with multiple relaxation processes is given as equation (3). Compared with Cole-Cole model, the modified model takes DC conductivity (σ_{dc}) and hopping conductivity (σ_{ho}) of the material into consideration. Where ε_{hf} is the dielectric permittivity at high frequencies, ω is angular frequency, n is distribution parameters of relaxation process, m is the number of multiple relaxation processes, χ_s is the static dielectric permittivity ($\omega = 0$),_τ is relaxation time; ε_0 is vacuum dielectric constant, $\varepsilon_0 = 8.85\times10^{-12}$F/m, γ is a constant, $0\leq \gamma \leq 1$.

$$\chi^*(\omega) = \varepsilon_{hf} + \sum_{i=1}^{m} \frac{\chi_s}{1+(j\omega\tau_i)^n} + \frac{\sigma_{dc}}{\varepsilon_0 j\omega} + \frac{\sigma_{ho}}{\varepsilon_0 (j\omega)^\gamma} \quad (3)$$

Taking dielectric curve of XLPE samples and fitting accuracy into consideration, a modified Cole-Cole model with α and β relaxation processes was considered and given as equation (4).

$$\chi*(\omega) = \varepsilon_{hf} + \frac{\chi_{s\alpha}}{1+(j\omega\tau_\alpha)^{n_\alpha}} + \frac{\chi_{s\beta}}{1+(j\omega\tau_\beta)^{n_\beta}} + \frac{\sigma_{dc}}{\varepsilon_0 j\omega} + \frac{\sigma_{ho}}{\varepsilon_0 (j\omega)^\gamma} \quad (4)$$

With equation (4), ten different characteristic parameters ($\varepsilon_{hf}, \chi_{s\alpha}, \chi_{s\beta}, \tau_\alpha, \tau_\beta, n_\alpha, n_\beta, \sigma_{dc}, \sigma_{ho}, \gamma$) can be used to fit the complex permittivity curves of XLPE samples with different aging conditions under 70 °C (FIGURE 11). These parameters can be extracted by nonlinear fitting of multivariate functions as the fingerprint characteristic parameters, which can be applied in the quantitative analysis of XLPE insulation with different aging conditions. The parameters of the modified Cole-Cole model can be obtained by curve fitting and optimization of multivariate function parameters. TABLE 2 shows the fitting parameters of the modified Cole-Cole model with α and β relaxation progresses for the FDS curves of samples with different aging degrees at 70 °C.

As can be seen from TABLE 2, with the development of aging progress, different parameters show different changes. The ε_{hf} shows the same trend with ε', it decreases slightly to 2.59 and soars to 4.47, nearly twice as much as the minimum value. σ_{dc} and σ_{ho} increase significantly from 7.61×10^{-17} and 1.13×10^{-16} to 2.83×10^{-10} and 6.51×10^{-14} respectively. During the α relaxation progress, there are no significant changes in n_α, but $\chi_{s\alpha}$ increases from 6 to 30.61 while τ_α drops significantly from 80000 to 100. However, no significant changes can be found in the parameters of β relaxation progress.

From the above analysis, it can be found that the effect of aging on dielectric properties is mainly reflected in parameters of α relaxation process and the changes of σ_{dc} and σ_{ho}. During the aging process of XLPE materials, oxidative cracking and other reactions occur inside the materials, resulting in crystallization cracking and a large number of polar substances. On the other hand, the aging state of materials leads to the uneven distribution and disorder of these materials, which makes the polarization parameters change more dramatically. $\chi_{s\alpha}$, τ_α, σ_{dc} and σ_{ho} are sensitive to the changes of XLPE material aging stages. Dielectric properties are caused by complex polarization processes. The polarization process is a physical change of matter. The model profoundly reveals the changes of the corresponding physical parameters in the polarization process. These four fitting parameters can be extracted as dielectric characteristic parameters in frequency domain and the insulation state of XLPE material can be quantitatively analyzed.

TABLE 2. corresponding fitting parameters of samples with different aging conditions.

Parameter	0-day	40-day	95-day	125-day	135-day	160-day
ε_{hf}	3.04	2.82	2.59	4.60	4.76	4.47
$\chi_{s\alpha}$	6.00	13.17	15.80	18.49	20.10	30.61
τ_α	80000	50000	40000	300	250	100
n_α	0.75	0.76	0.75	0.78	0.78	0.78
$\chi_{s\beta}$	0.05	0.1	0.05	0.6	0.6	0.6
$\tau_\beta(10^{-4})$	1	1	1	1	1	1
n_β	0.08	0.08	0.08	0.08	0.08	0.08
$\sigma_{dc}(\times10^{-16})$	0.76	133	532	1600	846000	2830000
$\sigma_{ho}(\times10^{16})$	1.13	1.85	1.90	63.8	69.4	651
γ	0.19	0.19	0.19	0.81	0.81	0.81

F. AC BREKDOWN VOLTAGE

FIGURE 12 is the average AC breakdown electrical field of samples with different aging conditions. It can be found that the AC breakdown electrical field strength gradually decreases as the aging process progresses. The AC breakdown electrical field decreases approximately linearly during the beginning 125 days and the downward trend gradually flattened out from 125-day samples to 160-day samples.

IV. NEW MUITI-FACTOR DETERIORATION CONDITION ASSESSMENT METHOD BASED ON GREY CORRELATION ANALYSIS

For the traditional method, the elongation at break retention rate of 50% is the criteria to indicate the end lifetime of XLPE sample. The traditional method only takes the mechanical strength of XLPE material into consideration. However, the thermal aging process of XLPE material is accompanied by a variety of changes in physical, chemical and dielectric properties. It can be found that these changes are not consistent, so only considering the mechanical strength is not enough to characterize the deterioration of materials. Therefore, it is necessary to establish a deterioration condition assessment model considering multi-physical quantities.

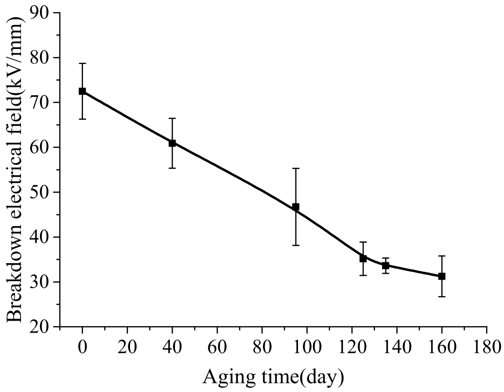


FIGURE 12. Changes in AC breakdown electrical field of samples with different aging conditions.

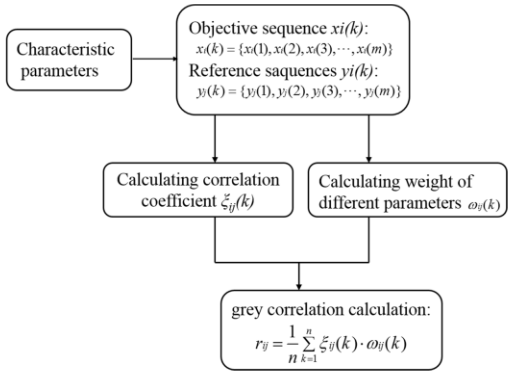


FIGURE 13. Gray relational calculation flow chart.

A. GREY CORRELATION METHOD

Grey relational analysis is a method of describing the strength of the relationship between factors in the order of grey relational degree. It can sort out the correlation extent or effect factors in a system with uncertain information [33]. The relational grade r_{ij} can be used to quantitatively describe correlation degree between an objective sequence $x_i(k)$ and a reference sequence $y_j(k)$. These sequences can be represented by series:

$$xi(k) = \{xi(1), xi(2), xi(3), \ldots, xi(m)\} \tag{5}$$

$$yj(k) = \{yj(1), yj(2), yj(3), \ldots, yj(m)\} \tag{6}$$

where m represents the total numbers of different properties.

Correlation coefficient $\xi_{ij}(k)$ is defined as:

$$\xi ij(k) = \frac{\min_i \min_j |yj(k) - xi(k)| + \alpha \max_i \max_j |yj(k) - xi(k)|}{yi(k) - xi(k) + \alpha \max_i \max_j |yj(k) - xi(k)|} \tag{7}$$

where α is an index for distinguishability, $\alpha \in (0, 1)$. $\xi_{ij}(k)$ represents the relational degree of different properties. $\max_i \max_j |yj(k) - xi(k)|$ represents the maximum value of $|yj(k) - xi(k)|$ while $\min_i \min_j |yj(k) - xi(k)|$ represents the minimum value of $|yj(k) - xi(k)|$.The r_{ij} can be calculated based on the $\xi_{ij}(k)$ and the weight corresponding to each property:

$$rij = \frac{1}{n} \sum_{k=1}^{n} \xi ij(k) \cdot \omega ij(k) \tag{8}$$

where $\omega ij(k)(k = 1, 2, 3 \ldots)$ represents the weight corresponding to each property. The weight of entropy is proportional to the function of characteristic quantity in the evaluation system. Assuming that the number of XLPE samples with different aging States is n and the number of deteriorating characteristic quantities is m, matrix X is constructed as $X = (x_{ij})_{n \times m}$ $(i = 1,2,\ldots,n; j = 1,2,\ldots,m)$. Dimensionless treatment of deteriorating characteristic quantities of XLPE samples was carried out and standardized index characteristic quantities P_{ij} were obtained:

$$P_{ij} = \frac{x_{ij}}{\sum_{i=1}^{n} x_{ij}} \tag{9}$$

Entropy weights E_j of deteriorating characteristic quantities of XLPE samples are shown in formula (10).

$$Ej = \frac{\sum_{i=1}^{n} P_{ij} \ln P_{ij}}{\ln n} \tag{10}$$

$\omega_{ij}(k)$ is the weight of each index characteristic quantity, as shown in formula (11).

$$w_{ij}(k) = \frac{1 - Ej}{\sum_{j=1}^{m} (1 - Ej)} \tag{11}$$

The flow chart shown in FIGURE 13 describes the process of evaluating the thermal deterioration condition by gray correlation method in detail.

B. NEW-FACTOR DETERIORATION CONDITION ASSESSMENT MODEL

As shown in FIGURE 14, the thermal aging process of XLPE materials is divided into four stages including enhanced stage (I&II), wear-out stage and disposal stage according to the retention rate diagram of elongation at break. The start and end times corresponding to each stage are marked in the graph. Enhanced stage (I&II) means some properties of XLPE material are enhanced and the XLPE is in a good condition. Wear-out stage means properties of XLPE begin to decline continuously, but mechanical strength has not reached the criterion of end of life. When the XLPE material reaches the disposal stages, the XLPE material has been deteriorated seriously.

According to the test results and fitting curves of the above test samples, reference samples and reference sequences corresponding to the start and end times of different stages can be established. Table 3 is a table of physical and chemical

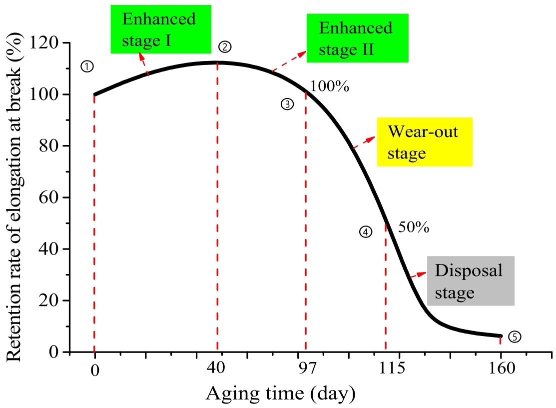


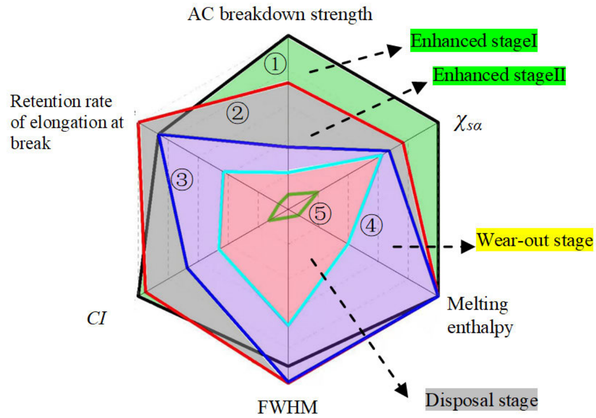
FIGURE 14. Schematic diagram of deterioration condition.

TABLE 3. Parameters of reference sequences for the multi-factor deterioration condition assessment model.

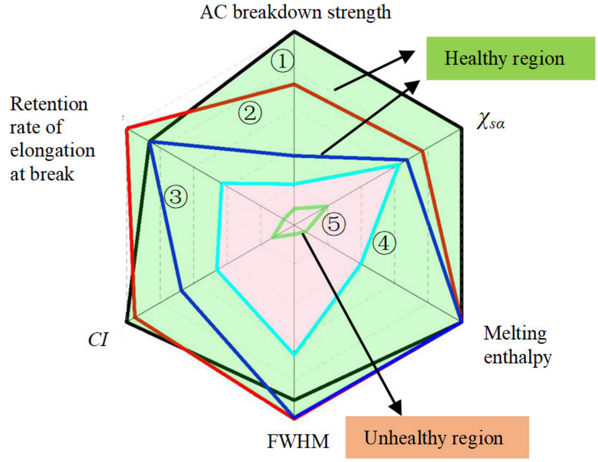
Boundary	Mechanical property	Chemical properties		Thermal property	Dielectric property	
	Retention rate of elongation at break /%	*CI*	FWHM /°	Melting enthalpy /J/g	$\chi_{s\alpha}$	AC breakdown strength /kV/mm
①	100	0	0.53	86.77	6	72.48
②	115.72	1.39	0.51	91.55	13.17	60.9
③	100	12.2	0.51	87.05	15.98	45.22
④	50	20.44	0.58	49.61	17.51	38.99
⑤	7.1	33.38	0.72	29.24	30.61	33.62

parameters and numerical values corresponding to the reference samples. As can be seen from the TABLE 3, six parameters were selected from four different aspects including mechanical, chemical, thermal and dielectric properties for each reference sequence: retention rate of elongation at break, *CI* based on FTIR to characterize the production and growth of new substances, FWHM based on XRD to characterize the phase, orientation of XLPE, melting enthalpy based on DSC, $\chi_{s\alpha}$ of modified Cole-Cole model based on results of FDS and AC breakdown strength. They are the reference sequence.

As shown in FIGURE 15, these reference parameters can be shown in a radar chart (FIGURE 15a). The radar chart shows the changes in mechanical, chemical, thermal and dielectric properties of XLPE samples with the development of the aging process. Four different deterioration conditions are divided in the radar chart. Parameters of deterioration condition ①, ② are the best conditions in the aging process of test samples, forming the boundary of the radar chart. The closed region formed by the boundary condition and parameters of deterioration condition ③ is the enhanced stage (I&II). In particular, reference sample ④ is regarded as a health threshold. The closed region formed by deterioration condition ③ and deterioration condition ④ is the wear-out stage. The closed area formed by deterioration condition ④ alone corresponds to the disposal stage. Regarding



(a) sketch of boundary threshold for deterioration condition



(b) sketch of healthy and unhealthy region

FIGURE 15. A sketch of boundary and threshold for the new multi-factor deterioration condition assessment.

TABLE 4. parameters of objective sequences.

Aging time/ day	Mechanical property	Chemical properties		Thermal property	Dielectric properties	
	Retention rate of elongation at break /%	*CI*	FWHM /°	Melting enthalpy/ J/g	FDS ($\chi_{s\alpha}$)	AC breakdown Strength /kV/mm
95	106.9	11	0.51	89.45	15.8	46.73
125	21.4	25	0.62	29.47	18.49	31.24
135	7.6	25.4	0.71	29.31	20.1	35.168

deterioration condition ④ as the threshold condition, regions of enhanced stage and wear-out stage are collectively referred to as healthy region while regions of disposal stage are referred to as unhealthy region (FIGURE 15b). Based on the equation (10-11), the weight $\omega_{ij}(k)$ of each index characteristic quantity is shown in FIGURE 16.

C. VERIFICATION OF THE NEW-FACTOR DETERIORATION CONDITION ASSESSMENT MODEL

The test results of 95-day, 125-day and 135-day samples were used to verify the new multi-factor deterioration condition

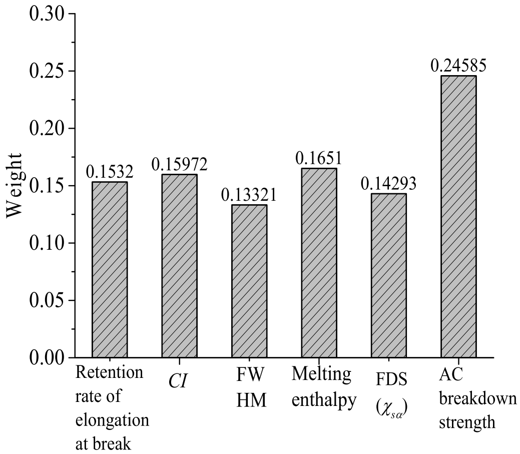


FIGURE 16. The weight ω_{ij} (k) of each index characteristic quantity.

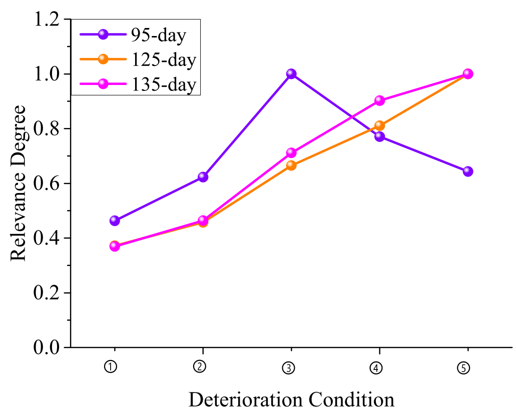


FIGURE 17. Gray relevance comparison of objective samples and its deterioration condition assessment.

assessment model shown above. Table 4 shows the objective sequences. The results of grey correlation between the 95-day, 125-day, 135-day objective sequences and the five reference sequences are shown in FIGURE 17. It can be found that the correlation between 95-day sample and deterioration condition ③ is the highest, and the deterioration degree of both is close. The correlation between 125-day sample and deterioration condition ⑤ is higher than other deterioration conditions, which means the deterioration condition of 125-day has been serious. The result of correlation degree of 135-day sample is the same as 125-day sample. Therefore, 95-day sample is located in the healthy region area of the radar, while 125-day sample and 135-day sample are located in the unhealthy region area. The verified result is consistent with the experimental result.

V. CONCLUSION

The changes of morphological, structural and dielectric properties of 500 kV XLPE submarine cable insulation material during accelerated thermal aging at 130 °C were introduced in this paper. According to the physicochemical and dielectric characteristic parameters, a multi-parameter material deterioration condition assessment model was established based on grey correlation method. The experimental and analytical conclusions are as follows.

With the increase of thermal aging degree, the surface color of the sample deepens and bubbles appear inside the XLPE samples. In terms of chemical structure, the increase of carbonyl content was accompanied by the decrease of crystalline part. The mechanical properties increased slightly at first, then decreased rapidly. The phase transition temperature decreases and the thermal performance decreases seriously. Four parameters including retention rate of elongation at break, *CI* based on FTIR, FWHM based on XRD, melting enthalpy based on DSC are chosen as sensitive to deterioration condition of XLPE material.

For dielectric properties, $\varepsilon' r$ and $\tan\delta$ increase. However, AC breakdown strength decreases rapidly with the thermal aging conducted. Cole-Cole analysis shows that the parameter $\chi_{s\alpha}, \tau_\alpha$, σ_{dc} and σ_{ho} are sensitive to the aging degree of XLPE samples. Parameters $\chi_{s\alpha}$ and AC breakdown strength were selected for deterioration condition assessment.

According to the change of elongation retention at break, the deterioration condition of XLPE sample was divided into enhanced stage (I&II), wear-out stage and disposal stage. The healthy and unhealthy regions of the samples are divided in the radar chart by taking the six characteristic parameters (retention rate of elongation at break, *CI* based on FTIR, FWHM based on XRD, melting enthalpy based on DSC, $\chi_{s\alpha}$ of modified Cole-Cole model based on results of FDS and AC breakdown strength) of the corresponding samples.

A new multi-factor deterioration condition assessment model including six characteristic parameters was established by grey correlation analysis. The new model can reflect the property of mechanical, chemical, thermal and dielectric. The deterioration condition of samples was judged by model matching. Results show that the new multi-factor deterioration condition assessment model has prominent effect in differentiating deterioration degree. To an operated cable, XLPE materials can be obtained during cable maintenance. These materials can be used to analyze the characteristics of materials and establish a grey relational model. The characteristic parameters selected in the evaluation can be optimized and upgraded according to the actual situation as well.

REFERENCES

[1] X. Wang, Z. Gao, Z. Liu, Z. Zhu, S. Li, J. Hao, and Z. Ma, "Chemical structure and breakdown behaviors of a new AC 500kV XLPE submarine cable insulation with different thermal aging conditions," in *Proc. IEEE Conf. Elect. Insul. Dielectric Phenomena (CEIDP)*, Cancun, Mexico, Oct. 2018, pp. 354–357.

[2] Y. Zhang, D. Liu, J. Wu, and Y. Yin, "A modified algorithm for the simulation of charge behavior in water tree aged cross-linked polyethylene cable," *IEEE Access*, vol. 6, pp. 23929–23938, 2018.

[3] X. Wang, Z. Gao, Z. Zhu, S. Li, Z. Liu, J. Hao, and Z. Ma, "Physicochemical and electrical property of a new AC 500kV XLPE submarine cable insulation with different thermal aging condition," in *Proc. 12th Int. Conf. Properties Appl. Dielectric Mater. (ICPADM)*, Xi'an, China, Mar. 2018, pp. 479–482.

[4] Z. Zhang, P. D. S. Assala, and L. Wu, "Residual life assessment of 110 kV XLPE cable," *Electr. Power Syst. Res.*, vol. 163, pp. 572–580, Oct. 2018.

[5] D. He, J. Gu, W. Wang, S. Liu, S. Song, and D. Yi, ''Research on mechanical and dielectric properties of XLPE cable under accelerated electrical-thermal aging,'' *Polymer Adv. Technol.*, vol. 28, no. 8, pp. 1020–1029, Aug. 2017.

[6] Y. Wang, J. Wu, and Y. Yin, ''Space charge behavior in low density polyethylene at low temperatures,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 24, no. 6, pp. 3860–3868, Dec. 2017.

[7] L. Boukezzi, A. Boubakeur, C. Laurent, and M. Lallouani, ''DSC study of artificial thermal aging of XLPE insulation cables,'' in *Proc. IEEE Int. Conf. Solid Dielectrics*, Winchester, U.K., Jul. 2007, pp. 146–149.

[8] L. Boukezzi, A. Boubakeur, and M. Lallouani, ''Effect of artificial thermal aging on the crystallinity of XLPE insulation cables: X-ray study,'' in *Proc. Annu. Rep. Conf. Elect. Insul. Dielectric Phenomena*, Vancouver, BC, Canada, Oct. 2007, pp. 65–68.

[9] F. Khabbaz, A.-C. Albertsson, and S. Karlsson, ''Chemical and morphological changes of environmentally degradable polyethylene films exposed to thermo-oxidation,'' *Polymer Degradation Stability*, vol. 63, no. 1, pp. 127–138, 1999.

[10] K. Anandakumaran and D. J. Stonkus, ''Assessment of oxidative thermal degradation of crosslinked polyethylene and ethylene propylene rubber cable insulation,'' *Polymer Eng. Sci.*, vol. 32, no. 18, pp. 1386–1393, 1992.

[11] W. Nelson, ''Accelerated life testing–step-stress models and data analyses,'' *IEEE Trans. Rel.*, vol. R-29, no. 2, pp. 103–108, Jun. 1980.

[12] M.-Y. Hu, C.-C. Zhang, F.-L. Ma, and B.-Z. Han, ''Influence of short-time thermal aging on XLPE insulation properties of HVDC cable,'' in *Proc. Mater. Eng. Mech. Eng.*, Hangzhou, China, 2016, pp. 1267–1274.

[13] A. Tzimas, S. Rowland, L. A. Dissado, M. Fu, and U. H. Nilsson, ''Effect of long-time electrical and thermal stresses upon the endurance capability of cable insulation material,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 16, no. 5, pp. 1436–1443, Oct. 2009.

[14] Z. Duan, G. Wu, and B. Gao, ''Impacts of copper products on characteristics of frequency domain dielectric spectroscopy of oil-paper insulation,'' in *Proc. Annu. Rep. Conf. Elect. Insul. Dielectric Phenomena*, Oct. 2013, pp. 60–64.

[15] J. Liu, Y. Xu, X. Wei, and X. Li, ''Research of dielectric spectroscopy on insulation ageing assessment of XLPE cables,'' in *Proc. Annu. Rep. Conf. Elect. Insul. Dielectric Phenomena*, Shenzhen, China, Oct. 2013, pp. 140–143.

[16] M. Nedjar, ''Effect of thermal aging on the electrical properties of crosslinked polyethylene,'' *J. Appl. Polymer Sci.*, vol. 111, no. 4, pp. 1985–1990, 2009.

[17] C. Kim, Z. Jin, P. Jiang, Z. Zhu, and G. Wang, ''Investigation of dielectric behavior of thermally aged XLPE cable in the high-frequency range,'' *Polymer Test.*, vol. 25, no. 4, pp. 553–561, 2014.

[18] P. Geng, J. Song, M. Tian, Z. Lei, and Y. Du, ''Influence of thermal aging on AC leakage current in XLPE insulation,'' *AIP Adv.*, vol. 8, no. 2, Feb. 2018, Art. no. 025115.

[19] G. C. Montanari, A. Motori, A. T. Bulinski, S. S. Bamji, and J. Densley, ''Application of oxidation induction time and compensation effect to the diagnosis of HV polymeric cable insulation,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 3, no. 3, pp. 351–360, Jun. 1996.

[20] J. C. Fothergill, G. C. Montanari, G. C. Stevens, C. Laurent, G. Teyssedre, L. A. Dissado, U. H. Nilsson, and G. Platbrood, ''Electrical, microstructural, physical and chemical characterization of HV XLPE cable peelings for an electrical aging diagnostic data base,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 10, no. 3, pp. 514–527, Jun. 2003.

[21] C.-H. Lin, C.-H. Wu, and P.-Z. Huang, ''Grey clustering analysis for incipient fault diagnosis in oil-immersed transformers,'' *Expert Syst. Appl.*, vol. 36, no. 2, pp. 1371–1379, Mar. 2009.

[22] Y. Zhang, R. Liao, L. Yang, X. Deng, H. Cheng, and C. Lv, ''A cost-effectiveness assessment model using grey correlation analysis for power transformer selection based on life cycle cost,'' *Kybernetes*, vol. 43, no. 1, pp. 5–23, Jan. 2014.

[23] R. J. Liao, J. P. Bian, L. J. Yang and S. Grzybowski, ''Cloud model-based failure mode and effects analysis for prioritization of failures of power transformer in risk assessment: Failure prioritization of power transformer,'' *Int. Trans. Electr. Energy. Syst*, vol. 23, no. 7, pp. 1172–1190, May 2012.

[24] W. Hang, G. Tan, Y. Tan, L. Zhou, F. Zhou, G. Liu, and Y. Lu, ''Analysis of thermal aging life and physicochemical properties of crosslinked polyethylene seabed cable insulation,'' *Polymer Mater. Sci. Eng.*, vol. 31, no. 3, pp. 71–75, 2015.

[25] Y. Wang and W. Can, ''Anti-thermal aging properties of low-density polyethylene-based nanocomposites,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 3, no. 25, pp. 1003–1013, Jun. 2018.

[26] V. Drits, J. Środoń, and D. D. Eberl, ''XRD measurement of mean crystallite thickness of illite and illite/smectite: Reappraisal of the Kubler index and the Scherrer equation,'' *Clays Clay Minerals*, vol. 45, no. 3, pp. 461–475, 1997.

[27] A. Xie, S. Li, X. Zheng, and G. Chen, ''The characteristics of electrical trees in the inner and outer layers of different voltage rating XLPE cable insulation,'' *J. Phys. D, Appl. Phys.*, vol. 42, no. 12, 2009, Art. no. 125106.

[28] D. Saurabh, B. Arijit, A. K. Pradhan, and S. Chakravorti, ''Effect of measurement temperature on power transformer insulation diagnosis using frequency-domain spectroscopy,'' *IET Sci. Meas. Technol.*, vol. 11, no. 6, pp. 773–779, Sep. 2017.

[29] S. K. Ojha, P. Purkait, and S. Chakravorti, ''Modeling of relaxation phenomena in transformer oil-paper insulation for understanding dielectric response measurements,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 23, no. 5, pp. 3190–3198, Oct. 2016.

[30] D. K. Das-Gupta and P. C. N. Scarpa, ''Modeling of dielectric relaxation spectra of polymers in the condensed phase,'' *IEEE Elect. Insul. Mag.*, vol. 15, no. 2, pp. 23–32, Mar. 1999.

[31] J. Liu, X. Fan, Y. Zhang, H. Zheng, H. Yao, C. Zhang, Y. Zhang, and D. Li, ''A novel universal approach for temperature correction on frequency domain spectroscopy curve of transformer polymer insulation,'' *Polymers*, vol. 11, no. 7, p. 1126, Jul. 2019.

[32] C. Qi, L. Yang, W. Li, J. Hao, Z. Ma, and J. Bai, ''Extraction method for frequency domain characteristic parameters of oil impregnated insulation paper,'' in *Proc. Int. Conf. High Voltage Eng. Appl.*, Shanghai, China, Sep. 2012, pp. 110–114.

[33] S. Wolny, A. Adamowicz, and M. Lepich, ''Influence of temperature and moisture level in paper-oil insulation on the parameters of the cole-cole model,'' *IEEE Trans. Power Del.*, vol. 29, no. 1, pp. 246–250, Feb. 2014.

[34] J. Gao, L. Yang, Y. Wang, C. Qi, J. Hao, and J. Liu, ''Quantitative evaluation of ageing condition of oil-paper insulation using frequency domain characteristic extracted from modified cole-cole model,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 22, no. 5, pp. 2694–2702, Oct. 2015.

[35] C. Fu, J. Zheng, J. Zhao, and W. Xu, ''Application of grey relational analysis for corrosion failure of oil tubes,'' *Corrosion Sci.*, vol. 43, no. 5, pp. 881–889, 2001.

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