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Effects of electrical aging on the structural and physicochemical properties of crosslinked polyethylene (XLPE) cable insulation material

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Abstract

The aim of this work is to highlight the evolution of physico-chemical properties of peroxide crosslinked polyethylene (XLPE) insulation material exposed to electrical aging. Specific analysis and characterization studies were carried out to determine the effect of electrical aging on different samples at micro and macro levels. Based on the analytical investigations, it was shown that electrical aging yielded significant changes in the chemical and thermal characteristics of XLPE. It was also showed that property loss increased with the aging time.

Keywords: Crosslinked polyethylene (XLPE), Insulation, Aging, Characterization.

Introduction

Sustained efforts have been conducted for developing new materials, production methods, and testing techniques in electrical cable and device industries as the essential role of signal and power transmission in technology [1]. Thermoplastic polymers are preferably used in manufacturing insulating layers of underground electrical cables, due to their superior chemical resistance and mechanical durability. In addition, commercial thermoplastics are cheap and easily processable materials [2].

The most extensively used thermoplastics in cable insulating applications are poly (vinyl chloride) (PVC) compounds, polyethylenes (PEs), ethylene-vinyl acetate (EVA) copolymers, and other elastomeric polymers. These thermoplastics provide excellent physical and insulation performance for low voltage (LV) cables. Medium and high voltage (MV and HV) applications require some extra physical properties. Specifically, long-term mechanical and electrical performances of insulating materials become much more important features for MV and HV applications. Thus, structural properties of thermoplastics are modified by
various physical and/or chemical methods. Cross linking is the most versatile solution to dramatically improve the physical properties of PEs. Cross linked polyethylene (XLPE) is the most widely used materials in MV cable industry [3] because it possesses many advantages such as high dielectric strength, low dielectric permittivity and loss factor, good dimensional stability, excellent resistance to solvents, chemicals, and oxidative species, and superior thermo-mechanical behavior [4]. Nevertheless, XLPE cable can be subjected to highly erosive and extreme thermal, mechanical, electrical, and environmental effects, which lead to forming irreversible structural damages yielding significant reduction in cable service life [5]. It is well established that the effects of environmental and electrical constraints are mainly responsible for the degradation of XLPE insulation and cable rupture [6]. The effects of temperature and radiation on the aging of XPLE have been previously studied. Ouyang et al. [7] studied the role of microstructural changes on the space charge distribution of XLPE during thermo-oxidative aging. Boukezzi et al.[8] showed the effect of thermal aging on the electrical characteristics of XLPE. The impact of ionizing radiation on XLPE has also received special attention. As such, Fifield et al. [9] have shown that gamma rays induce changes in the XLPE structure. Furthermore, Shimada et al.[10] reported that gamma radiation induces changes in XLPE performance. XLPE cables must withstand nominal alternating voltage and charging current to avoid failure during service [11],[12]. Electrical aging has different effects on the insulation properties of XLPE. Liu et al. [12] studied the growth characteristics of trees in the insulation of XLPE cables under DC conditions. Bao et al. [13] studied the influence of electrode frequency, voltage, and distance on the propagation of trees in XLPE cables. Abou Dakka et al. [14] suggested a correlation between DC polarization and the failure characteristics of aged XLPEs under AC voltage. A great attention has been paid to the monitoring and evaluation of power cables [15]. Several approaches for assessing the insulation performance of such materials were established. However, it should be noted that the physico-chemical alterations in insulating materials induced by electrical stress have not been studied yet. Moreover, underlying mechanisms for molecular and/or structural changes are still not sufficiently understood.

This study aims to evaluate the effects of electrical aging on the some properties of XLPE insulating power cables, comprehensively. It proposes quantitative relationships between aging time and physico-chemical characteristics of specimens based on various structural analysis and instrumental techniques such as Attenuated Total Reflectance-Fourier Transform Infrared (ATR-FTIR) spectroscopy, Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA), X-Ray Diffraction (XRD), and Scanning Electron microscopy (SEM).

Material and methods

The studied material was kindly obtained from Boracal as a commercial grade crosslinkable polyethylene compound (XLPE, HFDELE4201R containing 2% of dicumyl peroxide (DCP) as crosslinking agent). Square plates with a length of 130 mm and a thickness of 2 mm were prepared by compression molding at 180 °C under a pressure of 300 bar for 2 minutes. The molding condition ensured to completion of crosslinking of PE. Then the plate was cooled down to 45 °C. Electrical aging test set up was schematized in Figure 1. The procedure consists of placing the XLPE sample between two electrode planes with a Rogowski profile of 50 mm diameter under an AC electrical field of 5 kV/mm. The upper electrode was connected to the voltage source and the lower one to the ground. A 100-kV, 50-Hz test transformer of 10-kVA power delivered the 100-kV-applied voltage controlled by an autotransformer.

![Experimental setup for electrical aging test.](image)

**Figure 1.** Experimental setup for electrical aging test.

FTIR spectra were recorded between 4000 and 650 cm⁻¹ with an accumulation of 50 spectra with a resolution of 4 cm⁻¹ using a Shimadzu 8400s spectrometer; Furthermore, DSC analysis was performed using DSC 204-F1 Phoenix, Netzsch Co. equipped with a data acquisition system under a constant flow of nitrogen gas. Melting and crystallization behaviours of samples were measured by applying heating and cooling cycles between 20 and 160 °C with a heating/cooling rate of 10 °C/min. Thermal stability of XLPE specimens before and after electrical aging was studied with TGA analysis in a TGA analyzer Tarsus type (Netzsch Co model TG 209 F3) under a nitrogen atmosphere. The TGA analysis was carried out by heating the samples from room temperature to 600 °C with a heating rate of 10 °C/min. Furthermore, X-ray diffraction spectra were obtained using Cu-Kα radiation with a wavelength of 1.5406 Å in a D8 ADVANCE, with a Brucker co X-ray diffractometer in the 2θ range of 10-70° by
a step of 0.05° with a 0.5 s/step scan rate. Finally, the morphological analysis was performed in a scanning electron microscope (Phillips XL30) with a magnification of 2000 under a voltage of 20kV.

Results and discussion

![Figure 2. ATR-FTIR spectra of XLPE samples at different aging time.](image)

Figure 2 shows the FTIR spectrum of unaged and aged specimens in the wavenumber range of 4000-650 cm⁻¹. Unaged XLPE, exhibited characteristic peaks of polyethylene structure at 720 cm⁻¹ corresponds to the methylene group balance (-CH₂) [16] while those observed at 1471, 2856 and 2937 cm⁻¹ correspond to valence, symmetric, and asymmetric vibrations of -CH₂, respectively [17]. The peaks at 1370 and 2150 cm⁻¹ are attributed to methyl group related to symmetric deformation vibration and symmetric valence vibration, respectively [18]. The absorption band around 1078 cm⁻¹ is attributed to the presence of an antioxidant, such as Irganox [19], which could have been introduced into the polyethylene resin by the manufacturer. In addition, the presence of ester or aldehyde supported by the presence of the peak at 1741 cm⁻¹ in the unaged sample indicates that oxidation process has taken place during manufacturing process. The diminution in the peak intensity after 10 hours of aging may be due to chain breaks induced by thermolysis of chain bonds or by oxidation. After 20 hours of aging, the peak intensity increases, indicating the apparition of a competitive process to oxidation, namely, crosslinking. This phenomenon comes from the formation of covalent bridges between the chains of the XLPE polymer.

Finally, absorption zone of hydroxyl (-OH) groups is seen in the wavenumber range of 3300-3500 cm⁻¹. As seen in Figure 1, the FTIR spectra of samples exhibits some differences. The peaks between 2800 and 3000 cm⁻¹ strongly decreased on the spectra of aged samples which clearly indicated that electrical aging affected the chain dynamics of XLPE samples. On the other hand, the absorption peak of carbonyl bonds (C=O) at around 1700 cm⁻¹ appeared at the spectrum of aged samples and increased with aging time and shifted to higher wave numbers. This was due to the formation of oxidized species such as carbonyl groups, ketone, carboxylic acid and ester via chemical interaction between carbon radicals and air. This change in chemical structure and intensity of the peaks could be related to surface erosion or roughness phenomena as discussed in next parts.

The carbonyl index (CI) and trans-vinylene index (TVI) were calculated to quantify level oxidation as a function of electrical aging based on the FTIR spectra as follows:

$$CI = \frac{Abs_{1741}}{Abs_{1471}}$$

$$TVI = \frac{Abs_{965}}{Abs_{1370}}$$

Where Abs₁₇₄₁, Abs₁₄₇₁, Abs₉₆₅, and Abs₁₃₇₀ are the peak areas of carbonyl (aldehyde), methylene, trans-vinylene and trans-methylene absorption bands at 1741, 1471, 965 and 1370 cm⁻¹, respectively.

Figure 3 shows the presence of carbonyl and double band indexes in aged and unaged samples. This figure also implied that the CI and TVI indexes increased with the electrical aging time. The CI and TVI indexes were calculated as 0.09 and 0.34, for the unaged sample, respectively, while such values were 0.12 and 0.46 for the 10-hour aged sample. These values increased to 0.29 and 0.48 by the electrical aging of 20-hours. The calculated TVI values are consistent with the previously reported ones in the literature. As reported in the relevant literature, the process of splitting the polyethylene chain is favoured by the presence of oxygen during thermo-oxidation reactions and contributes to the formation of unsaturated groups [20],[21]

![Figure 3. Carboxyl and Trans-vinylene indices of XLPE samples.](image)

Figure 4 shows the DSC heating-cooling thermograms of unaged and aged samples. Figure 5 represents the melting and crystallization peaks of samples within a narrow phase transition range.
Figure 4. DSC heating and cooling thermograms of aged and unaged XLPE samples

Figure 5 shows intensity of thermograms (exothermic and endothermic peaks) with a scale-bar of 0.3 mW/mg. It can be noted from this figure that unaged XLPE shows a characteristic melting endotherm with a peak maximum temperature of 104.8 °C. After aging, the melting peak temperature ($T_m$) of XLPE shifted to 108 °C. This is probably due to the increasing the polarity of material as a result of oxidative reactions and improving the interactions between PE chains [3]. On the other hand, it was observed that electrical aging stress caused a slight decrease in the intensity and broadening of the endothermic peak. This broadening indicates the presence of smaller chains segments resulting from the XLPE bond scission associated to the oxidative process [18]. It was also found that the crystallization peak temperature ($T_c$) decreased from 88.8 °C to 86.7 °C as a result of 20-hours aging. This slight difficulty in chain ordering behavior suggests an oxidative degradation in the molecular structure [16].

Figure 5. DSC endothermic (a) and exothermic (b) peaks

The degree of crystallinity $\chi_{DSC}$ (%) of XLPE was calculated using the following relationship:

$$\chi_{DSC} \% = \frac{\Delta H_m}{\Delta H_0} \times 100$$

Where $\Delta H_0$ is the melting enthalpy values of 100% crystalline and $\Delta H_m$ is the melting enthalpy of samples. $\Delta H_0$ was taken as 280 J/g in this study [3].

Figure 6 shows the XRD patterns of aged and unaged samples. It can be seen that the characteristic diffraction peaks of PE crystals do not change their positions and yielded the same 2θ values. This implies that no new crystalline phase is produced in the material.

The crystallinity values of XLPE that is generally described as “crystallinity index” can be calculated using the Hinrichsen’s method [22]. By fitting the diffractogram into three Gaussian curves, the crystallinity index $\chi_{XRD}$ (%) is given by the following relationship [23]:
$X_{SD} = \frac{Area2 + Area3}{Area1 + Area2 + Area3} \times 100$

Where $Area1$, $Area2$, and $Area3$ are the areas of amorphous halo, principal crystalline peak at 20 of 21.5° and secondary crystalline peak at 20 of 23.8°, respectively.

The evolution of crystallinity is illustrated in Figure 6. The slight increase in crystallinity can be attributed to the thermal oxidation occurring during the electrical aging. Similar results were previously reported [24], when studying the impact of accelerated water tree aging (AWTT) on XLPE. By comparing the degree of crystallinity values obtained with XRD and DSC methods, it can be observed that the values are not the same; however, show the same trend with electrical aging. The difference may be due to the fact that the DSC measurement only concerns the crystalline phase of polymer while the XRD measurement includes the total crystallinity as sum of polymer, fillers, and additives.

The DSC and XRD parameters of samples are also listed in Table 1. From DSC results, it can be noticed that electrical aging induce an increase in melting temperature and melting enthalpy, and a decrease in crystallization temperature and crystallization enthalpy. DSC and XRD show that electrical causes a decrease in crystallinity. If the results obtained with XRD and DSC does not present the same values of crystallinity; however, they show the same growth trend with the electrical constraint. Therefore, it is possible to correlate the crystallinity obtained by XRD with that found by DSC.

**Table 1.** Thermal properties and degree of crystallinity values of samples obtained from DSC and XRD analysis.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$T_m$ (°C)</th>
<th>$\Delta H_m$ (J/g)</th>
<th>$T_c$ (°C)</th>
<th>$\Delta H_c$ (J/g)</th>
<th>$X_c$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unaged</td>
<td>104.8</td>
<td>68.04</td>
<td>88.8</td>
<td>93.0</td>
<td>24.3</td>
</tr>
<tr>
<td>10 h</td>
<td>108</td>
<td>65.10</td>
<td>87.7</td>
<td>91.4</td>
<td>23.2</td>
</tr>
<tr>
<td>20 h</td>
<td>108</td>
<td>64.90</td>
<td>86.7</td>
<td>91.6</td>
<td>23.1</td>
</tr>
</tbody>
</table>

Figure 7 reports TGA weight loss thermograms of samples. The first weight loss stage, about 1% between the temperatures of 235 and 300 °C may corresponds to removing of volatile components of fillers and/or additives. ($O_2$ incorporation should increase the weight). The second weight loss stage, about 8% in a temperature range of 300-420 °C could be originated degradation of the sensitive components, probably organic groups of peroxide linkers. It should also be noted that the rate of second step is quite low [25]. In the third stage, all samples lost about 86% of their initial weightin the temperature range of 420-460 °C. This step is the main decomposition stage occurred in a very narrow temperature range and corresponds to quick formation of volatile hydrocarbones by random scissions of polyethylene macromolecules. It was found that the electrical aging did not affect thermal decomposition behavior of polyethynene at high temperatures.
Figure 8 shows that electrical stress affects the morphological features of XLPE. The surface of the virgin sample (Figure 8(a)) is smooth and homogeneous. Per contra, the surfaces of the aged samples are rough and show signs of degradation on the whole surface. After 10-hours aging, a considerable amount of defects appears, and the surface topology becomes gritty having a few holes. After 20 hours, the surface appearance looks like more damaged and lumpy showing more cracks and larger holes. The holes appearance can be attributed to the evaporation of volatile components during the aging process [26]. In fact, it has been reported that the distribution of the surface into blocks and the appearance of cracks is a sign of aging of the material [27]. These observations are very consistent with those reported in previous works [18],[19],[26].

What’s new?

Several studies in this subject are mainly interested in the effects of environmental parameters on XLPE used as power cable insulation. However, some investigations have evaluated the electrical degradation of XLPE. From physico-chemical point of view, the structural changes of crosslinked polymers induced by electrical constraint are not well understood. Thus, this study constructed a different approach from the literature considering only an electrical stress as an aging factor for XLPE materials rather than conventional environmental constraint such as service temperature, UV, mechanical stresses and deformations, water, humidity etc.). In this work, it has been proved that electrical constraint is responsible of physical alterations and structural changes in XLPE insulation materials. Thus, the technical data and results show that electrical aging induces oxidation, decreases crystallinity and changes the surface topography of XLPE. These alterations may induce a degradation of the electrical and mechanical properties, and consequently failure of the insulation.

Conclusion

The effects of electrical stress on the aging of XLPE has been experimentally shown via various thermodynamic and physico-chemical characterization tests. The theoretical hypothesis and expectations guide that electrical stress would be one of the topmost factor of current XLPE aging. This study has verified this hypothesis well. From these investigations, the main conclusions can be summarized as follows:

- The FTIR showed the presence of carbonyl groups in aged XLPE insulation indicating that oxidation process occurred during electrical aging process.
- In DSC analysis, it was observed that the aging of the XLPE insulation causes a slight decrease in crystallinity that can be attributed to the molecular chain scission and oxidation. Furthermore, from the TGA analysis, it was observed that the decomposition temperature of XLPE insulation is not significantly affected.
- XRD patterns indicated that no new crystalline phase is produced in the material during the aging.
- Finally, SEM analysis clearly shows morphological changes caused by electrical aging. This study could be used to develop new insulating materials based on XLPE with better performance.
REFERENCES


