BEHAVIOR OF CROSSLINKED POLYETHYLENE INSULATION OF MEDIUM AND HIGH VOLTAGE POWER CABLES UNDER UV RADIATIONS

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Abstract: In this study, an experimental investigation of ultraviolet (UV) radiations effects on the high voltage cable crosslinked polyethylene (XLPE) insulation characteristics is presented. Measurements of mechanical, physical and electrical properties are also performed. In order to highlight the surface damages after ageing, different complementary techniques such as attenuated total reflectance - Fourier transformer infrared spectroscopy) and scanning electron microscopy are conducted. Obtained results show that several of the XLPE insulation properties are affected by exposition to UV radiations. Moreover, a correlation between mechanical, electrical and physical properties, and physico-chemical alterations was noticed. Furthermore, our study displayed prominently that there was a concurrence between crosslinking and chain breaking.

Key words: Ultraviolet (UV) radiations, Crosslinked polyethylene (XLPE), Insulation.

1. Introduction

Owing to their excellent properties, polymeric insulating materials are widely used in electrical power equipment [1]. With age, like all materials, devices made of polymers undergo dramatic effects on their properties [2]. Actually, it is well known that the most significant degradation occurs during exposure to an aggressive environment. This will result in several modifications of polymer mechanical, physical and chemical properties.

Generally, polymers which are extensively used in outdoor applications are exposed to solar ultraviolet (UV) [3]. Radiation reaching the earth surface is characterized by wavelengths ranging from approximately 295 nm up to 2500 nm. The actinic range of solar UV radiation representing UV-A and UV-B is about 6% of the total radiation [4]. Previous studies have shown that UV radiations involve crucial degradations in the properties of polymer materials [5]. The recent studies have adopted several methods to monitor the aging condition of cable materials. They have reported that UV radiations can cause several evolutions in physical properties of polymers, like hydrophobic stability [6] or weight loss [7], and mechanical characteristics [8]. It has also demonstrated that all these changes are a consequence of material chemical modifications, such as chain scission and crosslinking [8].

Polyethylene in its crosslinked form (XLPE) is widely used as an electrical insulation in extruded power cables [9] and preferred by utilities worldwide [10]. It is gradually replacing other types of polyethylenes due to its greater mechanical and thermal stability [11]. The effect of the environmental constraints (temperature, humidity and pressure) on the XLPE characteristics has received considerable attention and several studies have been carried out on this topic. However, few works are dedicated to its photodegradation phenomenon [11], [12], [13]. Guimine and Akcelrud [11] have evaluated the effect of UV-ageing in its appearance and chemical properties. Wu et al. [12] have shown that the products of XLPE photooxidation are mainly hydroperoxides and various carbonyl groups. The authors in [13] demonstrated that the kind of crosslinking agent (silane and peroxide) influences the XLPE’s degradation mechanism under UV.

The focus of this work is to investigate the possible changes in the properties of XLPE insulation subjected to UV. These changes are investigated using mechanical tests (tensile strength, elongation at break and surface hardness), physical experiments (surface wettability and water retention), electrical surface resistivity and Physico-chemical characterizations using attenuated total reflectance - Fourier transformer infrared spectroscopy (ATR-FTIR) and scanning electron microscopy (SEM).

2. Experimental Setup

2.1. Samples and UV Ageing Conditions

Square plates of 130 mm x 130 mm and 2 ± 0.2 mm thickness were moulded and crosslinked at 180°C under a pressure of 300 bars from granules of Borealis natural crosslinkable polyethylene compound HFDE LE4201R, using pressurized heat press. This material, in its granule form, contains polyethylene blended with 2% of dicumyl peroxide (DCP) as cross-linking agent and an antioxidant. The studied material is used as high voltage cable insulation. Artificial UV ageing
undergone by the XLPE samples was carried out in an accelerated UV-aging chamber, designed and built specially for this investigation. The samples irradiation is accomplished using ten low-pressure vapor fluorescent lamps of 36 Watts characterized by wavelength irradiation ranging mostly (98%) from 350 nm to 400 nm. The lamps were placed 10 cm above the XLPE’s top surface. The samples are subjected to the various experiments after each 20 hours of exposure. The maximum aging time is 240 hours. The ageing is performed under a temperature of 55±5°C without controlling the humidity.

2.2. Mechanical Properties

Mechanical tests were carried out to determine the evolution of the elongation at break, tensile strength and surface hardness as well to assess the general relaxation behaviour of the material under mechanical load. According to IEC 60811.1.1 (International Electrotechnic Committee) standard, the elongation at break and tensile strength tests are performed on samples having a dumbbell shape of 5 cm length, cut from the obtained plates. After each 20 h aging time, 7 samples were taken, the first five are intended for testing elongation at break and the tensile strength, and the two others are reserved for measuring the surface hardness.

2.2.1. Elongation at Break and tensile strength

Tensile testing (elongation at break and tensile strength), was carried out on a Schnek-Trebel testing machine. The experiments consist in breaking the sample, at ambient temperature, using a dynamometer which moves with speed of 50 mm/min. Elongation at break and tensile strength were measured simultaneously.

2.2.2. Surface Hardness

Surface hardness is a versatile tool for evaluating the mechanical properties of polymers [14]. It is a property directly related to the efficiency of polymerization and to the mechanical transitions, for instance, glass transition [15]. In our study, the measurement of this property was performed using an Amsler hardness tester.

2.3. Physical Properties

From the obtained plates, specimens in square form (6cm × 6cm) were cut. After each 20 hours of UV-exposure ten samples were removed from the UV-irradiation chamber. The first five are intended for evaluating the water absorption and the others five for measuring contact angle and work of water adhesion to XLPE surface evolutions.

2.3.1. Water Contact Angle and Work of Water Adhesion

The water contact angle measurement and work of water adhesion evaluation are frequently used in characterizing surface hydrophobic properties of various materials. Static contact angle is the equilibrium angle at which a liquid interface meets at the solids surface. Usually, the concept is illustrated with a small liquid droplet resting on a flat horizontal solid surface, which can identify the surface hydrophobicity (water repellant) and surface hydrophilicity (water attraction) of the materials [16].

In order to study the UV radiation effect on the hydrophobicity, a test ring was carried out. It comprises a micro-syringe, a torch, a samples holder and a high-resolution camera. An experimental procedure was thus followed to determine the contact angle value: it is to take pictures using the digital camera for a droplet of 10 ml of distilled water, deposited on the solid insulation surface. Contact angles are then determined. The measurement procedure with this device complies with the operation principle of a goniometer. The total water adhesion (Ws) calculation is related to the contact angle measurement, it is given by the following equation:

\[ W_s = \gamma_{sw} \cdot (1 + \cos \theta_w) \]  

(1)

Where \( \gamma_{sw} \), 72.8 mJ/mm² is the surface tension of water and \( \theta_w \) is the contact angle.

![Fig.1 Contact angle of water droplet on the material surface](image)

2.3.2. Water Absorption

The UV-exposed and Unexposed samples to UV were immersed into distilled water for 24 h to be saturated, and then dried in an oven for 2 hours at 105 °C. Because the samples had different volumes and weights, water absorbance, \( W_r (%) \) was adopted to quantify the amount of absorbed water. The weights of the samples before and after drying were recorded using an electronic balance. Water absorption is calculated using the formula:

\[ W_r (%) = (W_d - W_i / W_i) \times 100 \]  

(2)

Where \( W_i \) and \( W_d \) are, respectively, the mass of wet sample and the mass of dry sample.

2.4. Electrical Surface Resistivity

Surface resistance was measured according to IEC 60093 standard, using a Teraohmmeter and a resistivity
measurement cell (three electrode method) as shown in Fig.2. Fig.3 is a sketch of the surface resistivity measurement cell; it is containing a guarded or measuring electrode (diameter, \(D_1=50\) mm), a ring electrode (inner diameter, \(D_2=60\) mm) and a counter electrode (diameter, \(D_3=110\) mm).

Surface resistance measurement was carried out at ambient conditions. It was obtained by applying a DC voltage of 500 V between the guarded electrode and the ring electrode. The value obtained after 60s of voltage application was recorded as the suitable resistance. Surface resistivity is related to the configurations and dimensions of the used electrodes. It is calculated by:

\[
\rho_s = R_s \frac{P}{g}
\]

Where \(R_s\) is the measured resistance in ohm, \(P\) is the effective perimeter \((P = \pi (D_1 + D_2) / 2)\) of the guarded electrode and \(g\) \(((D_2-D_1) / 2)\) is the distance between the guarded and ring electrode.

2.6. Scanning Electron Microscopy (SEM)

In order to study, the changes in morphology of UV- aged XLPE; micrographs of samples in different aging states were investigated on the surfaces by scanning electron microscopy (SEM). SEM has become a powerful technique in materials research [7]; it is based on the principle of electron interaction with matter. It allows obtaining images of the sample surface with high resolution and providing information about the morphology and the chemical composition of a solid material. In our work, the exposed XLPE’s surfaces morphology were studied using Scanning Electron Microscope (SEM, Phillips XL30-type) operating at 20 kV in high vacuum.

3. Experimental Results and Discussions

3.1. Mechanical Properties

3.1.1. Elongation at Break and Tensile Strength

In general, the mechanical properties (elongation at break and tensile strength) of the polymers depend on their molecular weight, the presence of branches, crosslinks [19], crystallite thickness and crystalline – amorphous ratio [20]. These properties are controlled more by the changes occurring in the amorphous phase (phase that chains are in messy) [19]. Many authors have demonstrated that the changes of mechanical characteristics under several constraints are a consequence of chemical changes. However, a correlation between the evolution of elongation at break and tensile strength has been often excluded.

![Fig.4 Evolution of elongation at break according to aging time](image-url)
Figs 4 and 5 depict respectively the elongation at break and tensile strength evolutions. As it can be seen in these figures, the mechanical properties of XLPE insulation decrease with UV-exposure time. The elongation at break dropped from 423% to 350% after 120 hours of UV-exposure and reaches 336% at the aging end. For tensile strength, the decrease is slighter, so it decreases from 11.77 MPa to 11.5 MPa after 120 hours and close the value of 10.20 MPa at the experiment end.

According to the literature [21], [22], [23], [24], [25], the observed facts can be attributed to the XLPE insulation photodegradation which is followed by chemical changes such as chain scission. These alterations lead to the decrease of both amorphous region size and molecular weight which lead to weakness and embrittlement of the material, to which can be attributed the decrease in the elongation at break and tensile strength. Kaczmarek et al. [21] have shown that the mechanical properties deterioration of polypropylene after UV irradiation is a consequence of main chain scission, which takes place mainly in its amorphous phase. Jin et al. [22] have proved that the reduction of mechanical properties can be attributed to the combination of growth in crystallinity and chain scission which gives rise to the stress concentration and crack propagation in material. Eve and Mohr [23] have correlated the mechanical parameters decrease to the reduction of the molecular weight of the polymer material, resulting from the photo-induced chain breaking. Cui et al. [24] have concluded that the embrittlement caused by chemical evolutions is a factor which affects the tensile properties. Al-Salem et al. [25] have deduced that the loss of amorphous regions due to the chemical alterations leads to the change in mechanical characteristics of polymers.

3.1.2. Surface hardness

Generally, surface hardness evolutions are often correlated to degradation. Under UV exposure, it was observed that this characteristic is sensitive to UV-induced degradation [26]. Considering this fact the hardness studies of the UV-exposed and unexposed samples were carried out. Fig.6 represents the XLPE insulation hardness evolution. As it can be seen in this figure, the surface hardness increases slightly. It grows from 54 shD to reaches 58 shD after 100 hours of UV-exposure and remains relatively constant after this period.

3.2. Physical Properties

3.2.1. Water contact angle and work of water adhesion

The contact angle measurement indicates that UV exposure increases the wettability (Fig.7) of the XLPE insulation. Fig.8 shows that the contact angle decrease (wettability increase) is dramatic at the beginning of exposure, it decreased from 92.4° to 54.3° in the first 120 hours of treatment. After this phase, the decrease becomes less marked by increasing the length of UV exposure. The contact angle reaches the value of 38.88° at the end of UV-aging. Identical wettability behaviour was observed by many authors. Waddell et al. [28] have shown that the wettability of UV-irradiated polydimethylsiloxane elastomer increased with an increase in exposure time until the angle reached a plateau; in which no significant changes in wettability were observed. Gaubner et al. [29] have
also noticed a substantial increase in wettability polydimethylsiloxane irradiated at 172 nm.

The contact angle decrease can be assigned to the creation of new polar groups and to the increase of surface roughness. The contact angle measurements results are in good agreement with those reported in previous works. Jofre-Reche et al. [30] ascribed the contact angle decrease to the creation of carbon–oxygen groups and to surface morphology changes (roughness). O’Connell et al. [31] have shown that both polar and non-polar group’s rise leads to the decrease of contact angle in non-polar polymers. Fig. 9 shows that the work of water adhesion behaviour correlates with the wettability increase. So, it increased considerably at the beginning of ageing and slowed thereafter. The rise of work adhesion can be attributed to the photooxidation phenomenon which leads to the increase of surface free energy and polar contents.

2.2.2. Water absorption

Fig.10 displays the evolution of retained water with irradiation time. As it can be seen in this figure, the water content increased rapidly at the beginning of treatment and after that, the absorption process inside the XLPE insulation becomes less important. This behaviour is normal; it has been noted and explained
by several authors. Asmatulu et al. [32] have attributed the increase of polymer coating water retention under UV to the increase of wettability (decrease of contact angle), which lead to water spread on the coating surface. Rosu et al. [33] have assigned the increase of water retained amount to the formation of chemical structures with high polarity as a result of photochemical and photo-oxidation processes.

3.3. Electrical surface resistivity

Surface resistivity or conductivity is one of properties that represent significantly the insulation state. It is frequently used as a diagnostic tool for characterizing aging of polymers under several constraints, in particular under UV radiations [34]. The change of this property is an indication of the physical and chemical properties evolutions.

Qualitatively, we notice that surface resistivity underwent variations with respect to UV-exposure time. Globally, the surface resistivity of XLPE decreases with exposure time. This decrease can be correlated with the decrease of hydrophobicity and the change in the surface chemical structure. Indeed, the photooxidation phenomenon leads to change in the surface chemical structure and to creation and accumulation of surface charges. These charges could lead to an increase of both wettability and surface conductivity.

3.4. Attenuated Total Reflectance (ATR)

In order to identify and to understand the structure changes of XPLE induced by the UV radiations, infrared spectroscopy ATR-FTIR investigations are conducted. Figure 12 shows the transmittance FTIR spectra of unaged and UV-aged XPLE. Globally, spectra of virgin and irradiated samples display similar characteristics, but indicate changes in the intensities of several peaks. The main results are summarized as follow:

a. The observed peaks at 721, 2847 and 2915 cm\(^{-1}\) assigned to the methylene \(\text{CH}_2\) of polyethylene [35], and the \(\text{CH}_3\) peak at 1369 cm\(^{-1}\) decrease,

b. The intensities of the small peaks at 1186, 1264 and 1315 cm\(^{-1}\) assigned, respectively, to C=C bond, Methyl group and Acetophenone group decrease and then disappear for higher expose time,

c. The small peak corresponding to antioxidant at 1129 cm\(^{-1}\) decreases then disappear for higher expose time. Indeed, the antioxidant loss may be responsible of the polymer degradation,

d. The peak at 2361 cm\(^{-1}\) assigned to \(\text{CO}_2\) increases with exposure time. This increase indicates an increase in the photo oxidation activity,

e. The two narrow and weak absorption peaks at 3600 and 3626 cm\(^{-1}\) are attributed to non-hydrogen-bonded alcohols and hydroperoxides [12].

These changes in peaks intensities and the scission of several bonds reveal a degradation of the XPLE after irradiation. Indeed, the chains scission leads to the diminution of the mechanical properties (elongation-at-break and tensile strength). On the other hand, chains scission reactions contribute to the formation of vinylidene and vinyl groups and other unsaturated groups in XPLE chains. These groups are responsible for the color changes of the material.

3.5. Scanning Electron Microscopy (SEM)

Fig. 13. (A-D) shows the micrographs of aged and unaged XLPE samples. As it is observed, no significant changes in surface morphology of the sample are observed. Thus, the surface appearance of all the samples is almost identical. By comparing these micrographs, it is observed that UV-exposed samples showed neither pores nor cracks, but with less smooth and homogenous surfaces than the exposed one. So, the aged samples exhibit slight changes of roughness and color. For longer exposure time, oxidation induced by UV may take place in the amorphous region of the semi-crystalline studied material (XLPE), because the
crystalline region is impermeable to oxygen. On the other hand, the superficial layer crystallizes and shrinks, inducing craters which lead to the material weakness and fragility.

![SEM micrographs of the samples: (A): Unaged, (B): Aged 80h, (C): Aged 160h, (D) Aged 240h.](image)

Fig. 13 SEM micrographs of the samples: (A): Unaged, (B): Aged 80h, (C): Aged 160h, (D) Aged 240h.

The results are in good accordance with those reported previously [36]-[37]-[38]. Many works have shown that no significant changes in occurs in the UV-aged insulators morphology [36]. Venkatesulu and Thomas [37] noticed that UV radiations lead to the roughness increase of the polymeric insulators. Woo et al. [38], when studying the UV-ageing of certain composites, have shown that microcracks started to appear on the material surface after about 300 hours under UV-exposure.

4. Conclusions

In this work, the XLPE insulation behaviour under UV irradiation has been studied. Typical mechanical parameters ( elongation at break, tensile strength and surface hardness), electrical surface resistivity and physical properties (water retention, contact angle and work of water adhesion) have been determined for the high voltage cable insulation. The evolution of these properties has been correlated to the physico-chemical photo-degradation of the insulating polymer. Also, it has been shown that UV exposure of XLPE insulation leads to evolutions in the mechanical properties. The contact angle and work of water adhesion measurements show that UV irradiation increases the wettability of the XLPE insulation. The reduced hydrophobicity is supported by a decrease in the contact angle and an increase of the total water adhesion. The physical evolutions lead to the rise of retained water amount inside the material and to the decrease of electrical resistivity. The surface morphology changes (increase of roughness) showed by SEM micrographs and chemical alterations confirmed by ATR analysis substantiate the mechanical (decrease of elongation at break and tensile strength and increase of surface hardness) and physical (increase of wettability and water absorption) behaviors. Generally, it was showed that UV irradiation leads to an increase of some of the XLPE insulation properties, associated with deterioration of others.

References


