Thermal Degradation Precursor of XLPE Insulation Assessed by Leakage Current Behavior : Effect of Thermal Aging and e-beam Energy in SEM

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Abstract **-** Thermal degradation precursor of XLPE insulation assessed by leakage current behavior has been analyzed in this paper. The effect of thermal aging and e-beam energy levels in SEM has been pointed out. The analysis of our results shows that aging process under high temperatures has a considerable effect on the conduction process of XLPE insulation. We have found that the evolution of leakage current according to the aging time presents a decrease trends at first step of aging and increases at the end of aging time. The decrease of leakage current has been observed after the first 1500 h of aging at 120°C and after 1000 h of aging at 140°C. The same behavior was observed when we have studied the effect of irradiation energy levels. The decrease of leakage current at the beginning step of aging might be assigned to crystallinity increase and elimination of some manufacturing defects. However, the rapid augmentation of leakage current when the aging process is deepening is caused by the augmentation of the conductivity of XLPE. In the other hand, the creation of outside imperfections and oxidized pieces (carbonyl groups) caused by the thermo-oxidation degradation aging process can contribute also in this increase.

Keywords **-** Leakage current, SEM, Thermal aging, XLPE

I. INTRODUCTION

After its first industrial synthesis in 1933, the polyethylene which is the base resin of cross-linked polyethylene (XLPE) takes a big place in the manufacturing of the high voltage cables insulation [1]. The importance of XLPE as an electrical insulation material was proved over the years and its superiority position is remained to this day. These advances were fundamentally built upon materials enhancements and, in particular, introducing new strategies to reduce contamination levels within the XLPE insulation [2].

Whereas contaminations and impurities can be found initially during the manufacturing process, the processes of aging and degradation during exploitation will also contribute to the structure changes which are closely related to the macromolecular architecture and chemical make-up of the polymer. Therefore, each chemical reaction or physical changes lead to changes in macroscopic characteristics.

Thermal aging is one of the harmful phenomena affecting the state of the insulation during the use of the HV cables. Therefore, the almost of its initial dielectric behavior is lost at the end of its lifetime. Many scholars highlight that thermal aging largely generates a big modifications in the chemical and physical structure of XLPE insulation [3-7]. Kim et *al*. [8] highlight in recent study that the undertaken modifications in the chemical and physical behaviors, under thermal aging, leads to many changes in the dielectric characteristics. Many scholars also have been noted that electrical conductivity and dielectric losses factor increase together with aging time under high temperature [9, 10]. In the other hand, Boukezzi et *al*. [11] and Kim et *al*. [8] have been reported that the mechanical properties present some decrease. From another point of view, the leakage current can be considered one of the important indicators of high voltage insulation degradation. It is well known that

the leakage current can flow through the surface and/or the bulk of the materials and this current can have a solid relation with the aging process [12]. Moreover, the leakage current in dielectric materials can be related to the polarization and relaxation phenomena.

The existing correlation between the breakdown of the cables insulation and the leakage and flow of charges under environmental constraints was considered by many researchers [13-15]. The goal of these studies is to find the probably existing relationship between the macroscopic properties (electrical, mechanical,…etc) and the trapping/ detrapping and conduction phenomena in the dielectric insulation. One of the suggested relationships is that the presence of infection and/or chemical or physical imperfections in both surfaces and/or inside the material could be supposed to be a very probable catcher or charge carriers of space charges [16, 17].

Furthermore, beside the manufacturing process of XLPE, which leads to the formation of cross-linking by products, the exposure to elevated temperatures undergoes the thermo-oxidation degradation and generates oxidized pieces like ketones, esters, carboxylic acids and aldehydes [18]. All these imperfections and impurities generated during the manufacturing or aging process lead to the formation of degraded surface layer and can act as traps of space charges or charge carriers (paths) of leakage current.

Many characterization techniques to study the evolution of leakage current and conduction process in dielectric materials have been developed and used by scholars through the word and scanning electronic microscopy (SEM) is one of them. The leakage current measurements were performed with SEM under electron beam irradiation in high vacuum and the charging and discharging processes were observed by time response of current measurements. The main advantageous of this characterization technique is its non-destructive aspect and its principle is based on the evolution of the currents with time. The dynamic feature of this technique authorizes the computation of numerous issues and amounts that describe the conduction procedure in e-beam charged polymers [19, 20].

The presented study in this paper is focused on the analyzing the effect thermal aging under elevated temperatures (thermal constraint) and electron beam energy level on the behaviour of leakage current generated under e-beam irradiation. The leakage current is modulated with the injected electrons by e-beam irradiation and its behaviour is a precursor of the material degradation under thermal aging. The aging process is done in ventilated oven fixed at constant temperature. Two temperatures have been selected: 120°C and 140°C. Both fresh and aged samples were introduced in SEM and irradiated with e-beam energy. Different levels of energy have been applied and ranged from 5 keV to 30 keV. The obtained results will be analysed and discussed in this paper.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURES

A) Samples preparation

The considered samples in this study are prepared from the XLPE commercial insulation used in high voltage cables manufacturing. With a heat press device, samples of 2 mm thickness were prepared from granules of XLPE Extra clean material blinded with 0.2% of Irganox 1035 as an antioxidant and 2% of DCP peroxide as cross-linking reactive.

B) Aging procedure under high temperatures

As we have said in end of the introduction section, the aging process is performed in an air circulating oven fixed at two temperature levels: 120°C and 140°C. These temperatures were chosen to be above the melting temperature of XLPE insulation in order to exercise severing aging condition and accelerate the degradation process. For temperature 120°C the maximum aging time reaches 2500 h, however for 140°C the aging process is performed until 1500 h. After each specific period, 3 samples are used and irradiated in SEM.

C) Leakage current evaluation with SEM

The measurements of the leakage current in SEM are done at room temperature and high vacuum condition near to 10^{-6} Torr. Samples have been introduced in SEM and to electron beam irradiation generated with relatively high energy. The chosen levels of energy in this study to show the effect of e-beam energy on the leakage current behavior are: 5 keV, 15 keV and 30keV under primary current I_0 = 1.5 nA. The used SEM is associated with an adaptive system (Fig. 1) to collect the leakage current. The leakage current I_L is recorded by use of picoammeter

having 10^{-15} A as sensitivity. The description of different parts of the used system can be found and consulted in previous published papers in the literature [16, 21].

Theoretically, when the samples are subjected to the e-beam irradiation in SEM, several currents can be present conjointly in the sample and linked between them with the next equation:

$$I_0 = \sigma I_0 + \frac{dQ_t}{dt} + I_L \quad (1)$$

From equation (1) and Fig. 2, the total incident electrons generate the primary current I_0. Some of the incident electrons are trapped in the materials progressively leading to the variation in the total charge Q_t and hence the creation of the displacement current ($I_d = dQ_t/dt$). Other electrons are created by the secondary electron emission and the backscattered electron emission, and the creation of these electrons leads to another current (σI_0) proportional to I_0 with the so-called electron emission yield σ. Another part of electrons is flown on the surface of the material creating leakage current I_L.

III. RESULTS AND DISCUSSIONS

As the leakage current can be take a big part to mimic the degradation process of high voltage insulations under thermal aging, some results on the behavior of this characteristic will be presented in this section. Two parameters will be considered namely the aging time and the e-beam energy level. First all samples have been well cleaned carefully with ultrasonic vibration to eliminate all contaminations, after that with electron beam generated in SEM, surface of 28 mm² of the samples were irradiated. In Figs. 3, 4 and 5 we have presented the results obtained for 120°C and in Figs. 6, 7 and 8 the results obtained for 140°C. The recorded leakage current with the picoammeter is presented according to the irradiation time.

In the case of 120°C, the depicted results in Fig. 3 show the evolution of I_L according to the time of irradiation. It is clear from this figure that the leakage current is negative for all applied e-beam energy and for different aging times.

Moreover, negative sign of leakage current means that negative charges (electrons) crossing the picoammeter come from the sample to reach the ground. In all cases, the absolute values of the leakage current I_L start from zero when the incident electrons start to reach the surface of the samples and get a saturation state after a very short time when the part of oncident electrons flowing through the surface of the sample is remain constant.

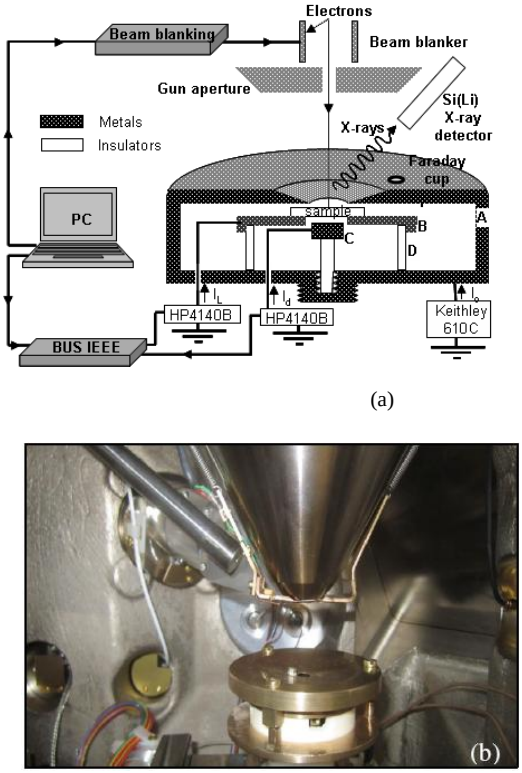


Fig. 1. View of the adopted system measuring the leakage current

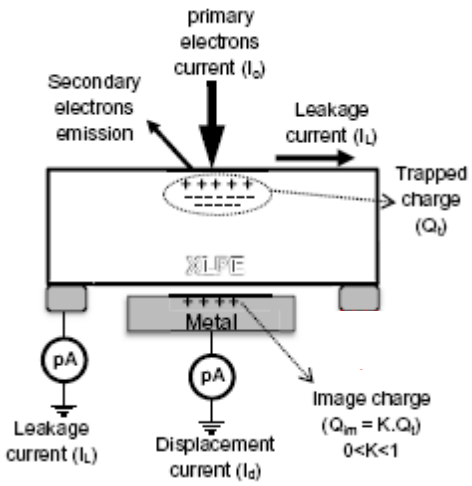


Fig. 2. Generation and measurement of different currents

The same behavior can be observed in the case of aging at 140°C (Fig. 6) with only different values at the saturation.

Obviously, the quantity of the I_L measured with the used system results from the flow of electrons

through the surface or/and through the bulk of the material. The separation between both contributions can't be made easily here. However, the calculated penetration depth of incident electrons *r* using Kanaya & Okayama expression (equation 2) shows that values of *r* are in order of µm which are very less than sample thickness which is 2 mm. Therefore the almost part of I_L results from the drift of electrons through the surface of the insulation. The Kanaya & Okayama expression is given as follow [22]:

$$r(\mu m) = \frac{2.76.10^{-2} A.E_o^{1.67}}{\rho Z^{0.89}} \quad (2)$$

Where : A and Z represent the atomic weight and number respectively and ρ and E_o are the density and the applied energy respectively.

The evolution of the leakage current at the study state with respect to aging periods and energy levels of electron beam is presented in Figs. 4 and 5 for the aging temperature 120°C and in Figs. 7 and 8 for 140°C. Concerning the aging temperature 120°C, the absolute values of leakage current present a few reductions after 1500 h of aging and increase after that (Fig. 4). This behaviour was observed only for 5 keV and 15 keV e-beam energy levels. However for 30 keV level, the leakage current presents a monotonic increase over the aging time. The monotonic increase is probably governed by the enlargement of penetration depth and more charge carriers are reached by the incident electrons and more charges are evacuated through the sample. These charge carriers are enhanced with aging time.

For 140°C the evolution of I_L is presented in Fig. 7. It is very clear from this figure that the leakage current decreases after 1000 h of aging and increases after 1500 h of aging. This behaviour is observed for all applied e-beam energies. The decreasing of I_L after 1000 h of aging at 140°C and 1500 h at 120°C is might be due to the enhancement of crystallinity degree [23]. Furthermore, Teyssedre and Laurent [24] have highlighted in their works that conduction phenomenon in amorphous polymers can happen easier than in crystal polymer. The main cause of this conduction is the presence of more localized levels of carriers in the amorphous parts than crystalline parts.

At the end of aging period, we have observed that I_L presents some increases. The main causes of these increases can be: (i) the conductivity augmentation, (ii) surface imperfection caused by the long-term high temperature application, (iii) the formation of a thickness degraded surface layer and (iv) decrease of viscosity leading to the increasing of the mobility of the charge transporters.

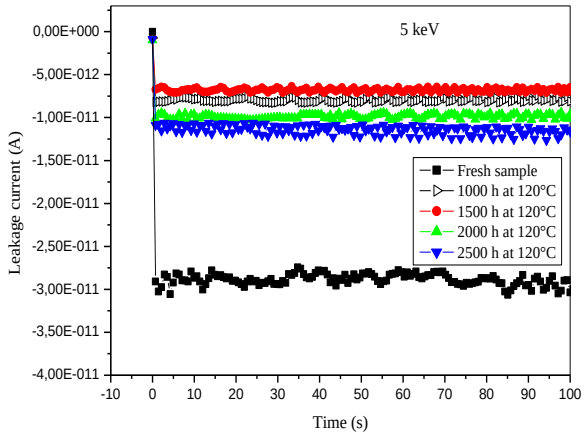
On the other hand, it is well known that during aging process of polymer under high temperatures chemical and physical modification and reactions can be occurred and accelerated as the aging time goes on. One of the important chemical modifications is the creation of carbonyl species, due to the thermo-oxidation degradation, which present a conductive character; hence the significant increment of leakage current after 1500 h of aging at 140°C and 2500 h at 120°C could be ascribed to these created conductive groups.

Figs. 5 and 8 showed the variation of leakage current according to the applied energy level for aging temperatures 120°C and 140°C respectively. By increasing the energy from 5 keV to 15 keV, the leakage current decreases for all aging periods except the case of 2500 h of aging at 120°C. However, when the energy level increases to reach 30 keV the leakage current presents increasing trends. This behavior is monitored by the existing of shallow and deep traps. The detraping of electrons from the shallower traps is easier than the deeper traps.

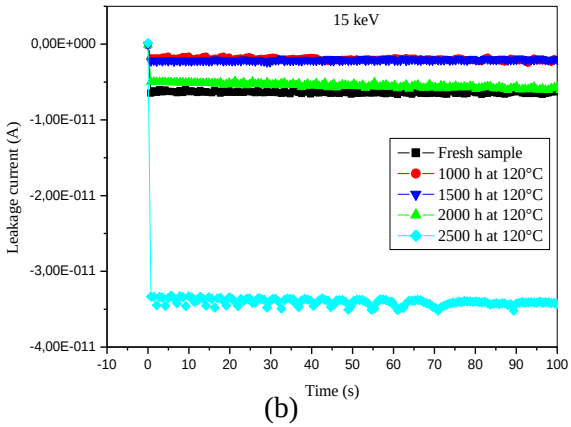
The created impurities due to the aging procedure can provide various kinds of traps. Many scholars [25-27] have evidenced that carbonyl groups generated by the thermo-oxidation degradation can provide traps energy near to 0.90 eV (shallow traps) [25]. The crystal/amorphous interfaces are an important trap sites and they can provide traps energy level of 1 eV (deep traps) [26]. Thus, it is considered that the increase of shallow traps is mainly due to the generation of carbonyl groups during aging. These shallow traps are situated deeper in the surface of the material and when the energy level is higher (example 30 keV) the trapped electrons in these traps are detrapped rapidly and contribute to the enhancement of the leakage current. However, if the energy level is small (case of 15 keV), the reached traps are situated shallower in the surface of the material and these traps are deep traps. The detrapping of trapped electrons in this case is very difficult leading to the reduction of the leakage current.

According to the overall analysis, it has been found that in the early stage of thermal aging, the leakage current decreases in a certain extent. Nevertheless, with the increasing of aging time, the thermal decomposition of the material is accelerated, the

number of small molecules and density of defects and traps in XLPE insulation increase, the electron barrier height decreases and more carriers are excited, which lead to the increase of conductivity. All of these result in the enhancement of relaxation polarization and increase of leakage current.



(a)



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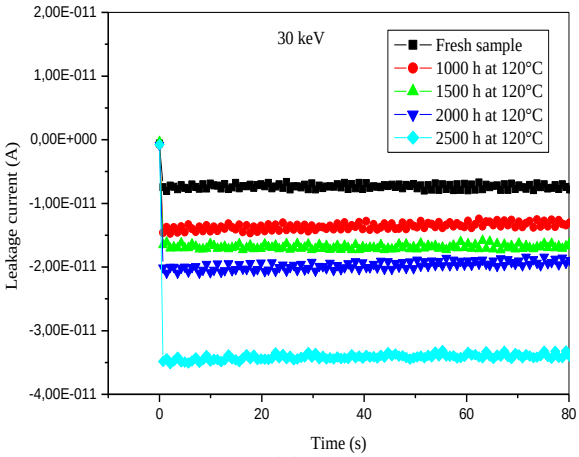


(c)

Fig. 3. Plot of leakage current vs time before and after aging at 120°C for e-beam energy equal to 5 keV (a), 15 keV (b) and 30 keV(c).

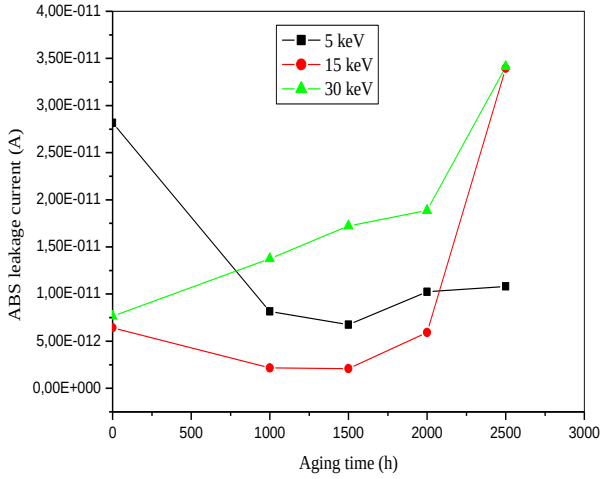


Fig. 4. Evolution of leakage current at steady state vs the aging time for aging temperature 120°C

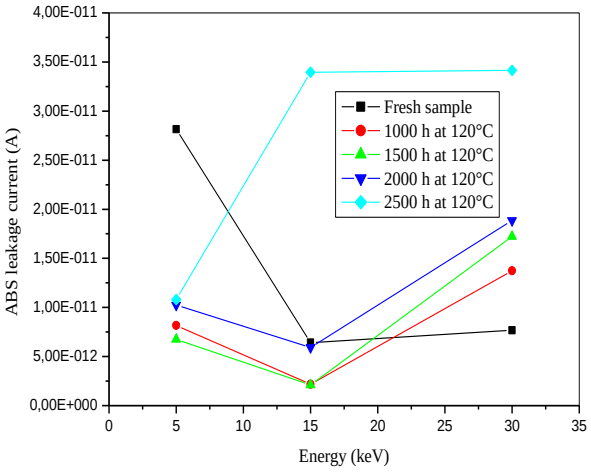
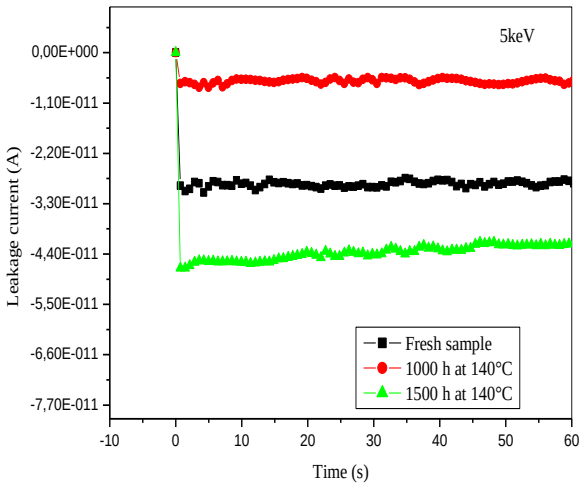
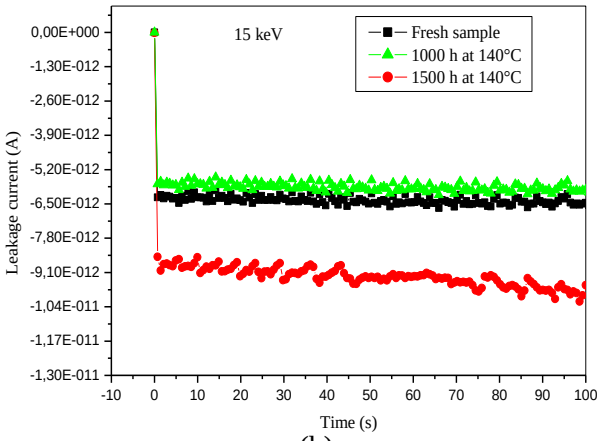


Fig. 5. Variation of leakage current at steady state vs energy levels for aging temperature 120°C



(a)



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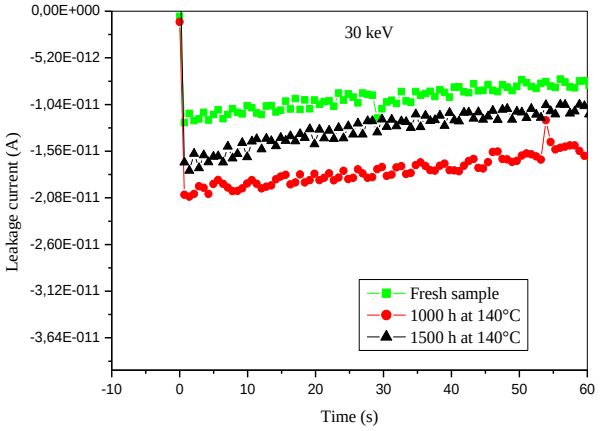

(c)

Fig. 6. Plot of leakage current vs time before and after aging at 140°C for e-beam energy equal to 5 keV (a), 15 keV (b) and 30 keV(c)

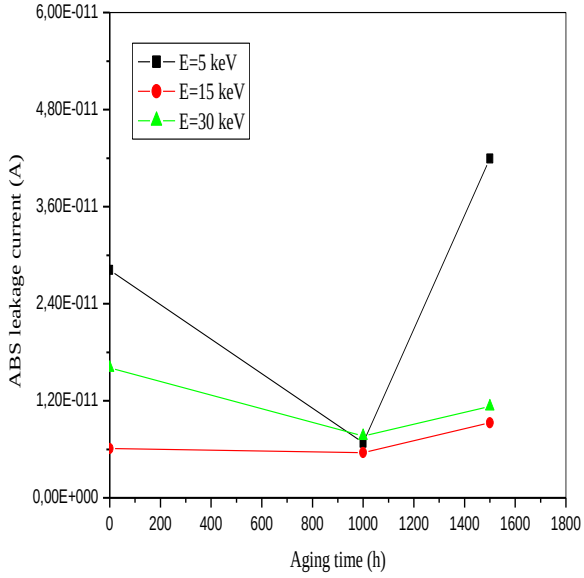

Fig. 7. Evolution of leakage current at steady state vs the aging time for aging temperature 140°C

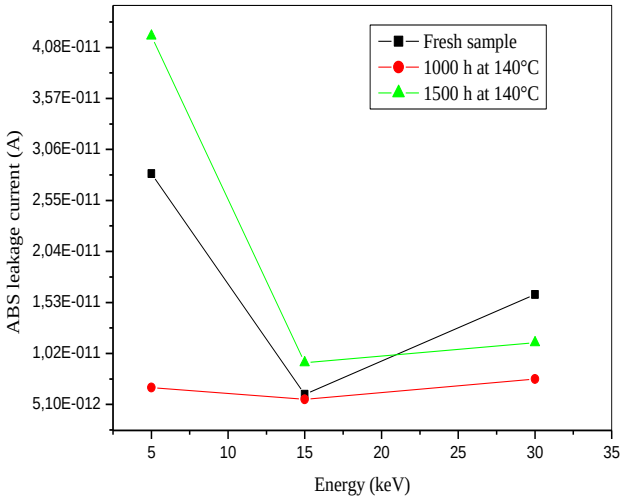

Fig. 8. Variation of leakage current at steady state vs energy levels for aging temperature 140°C

IV. CONCLUSIONS

The presented study in this paper aimed with the analysis of behavior of leakage current in the XLPE under thermal aging to assess the thermal degradation of the material. The leakage current was generated and measured with an adaptive system used with SEM. The obtained results pointed out some interesting aspects. It has been found that the evolution of leakage current according to the aging time presents a decrease trends at first step of aging and increases at the end of aging time. The decrease of leakage current has been observed after the first 1500 h of aging at 120°C and after 1000 h of aging at 140°C. The same behavior was observed when we have studied the effect of irradiation energy levels. The decrease of leakage current at the beginning of aging maybe assigned to crystallinity increase and elimination of some manufacturing defects. The main causes of the leakage current increase at advanced stage of aging can be: (i) the conductivity augmentation, (ii) surface imperfection caused by the long-term high temperature application, (iii) the formation of a thickness degraded surface layer and (iv) decrease of viscosity leading to the increasing of the mobility of the charge transporters. Consequently, the evolution of leakage current under temperature and electron beam energy can be considered as one of the important way and powerful precursor tool to monitor the XLPE thermal aging.

V. ACKNOWLEDGMENT

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