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ELECTROLUMINESCENCE AND SPACE CHARGE MEASUREMENTS FOR THE STUDY OF PEN AGEING UNDER UV IRRADIATION

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UV irradiation is one of the possible sources of electrical insulator degradation. Their origin can be partial discharges, parasitic discharges (environmental) or ambient light. Our intention is to determine the degree of UV interaction, according to the material photosensitivity, on the electric properties in the bulk as well as at the surface, and on the space charges creation. The presence of space charge is now recognized as one of the causes of the electrical insulating material ageing leading to their dielectric breakdown. To highlight this phenomenon, we used a space charge measurement technique in association with the results obtained by electroluminescence in the study of PEN used in the insulation of the certain rotating machines subjected to an UV irradiation. Space charge measurements were carried out using FLIMM (Focused Laser Intensity Modulation Method). This non-destructive technique permits realization of 3D cartographies [1] and localized space charge detection in a solid dielectric. Moreover, electroluminescence (EL) measurements carried out simultaneously emphasize the irradiated zone emission level variations compared to that of unirradiated part. Photoluminescence measurements confirm that the irradiation mainly involves surface modifications of the treated samples.

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1. Introduction

According to the system environment, various mechanisms can be involved in the ageing of insulating polymers under electrical constraints. Among those, UV irradiations are recognized for their degrading effect on many insulating polymeric materials. In the systems such as transformers or rotating machines, the main sources of UV emission are partial discharges. These UV rays will induce chain scission and possibly reticulations and consequently, changes in the chemical properties resulting in the modification of the electrical properties. Poly (ethylene 2.6-naphthalate) films – PEN – were irradiated under controlled conditions.

The main objective of this work was initially to highlight the complementarity of FLIMM technique and electroluminescence in the detection of the irradiated zones, and in a second time to use them in order to evaluate the way of the material electrical property modification induced by UV irradiation, and the probable relationship with chemical degradation. In previous studies [2] on PEN, we have shown that some spectral characteristics of the light emitted under strong fields could be associated with the material degradations. It seemed interesting to be able to use UV photons as material physicochemical property probes. So long as the nature of the chemical mechanism of degradation cannot be easily identified, interest in using various excitations (more or less "strong") probably able to produce significant degradations [3], and the study of their effects with various techniques (FLIMM, electroluminescence or photoluminescence) can only be an advantage.

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2. Experimental procedures

Samples used in these studies were Poly (ethylene 2.6-naphthalate) (PEN) films supplied by Dupont of 25 µm thick from space charge measurements and 50µm thick for electroluminescence investigations. UV aging has been undertaken using a 30W lamp designated for resins exposure and whose spectral characteristics are given in Fig. 1. Some narrow bands appear in the visible range. In the UV one, there is a broad band peaking at 350 nm and half width at mid-height of 20 nm. The transmission spectrum of a reference PEN film is also shown in Fig. 1. As PEN absorbs only in the UV range, the broad band of the lamp spectrum will essentially induce photo-degradation of the material.

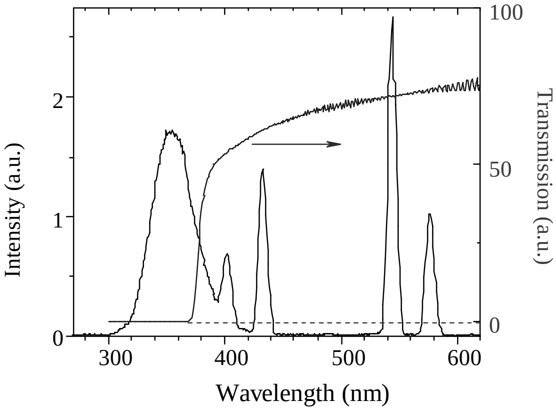

Fig. 1. Spectrum of the lamp used for irradiation (a) and transmission spectrum (b) of a 25 µm-thick PEN film

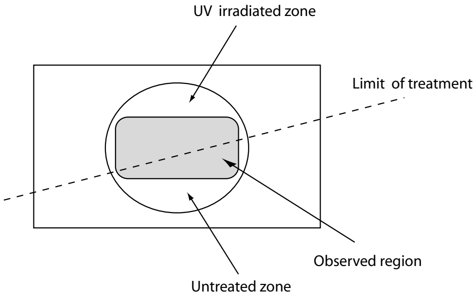

Fig. 2. Samples configuration for EL measurements

Concerning *electroluminescence* measurements, two samples (A and B) were irradiated on only one half surface during 48 hours. Their configuration is shown in Fig. 2. The third sample (C) was totally exposed with the exception of a protected zone of 5 mm diameter.

Once the irradiation carried out, 30 nm-thick semitransparent gold electrodes were deposited on the films. Sample was sandwiched between two brass electrodes with polished surfaces as shown in Fig. 3. The lower electrode was a disk, and the upper one a ring with an inner diameter of 20 mm. In order to minimise the edge effect and avoid discharges, the edge of the metallized area was covered with silicone rubber. Moreover, a plastic cover was used to detect the light coming only from the central part of the sample. The tests were performed in a light-tight chamber under secondary vacuum (<10-4 Pa) in order to avoid discharges.

Imaging of the emitting area was made by using a liquid nitrogen cooled charge-coupled device (CCD camera from Princeton, with a resolution of about 5 nm in the range 300-850 nm). The light was collected along a direction perpendicular to the plane of the sample film. It was either focused on the photocathode of a photomultiplier for photon counting, or deflected by a mirror towards the imaging camera (Fig. 4).

For *space charge* measurements, sample was exposed to UV according to a particular configuration as shown in Fig. 3. The sample was protected by a mask and only three zones were irradiated for different times, respectively 12, 24, 48 hours. Irradiated zones were circles of 1mm diameter.

A semi transparent gold layer (50nm) was deposited on both sample surfaces. In order to make visible the irradiated zones, in accordance with the mask, a very thin carbon layer was next evaporated. The space charge measurements were performed with FLIMM technique. Based on the LIMM technique [4-7], the FLIMM uses an intensity and frequency modulated laser diode ($45mW, \lambda = 658nm$) which can be focused on the sample to be studied. The working frequencies range between 1Hz to 100 kHz. FLIMM set-up is shown in Fig. 5.

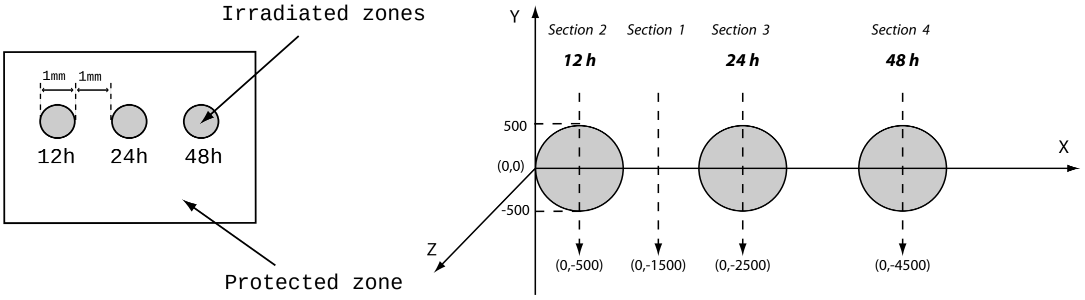

Fig. 3. Sample configuration for space charge measurements.

As a consequence of the created thermal gradient, the induced periodical and local expansions cause a relative charge displacement within the irradiated volume. The lower the frequency, the deeper the irradiated zone. Varying the laser beam modulation frequency, one can control the thermal waves diffusion length and then emphasize the contribution of the corresponding depth into the sample to the total current signal $I(f)$. Under short-circuit conditions, its expression is given by [8]:

$$I(f)=\frac{A}{L}2j\pi f\int_0^L r(z)T(z,f)dz \quad \text{and} \quad r(z)=p(z)-(\alpha_x-\alpha_\varepsilon)\varepsilon E(z) \qquad (1)$$

where $A, L, p(z), E(z), \alpha_x, \alpha_\varepsilon$ are the surface of metallic electrodes, the sample thickness, the pyroelectric coefficient in the direction of the sample thickness Z, the electric internal field, the thermal dilatation coefficient of material and the temperature dependence coefficient of the material permittivity respectively. *T(z,f)* describes the variations of the thermal gradient with respect to frequency and in the direction of the sample thickness Z. Because of its weakness (several pA typically), this current must be electronically well-conditioned by a low-noise wide-bandwidth current-to-voltage converter. The signal is extracted from noise by the mean of a lock-in amplifier and recorded via a PC. Finally, a mathematical treatment allows the reconstruction of the polarization or charge profiles in the direction of the sample thickness [9].

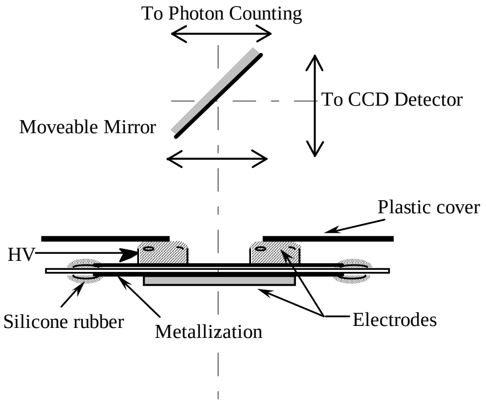

Fig. 4. Schematics of the sample configuration and optical arrangement for EL measurement.

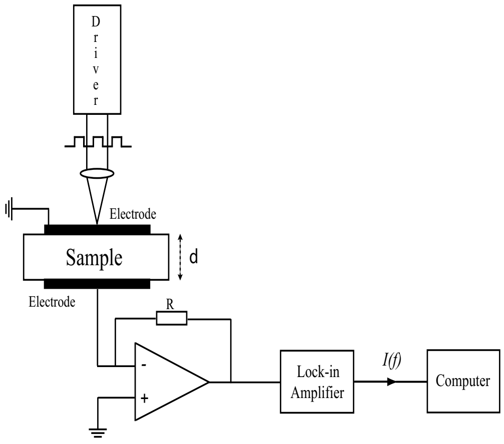

Fig. 5. FLIMM set-up.

3. Results and discussion

Electroluminescence: Samples were submitted to an electric stress (AC for sample A and DC for sample B) by steps of 500V. In this paper, only the more significant images are shown, corresponding to a spaced voltage level. The step duration was chosen in order to allow the acquisition by our measurement set-up (~15 min). Fig. 6 shows several EL images obtained for samples A and B. An important difference in the EL emission can be noted between the irradiated zone and the untreated one, according to the sample configuration (Fig. 2). This phenomenon was observed for both AC and DC stress. According to measurements obtained on untreated samples [10], the emission level under AC is weaker than under DC stress. This result remains valuable for our measurements (Fig. 6). Furthermore, under DC stress, the emission of EL is more homogenous than in the AC situation.

The EL emission for sample C is shown in Fig. 7. For reasons of signal level and facility, the EL measurements carried out on this sample were made under DC field. We notice that the untreated central zone appears very clearly with the voltage increasing and the EL yield presents a higher level than the rest of the surface for all fields. It was shown that the decreasing in the EL emission observed for the UV treated zone can be explain by a decreasing in the conduction current [11].Charges are probably efficiently injected and trapped at the interfaces, so that the interface field is reduced and the injection becomes self-limited. In order to bring an answer to this question, space charge measurements were undertaken.

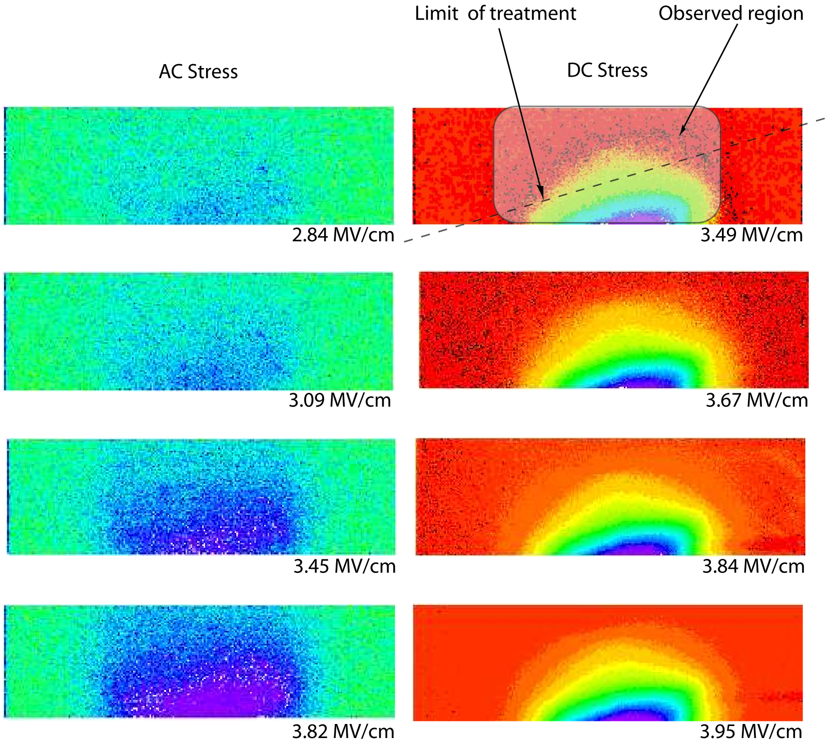

Fig. 6. EL imaging on sample A under AC stress (left) and on sample B under DC stress (right).

Space charge measurements were performed in X and Y directions with a step of 50µm (Fig. 3). The results obtained in both directions are very similar, consequently, only the results got in the Y direction are presented in this paper. Z direction is related to the thickness. Space charge cartographies are shown in Fig. 7. In Section 1, space charge distribution for an unirradiated zone is presented and we consider it as our reference profile. By a visual analysis, one can observe that there is no major difference between Section 1 and Section 2. That means that for low exposure times (<12h), the material is not affected by the UV irradiation. On the other hand, we can notice that the space charge level increases with the exposure time. Starting to 24h irradiation time, negative charges appear in the electrode vicinity, but in an inhomogeneous way. For an irradiation of 48h, space charge level becomes significant compared to the reference profile and the accumulation zone increase.

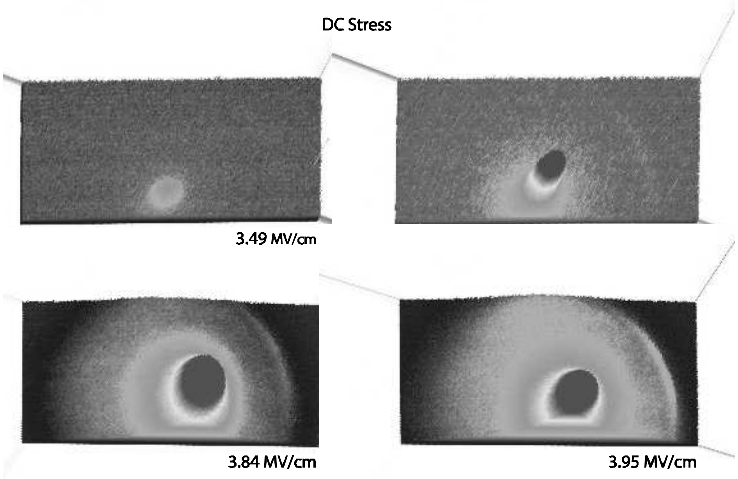

Fig. 7. EL imaging on sample C under DC stress.

Other important observation concerns the space charge depth. An accumulation of space charge near the sample surface is observed in all sections, its amplitude depending on the exposure time. This result was expected considering that the photo-oxidation induced by the UV is a predominantly a surface effect [12]. It was shown, by IR microprofilometry, that the photo-degradation of the material does not exceed 10 µm. This corresponds to our measurements, space charge distribution being confined to the first 7 µm of the PEN sample.

Besides, the degraded surface layer brings further absorption, leading to yellowing, which constitutes a self-screening effect for the polymer bulk. This surface phenomenon was also illustrated in a previous work [11] by photoluminescence measurements.

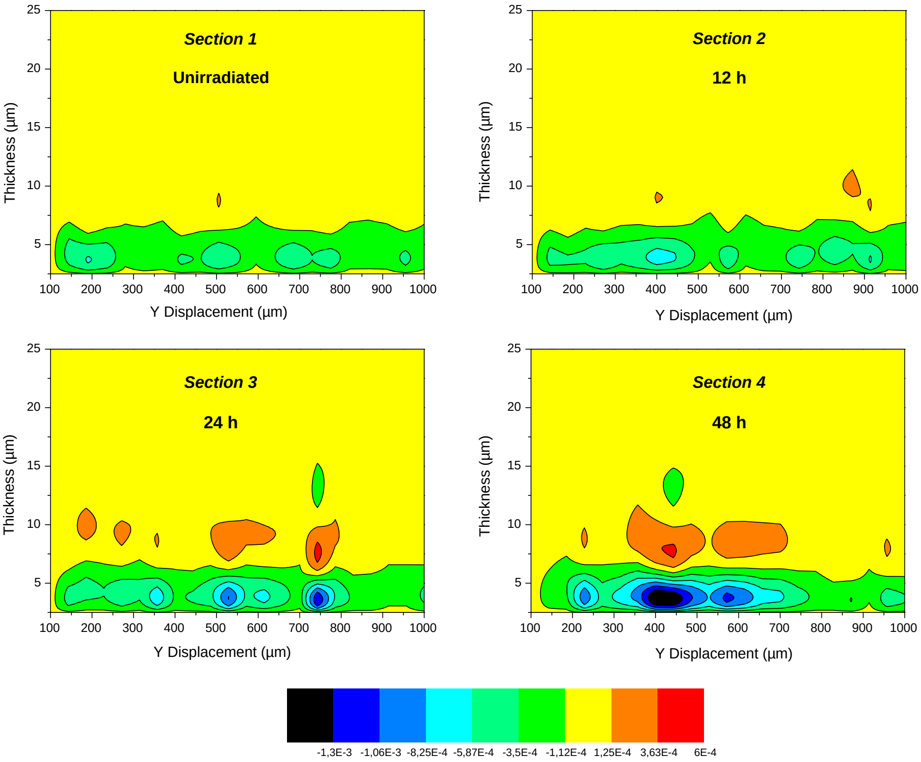

Fig. 8. Space charge cartographies in Y direction for different irradiation times.

4. Conclusion

Electroluminescence and space charge measurements are two complementary techniques used for the detection of UV irradiated zones in polymeric materials. EL imaging under AC and DC stress allows a quickly detection of treated zones. Under DC stress, the EL emission is more homogenous and its level is higher than under AC constrain.

Space charge measurements confirmed the photo-degradation of the treated zones. Under UV irradiation, negative charges are accumulated in a zone near the surface sample. The depth accumulation does not go beyond 7 µm from the irradiated surface. This result is in agreement with photoluminescence measurements and IR microprofilometry. The presence of space charge in the interface vicinity could explain the decreasing in the EL emission for the irradiated zones.

In this paper, we have showed that pertinent analysis of polymer photo-degradation can be performed by these two techniques. Further measurements are under way in order to determine, in a quantitative way, the detection limits (minimal treated surface, irradiation intensity, time exposure).

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