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Study of Trapping Process in BOPP by Coupled Space Charge and Photo-stimulated Discharge Techniques

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Abstract- The global market of biaxially oriented polypropylene (BOPP) film capacitors is growing constantly, as part of energy transition needs. The formation of physical or chemical defects in BOPP can lead to insulator failure due to charge build-up. It is therefore necessary to determine the amount and location of charges and to evaluate the trap energy depths to better understand conduction and aging phenomena. A test bench allowing to do combined space charge measurements by the laser intensity modulation method (LIMM) and optically excited current acquisitions by the photo-stimulated discharge method (PSD) is used to monitor charge accumulation, trap population and de-trapping scenarios. In this investigation, homocharge enhancement near the probed electrode is evidenced in a blank sample. Under light exposure for PSD measurements, charge de-trapping or reactions of BOPP additives modify the charging process.

I. INTRODUCTION

Energy storage and conversion devices are essential in electrical systems and their evolution has been evident as market needs have increased. One particular category is the film capacitors, energy storage elements made of metallized polymer thin films which are typically used in electronic equipment as well as in power applications. A large number of polymeric materials are suitable as insulators in these components, but the most used is BOPP (biaxially oriented polypropylene) because of its self-healing characteristics, high breakdown resistance and low loss factor [1].

The quality of a film capacitor depends on the properties of its components. It is therefore necessary to characterize the insulators to understand their nature, operating mechanisms and the causes of their degradation. Formation of physical or chemical defects affect polymer structure and hence their electrical behavior by modifying the energy levels in the dielectric band gap [2]. Thus, under the influence of an electric field stress, these imperfections can increase carrier mobility and contribute to the formation of shallow (0.1 - 1 eV) or deep (>1 eV) traps [3]. The electrical breakdown of dielectrics is promoted by these microscopic imperfections where charge buildup distorts the electric field and creates electro-mechanical effects that can lead to insulator failure [4]. Reducing trapping phenomena is therefore a way to improve reliability.

Many studies on space charge phenomena focus on the amount and location of charges within the material [5]. For instance, space charge measurements in BOPP usually give the in-depth charge density profile and its analysis allows inferences about its kinetics [6]. However, space charge experiments do not provide information on the energetic state

of the molecular groups acting as traps. In addition, few investigations have explored in the same experimental setup the relationship between charge build-up and energetic aspects of traps. Indeed, alternative techniques allow the estimation of localized states in the energy band model of the insulator which are related to deep and shallow traps. Thermally stimulated discharge currents (TSDC) revealed trap depths in PP the range of 0.3 - 1.5 eV and allowed relating the trapped charges to the structural transitions of the material [7]. Those energy levels reflect physical and chemical disorder in the material, control charge generation, assist transport and explain long-lasting storage of charges.

This research work proposes a test bench that allows the laser intensity modulation method (LIMM) and the photo-stimulated discharge (PSD) measurement to be made without modifying the sample conditions. In this way, electric charge conditions are not affected by extraneous contacts or by the effect of time. Thus, not only space charge formation may be monitored but trapping and de-trapping phenomena.

The impact of irradiation achieved for PSD measurements is considered in this work, revealing sensitivity of BOPP to UV-light. Combined LIMM-PSD acquisitions demonstrate that the amount and distribution of charges near the electrode changes when irradiations between 600 and 200 nm are done. Optically stimulated currents and space charge cartographies are presented, and its evolution is analyzed.

II. EXPERIMENTAL DETAILS

A. Coupled measuring bench

Fig. 1 presents the schematic of the system combining the LIMM and PSD techniques. The controlled switch that allows the selection between each one of the methods has been designed and tested to prevent electrical noise addition on the measured signals. The samples under study are charged by applying a DC electric field through a high voltage source. A RC filter is used to shield and condition the input signal.

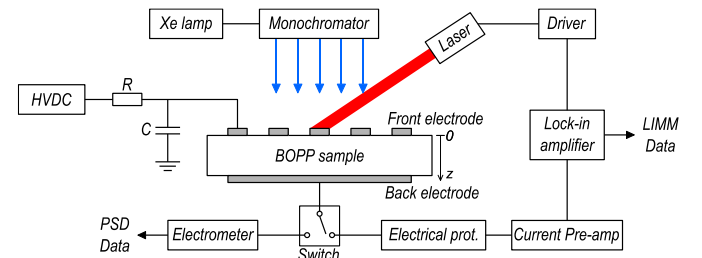


Fig. 1. Schematic of the LIMM-PSD coupled measurement setup.

On the LIMM side [8], a 100 mW 660 nm modulated laser diode is used in the frequency range from 10 Hz to 10 kHz. The back electrode is connected to the switch which, in LIMM mode, connects the electrical protection circuit, the current pre-amplifier and the lock-in synchronous amplifier. The latter allows measurement of the periodic current (decomposed into real and imaginary parts) and transmission of data to a computer for processing. With regard to the PSD technique [9], the monochromatic light is produced by a 150 W xenon arc lamp associated with a 1200 gr/mm monochromator, with entrance and exit slits of 2 mm producing a spectral bandwidth of 7 nm. The incident irradiation is perpendicular to the face of the sample and the irradiated surface is approximately 8 cm². The discharge currents are measured using a Keithley 617 electrometer connected to the back electrode.

B. Material

The samples used in this study are BOPP thin films with a thickness of 17.8 μm and a diameter of 8 cm. 50 nm thick gold electrodes were deposited on each face by sputtering method. The front electrode is connected to the HVDC source and receives the thermal and optical disturbances. Its surface has 4 mm fingers shape leaving areas of the polymer uncovered, so as to improve penetration of incident light in PSD. The back electrode shape is full.

C. LIMM-PSD Protocol

In order to follow charge accumulation and de-trapping phenomena in a sample, the protocol of Fig. 2 is repeated consecutively with varying voltage levels in each cycle. First, six space charge measurements of 10 min each (step A) are performed before charging. Then, the sample is charged for 1 h, under a voltage in the range of 1 - 3.5 kV (step B). The voltage is then set to zero to perform six LIMM measurements (step C). In step D, the PSD spectrum is recorded by scanning from low (2 eV, $\lambda = 600$ nm) to high energy (6.5 eV, $\lambda = 190$ nm) at a scanning speed of 24 nm/min. PSD vs. time at a characteristic wavelength is finally recorded (step E). The procedure is repeated with a different voltage, then with changing the voltage polarity.

III. RESULTS AND ANALYSIS

A. Reference measurements on non-irradiated samples

The current density as a function of the electric field in a blank sample is illustrated in Fig 3. Conduction currents were

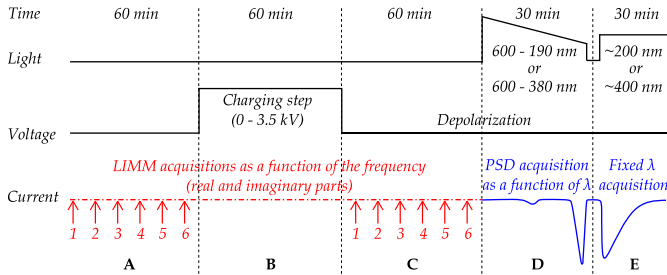


Fig. 2. LIMM-PSD measurement protocol for a single voltage level.

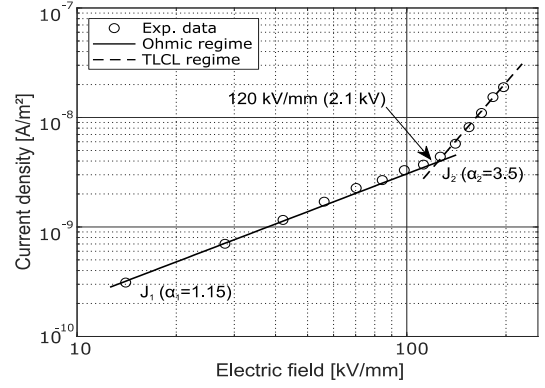


Fig. 3. Current density as a function of field for non-irradiated sample.

measured after 1 h positive polarization steps which were separated by 1 h depolarization periods. Previous reports on conduction in BOPP dealt with hopping conduction at high field [10], and Schottky effects for fields below 100 kV/mm [11]. Considering Fig. 2, two linear regions can be clearly observed in the characteristic. The first region (J_1) is close to an ohmic behavior (slope $\alpha=1.15$ near 1), pointing to negligible injected carrier density compared to the intrinsic conduction. The fast increase of the current density (J_2) for a charging field greater than 120 kV/mm may represent the trap-limited-charge-limited regime (TLCL) with a slope typically between 2 and 4 for traps located in different energy levels. The injected charges become dominant as they fill the traps or contribute to the conduction.

Fig. 4 shows the space charge cartography for all the depolarization steps in a protocol that intersperses charge and discharge steps of one hour each without PSD measurements. Homocharge formation is detected near the upper electrode, where the thermal disturbance is generated. An increasing density of positive charges is observed as the applied field is increased from 2 to 3.5 kV. Then, when the polarity of the source changes, the positive charges gradually disappear, and negative charges take their place. The space charge remains relatively stable on each of the Volt-off steps and the charges get trapped near the surface. Results are in agreement with the injection threshold at 2.1 kV presented in Fig. 3. The space charge near the electrode travels to the deeper bulk as the field is enhanced. This can be explained by the traps which have been filled and allow the passage of carriers to deeper regions. Therefore, the hopping conduction may be favored in the charged region.

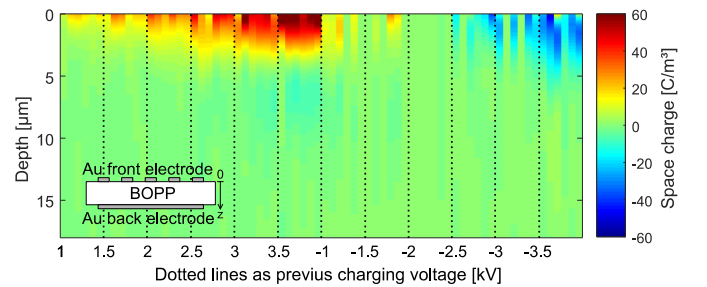


Fig. 4. Volt-off space charge cartography of a non-irradiated sample that was polarized at different voltages for 1 hour each step.

B. UV-vis absorption spectra

In order to investigate the interaction between the irradiated light and the dielectric material, UV-vis absorption spectra were recorded on a BOPP sample. Fig. 5 shows the absorption spectra of a non-metallized as-received sample and illustrates the effect of constant 200 nm light irradiation for 60, 120 and 180 min. The fresh sample presents a narrow, prominent absorption peak around 200 nm and a lighter peak at 280 nm. After irradiation for 60 min, the absorbance peak at 200 nm decreases by about 20%. Moreover, the region from 210 to 380 nm presents an absorption increase. Other irradiations cause slight modifications in the highest peak, but it is clear that the first one caused the greatest impact. These results are consistent with the work of Ho et al. which reveals the presence of antioxidants in BOPP that absorb primarily in the UV spectrum, resulting in the creation of by-products that improve the dielectric strength of the material [12].

C. PSD-LIMM measurements

LIMM acquisition at 0 V on a blank sample (step A in Fig. 2) does not give current signals of sufficient amplitude to perform a correct deconvolution. In fact, measurements with bias voltages below 1 kV do not show significant charge densities and are therefore not presented here. However, the PSD spectrum when scanning from 600 to 190 nm on a fresh sample shows slight peaks at irradiation wavelengths close to 400 nm and 200 nm, the latter being the most intense. Considering that the material has not yet been charged, both of the photocurrents could be explained by one or more of the following phenomena [13]: photoinjection effects where carriers are emitted from the electrode to the polymer when light illuminates the metallic contact; photocarrier generation when excitons are dissociated and interact with trapped charges or impurities; direct photoexcitation of carriers (trapped or not) jumping to the conduction or valence band.

Fig. 6 shows the PSD spectra measured one hour after each charging step (in the order shown in the inset). A first peak is observed between 420 and 380 nm with a maximum between 404 and 396 nm which does not coincide with any absorption band of the material. As long as the involved wavelength is not in the same region as the absorption spectrum, photogeneration of carriers cannot be considered as the mechanism describing

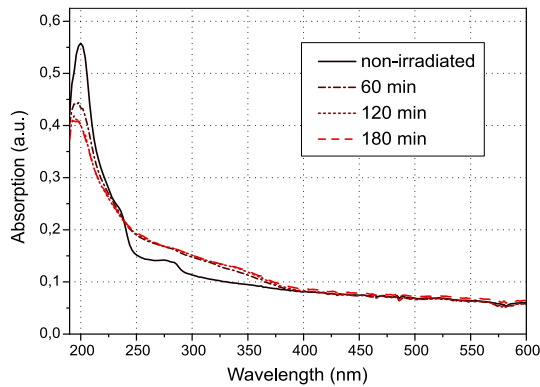


Fig. 5. Absorption spectra from 190 to 600 nm of BOPP film before and after 200 nm light irradiation.

this current response [14]. At this stage, carrier photoemission from the gold electrode or direct photoexcitation can be responsible of the peak near 400 nm. A second, much larger peak is found in the 250-190 nm range, with maximum values between 226 and 200 nm. These PSD peaks near 200 nm could be related to all the mechanisms described above. The fact that it appears in the absorption band reveals that it can be associated to the material photodegradation when exposed to UV rays or to $\pi \rightarrow \pi^*$ transitions in the double bonds of the carbon chains [15]. It is also believed that the dissociation of excitons by the absorption of radiation can explain the PSD phenomena as generated excitons interact with trapped charges or impurities [13]. The illustrated peaks due to the optical disturbance correspond to energies of ≈ 3.3 and ≈ 6.2 eV, respectively.

The inset in Fig. 6 shows the evolution of the two peaks as a function of the applied voltage. After applying the positive voltage steps, the PSD associated to 200 nm shows a significant increase when charging the insulator with 1.5 kV without further growth after. When making the change of polarity, the PSD decreases and does not show additional progress. The peak at 400 nm presents a similar behavior though with lower amplitude. These evolutions in the PSD results are certainly linked to the history of the sample (field strength, polarity, charging time). However, it is evident that a regime change takes place at the charge injection threshold near 2 kV. De-trapping of injected charges in the positive polarization step can be the reason of high amplitude current peaks.

To evaluate the effect of irradiation at a fixed energy level, the PSD current was recorded under monochromatic irradiation at 200 nm as shown in Fig. 7. UV irradiation produces a transient current which tends towards zero when the polarization voltage is less than 2 kV. Beyond 2 kV of voltage, the initial current is practically that measured at the peak of the spectrum of Fig. 6, then it decreases gradually without canceling out after 30 minutes. According to this behavior, subjecting the sample to sufficiently high fields produces more persistent charge trapping. However, the impact of cumulative irradiation remains to be assessed.

The distribution of charge density after each polarization step is shown in the cartography of Fig. 8. Considering Fig. 4, the charge amount in the non-irradiated sample gradually

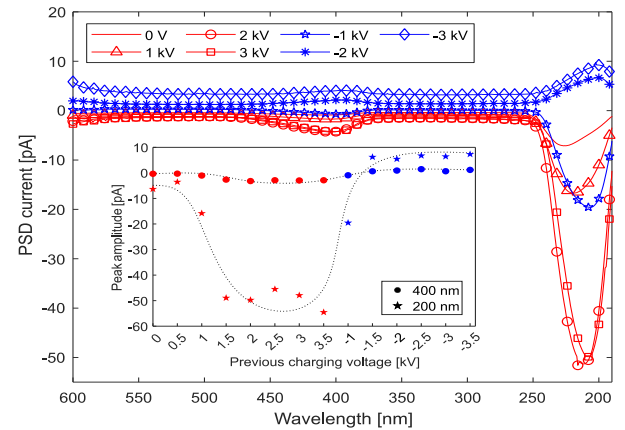


Fig. 6. Evolution of the PSD spectrum vs. previously applied charging voltage.

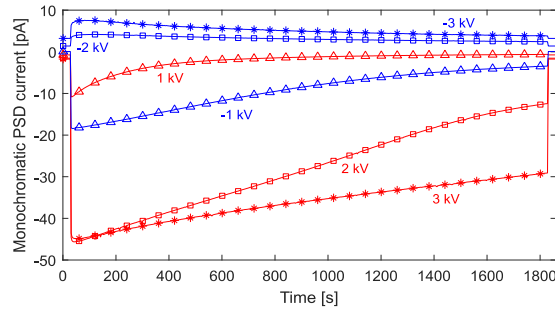


Fig. 7. PSD current under monochromatic irradiation at 200 nm for different previously applied charging voltages.

increases on the front electrode side as the field increases and quickly disappears to give way to negative charges after polarity reversal. In the progressively irradiated sample (by PSD measurements), the homocharge formation is less efficient, as well as the evacuation of the trapped carriers. After polarity reversal (which has been done one week later), positive charges appear in the depth range from 2 to 4 μm in the irradiated sample, contrasting with the results in the reference sample. The space charge accumulation beyond the surface could be explained by the BOPP surface photodegradation upon irradiation at 200 nm. At this energy level, photochemical reactions take place at the surface, and gives rise to other radiation-induced processes across the specimen thickness [14].

The excitation at 200 nm energy can increase the mobility of the previously injected carriers and promote the charge migration, thus causing their stagnation in deeper spaces. While the reference sample shows an accumulation of negative charges at the end of the protocol, this is not the case for the irradiated sample. This indicates that the UV excitation of the material promotes a longer residence time of positive carriers despite the presence of electric fields of opposite polarity. In addition, the transient currents at 200 nm from -1.5 to -3.5 kV (Fig. 7) turn out to be almost zero, thus demonstrating that irradiation under these negative polarity conditions do not generate any perceptible dynamics. Further analysis of the memory effect needs to be done in order to consider the time interval between positive and negative polarization sequences.

III. CONCLUSIONS

The conduction and charge trapping phenomena were investigated on a 17.8 μm BOPP sample. In the range of applied fields, the predominant conduction mechanism is the SCLC

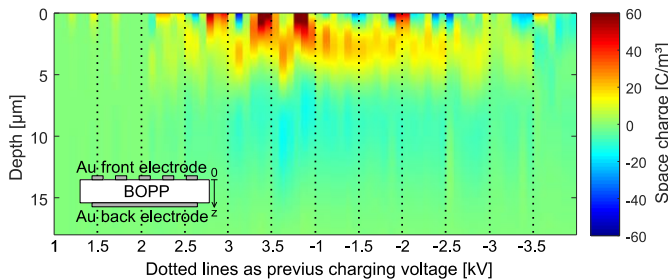


Fig. 8. Volt-off space charge cartography of a PSD (600 to 190 nm) irradiated sample that was polarized at different voltages for 1 hour each step.

model with an injection threshold of 120 kV/mm and traps located in different energy levels. Space charge measurements on a non-irradiated sample show essentially the build-up of homocharges due to charge trapping when the threshold is exceeded. In the combined test bench, PSD current reveal two peaks with apparent trap energies of 3.2 and 6.2 eV. The larger current peak at 6.2 eV could be related to carrier energy transfer processes caused by the transformation of the antioxidant. Irradiation at this energy level promotes deep and lasting charge displacements as revealed by LIMM results. The peak at 3.2 eV can be related to photoemission phenomena or direct carrier excitation with less impact in the current signal. The analysis is still complex as the UV light irradiation used in PSD produces photochemical reactions that may affect trap properties.

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