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Space charge and conduction measurements in gamma-irradiated polyethylene terephthalate

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Abstract—We report on the impact of irradiation on the dielectric properties of Poly(Ethylene Terephthalate)(PET).Thin films of PET were irradiated in air at room temperature by means of a ^{60}Co gamma source at a dose rate of approximately 31 Gy/min. Total doses of 30 kGy and 150 kGy were adopted in this work. The space charge profiles and residual electric field of virgin material and material exposed to these representative doses, obtained after 30 min of depolarization following field application in the range 40–200 kV/mm, have been examined by Laser-Intensity Modulation Method (LIMM). The dependence of space charge density with electric field values and irradiation dose is analysed. Under high DC voltages and after irradiation, the space charge stored in PET tends to be less than in reference PET. The charging and discharging currents tend to increase with the irradiation, considering both the transient contribution to the current and the conductivity contribution. Space charge and conduction measurements exhibit good agreement in the fact that higher conductivity would prevent charge accumulation.

Keywords—Space charge, LIMM, gamma irradiation, PET.

I. INTRODUCTION

Owing to their excellent properties, polymeric materials have a great potential in many important applications with severe service conditions such as in electrical engineering, nuclear reactors or space equipment for examples. In these environments, they are often exposed to X-rays, gamma-rays, UV, protons, electrons, etc [1]. Therefore, much attention has to be given to the study of the behaviour of irradiated polymers, with the aim to evaluate the change of some properties, such as insulation properties of insulated cables, and also mechanical and chemical integrity of the materials, etc. Some of the above stresses are ionizing radiations, meaning that the deposited energy is high enough to extract electrons from atoms of the material and induce chemical changes. Radiation effects on polymers consist in the generation of high-energy electrons causing ionization, bond breaking, the creation of radicals, and the possibility of cross-linking [2].

Through its good electrical, chemical, physical properties, Poly (ethylene terephthalate) has become one of the most used industrial polymers. Exposition of PET to gamma radiation results in the formation of gaseous products such as carbon monoxide, Carbone dioxide (main products), hydrogen, and methane, accompanied by the formation and the scission of intermolecular bonds in the main polymer chain and in side chains [3]. Several experimental techniques have been implemented to understand the changes in electrical properties of polymeric materials (e.g.Isotactic Polypropylene - Poly(ethylene terephthalate) – PET, Poly(ethylene naphthalate) –PEN, low density Polyethylene –LDPE, etc.) after irradiation [2-6], showing that polymers containing aromatic moieties are in general more radiation resistant than those with aliphatic chains.

This paper reports on experimental research on the effect of gamma radiation on Poly (ethylene terephtalate), PET, films, particularly with regards to the space charge behavior and conduction properties. The space charge measurements were performed by the LIMM technique (Laser Intensity Modulation Method), which is a technique suited for the determination of space charge density profiles inside relatively thin (a few tens of micrometers thickness) insulating materials. The investigation is complemented by charging–discharging current measurements aimed at analyzing transport properties of irradiated PET films.

II. EXPERIMENTAL

A. Materials and irradiation conditions

All measurements were performed on 25 µm-thick semi crystalline polyethylene terephthalate (PET) films, supplied by Dupont de Nemours, Luxembourg. The chemical structure of PET is shown in Fig. 1. The samples were exposed to ^{60}Co source using a gamma cell available in Nuclear Research Center of Algiers (CRNA), Algeria, with a source dose rate of approximately 31 Gy/min. Total doses of 30 kGy (16 h irradiation time) and 150 kGy (81 h) were adopted in this work. For all dielectric measurements, 30 nm-thick gold electrodes were deposited on the films. Gold layers were deposited by cold sputtering: each face of the film was exposed to a DC-

powered low pressure argon discharge for 300s. The electrode diameter was 16 mm for LIMM measurements and 10 mm for charging/discharging current measurements. All the experiments were carried out at room temperature.

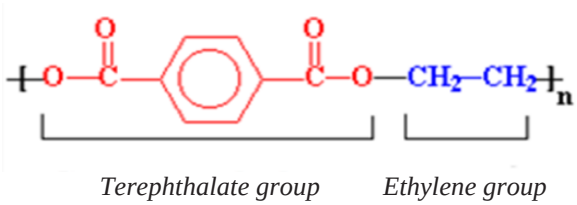

Fig. 1. Chemical formula of Poly(Ethylene Terephtalate)

B. Space charge measurements

Derived from the LIMM technique, the FLIMM (Focused LIMM) technique consists of heating the surface and/or volume of the sample to be studied, using an intensity and frequency modulated laser diode [7,8]. In presence of space charge and/or polarization in the sample, the interaction between the thermal source and the material creates a pyroelectric current resulting from the rebalancing of charges on the electrodes. This current, called LIMM current is detected and analyzed. The spot size can be varied from 1µm to several millimeters [7]. In this work, as our purpose was not to get high in-plane resolution, the spot size was set to about 2 mm, which means that we essentially worked in the LIMM configuration. The modulation frequency range was 10^2–10^4 Hz. The LIMM set-up is shown in Fig. 2. Details of the signal conditioning procedure consisting in converting the LIMM current into the charge profile and field profile based on a modelling of the temperature distribution in the sample is available elsewhere [7, 8], with a recent declination of the LIMM analysis on Polyimide thin films proposed in [9].

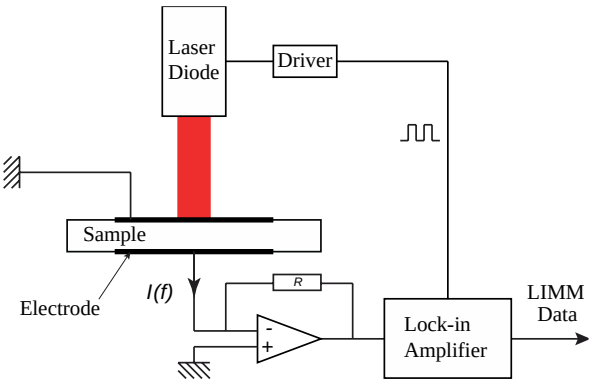

Fig. 2. Schematic of the LIMM set-up

All LIMM experiments were carried out in air at room temperature. Samples were polarized for 30 min with a DC field at different levels between 40 kV/mm and 200 kV/mm. After each polarization step, the sample was short-circuited for 30 min and then the LIMM measurement was carried out so that only residual stored charges were detected.

C. Current measurements

For conductivity measurements, PET samples were prepared as explained above. Polarization and depolarization times were 30 min, and the same field range as for LIMM measurements was considered. The current was continuously recorded and sampled in 2 s intervals using a Keithley 617 programmable electrometer driven by a microcomputer. Charging and discharging current measurements were realized under secondary vacuum at room temperature.

III. RESULTS AND DISCUSSION

A. FLIMM measurements

The space charge profiles and residual electric field obtained after 30 min of polarization followed by 30 min of depolarization are plotted in Fig. 3.Plots (a), (b) and (c) refer to virgin PET and PET subjected to 30 kGy and 150 kGy of gamma radiation, respectively. Data are provided for 5 values of the field up to 120 kV/mm except for the 150 kGy where early breakdown occur. We cannot associate for certain the early breakdown to the irradiation. Positive voltage was applied on the left electrode (therefore anode). The charge profiles were obtained by measurement made on the same electrode. In this zone (at the anode), an accumulation of homocharges (positive charge) can be noticed, for all samples, when the applied field is sufficiently high. It can be noticed that the charge profile recorded under 0-field, i.e. prior to any field application, is not flat, and is of the shape for the three tested samples: possibly the investigated PET films had specific surface properties due to its processing conditions.

For a virgin sample, at low field, the charge density is very weak and can be associated with a dipolar orientation. On the other hand, a massive charge injection (120 C/m^3) appears at 200 kV/mm. The penetration depth of positive charges is low, about to 3 µm from the surface: the depth is not clearly dependent on the applied field. Negative charges are also present in the bulk of the insulation and apparently spread more than positive charges. However, it must be stressed that the exact position of negative charges cannot be easily identified. The spatial resolution of the LIMM method, as for any thermal method, decreases when going from the surface to the middle of the sample, and therefore all interpretations are limited close to the left electrode.

A noticeable decrease in stored positive charge quantity is observed after irradiation of the sample. For a field of 120 kV/mm, the charge density is halved after irradiation. However the evolution with the dose is not obvious. What changes with the highest dose (150 kGy) is a deeper penetration of the positive charge and a boarder space charge peak.

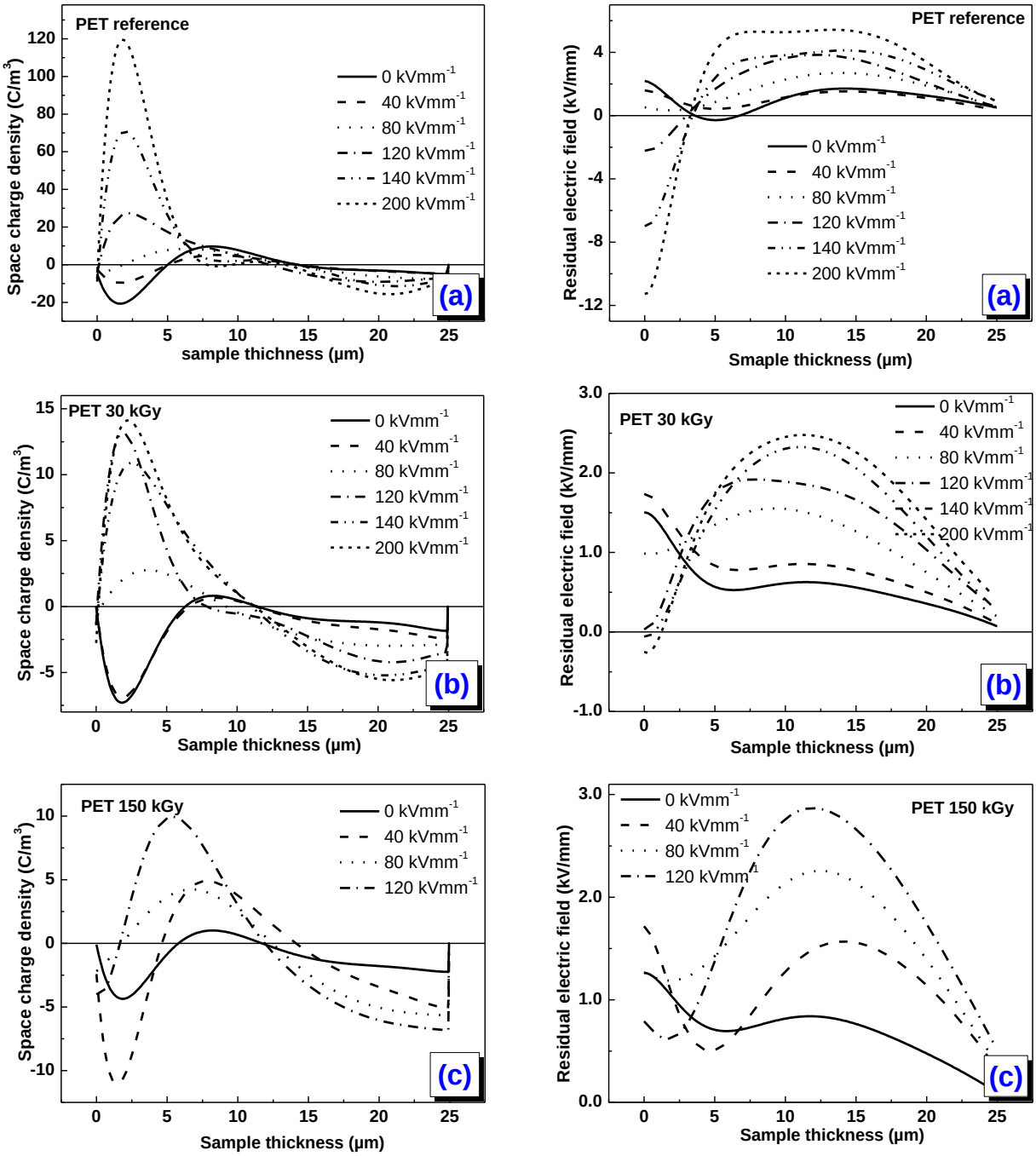

Fig. 3. Space charge density (left) and residual electric field (right) profiles for different applied electric fields (a) reference PET, (b) 30 kGy and (c) 150kGy irradiated samples.
Anode and cathode were respectively at x=0 and x =25 µm during polarization.

Residual electric field values estimated from these charge profiles decrease with the irradiation dose if one consider the field at the anode. However, in the middle of the insulation the variation is mild: the residual field is in the range 2-4 kV/mm for an applied field of 120 kV/mm. In all cases, it represents only a small fraction (<1-3%) of the applied field. This is presumably due to the fact that measurements were achieved a relatively long time (30 min) after field removal.

B/ Charging/discharging current measurements

Charging/discharging current measurements were carried out under vacuum conditions at room temperature. An example of transient current measurements is presented on Fig.4 for the reference PET and PET irradiated with doses of 30 and 150 kGy. Both charge and discharge current are plotted as a function of time, for different values of applied electric field. The superposition of these two currents for low electric field is a sign of

dominant dipolar mechanism which is not the case for high electric field, and for long times.The difference between the charging and the discharging current is more and more evident as time increases. This asymmetry is related to the contribution from conduction process to the charging current.

The transient currents are also defined by the Curie-von Schweidler law with values of the exponent ***n*** close to 1 (cf. Table I). Values of ***n*** were estimated considering the linear part of the decay (in log-log plot). The slope is higher in the discharging than in the charging steps due presumably to conduction processes.

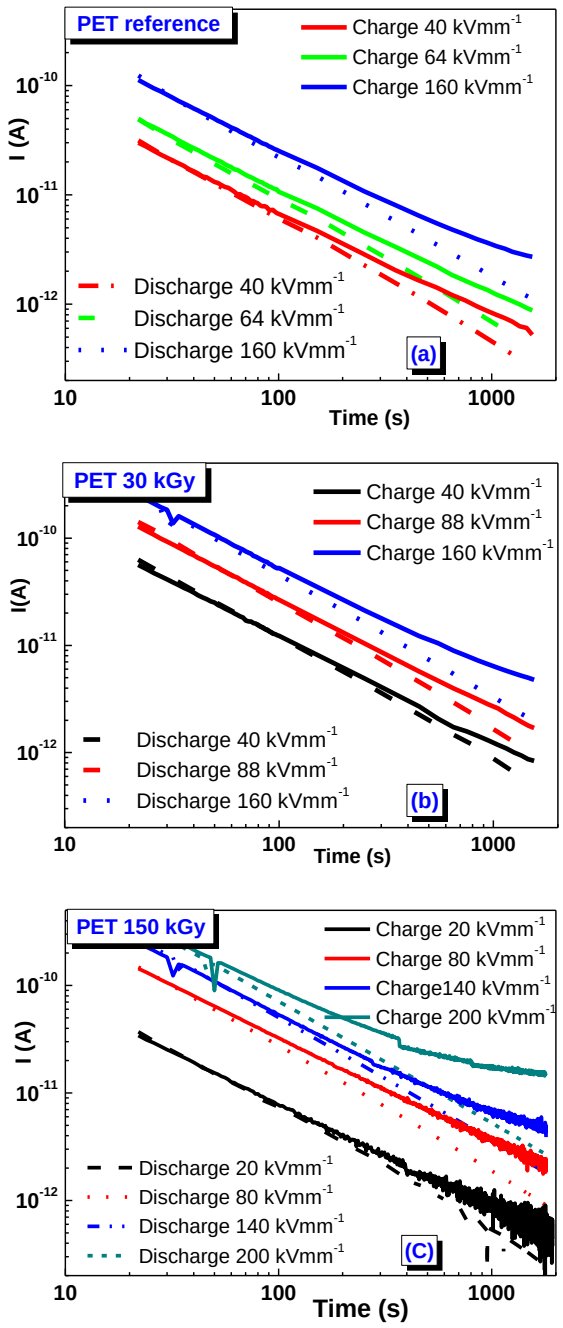

Fig. 4. Time dependence of charge and discharge currents at various electric fields for (a) reference PET, (b) 30 kGy and (c) 150kGy irradiated samples.

It is observed that an increase occurs in the value of the current due to gamma irradiation. The increase is significant considering the amplitude of the transient current. It is also visible on the steady state current viewed as the difference between the charging and discharging currents for a given field. This increase in the charging current can be considered a result of the chain scission of the macromolecules induced by gamma radiation [10] .This chain scission leads to a reduction in the average molecular weight of the PET, which, in turn, increases molecular mobility. Radwan [11] reported that chain scission results in a decrease in the number of valence electrons in PET [11]. Therefore, an increase in the free charge carriers occurred due to this scission, which increased the DC current.

Table I
Values of ***n*** for charging and discharging currents at a field of 40 kV/mm

n / Dose	0 kGy	30 Gy	150 kGy
Charging current	1.10	1	0.94
Discharging current	1.06	1.16	1.16

IV. CONCLUSION

Space charge and conductivity measurements were carried out on samples of virgin and gamma irradiated poly (ethylene terephthalate). The results from the irradiated sample show some differences from the virgin sample. A decrease in the stored space charge density with irradiation dose has been observed along with an increase in conductivity. DC current voltage measurement for the irradiated sample showed a significant increase in current densities for the same applied field. Besides the increase in conductivity, changes in the field dependence of conductivity were observed, conductivity being more dependent on field after irradiation. This constitutes important results, as the dose considered here (150kGy) is mild relatively to that reported (1000kGy) for noticeable degradation of PET. The explanation for the change in electrical properties is believed to be radiation-induced chain scission in PET films.

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