

Charging Properties and Time-temperature Stability of Innovative Polymeric Cellular Ferroelectrets

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ABSTRACT

After appropriate mechanical and electrical treatments, some cellular polymers become able to retain space charge for a long time, i.e. they acquire electret behavior. The electrical treatment consists of charging under high levels of DC electric field. The mechanical treatment, based on the application of stretching forces to cellular polymer slabs that were before expanded under pressurized gas, affects the cavity size and shape, and therefore also the effectiveness of the charging process itself. An investigation of charging mechanisms, as well as of mechanical treatment, is therefore fundamental for optimizing the ferro- and piezo-electret properties. The aim of this paper is to discuss the effect of the physical dimension of the cavities on the charging behavior of cellular ferroelectrets and to focus on the time-temperature stability for two families of polymeric cellular ferroelectrets based on polypropylene (PP) and on a cyclo-olefin copolymer (COC). Emphasis will be given to the stretching process and in particular to the expansion rate applied during the manufacturing process (which affects the radial dimension and the height of the cavities, respectively). Space-charge and partial-discharge measurements as a function of time and temperature are the main tools to infer the influence of the cavity size on charging and stability characteristics.

Index Terms — Ferroelectric materials, partial discharges, piezoelectric films, space charge

1 INTRODUCTION

CELLULAR polymeric materials may acquire piezoelectric and ferroelectric behavior after suitable high-electric-field charging procedures, [1-3]. These kinds of materials are obtained by means of additives (e.g. talc, calcium carbonate, mica, barium sulphate) in polymeric films, which cause, after appropriate thermo-mechanical treatments [1], the formation of micro-cavities. Charging under high electric field will provide space-charge accumulation in these materials and thus so-called ferroelectret and, possibly, also piezoelectret behavior [4-8]. The onset of stable piezoelectric properties depends on the base polymer characteristics, as well as on cavity density and size.

A European Union project (DURASMART: DURABLE cellular polymer films with giant electromechanical response for SMART transducer applications), which has just been concluded, was dedicated to the development and the characterization of expanded cellular polymers that present quasi-piezoelectric properties. The aim of this project was to improve the long-term stability and the operating temperature of these materials, with the purpose to promote their broader application in sensors and actuators.

Interesting results were obtained starting from polymeric materials based on polyesters, cyclo-olefins and polyolefins. In particular, it was found that ferroelectrets obtained from polypropylene exhibit the largest piezoelectric coefficients, which are stable over time up to temperatures close to 50°C, [8-11], whereas electrets obtained from cyclo-olefin copolymers show lower piezoelectric coefficients, but are stable at higher temperatures (up to 110-120 °C), [12].

In the following, results of an investigation relevant to the above materials, based upon space-charge and partial-discharge measurements [13-17], on charge-injection and accumulation mechanisms, as well as assessments of the temperature and long-term stabilities of the electret charge, are presented. In addition, the dependence of the ferroelectret properties on the physical dimensions of the cavities is studied, focusing on the effects of stretching stress and expansion rate applied during the manufacturing process, which affect the radial dimensions and the height of the cavities, respectively.

2 TEST PROCEDURES

Two types of cellular films are taken into account here:

- 100 μm -thick polypropylene (PP) films obtained after an expansion of 200%;
- 60 to 105 μm -thick copolymer (COC) films (a compound including 90% of Topas 6015 and mineral filler), whose expansion rates ranged from 10 to 49%.

Figure 1 displays scanning-electron-microscope (SEM) pictures of a PP specimen (Figure 1a) and of a COC specimen

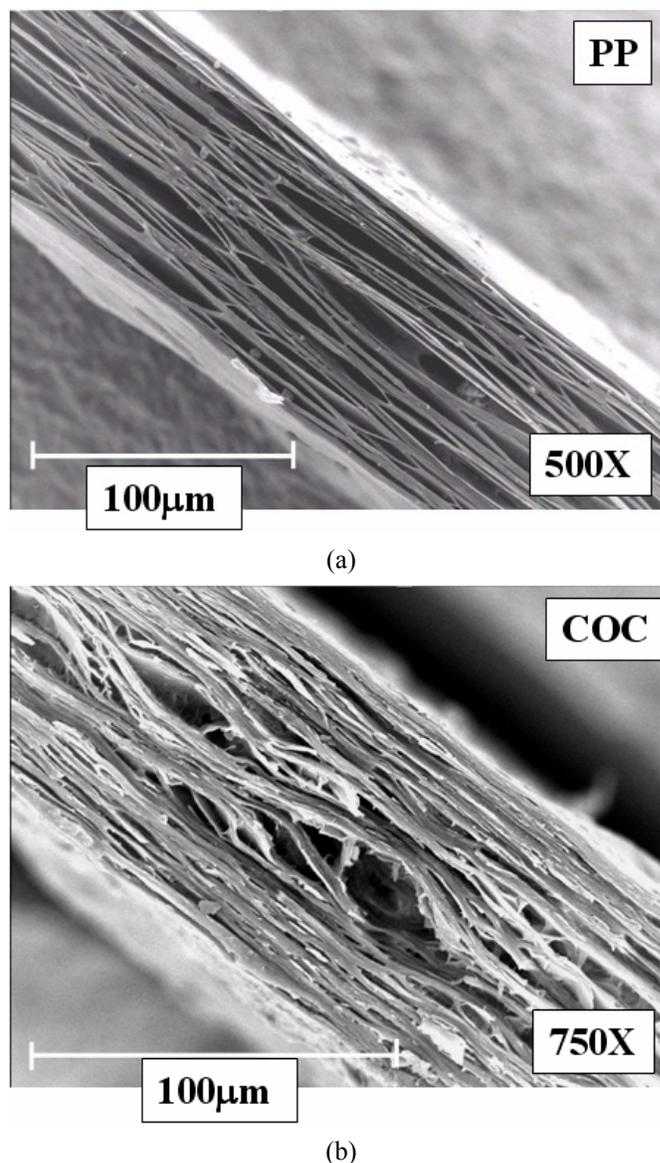


Figure 1. Example of SEM picture of polypropylene (PP) and cyclo-olefin copolymer (COC) films, (a) and (b), respectively, magnification 500 X for PP and 750 X for COC.

(Figure 1b) (expansion rate 24%), showing a large distribution of cavity heights and thicknesses originating from the thermo-mechanical treatment. On the average of several observations, it was seen that COC films exhibit a quite uniform void structure, with relatively few large cavities. The average cavity height for COC is around 1 μm , with a few cavities that can be as high as 10-20 μm . PP specimens display longer cavities, with larger mean height, but a smaller amount of cavities having heights below 10 μm .

Space-charge measurements can be employed to assess the efficiency and the extent of the charging process, by direct measure of the space charge accumulated in the specimens. They permit also to infer the poling field at which a material starts storing charge (threshold for space-charge accumulation, E_{ta}) and the poling field at which the material becomes an electret, i.e. at which steady electret behavior is

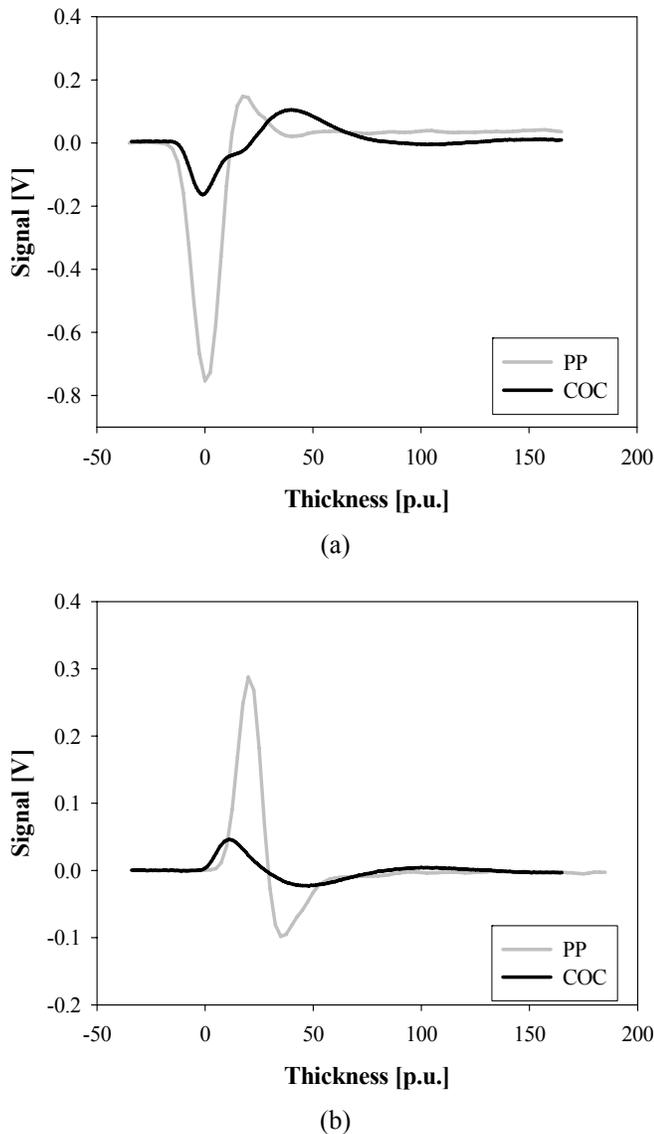


Figure 2. Example of space charge profiles in PP and COC at the end of the polarization (100 kV/mm) (a) and 10000 s after depolarization (b).

established (threshold for the inception of electret behavior, E_{tc}). Space-charge measurements were performed at room temperature, in controlled atmosphere, using a PEA (Pulsed Electro-Acoustic) system [18-20]. The top electrode is a round carbon-black filled polyethylene with a diameter of 10 mm. The bottom electrode is an aluminium plate larger than the specimen. A drop of silicone oil was used to form a film between the specimen and the aluminium plate to ensure good acoustic coupling. The system resolution is better than $0.1 \text{ C}\cdot\text{m}^{-3}$ in charge density and $8 \mu\text{m}$ spatially.

Polarizations at different DC electric fields (ranging from 10 to 100 kV/mm, and lasting 10000 s) were used in order to establish the values of E_{ta} , [13]. After polarization, long-term depolarization tests (with supply voltage removed and electrodes short-circuited) were performed with the purpose to measure the variation of space charge with the depolarization time. These tests allowed an estimate of E_{tc} (i.e. the field

above which charge depletion becomes very small during the test time).

As expected, it was noticed that a space-charge signal is strongly dumped going from the low voltage electrode (the closest to the piezoelectric sensor) to the bulk of the cellular-polymer films, due to attenuation of the pressure wave perpendicular to the voids (caused mainly by the air-filled voids themselves). Therefore, it was possible to obtain information on accumulated charge during polarization and depolarization through the electrode charge and the interface charge near to the electrode closest to the piezoelectric sensor of the PEA cell. As a consequence, calibration was not possible and space-charge signals can be given only in mV. Figure 2 shows an example of space-charges profiles, acquired for both materials, at the end of the polarization (Figure 2a) and 10000 s after depolarization (Figure 2b). As can be seen, the signal is significantly larger in PP than in COC, which would indicate more charge stored in the former than in the latter.

The inversion of the profile between polarization and depolarization can be explained by the example of Figure 3. During depolarization a negative charge peak appears that was hidden under polarization by the electrode charge. This negative interface charge, close to the low voltage electrode, is depleted from the COC, thus reducing interface and image (positive) charge. After some time, positive interface charge is remaining (likely less mobile than negative), together with negative charge spread in the bulk (but still quite close to the low voltage electrode), the sum of which causes almost negligible image charge in quasi-steady state conditions (see Figure 2 and discussion later).

Since the process of cellular-polymer charging is due to micro-plasma discharges occurring in cavities, it was speculated in [13-17] that values of thresholds can be estimated also by means of partial-discharge measurements. Partial-discharge detection was carried out using a direct

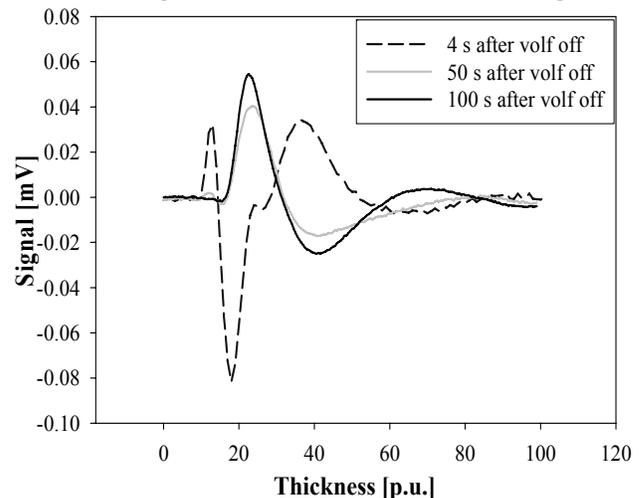


Figure 3. Space charge profiles at the end of polarization and at different depolarization times. COC, 100 kV/mm.

circuit, i.e. connecting the measurement impedance (50 ohm anti-inductive resistor) in series to the specimen and placing in parallel 1 nF coupling capacitor [21,22]. The electrodes of the PD test cell are stainless steel cylinder with pseudo-Rogowski profiles (this profile has the purpose to minimize discharges on the surface of the specimens at electrode boundaries). The sensitivity of the PD detection device is better than 1 pC. Measurements were performed under DC field, from 10 to 120 kV/mm, applying the supply voltage step wise with each step lasting 1500 s.

3 SPACE-CHARGE ACCUMULATION

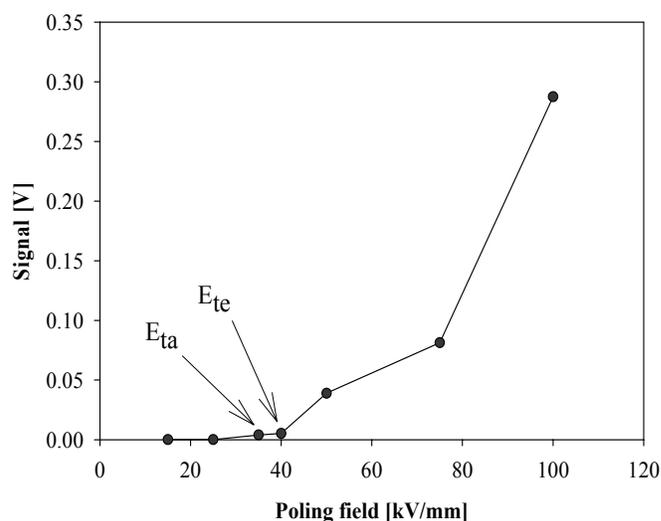
3.1 SPACE-CHARGE MEASUREMENTS

Space-charge accumulation was investigated with the purpose to understand the behavior of PP and COC electrets. Since space-charge accumulation affects the amplitude of the piezoelectric coefficient d_{33} , it can be speculated that the behavior of the piezoelectric coefficient as a function of temperature and time can be evaluated indirectly measuring space-charge accumulation. Actually, the relationship between the stored space charge and the d_{33} coefficient is given by [8]:

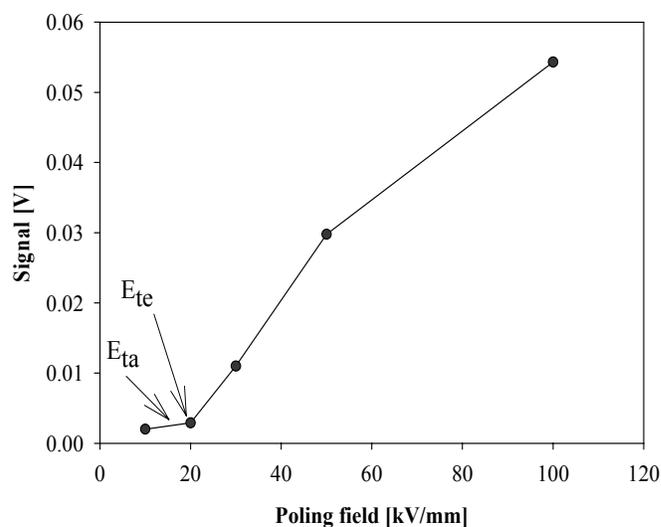
$$d_{33} = \frac{\varepsilon \cdot \sigma}{Y} \cdot \frac{1 + \frac{s_2}{s_1}}{\left(1 + \varepsilon \cdot \frac{s_2}{s_1}\right)^2} \quad (1)$$

where ε is the permittivity of the solid material, Y is its Young modulus, s_1 and s_2 are the total thicknesses of the solid and gaseous layers respectively and σ is the charge density on the surface of each gas-filled cavity. Equation (1) has been obtained under the assumption that the cellular ferroelectret consists of plane-parallel solid and gas layers, with the solid layers charged to one polarity on their top surface and to the other polarity on their bottom surface. Even if space charge measurements can not provide information on mechanical properties of the cellular material or on the actual amount of accumulated charge, the direct proportionality between d_{33} and the stored charge (here quantified mostly by the charge measured at the electrode/dielectric interface during depolarization) may allow one to speculate that materials with larger space-charge accumulation after the charging process might possess larger d_{33} coefficients. In addition, it can be argued that time and/or temperature variations of the signal detected by PEA will indicate a similar variation of the piezoelectric coefficient.

Under this perspective, it is noteworthy that measurements performed at 20 °C show a greater amount of charge on PP specimens, even if COC starts to accumulate charge at lower values of the electric field (i.e. it has a lower E_{ta}). This difference between space-charge signals comes out clearly in the space-charge profiles of Figure 2 obtained during polarization at 100 kV/mm and during depolarization. More



(a)



(b)

Figure 4. Threshold characteristics: amplitude of the positive charge peaks at the electrode/dielectric interface as a function of poling field, 20°C. PP, (a), and COC, (b). The thresholds for the accumulation of space charge (E_{ta}) and for the inception of steady electret behavior (E_{te}) are indicated by arrows.

generally, Figure 4 depicts the amplitude of the positive charge peaks at the electrode/dielectric interface, 1000 s after the beginning of depolarization, as a function of the poling field for PP (Figure 4a) and COC (Figure 4b) (threshold characteristic). As can be seen, PP specimens accumulate a larger amount of space charge compared to COC for poling fields higher than 50 kV/mm. The threshold for space-charge accumulation E_{ta} is about 35 kV/mm for PP and about 15 kV/mm for COC. The thresholds for electret behavior shown in Figure 4 are obtained from the depolarization characteristics, that is, from the graphs which reveal the amplitude of the positive charge peaks at the electrode/dielectric interface during depolarization as a function of time: see Figures 5a and 5b for PP and COC, respectively. As can be seen, above 40 kV/mm for PP and about 20 kV/mm for COC, charge is not deployed anymore

from the specimens, which are thus stable for the whole test time. An exception is apparently found for COC at fields > 30 kV/mm (Figure 5b), where an initial enhancement of space charge can be noted. As mentioned before, this is likely due to the simultaneous initial presence of positive and negative charges having different depletion rates. Since the PEA system can measure only the net charge, a decrease of negative charge is detected as an increase of positive charge.

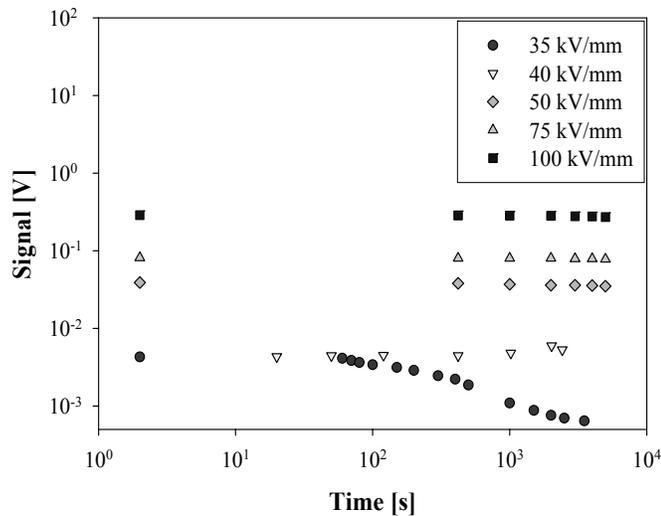
In order to understand which is the prevailing cause of such space-charge accumulation that leads to ferroelectret properties, e.g. cavity discharges or injection, partial-discharge measurements were performed.

3.2 PARTIAL-DISCHARGE MEASUREMENTS

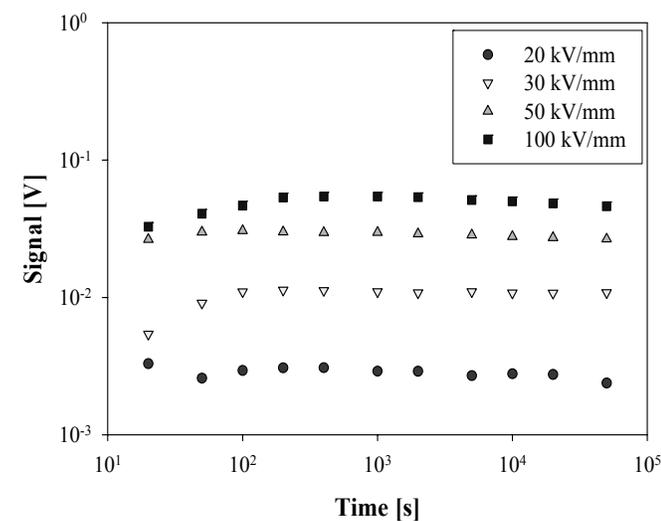
If the process of cellular-polymer charging is due prevalently to micro-plasma discharges occurring inside the

cavities [13], the values of E_{ta} can be determined by PD measurements and should coincide with those obtained from space-charge measurements. Figure 6 shows the number of PDs detected over a period of 1500 s during DC polarization, at different poling fields at 20 °C, for PP and COC, respectively. Comparing Figures 5 and 6, it can indeed be noted that the values of E_{ta} determined from PD measurements coincide with those estimated from space-charge measurements.

On the basis of space-charge measurements, it can be concluded that COC films store less space charge than PP films (and would thus very likely have lower piezoelectric coefficients, see also the next section), but the threshold for the inception of steady electret behavior (E_{tc}) is smaller in COC than in PP (20 kV/mm vs. 40 kV/mm). This might be associated with the distribution of cavity dimensions. As observed from Figure 1, PP shows cavities with larger mean

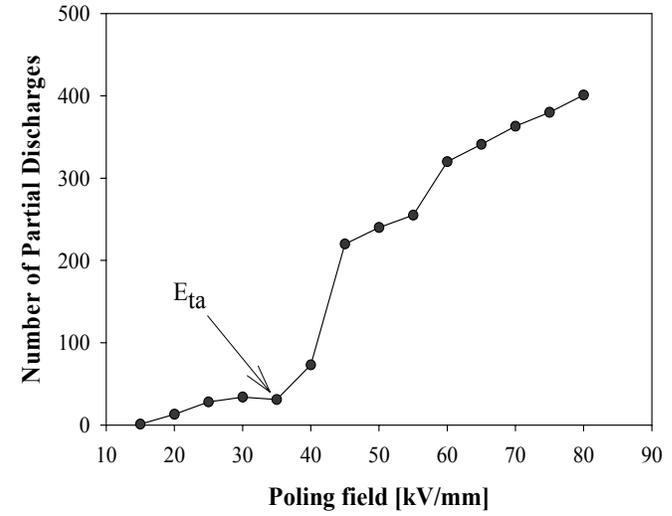


(a)

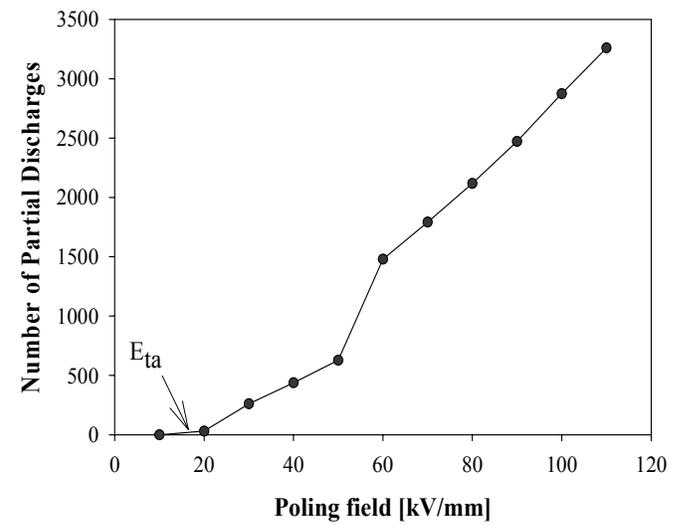


(b)

Figure 5. Depolarization characteristics: amplitude of the positive charge peaks at the electrode/dielectric interface as a function of the time (during depolarization), 20°C. PP, (a), and COC, (b).



(a)



(b)

Figure 6. Number of partial discharges detected in 1500 s during DC polarization, at several values of poling field, 20°C. The thresholds for the accumulation of space charge (E_{ta}) are indicated by arrows. PP, (a), and COC, (b).

height, with respect to COC, but COC has a number of cavities with height $\geq 10 \mu\text{m}$ (i.e. suitable for the inception of partial discharges) larger than PP, thus it can be argued that micro-discharges would already start at lower electric fields [17]. Therefore it can be speculated that an appropriate mechanical treatment, including stretching and gas expansion, would be able to affect the charging properties and the stability.

4 TIME-TEMPERATURE STABILITY

4.1 THERMAL-ENDURANCE CHARACTERIZATION

The thermal endurance of COC was investigated according to the procedure specified in the IEC 1026 Standard (see [23]). It consists of measurements of the activation energy of the thermal degradation process through isothermal thermogravimetric analysis (ITGA), besides a conventional thermal endurance test at a given temperature, in order to be able to draw a thermal endurance line of the tested materials at relatively short times [24]. Considering the results of the conventional life tests carried out at 130°C , a decrease of weight (diagnostic property) of 2% (end point) corresponds to a mean life of 730 h. The analytical test (ITGA) yielded an activation energy of the thermal degradation process of 3.7 eV. Such data translated, through the Arrhenius law [25], into a Temperature Index (TI) for the tested COC of 131°C , with a Halving Interval (HIC) of 2.4°C . The same procedure was also carried out on expanded cellular PP, providing a temperature index of 95°C and a halving interval of 8.0°C . The thermal endurance characterization shows, therefore, that COC has a larger Temperature Index and a smaller Halving Interval than PP, so that it can be speculated that it has a better long-term thermal stability.

4.2 SPACE-CHARGE MEASUREMENTS

Temperature and time stability were also investigated by means of space-charge measurements, by recording the residual space charge after thermal treatment for long times in cellular specimens. After polarization at 100 kV/mm for 10000 s at 20°C , specimens were placed in an oven with their electrodes short-circuited, in the absence of an external electric field. Various specimens of COC were tested, differentiated by expansion rate (E) and stretching force (F) applied to specimens during manufacturing (the values of E and F for each specimen are given in Table 1). PP specimens were exposed to 50°C , whereas COC specimens were subjected to 110°C . Space-charge measurements were performed at fixed time intervals and 20°C , in order to detect the residual charge.

Table 1 shows the physical characteristics of the specimens (percent expansion rate and stretching force) and some of the electrical quantities measured during the tests, that is, threshold for the inception of steady electret behavior (E_{te}), number of partial discharges (N_{PD}) detected over a period of 1500 s during polarization at 100 kV/mm and amplitude of the

Table 1. Per cent expansion rate (E), stretching force (F), threshold for the inception of steady electret behavior (E_{te}), number of partial discharges (in 1500 s) detected during polarization at 100 kV/mm (N_{PD}) and amplitude of space charge signal after 1000 s of depolarization (V_{SC}) for ten specimens of the family COC.

Specimens	E [%]	F [N]	E_{te} [kV/mm]	N_{PD}	V_{SC} [mV]
# 1	22	46	20	750	98.0
# 2	10	51.5	25	406	95.9
# 3	11	36	25	325	94.0
# 4	27	68.5	25	377	124.0
# 5	49	47.5	20	1050	90.0
# 6	16	48	25	775	100.0
# 7	21	43.5	25	350	96.0
# 8	34	50.5	20	557	95.0
# 9	36	49	20	830	100.0
# 10	39	47	20	550	97.6

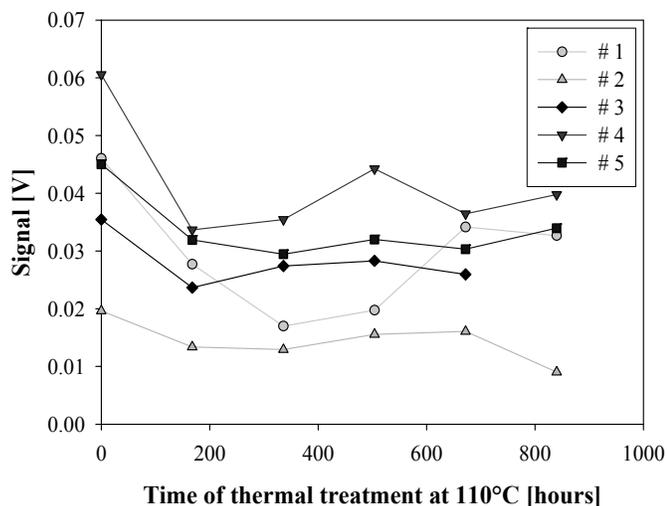
space-charge signal (V_{SC}) after 1000 s of depolarization, upon poling at 100 kV/mm .

Figures 7a and 7b show the results of such measurements. As can be seen in Figure 7a, space charge accumulated in PP quickly decreased when the charged specimen was kept at 50°C . After 220 h of exposure, only 10% of the initial charge was retained. The measurements performed on COC specimens, Figure 7b, show an initial decrease of space charge during the first week of heating, but afterwards the space charge appears to be stable with time. After 840 h of testing, more than 50% of the initial space charge is retained in all specimens. In particular, the sample # 5 maintains 75% of the initial space charge after 840 hours at 110°C . It can be noted that all specimens exhibit an enhancement of the positive charge peaks at the electrode/dielectric interface after some hundred hours in oven. As mentioned above, this can be explained by the faster expulsion of negative charge and the consequent enhancement of positive net charge (which is the quantity detected by the PEA system).

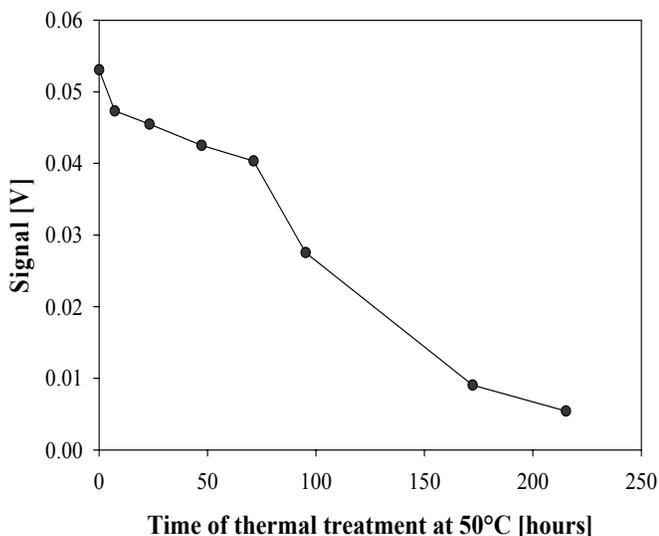
These results agree with [26] and seem to be consistent with the higher value of TI measured for COC. Therefore, space-charge measurements support a much better temperature-time stability of COC in comparison with expanded cellular PP.

4.3 PIEZOELECTRIC MEASUREMENTS

Studies of the time-temperature stability based on piezoelectric (d_{33}) measurements were performed on COC with the aim to confirm its good stability found through thermal endurance characterization and space-charge measurements. Long-term thermal stability tests of the electromechanical properties were performed at 80°C and 120°C on COC after suitable charging. Figure 8 displays the results of d_{33} measurements carried out on several specimens of COC (with different cavity-height distributions); in addition, the value of the d_{33} coefficient for PP measured at 20°C is reported as a reference. As can be seen from Figure 8a, the thermal treatment at 80°C does not cause a significant decrease of the electromechanical activity of COC films,



(a)



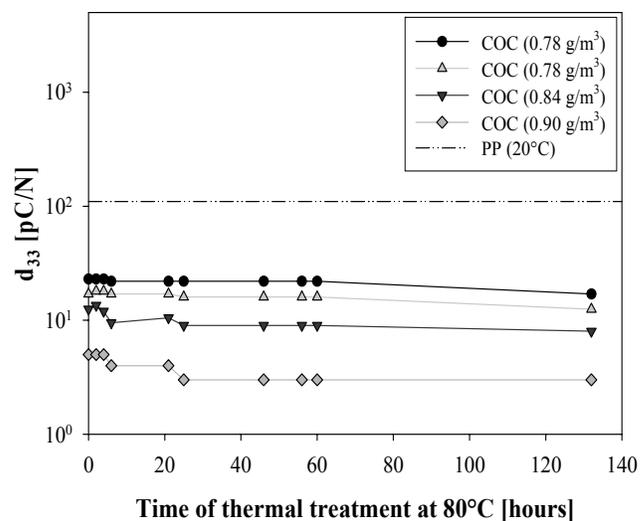
(b)

Figure 7. Amplitude of the positive charge peaks at the electrode/dielectric interface in COC specimens, as a function of expansion rate and stretching force applied during manufacturing (see Table 1), subjected to 110°C, (a), and in a PP specimen subjected to 50°C, (b), in the absence of external applied voltage (poling at 100 kV/mm).

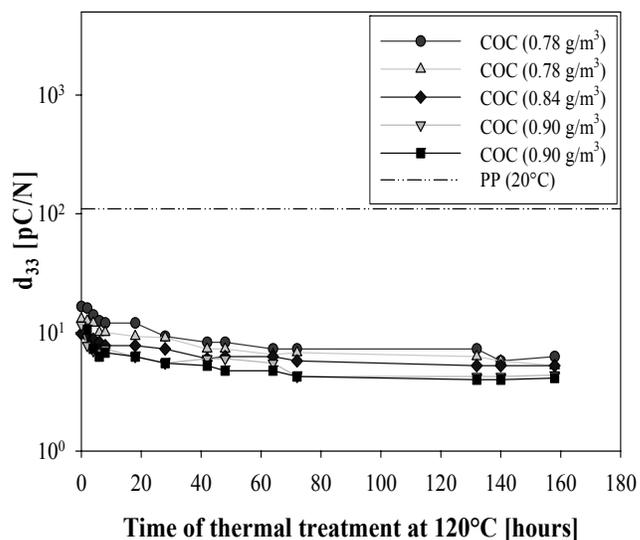
while the thermal treatment at 120 °C leads to a decrease of the electromechanical activity within the first hours, reaching then relatively stable values which are around 50% of the initial values measured after electric charging at room temperature, (Figure 8b). Therefore, for such materials an industrial pre-ageing would lead to thermally stable transducer films. However, at temperatures lower than 50 °C, the sensitivity of COC is smaller than that of PP.

4.4 CONDUCTIVITY MEASUREMENTS

The better time-temperature characteristics of COC with respect to expanded PP might be explained partly by the conductivity difference between COC and PP at high



(a)

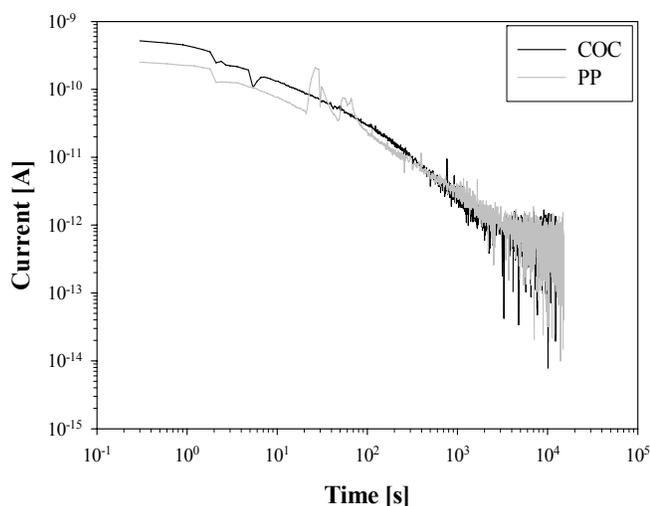


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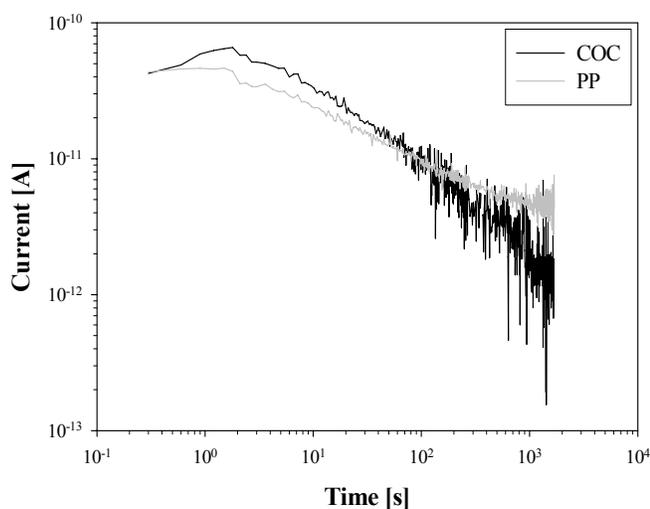
Figure 8. Long-term time-temperature stability of d_{33} coefficient in several COC specimens, subjected to 80 °C (a) and 120 °C (b), at different exposure times (as a reference the d_{33} of PP at 20°C is also reported).

temperatures, particularly by the slower increase of the conductivity of COC than PP when the temperature increases.

In order to verify this hypothesis, charging-current measurements were carried out on base PP and on COC at 20 °C and at 70 °C, at 10 kV/mm. Figure 9 displays the results of these measurements. As can be seen in Figure 9a, at 20 °C PP and COC show the same charging-current behavior and amplitude (the latter slightly larger for COC than for PP), whereas at 70 °C, after 1000 s of polarization, the current in PP is about three times of the one found in COC (Figure 9b). Therefore, it can be argued that due to its low conductivity, COC can keep charges deeply trapped also at high temperatures, thus preventing charges from being detrapped. This contributes to the better time-temperature stability of COC.



(a)



(b)

Figure 9. Charging current measurements on base PP and COC, poled at 10 kV/mm, 20 °C (a) and 70 °C (b).

5 EFFECTS OF THE PHYSICAL DIMENSIONS OF THE CAVITIES ON THE CHARGING BEHAVIOR

The effect of the physical dimensions of the cavities on the charging process was investigated focusing on the dependence of some electrical quantities on the stretching stress applied during the manufacturing processes (which is associated with the radial dimensions of the cavities) and on the expansion rate (which affects the height of the cavities). In fact, during the manufacturing of the electrets, the stretching force is applied in the direction parallel to voids, with the aim to increase the radial dimension of cavities. The expansion of the gas inside the voids produces an increase of the height of the cavities, since it acts perpendicularly to the voids surfaces. The expansion rate is calculated as the ratio between the

increase of the specimen thickness during the expansion process and the thickness before the expansion. Experiments were carried out on ten specimens of COC, differentiated by expansion rate (E) and stretching force (F), having all the same initial dimensions before application of stretching force.

The threshold values for the inception of steady electret behavior (E_{te}) reported in Table 1 were obtained by means of partial-discharge measurements. It is observed that the smallest values of E_{te} are detected on specimens whose expansion is higher. Indeed, for the dimensions and the pressure conditions of the tested specimens, it must hold that the larger the cavity height, the lower the partial-discharge inception field. No relation seems to exist between stretching force (which affects the radial dimension) and E_{te} .

Figure 10 shows the number of partial discharges (in 1500 s) during polarization at 100 kV/mm (N_{PD}) as a function of E and F . Increasing the expansion rate, at almost constant stretching force, a quadratic increase of the number of partial discharges (N_{PD}) occurring during polarization can be noted. The following relationship was obtained which is represented in Figure 10 as a hyperbolic paraboloid:

$$N_{PD} = -1864 - 9.74 \cdot E + 96.0 \cdot F + 0.342 \cdot E^2 - 0.923 \cdot F^2 \quad (2)$$

In the range of stretching force between 36 N and 69 N, an initial increase, then a decrease of N_{PD} occurs. This can be explained speculating that a certain level of stretching is needed to favor the expansion action, due to the stiffness of the wall between the voids. In other words, too small or too large stretching force will not promote creation of cavities of appropriate dimension for optimal expansion and, therefore, for partial-discharge activity inception.

The amplitude of the signal acquired during space-charge measurements (V_{SC}), after 1000 s of depolarization (upon

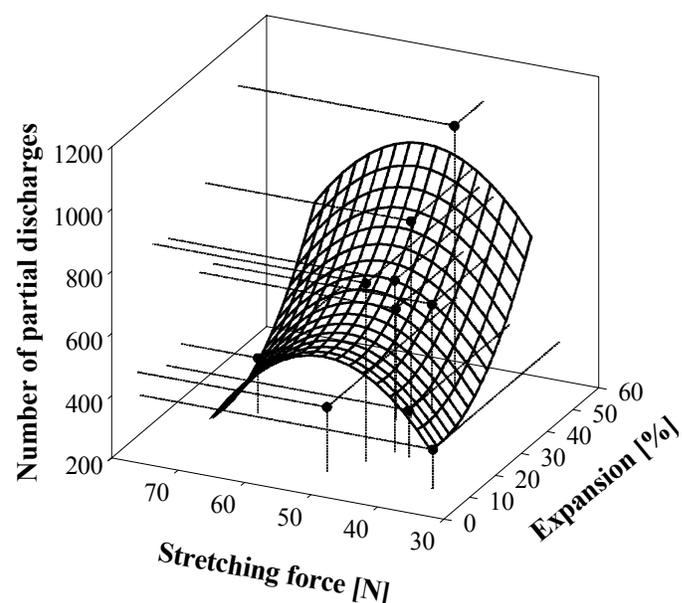


Figure 10. Number of partial discharges (in 1500 s) during polarization at 100 kV/mm (N_{PD}) as a function of E and F (see Table 1).

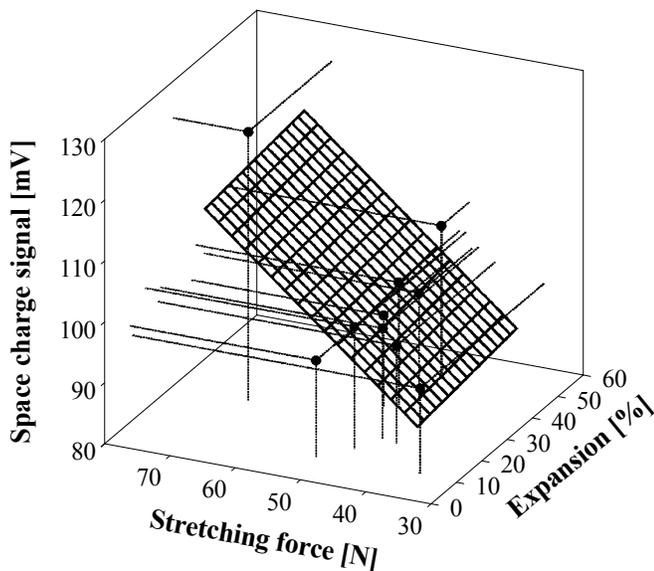


Figure 11. Amplitude of space charge signal 1000 s after polarization (V_{SC}) as a function of E and F (see Table 1).

poling at 100 kV/mm), as a function of E and F , is shown in Figure 11. V_{SC} increases with the stretching force, whereas the influence of the expansion rate seems to be limited. It may be speculated that specimens with large-radius cavities (i.e. specimens subjected to higher stretching force) offer a larger resistance to the deployment and recombination of space charge during depolarization, due to the greater radial dimension of their voids. Equation (3) represents the relation between E , F and V_{SC} , shown in Figure 11 as a plane.

$$V_{SC} = 55.0 + 0.0602 \cdot E + 0.902 \cdot F \quad (3)$$

A non-linear regression was applied to estimate the parameters of equations (2) and (3). The cross-correlation factors (R^2) obtained for the evaluations are 0.82 and 0.86, respectively.

6 CONCLUSIONS

Space-charge, partial-discharge and piezoelectric measurements show clear correlations. Thus these three measurement techniques can provide a global characterization of the new family of piezoelectric materials here investigated. The amplitude of space charge is associated with the size of the piezoelectric coefficient, as indicated by theory, and the partial-discharge inception level agrees with space-charge threshold and magnitude. These results support a mechanism of ferroelectret formation based on micro-plasma discharges in cavities, making evident how the mechanical treatment of polymer films during stretching and gas expansion can significantly change the ferro- and piezo-electret properties. However, the base material still has a fundamental role for both the mechanical properties (Young's modulus which affects piezoelectricity) and the electrical conductivity. The activation energy of the latter, in fact, plays a major role for the time-temperature stability of the materials.

Thermal-endurance characterization, space-charge and piezoelectric measurements show that at high temperatures (> 50 °C) COC can guarantee a piezoelectric coefficient which is stable with time up to 120 °C. PP, on the contrary, loses its electret behavior already at 50 °C. Indeed, conductivity measurements show that already at 70 °C COC exhibits lower conductivity than PP, and this fact, together with the larger temperature index, can explain the better thermal stability of space charge in COC at high temperatures. Therefore, cellular COC films can be used in more severe environmental conditions than cellular PP.

The dependence of quantities associated with charging, such as the threshold for the inception of steady ferroelectret behavior, the number of partial discharges detected during polarization and the amplitude of the space-charge signal, on the physical dimension of cavities (radial dimension and height, which are related to stretching force and expansion rate, respectively) has been inferred. With increasing expansion rates, the number of partial discharges is enhanced and the threshold E_{tc} is reduced. Increasing stretching force, space-charge accumulation increases, whereas the number of partial discharges increases for low values of the stretching force and tends to decrease for high values.

On the basis of such results and the relevant modeling, indications can be given for the design of sensors based on this new family of quasi-piezoelectric materials.

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