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Electrical Properties of Ultra-Thin Polyester Films: Method of Determination and Interpretation of Transient and Steady State Conductivity

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ELECTRICAL PROPERTIES OF ULTRA-THIN POLYESTER FILMS: METHOD OF DETERMINATION AND INTERPRETATION OF TRANSIENT AND STEADY STATE CONDUCTIVITY.

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Abstract The determination of transient and/or steady-state conductivity of PET ultra-thin films by means of electrode configuration involving any load or stress imposed to the measuring area during measurement usually leads to anomalous behaviors of the charging current and thus prevents a good evaluation of the electrical properties of the sample. This can be avoided by the use of a two-electrode system with lateral contacts, allowing to obtain reproducible results without any requirement for previous mechanical, thermal or electrical treatments (as is often recommended in the literature), which permits to characterize industrial films down to 1.5 μm thick in true storage conditions after production.

INTRODUCTION.

Measuring the transient or steady-state current flowing through a polymeric material after the application or removal of a step voltage generally involves the use of a three-electrode configuration including a guard ring around the low potential electrode in order to define accurately its area and to prevent surface leakage currents. Such a system is considered as a standard for determining the material conductivity (see for example ASTM[1] D-257). For thin films (some microns), the proper use of a guard system is somewhat delicate since the recommended condition $g \leq 2t$ where g is the gap width and t the film thickness is obviously not practical. However this fact is not really critical because the variations in conductivity determination due to changes of the effective area of the guarded electrode are small and usually negligible with regard to the variations observed from sample to sample. For ultra-thin films of PET down to 1.5 μm, a more drastic problem arises due to the necessary connections between the evaporated electrodes and the external circuit. Owing to the extreme fragility of the films, any external pressure exerted on the samples (by means of a spring electrode, for example) can lead to spurious and noisy currents, unexpected current jumps during the charging process and/or electrical breakdown at the

highest field strength values. Similar irregularities can even be observed by simply connecting thin copper wires to the evaporated electrodes with the help of conductive silver paste and improving the contacts with a thin coat of epoxy resin. It thus appears that any load or stress imposed to the sample in the measuring area prevents a proper evaluation of the electrical properties. It has also been often reported in the literature, especially for polyester Mylar* films, that unless a careful annealing procedure at high temperatures is previously used, the reproducibility of charging and discharging currents for a given sample is usually poor while large fluctuations occur from sample to sample[2-5]. Lilly and al.[6], among others, noticed that the samples had to be previously conditioned near the highest temperature to be employed and relatively high field strengths had to be applied for periods up to three weeks before measurements in order to obtain nearly reproducible steady-state currents (to ± 2.5 %). Such procedures are obviously not practical for characterizing industrial films in true storage conditions after production. Anomalies in insulating polymers, like anomalous discharge currents, reversal of current flow during TSC (thermally stimulated current) or charging current increasing with time have been encountered by a lot of authors[7].

In this paper, we show that a good reproducibility can be obtained for ultra-thin PET films under realistic conditions of temperature, electrical field strength and relative humidity, by using a two-electrode system with lateral contacts.

EXPERIMENTAL CONDITIONS.

Specimens 4x4 cm^2 were cut from rolls of Mylar* films ranging in thickness from 1.5 to 12 μm (1.5, 3, 6 and 12 μm) supplied by Du Pont de Nemours (Luxembourg). Whatever the configuration used (see below), the primary electrodes were obtained by evaporating 45 nm thick aluminium layers (occasionnally gold) in vacuum (P = 2 10^{-5} mbar). Before and after metallization, the samples were kept at room temperature in a controlled atmosphere of 52 % constant relative humidity obtained by means of a saturated salt solution ($Na_2Cr_2O_7.2H_2O$, ASTM E-104), which is close to the usual storage conditions of the industrial film rolls. They were then enclosed at the same RH value (52 %) in a grounded brass screen cell in a thermostatic bath whose temperature was controlled at 23 ± 0.1° C. Well stabilized field strengths ranging from 3 10^6 to 40 10^6 V/m were supplied by dry cells. Charging and discharging currents were measured by a programmable Keithley electrometer, model 617.

RESULTS AND DISCUSSION.

Charging and discharging currents were measured on Mylar* films with a classical three-electrode system connected to the external circuit either by means of thin copper wires fixed with silver paste and epoxy resin or by connecting the evaporated electrodes to the external circuit through lightly spring-loaded aluminium plates, polished to better than 0.3 μm. Typical results obtained for an applied field of 30x10^6 V/m with 12, 6, 3 and 1.5 μm thick films are shown in Figure 1.

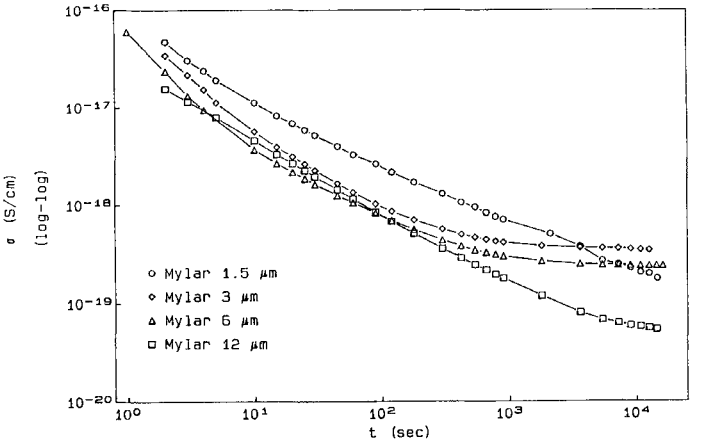

FIGURE 1. Transient conductivity observed in Mylar films of various thicknesses (12, 6, 3 and 1.5 μm) with the three-electrode configuration for times ranging from 1 to 10^4 sec. T = 23°C, $E_p = 3\ 10^7$V/m.

For the 12 μm thick films, the observed currents agree fairly well with a t^{-n} dependence up to 10^3 seconds, after which the steady-state conductivity progressively prevails. This is no longer true for the thinner samples, except in very limited periods of time. In addition, in the case of 6 and 3 μm thick films, the steady-state current is more rapidly reached and is approximately one order of magnitude higher than in the 12 μm thick specimens. It is also important to notice that the thinner the films, the more important was the number of experiments failing for unexplained reasons (high level noise, sudden current jumps during the charging process, dielectric breakdown at the highest field strength values,...). With the 1.5 μm thick films, in particular, it was found necessary to test more than twenty samples before obtaining a complete curve such as represented in Figure 1. The observed variation of the initial slope as a function of field strength or thickness seems to be complex and to disagree with the generally accepted opinion that n is a parameter characteristic of the material.

It thus appears that, in ultra-thin films, any stress imposed to the film in the measuring area unavoidably leads to an unexpected, non reproducible electrical behavior after application of a voltage step function. In the case of connections involving the use of epoxy resin, this could be due to the existence of mechanical heterogeneous stresses or even micro-tears resulting from shrinkage effects induced by the polymerization of the resin. With the spring electrodes, on the other hand, similar micro-tears or conducting paths could be created locally by the exerted pressure due to the particulate additive which provides the surface roughness required for high-speed film winding.

To overcome these difficulties, we have used a two-electrode configuration with opposite lateral contacts, the area of which was negligible with respect to the circular measuring area in order to minimize the field distortion (Figure 2).

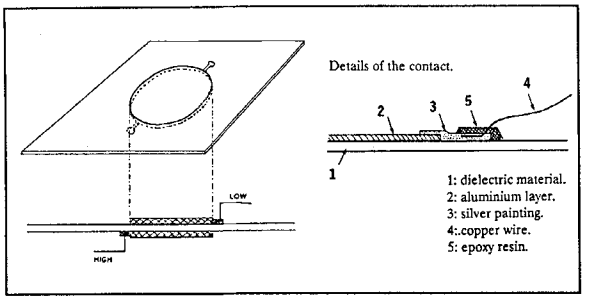

FIGURE 2. Two-electrode configuration including opposite lateral contacts.

A very good agreement between the two-electrode and three-electrode configurations was observed with simultaneously metallized 12 μm thick films (i.e. those films which are expected to be the less sensitive to contact effects), which proves the validity of the new system (it was already pointed out by several authors that, in the case of PET, measurements with and without guard-ring usually give very nearly the same results: see e.g. Lilly et al.[6]) and the possibility to use it with thinner films.

For the thinner films (from 6 to 1.5 μm), spectacular differences in results were obtained with the two types of electrode configurations (Figures 3 and 4).

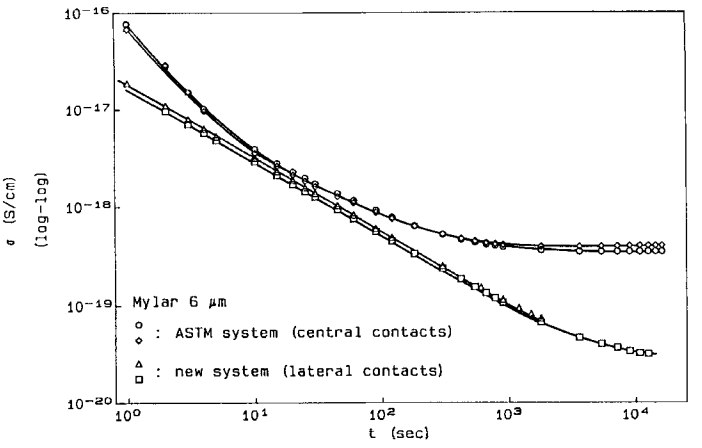

FIGURE 3. Transient conductivity observed in 4 samples of 6 μm thick Mylar film with the three- and two-electrode configurations for times ranging from 1 to 10^4 sec. T=23°C, $E_p = 4\ 10^7$V/m.

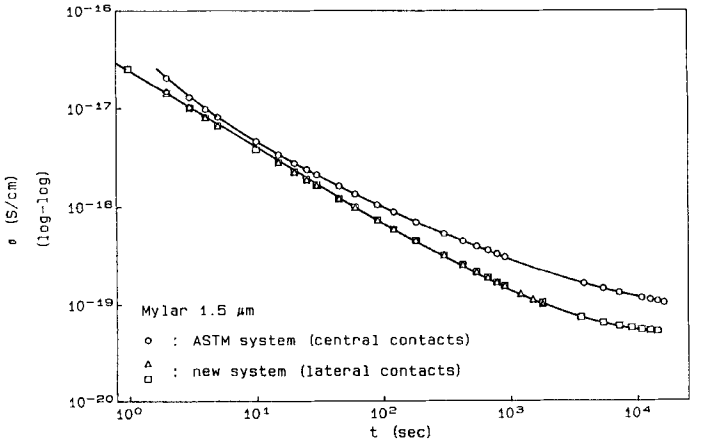

FIGURE 4. Transient conductivity observed in 3 samples of 1.5 μm thick Mylar film with the three- and two-electrode configurations for times ranging from 1 to 10^4 sec. T=23°C, E_p=4 10^7V/m.

Typical results obtained with the new configuration for four thicknesses down to 1.5 μm and a given field strength (4x10^7 V/m) are represented in Figure 5.

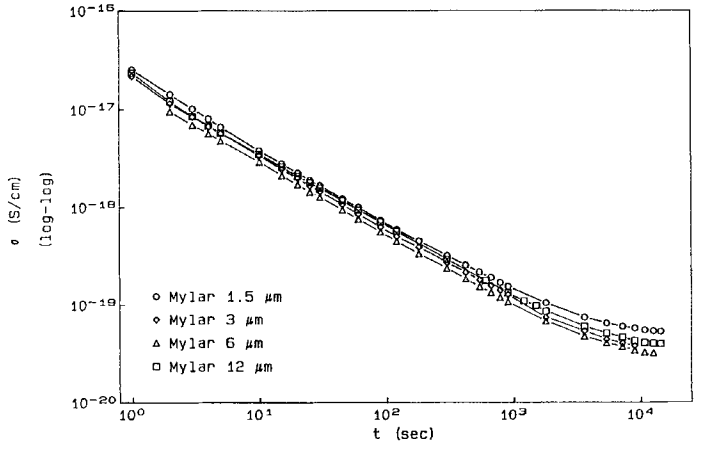

FIGURE 5. Transient conductivity observed in Mylar films of various thicknesses (12, 6, 3 and 1.5 μm) with the two-electrode configuration for times ranging from 1 to 10^4 sec. T=23°C, E_p=4 10^7V/m.

The log-log plots are now reproducible from sample to sample to a few percent and are all characterized by a good linearity over a time period longer than 10^3 secondes and the absolute values of conductivity as well as the exponent n of the Curie-Von Schweidler relationship are nearly independent of the film thickness ($n = 0.75 \pm 0.03$, which is close to the value obtained by Lengyel[8] for thicker films). Consequently, intrinsic properties of the films may be precisely characterized without previous conditioning.

CONCLUSIONS.

The present experimental evidence allows to define a simple procedure using a two-electrode configuration with opposite lateral contacts for obtaining reproducible and reliable measurements of the transient and steady-state electrical conduction in ultra-thin PET films without any previous electrical, mechanical or thermal treatments. Such treatments, which are often recommended and even claimed to be necessary to obtain reproducible results[2-5], induce in fact pronounced changes in the matrix morphology and/or the size and number of crystallites. This prevents a full characterization of industrial films under realistic storage conditions. It appears from our measurements that the previously encountered difficulties could arise from the existence of mechanical stresses or micro-tears induced in the films at the level of the connections to the external circuit. We have shown that this can be easily avoided by realizing opposite contacts outside the circular electrode area.

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