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Low-frequency dielectric functions of dense and chevronic thin films of parylene C

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ABSTRACT

Dense and chevronic thin films of parylene C were fabricated using a physicochemical vapor deposition technique and their dielectric functions were measured as functions of frequency (from 0.1 Hz to 0.1 MHz) and temperature (from $-140\,^{\circ}$C to $260\,^{\circ}$C) using an impedance meter. The real part of the dielectric function of the chevronic thin film was considerably higher than that of its dense counterpart. Higher dielectric strength, loss factor and conductivity for the chevronic parylene suggest an amorphous phase which are more present in this semi-crystalline polymer. X-ray diffractions are now envisaged to confirm this assumption.

1. Introduction

Dielectric relaxation spectroscopy (DRS) is a useful tool often applied to examine the influence of process parameters on the electrical properties of polymers [1,2].

Parylene C – also called poly(chloro-p-xylylene) or PPX-C – is a widely used polyxylene derivative in the electronics, medical, and aerospace industries [3]. Their uniformity, transparency, flexibility, and their high dielectric strength make thin films of parylene C especially attractive as dielectric insulators [4].

Grown by chemical vapor deposition (CVD) [5], these thin films are dense. However, a simple modification of the CVD reactor allows the fabrication of thin films of parylene C with fibrous volumetric morphology [6]. Nominally comprising parallel fibers – all of the same shape – these thin films have been shown to bind proteins and support cellular proliferation on their surfaces [7].

A good knowledge as a function of frequency of the dielectric constant ε' and dissipation factor tan δ of parylene C thin films present interests for many applications among them electrowetting on dielectric (EWOD) [8,9], organic field effect transistors [10] and biology [11]. Moreover, specific value of this dielectric constant of parylene C around 1 kHz is often required for applications. Let us quote for example biomedical applications as neural prosthetics [12], EWOD [9,13] and actuators in relation with mechanical vibrations [14].

In the present work, we conducted a comparative study of the real and imaginary parts of the complex-valued dielectric function of dense and chevronic thin films of parylene C, of the same thickness ($\sim 9\,\mu$m), as functions of temperature (from $-140\,^{\circ}$C to $260\,^{\circ}$C) and frequency (from 0.1 Hz to 0.1 MHz). Each fiber in a chevronic thin film has a zigzag shape and thus a chevronic thin film looks like a cascade of chevrons. Being fibrous, these thin films are not dense and therefore their dielectric properties should be quite different from those of their dense counterparts. In consequence, parylene C with fibrous volumetric morphology could be used for new applications for dielectric insulation. In a previous work, we investigated dielectric and ac-conductivity in dense parylene C films using conventional CVD reactor [15,16]. In this new study and in order to better compare and justify peculiar behaviors of chevronic compared to dense parylenes, these latter have been manufactured in the same modified CVD reactor [6] in order to limit possible dispersion in the results due to the process.

The plan of this paper is as follows. Section 2 describes a physicochemical vapor deposition method used to fabricate dense and chevronic thin films of parylene C, as well as the broadband dielectric spectroscopic procedure used to measure the complex-valued dielectric functions of these samples at different temperatures. The measured data are presented and discussed in Section 3. Concluding remarks are provided in Section 4.

2. Experimental and materials

Thin-film fabrication: All thin films of parylene C investigated for this study were fabricated by a physicochemical vapor deposition

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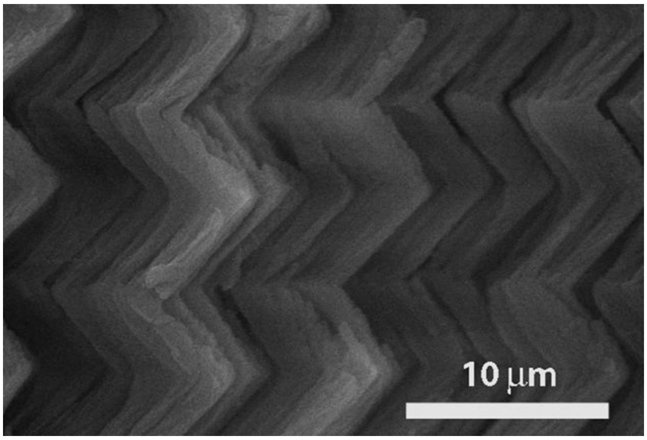

Fig. 1. SEM image of chevronic parylene thin films.

method [7,8] that is a modification of the standard CVD technique to grow dense thin films of the same material. To summarize the process, parylene-C dimers, the raw material, were first vaporized at 150 °C and then pyrolyzed into reactive monomers at 650 °C. A collimated flux of the reactive monomers was directed through a nozzle onto a planar platform on which a conducting brass plate had been mounted. Room-temperature polymerization of the reactive-monomer flux occurred on the exposed brass surface. The deposition pressure was set at 26 m Torr for both dense and chevronic thin films.

The orientation of the platform with respect to the direction of the collimated flux was adjusted by two motors. The rotation motor rotates the platform plane about the central normal axis, and the rocking motor rotates the platform about an axis tangential to it. For the deposition of a dense thin film, the rotation motor was shut off, while the rocking motor was used to fix the platform such that the collimated flux subtended an angle $\chi_v = 90^\circ$ on the platform plane. For the deposition of a chevronic thin film, the rocking motor was used to set the vapour incidence angle $\chi_v = 10^\circ$ whereas the rotating motor was abruptly rotated by 180° every 60 s. Both dense and chevronic thin films of thickness ~9 μm were fabricated. Each chevron of chevronic thin films was symmetric and the films had a period of a few microns in the thickness direction (Fig. 1).

Dielectric measurements: The complex-valued dielectric function

$$\varepsilon^*(f,T) = \varepsilon'(f,T) - j\varepsilon''(f,T) \tag{1}$$

of a planar sample is measured as a function of the frequency f and the temperature T by inserting the sample tightly between two parallel plates of a metal functioning as two electrodes across which a known sinusoidal time-varying voltage is applied while a uniform temperature is maintained; $j = \sqrt{-1}$. The amplitude and the phase of the sinusoidal time-varying current flowing across the sample are measured. The ratio of the voltage to the current equals the impedance of the sample, from which ε^* (f, T) can be calculated. A complex-valued conductivity

$$\sigma^*(f,T) = \sigma_{dc}(T) + j2\pi f \varepsilon_0 \varepsilon^*(f,T) = \sigma'(f,T) + j\sigma''(f,T) \tag{2}$$

is often used as well, with $\sigma_{dc}(T)$ denoting the dc conductivity and ε_0 the permittivity of free space (i.e., vacuum).

A silver electrode was deposited on top of each thin-film sample by low-pressure (3.75 m Torr) evaporation at room temperature to form a silver/parylene-C/brass capacitor that is leaky. Silver has been preferred contrary to gold avoiding a supplementary step of process for the adherence of metal on the surface of parylene. It must be paid attention that, in this case, thermal cycles on the device are not necessarily suitable. For our study, the sample was cooled to −140 °C and the measurement process was repeated from −140 °C to 260 °C. The cooling-and-heating cycle was repeated once more. No change in data occurred during the three runs. Dielectric measurements were carried out by means of a commercial Novocontrol Broadband Dielectric Spectroscopy impedancemeter. The electroded sample was always placed in a dark shielded cell containing dry nitrogen to avoid oxidation. The temperature was controlled by a Linkam TMS 94 hot stage to better than within ±0.1 °C and measurements were made in steps of 10 °C.

3. Results and discussion

Fig. 2 shows the variation of ε' with frequency at different temperatures for both the dense and the chevronic thin films of parylene C. Clearly for both types of thin films, ε' decreases with increasing f at fixed T, the decrease in ε' with increasing f being very prominent at both low frequencies and high temperatures. Due to dielectric relaxation – which is related to orientational polarization and therefore to the molecular arrangement – this trend at frequencies below the dielectric relaxation frequency is expected of the most dielectric materials [1,2].

The values of ε' for the chevronic thin film are higher than for the dense thin film at a fixed frequency and temperature. For instance, at 1 kHz and 20 °C, $\varepsilon' = 15$ for the chevronic thin film but $\varepsilon' = 3$ for its dense counterpart. Data measured for the dense thin

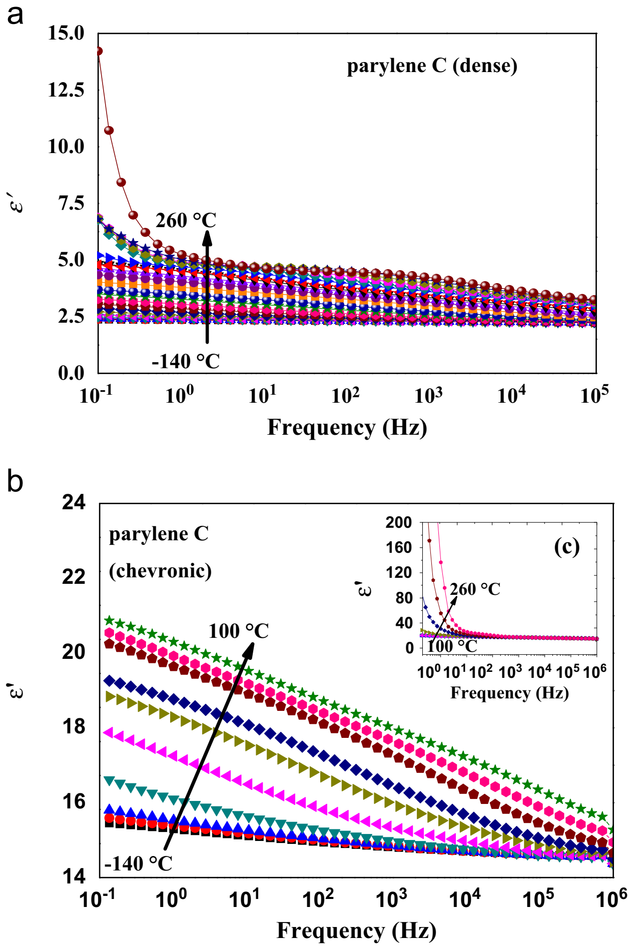

Fig. 2. Variation of ε' with frequency at various temperatures from −140 °C to 260 °C for (a) dense and (b and c) chevronic thin films of parylene C.