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# Dielectric function and degradation process of poly(triarylamine) (PTAA)

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#### 1. Introduction

During the last years, huge progress in basic research as well as in device related properties of organic electronics has been achieved. As benchmark for these achievements, the performance of organic field effect transistors (OFETs) can be used. OFETs have to rival with the classic (inorganic) FET. For this reason organic semiconductors with high charge carrier mobilities are necessary. One of these materials is the p-type organic semiconductor poly(triarylamine) (PTAA) with hole mobilities of  $10^{-3}$ up to  $10^{-2}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> [\[1\]](#page--1-0). PTAA forms amorphous films and has the huge advantage of air stability [\[2,3\].](#page--1-0) So far, the optical properties of this polymer in the infrared spectral region have not been published, however it can be used to further investigate devices. For application, the degradation process of organic thin films is of high

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## **ABSTRACT**

We investigated the optical properties and the degradation process of the organic p-type semiconductor poly(triarylamine) (PTAA) in the mid infrared region. The dielectric function of PTAA was determined in the spectral region of 700 up to 6000  $\text{cm}^{-1}$  by modeling ellipsometric measurements. Due to degradation at  $65^{\circ}$ C, PTAA thin films developed carbonyl and hydroxyl features in the infrared spectra. Degradation under 85% RH additionally led to absorption signals of water. A degradation of the bare gold substrates was also observed. For bare gold films, morphology changed upon degradation and adsorption of hydrocarbons from the ambience took place.

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relevance and its understanding contributes to an improvement of devices. So far, the influence of humidity [\[4–6\]](#page--1-0) and thermo-oxidation [\[7–9\]](#page--1-0) on different OFET and OLED structures or materials has been reported. We specifically investigated the organic semiconductor layer without top contacts and without a gate insulator. Our results may help to separate other degradation processes that may occur in an OFET.

#### 2. Experimental

In order to increase the infrared (IR) signal from the organic thin layer, gold substrates were used [\[10\].](#page--1-0) Gold was evaporated as a 180 nm thick film on glass with aluminum (12 nm) as adhesion layer. Thin films of PTAA were produced via spin coating on gold layers. For the spin coating, PTAA (Sigma–Aldrich, the structural formula is depicted in [Fig. 1\)](#page-1-0) was solved with  $2 \text{ wt}$  in mesitylene and spin coated for 60 s at 2000 rpm resulting in an average film thickness of about 50 nm (as measured with IR ellipsometry). IR spectra were taken using a







<span id="page-1-0"></span>

FTIR-RC-Ellipsometer (IR-VASE, J.A. Woollam Co., Inc.) with an incident angle  $\phi_1$  of 70° (to the surface normal) with  $4 \text{ cm}^{-1}$  resolution and under ambient conditions. In ellipsometry, the ratio

 $r_p/r_s = \tan \Psi \exp(-i \Delta)$ 

of the complex Fresnel coefficients  $r_p$  and  $r_s$  is measured without the necessity of a reference measurement. Furthermore, atomic force microscopy images were acquired using a tapping mode AFM (Digital Instruments Nanoscope IV). Samples were degraded on the one hand in an oven at 65  $\degree$ C and ambient air resulting in a low relative humidity (RH) at the elevated temperature. On the other hand, they were degraded in an environmental simulation chamber (ESC, Binder MKF 115) at 65 °C and 85% RH. During degradation the samples were stored in soda-lime glass petri dishes. For each condition six identical samples were prepared and each sample was degraded for a different time period of 10 up to 60 days.

### 3. Dielectric function of PTAA

Fig. 2 shows measured  $\Psi$ - and  $\Delta$ -spectra as well as the best-fit modeled data for a PTAA thin film on gold substrate. Because any crystallites in the dense layers are small compared to the wavelength, modeling was done within the Jones formalism where diffuse light scattering is neglected. The model was achieved in the spectral range from 700 up to  $6000 \text{ cm}^{-1}$  using 29 Gauss-Lorentz oscillators (see Supplement, Table S1) and a Drude oscillator for the gold substrate. Parameters of the Drude oscillator were chosen according to Ref. [\[11\]](#page--1-0). Since PTAA forms amorphous films [\[2\],](#page--1-0) an isotropic model was used. This was verified with FTIR transmission measurements under various incidence and substrate angels (not shown here). The surface roughness was not considered in the optical model since its scattering contribution is below the detection limit for the IR light. The dielectric background  $\varepsilon_{\infty}$ , that is the contribution of energetically higher oscillations, was determined to be  $2.9 \pm 0.1$ . Fig. 3 shows the real ( $\varepsilon_1$ ) and imaginary part  $(\varepsilon_2)$  of the modeled dielectric function  $\varepsilon = \varepsilon_1 + i\varepsilon_2$  of PTAA. The strongest absorption of PTAA at about 1490  $\text{cm}^{-1}$  could be assigned to the aromatic ring-stretch vibrations [\[12\].](#page--1-0) The absorption peak at 816 cm<sup>-1</sup>



Fig. 2. Measured ellipsometric variables  $\Psi$  and  $\Delta$  (black) for a 59 nm thick PTAA film spin coated on gold substrate and best-fit modeled data (red). The film was modeled as optically isotropic. All oscillations could be described with Gauss–Lorentz oscillators. The gold substrate was described with the Drude model [\[11\].](#page--1-0) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 3. Real  $(\varepsilon_1)$  and imaginary part  $(\varepsilon_2)$  of the dielectric function of PTAA. The dielectric function was derived from a 59 nm thick PTAA film on a gold substrate (Fig. 2).

can be assigned to aromatic  $C-H$  bending. Considering the structure of PTAA, the group of oscillations between 1250 and  $1320 \text{ cm}^{-1}$  involves mainly aromatic CN stretch vibrations  $[12]$ . So far, no IR spectra of PTAA have been published. In the following we will discuss the change in the  $\Psi$ -spectra as a measure of the IR absorption.

#### 4. Degradation results

#### 4.1. Degradation of gold substrates

In order to get an estimate of the contribution of substrates to the degradation, bare gold substrates were inspected. [Fig. 4](#page--1-0) shows the result of the two kinds of degradation experiments. Already in the spectra of the asdeposited gold substrate some vibrational signals appear in the typical organic fingerprint region  $(1164 \text{ cm}^{-1})$ 

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