Organic Electronics 14 (2013) 3453–3459

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Polymer dielectric layer functionality in organic field-effect transistor based ammonia gas sensor

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ARTICLE INFO

Article history:
Received 30 July 2013
Received in revised form 3 September 2013
Accepted 11 September 2013
Available online 25 September 2013

Keywords:
Organic field-effect transistors (OFETs),
Ammonia sensor
Polymer dielectric
Recovery property
Low detect limitation

ABSTRACT

Ammonia (NH_3) gas sensors based on pentacene organic field-effect transistors (OFETs) are fabricated using polymers as the dielectric. Compared with those incorporating poly(vinyl alcohol), poly(4-vinylphenol) or poly(methyl methacrylate) dielectric, a low detect limitation of 1 ppm and enhanced recovery property are obtained for OFETs with polystyrene (PS) as gate dielectric. By analyzing the morphologies of pentacene and electrical characteristics of the OFETs under various concentrations of NH_3, the variations of the sensing properties of different dielectrics based OFET-sensors are proved to be mainly caused by the diversities of dielectric/pentacene interfacial properties. Furthermore, low surface trap density and the absence of polar groups in PS dielectric are ascribed to be responsible for the high performance of NH_3 sensors.

1. Introduction

Environmental monitoring, especially the air quality monitoring has become a global issue due to terrible air pollution around the world [1,2]. Gas sensors as an effective way to monitor the gas quality have been developed for more than 30 years [3,4]. Among various gas sensors, organic field-effect transistor (OFET) based sensors have many advantages over semiconductor resistor sensors and optical gas sensors [5–7]. Apart from the low cost and light weight features of OFET [8,9], the sensing property of such devices can be modulated by changing the working conditions and integrating them in oscillators or amplifier circuits.

As is well known, organic semiconductor layer is a key factor in achieving high performance OFET-sensor. To date, many attempts in improving the sensing properties of organic semiconductor layers have been made [10,11]. For example, H. Katz et al. developed a sensor array with distinct response pattern and unambiguous recognition ability for individual analytes based on naphthalenetetracarboxylic diimide derivatives and copper phthalocyanine [12]. D. Yang et al. reported organic nitrogen dioxide sensors based on zinc phthalocyanine (ZnPc) nanofiber network with much better recovery characteristics and shorter response/recovery time compared with polycrystalline ZnPc film devices [13,14].

In addition, the dielectrics of OFETs also play an important role in sensing [15]. It is well known that proper dielectric materials or dielectrics surface modification can significantly enhance the performance of OFETs due to an improvement of the organic semiconductor/dielectric interface property or a better crystallization of the upper organic semiconductor [16–19]. In recent years, electrolyte-gate OFET sensors show great advantages in the detection of biomaterials in aqueous solution [20–22]. Q. Tang et al. designed sulfur dioxide sensors based on gas dielectric CuPc nanowire FET with detect limitation down to sub ppm levels (0.5 ppm) [23]. A. Klug et al. demonstrated a top-gate bottom contact OFET-sensor using pH-sensitive, xanthene-dye functionalized, active-sensing dielectrics

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http://dx.doi.org/10.1016/j.orgel.2013.09.018

with ammonia (NH_3) detect limitation of 100 ppm [24]. The above researches imply that dielectrics of OFETs, especially the dielectric surface properties have great influence on the sensitive characteristics of OFET-sensors. However, as polymer dielectrics hold the potential of being utilized in flexible and low-cost electronic devices, the influence of such materials on performance in OFET gas sensors is still lack of research.

Herein we report NH_3 sensors based on OFETs with four kinds of polymer dielectrics, including polystyrene (PS), poly(vinyl alcohol) (PVA), poly(methyl methacrylate) (PMMA) and poly(4-vinylphenol) (PVP). Through analyzing the electrical characteristics of the devices under different NH_3 concentrations and the surface morphologies of dielectric and pentacene, we identify the variations of sensing properties are mainly generated by the dielectrics/pentacene interface diversities. The low trap density and the absence of polar groups in PS dielectric contribute to the high performance of sensors based on pentacene OFETs, which show low detect limitation down to 1 ppm, high sensitivity and reliable recovery property.

2. Experimental

2.1. Device preparation

The OFETs were fabricated on indium tin oxide glass substrates. The architecture of the OFETs is shown in Fig. 1, along with the chemical structures of the polymer dielectrics. Before the dielectric layers were spin-coated, the substrates were successively ultrasonic cleaned in acetone, deionized water and isopropyl alcohol. PS (average Mw ~ 280,000), PVA (Mw 146,000–186,000, 99+% hydrolyzed), PMMA (average Mw ~ 120,000) and PVP (average Mw ~ 25,000) were dissolved in xylene, deionized water, anisole and butyl acetate, respectively. All the polymers are purchased from Sigma–Aldrich. The polymer dielectrics were spin-coated on ITO substrate at room temperature and dried in an oven under 100 °C for 1 h. The thickness of dielectrics was measured by a Dektak 150 stylus profiler. The finger source/drain electrodes (30 nm) were successively deposited under 3×10^{-3} Pa through a shadow mask. Subsequently, the devices were moved to another chamber for the deposition of 30 nm pentacene (Sigma–Aldrich) under 2×10^{-4} Pa at the rate of 0.2–0.3 Å/s. The devices with different dielectrics are referred as device A with PS dielectric, device B with PVA dielectric, device C with PMMA dielectric, device D with PVP dielectric. The electrical characteristics of the OFETs were carried out with a Keithley 4200 sourcemeter in nitrogen at room temperature. Charge carrier mobility (μ) and threshold voltage (V_{TH}) were extracted in the saturation regime from the highest slope of $|I_{DS}|^{1/2}$ vs. V_{GS} plots using the following equation:

$$I_{DS} = (W/2L)\mu C_i(V_{GS} - V_{TH})^2 \quad (1)$$

where L (100 μm) is the channel length, W (1 cm) is the channel width, C_i is the capacitance (per unit area) of the dielectric, V_{GS} is the gate voltage, and I_{DS} is the drain-source current.

2.2. Film characterization and sensor test

The morphologies of the dielectrics and pentacene films were characterized by atom force microscopy (AFM) (MFP-3D-BIO, Asylum Research) in tapping mode. The OFET-sensor was stored in an airtight test chamber (approximately 16 mL). Dry air and 500 ppm standard NH_3 gas were purchased from Sichuan Tianyi Science & Technology Co., and a mixture with the appropriate concentrations was introduced into the test chamber by mass flow controllers (S48 300/HMT, Beijing BORIBA METRON Instruments Co.). The flow rate in the test was fixed at 100 sccm (standard cm^3 per min). The testing environment with specific relative humidity was realized by utilizing a standard humidity generator (BSF-6X-2, Beijing Naisida New Technology Development Co.).

3. Results and discussion

Six cycles of the real-time I_{DS} responding to the dynamic switches in different NH_3 concentration (10–100 ppm) at room temperature for these four devices are shown in Fig. 2. The testing process of the time dependence of the drain-source current is not started until the drain-source current reached a stable state (the drain-source current

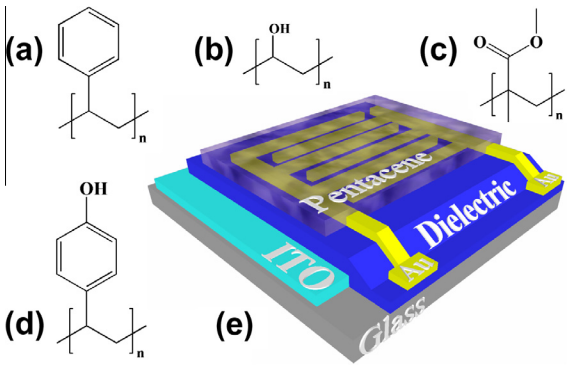

Fig. 1. (a, b, c and d) Molecular structures of PS, PVA, PMMA and PVP in this study, respectively. (e) Schematic of OFET based gas sensors.

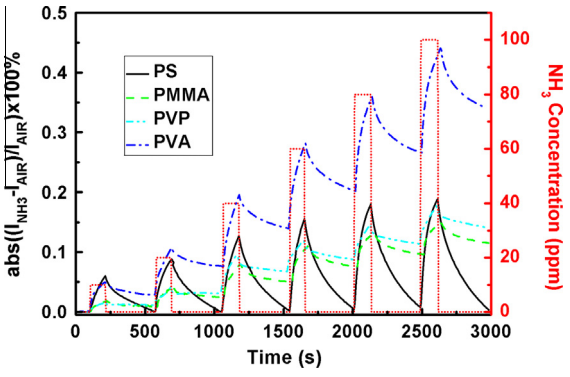

Fig. 2. Response curves of the four kinds of devices to NH_3 pulses.