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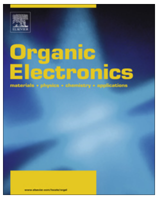

Insight into the charge transport and degradation mechanisms in organic transistors operating at elevated temperatures in air

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ABSTRACT

Operational stability of organic devices at above-room-temperatures in ambient environment is of imminent practical importance. In this report, we have investigated the charge transport and degradation mechanisms in pentacene based organic field effect transistors (OFETs) operating in the temperatures ranging from 25 °C to 150 °C under ambient conditions. The thin film characterizations techniques (X-ray photoelectron spectroscopy, X-ray diffraction and atomic force microscopy) were used to establish the structural and chemical stability of pentacene thin films at temperatures up to 150 °C in ambient conditions. The electrical behavior of OFETs varies significantly in above-room-temperature range depending on charge transport and material properties. Mobility, at temperatures below 110 °C, is found to be thermally activated in presence of traps and temperature independent in absence of traps. At temperatures above 110 °C mobility degrades due to polymorphism in pentacene or interfacial dielectric deterioration. The degradation of mobility is compensated with the decrease in threshold voltage at high temperatures and OFETs are operational at temperatures as high as 190 °C. 70 °C has been identified as the optimum temperature of operation for our OFETs where both device behavior and material properties are stable enough to ensure sustainable performance of pentacene based OFETs.

1. Introduction

Organic Semiconductor-based devices have shown a potential for use in a wide range of niche applications, such as the Radio frequency identity (RFID) tag [1], the backplane transistor matrix of flat panel display [2], and lighting applications [3]; these are either very expensive or impossible to fabricate through conventional silicon technology. As these devices nearing the reality a new problem pertaining to the reliability of organic devices has come to the light. The organic devices operating at high fields and prolonged hours are likely to get heated increasing the operating temperature of the device stack [4,5]. A large matrix of organic devices collectively generate heat increasing the operational steady-state temperature to 50–60 °C [4,5]. Further, the ambient temperature of the devices can contribute to excess heating, thus exacerbating the operational temperature requirements. Consideration of these practical limitations and identification of the factors that affect device operability at high temperatures are crucial for successful and durable deployment of organic devices.

There are fewer reports on the characterization of organic field effect transistors (OFET) at elevated temperatures and most of which have concentrated on the impact of annealing [6–8]. In other works the stability of OFETs at high temperatures is tested in inert conditions [9,10]. These works have been limited in their scope with the focus on the charge transport. Studies at high temperatures in organic semiconductors are not only important for understanding the charge transport but also can give insight into the degradation mechanism with temperature in ambient conditions.

In this manuscript, we have presented a comprehensive analysis to draw correlation between the material and electrical characteristics of pentacene-based devices in the temperature range of 25–150 °C – measured in ambient conditions. The chemical, thermal and the structural stabilities of pentacene (chemical structure shown in Fig. 1a) thin films have been investigated and their impact on electrical characteristics have been simultaneously analyzed. It has been found that pentacene is chemically stable and its

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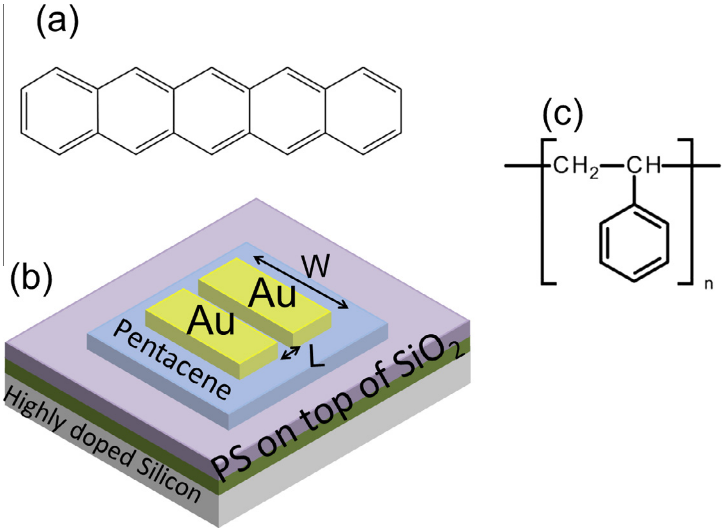

Fig. 1. (a) Chemical structure of pentacene (b) schematic of bottom gate top contact OFET with PS/oxide as gate dielectric (c) chemical structure of polystyrene (PS).

crystallinity is intact up to 150 °C. The stability of the current characteristic in OFETs (device structure is shown in Fig. 1b) at high temperatures is dependent on the thermal stability of the gate dielectric as well as the interfacial characteristics of the dielectric. The carrier mobility within the stable range of temperature has been found to be temperature-independent in the absence of traps. However, the mobility is thermally activated in the presence of interface defects. It has been demonstrated that pentacene OFETs work reliably under thermal stress and repeated cycles in the air and do not oxidize or degrade due to thermal mechanical stresses.

2. Experimental details

Pentacene films were characterized by X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD) and atomic force microscopy (AFM). The pentacene thin film was deposited on one large substrate. The substrate was cut into two sets, one was used for annealing and the other sample was used, as deposited, for comparison. The films were heated in ambient conditions at different temperatures for 1 h before being characterized. XPS studies allow us to quantify the different elemental compositions present in surface of the analyzed film. A surface scan of the pentacene film was carried out using the ULVAC-PHI system (Model: PHI5000VersaProbeII) with an Al anode as the source of the soft X-ray beam. A wide range scan was carried out to detect the presence of all possible elements. A narrow scan for C1s peak was done between the energy range of 275 eV and 290 eV with the step size of 0.08 eV. The change in the crystal orientation of pentacene films due to heat can be analyzed using XRD. The experiment was conducted using high resolution Rigaku system (Model: Smartlab 3KW) with a Cu-Kα ($\lambda = 1.541$ Å) X-ray source. The scanning range of 2θ is 3–30° in steps of 0.01°. Morphological imaging of the pentacene surface was carried out in the tapping mode using Veeco NanoScope IV Multi-Mode AFM. The image analysis was done using WSxM 5.0 software [11].

Pentacene-based OFETs (schematic shown in Fig. 1b) have been used as a test structure in this work for electrical study. A p-type highly doped silicon wafer was used as a substrate. An oxide of thickness 76 nm was thermally grown on the silicon wafer. The backside oxide of the wafer was etched in a BHF solution. Piranaha cleaning of the wafer was done to further clean the surface and make it hydrophilic. The substrates were heated in a clean environment for half an hour at 120 °C to eliminate the moisture from the surface. We fabricated two sets of OFETs; in the first set, pentacene was deposited directly on the oxide and in the other, the pentacene was deposited on a thin polymeric dielectric layer. A 1% polystyrene (PS—chemical structure shown in Fig. 1c) solution in toluene was spin coated (2000 rpm) on a piranaha-cleaned oxide wafer. A smooth and pin-holes-free PS film was observed under a high resolution (40× objective lens) optical microscope (Olympus). Pentacene, triple sublimed with 99.9% purity from Sigma Aldrich, was evaporated at room temperature using a shadow mask with a deposition rate of 0.1 nm/s in order to achieve a thickness of 50 nm. Au source/drain contact was deposited using a shadow mask with $W/L = 700\ \mu\text{m}/70\ \mu\text{m}$. After the complete OFET was fabricated, Al was deposited on the back side of the wafer to make a good contact with highly doped silicon. Electrical characterization was done using Keithley® SCS 4200. The devices were heated to a temperature of 190 °C on a Temptonic chuck with an inbuilt heating coil. All characterization was done in dark and ambient conditions with humidity of 40–50%. The results presented in this study represent a typical dataset measured over multiple samples in different runs.

3. Results and discussion

3.1. Chemical stability

In the line of different experiments, foremost is the stability of pentacene thin films in ambient condition at elevated temperature. Ambient stability of pentacene is disputable and as pentacene is often believed to get oxidized in air [7,8,12]. Such an oxidation phenomenon is likely to exacerbate at high temperatures. Consequently, first we have studied the chemical stability of 50 nm pentacene deposited on SiO_2 due to heating under ambient conditions using X-ray photoelectron spectroscopy (XPS). XPS probe core energy levels and provide information about the chemical composition in the pentacene film. As pentacene interacts with ambient moisture and oxygen [13], due to charge transfer, core level binding energies are likely to change [14]. A series of XPS scans of pentacene films were compared to investigate the impact of environmental moisture and oxygen at elevated temperatures, as shown in Fig. 2. The XPS scan of a freshly prepared pentacene film shows C1s peak at the binding energy (BE) of 284.5 eV, corresponding to the sp^2 hybridized state of carbon atoms in the pentacene structure similar to previous reports [14,15]. On heating

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