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Effect of temperature on hysteresis of dipolar dielectric layer based organic field-effect transistors: A temperature sensing mechanism

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ABSTRACT

This study investigates the effect of measurement temperature on the hysteresis of organic field-effect transistors (OFETs) under vacuum and humidity conditions (~65% RH). OFETs were fabricated using CuPc and PMMA/PVA/Al_2O_3 tri-layer dielectric materials to possess temperature-sensing capability through the controlled polarization of polar dielectric (PVA) layer with excellent stability, enhanced performance over a wide range of temperature. We report a novel temperature sensing mechanism of the OFETs by exploiting the temperature dependence of hysteresis, mobility and bias-stress. At room temperature, the device exhibited hole mobility of 0.004 $cm^2/V\,s$ and 0.016 $cm^2/V\,s$, threshold voltage of – 3.8 V and – 3.7 V under vacuum and ambient conditions, respectively. Over the temperature range of 150–370 K, the variation of mobility found to follow the Arrhenius behavior, supporting hopping charge transport. At 370 K, the mobility is enhanced by five times while switching the ambient from vacuum to humidity. However, there is a great enhancement in the mobility of ~30 times at 370 K compared to room temperature under ambient conditions. Under both the conditions, we observed a systematic variation of hysteresis from clock wise to anti-clock wise direction and its amount. Bias-stress experiments showed the enhanced stability in the performance of the OFETs under different ambient condition with temperature. We have demonstrated how the systematic polarization of polar dielectric layer can be exploited to fabricate OFET based temperature sensor, which is highly sensitive to temperature variation within 250 K–370 K ranges. The response/recovery times were found to be 25 and 15 s, respectively.

1. Introduction

Organic field-effect transistors (OFETs) have attracted immense attention for monitoring health and wellness parameters and other medical applications due to their compatibility with well-developed microelectronic fabrication techniques. In addition, such devices can be fabricated on transparent, stretchable and ultra-flexible substrates for large area applications with exceptional stability [1,2]. The OFETs are highly sensitive to the environmental conditions: moisture, light, organic vapors and temperature. Many attempts have been made to understand the effect of ambient conditions on the performance of the transistor [3]. At higher temperatures, the thermal instability is one of the major issues for organic devices because of their low melting temperatures and large thermal expansion coefficients that cause degradation of the devices [4]. Charge transfer mechanism in organic semiconductor materials and its temperature dependent changes are not understood completely. In organic semiconductors, the charge transport mechanism is explained mainly by two models (1) variable range hopping (VRH) and multiple trapping and release (MTR) models with a shallow distribution of trap states [5]. In MTR model, the free carrier mobility (μ_0) is diminished by recurrent charge carrier trapping and thermal release from shallow trap states

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below the conduction band edge [6]. The variable-temperature behavior of OFETs generally exhibits thermally activated mobility with the behavior consistent with the Arrhenius relationship $\mu = \mu_0 \exp\left[-E_a/k_BT\right]$ where E_a is the activation energy, k_B is the Boltzmann constant and T is the absolute temperature. Activation energy is the energy difference between the trap state and the conduction band edge, which is typically of the order of few tens to hundreds of milli-electron volts [7,8]. Nelson et al. reported that the temperature-independent transport in pentacene based transistors strongly influenced by the traps and contact effects [9]. The variation of contact to channel resistance with temperature was observed in the P3HT based OFETs by Hamadani et al. [10] Sakanou et al. investigated the origin of an apparent 'band-like', negative temperature coefficient of the mobility in derivatives of pentacene films [11]. We have observed reduction of contact resistance with gate voltage. As humidity increases, contact resistance decreases [Fig. S1]. In OFETs, most of the temperature dependent studies were confined to the devices fabricated on the standard Si/SiO_2 substrates. Organic semiconductors show temperature-dependent field-effect mobility at temperatures ranging from liquid helium temperature to room temperature (RT), but display very limited thermal sensitivity beyond RT. To overcome this limitation, the introduction of polar groups within the dielectric layer can provide an interfacial interaction with charge carriers of the organic semiconductor layer in a manner that can be influenced by the changes in temperature, which has been exploited for thermal sensing. Therefore, use of a polar polymer material as a component of the dielectric system in OFET fabrication can enhance the thermal sensitivity and stability for temperature sensing applications. Wu et al. reported the OFET based temperature sensors using biodegradable polymer containing carbonyl groups as polar component [12,13]. The operating voltage of these transistors is about 80 V and the measured temperature range is 25 °C–200 °C. To date, diverse active materials, which have nanowires, nanowalls, nanotubes, nanofilms, carbon based materials and composites have been employed to fabricate temperature sensors for different temperature ranges [14–17]. These devices are mostly resistive type sensor. However, OFETs reported as temperature sensors for flexible and stretchable applications are mostly operated at above 10V [18,19]. Though, bipolar junction transistors (BJT) based CMOS temperature sensors show better accuracy but such sensors are not suitable under flexible or stretchable platform [20–23]. A comparison of different temperature sensors is given in Table S1. Here, we report OFET as temperature sensors that cover 250 K–370 K (equivalent to −20 °C to 90 °C) with lower operating voltage of 10 V.

Polarization of dipoles in organic dielectric is not reversible under gate field and shows hysteresis during voltage swift. In this article, we present a detailed temperature dependent study of OFETs with polar dielectric system and have demonstrated how the temperature dependence on the hysteresis can be used to fabricate temperature sensors with exceptional ambient stability. We have used Copper phthalocyanine (CuPc) as active channel materials with tri-layer dielectric system consisting of PVA layer, which is sandwiched between PMMA and Al_2O_3 layer and studied the dependence of measurement temperature on the mobility, hysteresis and bias-stress under vacuum and ambient humidity conditions, which is about 65% RH. Due to its low cost and excellent ambient stability, CuPc is used as semiconductor channel in the OFETs. The reported mobility in CuPc based OFETs with different dielectric system varies from 0.02–0.06 cm^2/V s [24–27]. We observed extraordinary ambient stability, associated with the polarization of –OH groups present in PVA dielectric layer and a model was proposed to explain such behavior [28]. In addition, we have successfully fabricated OFET based humidity sensors by exploiting the variation of polarization of —OH groups in presence of moisture [29]. The temperature driven hysteresis in OFET transfer characteristics is studied by varying the measurement temperature from 150 K to 370 K. The devices exhibited the variation of hysteresis between clockwise hysteresis (CH) and anti-clockwise hysteresis (ACH). These studies revealed the important role of polarization of –OH groups in PVA dielectric layer on the carrier mobility, hysteresis and bias stress and observed that PMMA/PVA/Al_2O_3 is an excellent dielectric system in OFETs with exceptional stability under high humidity and temperature conditions. Some works are reported on the study of the effect of measurement temperature on the field-effect carrier mobility induced by the polarization of ferroelectric dielectric system [30–32]. In this work, we have exploited the polarization of the dielectric systems with permanent dipole moment to measure temperate. Furthermore, we have demonstrated temperature-sensing capability of such devices in a wide temperature range (243–353 K) with excellent stability and enhanced performance. These devices are likely to find applications in monitoring temperature for vaccines, biochemicals or transporting medicine and organs etc., on site or at desired location.

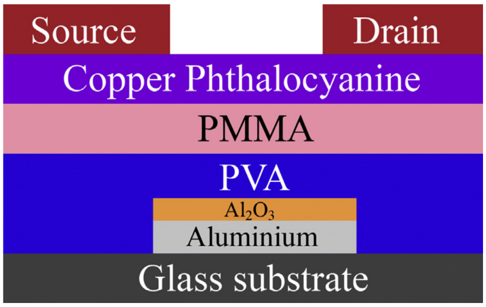

Fig. 1. Schematic diagram of the device fabricated using the top-contact and bottom-gate configuration on the glass substrate. The device contains PMMA/PVA/Al_2O_3 tri-layer gate dielectric material, aluminium as the gate electrode, CuPc as the organic semiconductor materials and copper as the source drain electrode material.

2. Materials and methods

2.1. Materials and design of OFETs

OFETs were fabricated on glass substrates with top-contact and bottom-gate configuration. The schematic diagram of the device structure is shown in Fig. 1. Thermally deposited ∼150 nm thick patterned aluminium layer has been used as gate electrode. In order to reduce the leakage current, the top surface of the aluminium was converted into Al_2O_3 (10 nm) by anodization process. The details of the anodization process and conditions were reported earlier [33]. Poly-(vinyl alcohol) (PVA, Sigma Aldrich, MW = 76500-81000 kg/mol) dielectric solution was prepared by dissolving 30 mg/mL in water and spun onto the anodized alumina substrates at 3000 rpm for 60 s, followed by annealing at 100 °C for 1h in vacuum oven. As third dielectric layer, PMMA (Sigma Aldrich, MW = 5,50,000 kg/mol, dissolved in anisole 10 mg/mL) film of ∼30 nm thick was grown on top of the PVA layer, which was vacuum annealed at 120 °C for 2 h to remove the residual solvent and to improve the film quality. A 60 nm thick CuPc (used as purchased from Sigma Aldrich) thin films was grown on top of tri-layer dielectric system at 80 °C substrate temperature with 0.2 Å/s. With the help of surface profilometer (Dektak), the individual layer thicknesses were measured. Device fabrication was completed by patterning 50 nm thick copper source-drain electrodes with defined channel width (W) and length (L) ratio of 30 using shadow masks.

Electrical characterizations of the OFETs were carried out in a probe station (Lake Shore) under vacuum ($<1 \times 10^{-4}$ mbar) with about 10% RH and under ambient humidity conditions (∼65% RH)