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Low-operating voltage and stable organic field-effect transistors with poly (methyl methacrylate) gate dielectric solution deposited from a high dipole moment solvent

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A low-operating voltage and stable pentacene field-effect transistor (FET) employing thin low-dielectric constant gate layer of poly (methyl methacrylate) (PMMA) dissolved in propylene carbonate (PC) has been realized. This device exhibiting high field-effect mobility, a threshold voltage of -1 V, and a small sub-threshold slope at operating voltages below -3 V is compared with an FET cast from PMMA film dissolved in a low dipole moment solvent. The negligible hysteresis and excellent electrical stability of FETs under gate bias stress with the use of PC are traceable to the low density of trap states in PMMA bulk and at the interfaces. © 2011 American Institute of Physics. [doi:10.1063/1.3669696]

Motivated by the need for low-cost, low-power, and large-area flexible electronics, the search for low-operating voltage and stable organic field-effect transistors (OFETs) has been of utmost attraction. In order to achieve stable and low-operating voltage OFETs, the dielectric surface has to be hydrophobic with a relatively high capacitance typically achievable via the use of high dielectric constant materials and/or ultra-thin layers. Poly (methyl methacrylate) (PMMA) is a great candidate for operationally stable and reproducible OFETs owing to its high hydrophobicity but has limitations in realizing low-operating voltage transistors due to its low dielectric constant ($k < 3.5$). Recently, low-operating voltage OFETs have been achieved through the use of high- k inorganic dielectric materials such as Al_2O_3 ,¹ $\text{Ti}_{1-x}\text{Si}_x\text{O}_2$,² and low- k /high- k dielectric hybrid bilayers such as PMMA/ ZrO_2 (Ref. 3) and PMMA/ Ta_2O_5 .⁴ Polymer dielectrics are still preferable over high- k inorganic dielectrics since they give rise to a high-quality interface.⁴ Low-operating voltage OFETs making use of high- k cross-linked polymer dielectrics such as poly (vinyl alcohol)^{5,6} and cyanoethylated pullulan⁷ have also been reported. Pentacene-based OFETs incorporating PMMA gate dielectric usually have high operating voltages (> -20 V) and PMMA films have been reported to have a high density of pin-holes⁸ and almost unusable without cross-linking for thicknesses below 100 nm.⁹ The lowest reported operating voltage (-8 V) for a PMMA-based OFET was achieved by using a 30 nm thick cross-linked PMMA layer.⁹

In this letter, we report on the study of the electrical characteristics and operational stability of low-operating voltage pentacene OFETs with thin pristine PMMA gate dielectric dissolved in propylene carbonate (PC) which has a large dipole moment (4.9 D at 20 °C). These results are compared with devices from pristine PMMA gate dielectric dissolved in butyl acetate (BTAc) with a low dipole moment (1.84 D at 22 °C). 8 wt. % of PMMA ($M_w = 996\,000$ g/mol)

were dissolved in PC and subsequently spincoated on aluminum-coated glass substrates in a nitrogen glove box. The PMMA dielectric film of thickness 70 nm was baked for 1 h at 100 °C. A 60 nm thick pentacene layer was thermally evaporated onto the dielectric surface at room temperature. Top contact source/drain (40 nm) gold electrodes defining a channel length and width of 500 μm and 50 μm , respectively, were thermally evaporated onto the pentacene film. The samples also supported device areas of $7.8 \times 10^{-3} \text{ cm}^{-2}$ for metal-insulator-semiconductor (MIS) diodes. Devices were also fabricated with PMMA using BTAc as the solvent (and denoted as PMMA-BTAc).¹⁰ OFETs/MIS structures cast from PC are simply denoted as PMMA.

The surface morphologies of PMMA and pentacene films on PMMA are shown in Figs. 1(a) and 1(b), respectively. The surface root-mean-square roughness of the PMMA film was found to be 1.0 nm over a $2 \mu\text{m} \times 2 \mu\text{m}$ area. The pentacene film on PMMA exhibited typical dendritic structure with layer-by-layer formation and large grain sizes ($\sim 1 \mu\text{m}$) comparable to other works,⁶ despite the relatively large PMMA film roughness which may be due to diminished polar groups on the PMMA surface. Polar groups are responsible for surface energy fluctuations on polymer dielectric surfaces, inhibiting surface diffusion and enhancing nucleation density.¹¹

The inset of Fig. 2 shows the leakage behavior of the PMMA insulator. A leakage current density of $9.8 \times 10^{-7} \text{ A cm}^{-2}$ at 0.45 MV cm^{-1} and a breakdown field exceeding 3.5 MV cm^{-1} , comparable with those reported for cross-linked PMMA,^{9,12} are observed. Capacitance-voltage (C - V) measurements from an MIS device (Fig. 2) show a typical p -type behavior. The device exhibits almost no hysteresis (no flat band voltage, V_{FB} , shift) when the gate bias is continuously swept from -3 V to 3 V at a step voltage of 0.05 V. The absence of hysteresis and near-zero occurrence of V_{FB} in the PMMA-based device are attributed to an improved dielectric layer with diminished trap states arising from polar groups, which usually give rise to slow polarization in PMMA bulk or at the semiconductor-PMMA interface.

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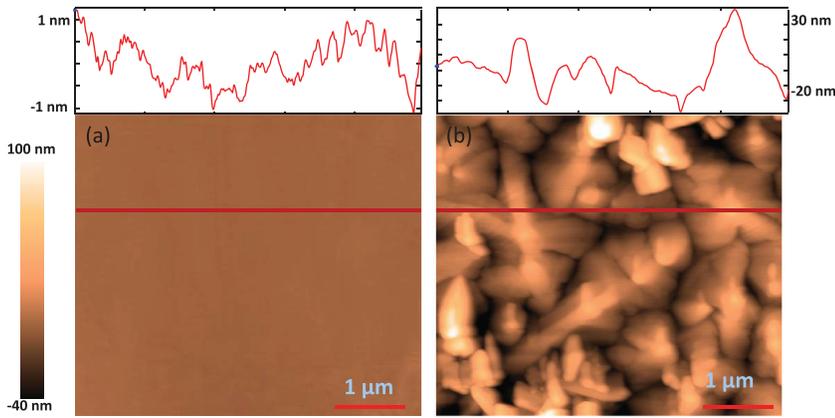


FIG. 1. (Color online) Atomic force microscopy images of (a) PMMA and (b) pentacene on PMMA are shown with cross sectional line scans above.

The improvements in dielectric properties observed with PC may be related to the mean square end-to-end distance, which increases with an increase in the dipole moment mainly due to an enhanced interaction of forces between the polymer and solvent cluster.¹³ As a result, the polymer chains remain more extended in solution. Furthermore, there may be an improvement in the degree of orientation of the polymer functional groups during deposition and baking.

Relevant extracted electrical parameters for both PMMA and PMMA-BTAc OFETs are summarized in Table I; every FET characteristic shows an enhancement with the use of PC over BTAc. Subsequent descriptions are focused on the PMMA-based OFET (with PC). Figures 3(a) and 3(b) show the output and transfer current-voltage (I-V) characteristics of an OFET measured under ambient conditions. These are typical characteristics of ~ 10 measured devices. The output characteristics could be operated below -3 V aided by its relatively high dielectric capacitance (20 nFcm $^{-2}$ at 100 kHz). The electrical parameters of OFETs were obtained from the saturation regime by using $\mu = \frac{2L}{WC_i} \left(\frac{\partial \sqrt{I_D}}{\partial V_G} \right)^2$, where C_i is the dielectric capacitance, W is the channel width, L is the channel length, V_G is the gate voltage, I_D is the drain current, and μ is the field-effect mobility. The subthreshold slope, S , controls the voltage swing required for the off-to-on switching of the transistor and ought to be as low as possible.⁷ The lower value of S here could be responsible for our

estimated near-zero OFET turn-on voltage (V_{on}) of -0.3 V. Based on the S value obtained from the OFET and using the relation, $N_{trap}^{max} \approx \left[\frac{qS \log(e)}{KT} - 1 \right] \frac{C_i}{q}$,⁷ we estimate a maximum trapped charge density of 7.1×10^{11} cm $^{-2}$.

In order to verify the reproducibility and operational stability the OFET, hysteresis, duty-cycle, and gate bias stress measurements were carried out. We investigated the hysteresis by fixing the drain voltage at -3 V and sweeping the gate voltage from 0 to -3 V (forward direction) and then back. As seen from Fig. 3(b), the OFET exhibits little off-regime hysteresis and a negligible positive shift of the threshold voltage (V_T) from -1 to -0.89 V ($\Delta V_T = 0.11$ V), consistent with the absence of hysteresis in the C-V characteristics of the corresponding MIS diode. We estimate an interfacial trap density (N_{trap}) of 1.38×10^{10} cm $^{-2}$ using the relation, $N_{trap} \approx C_i \Delta V_T / q$.⁷ Our N_{trap} value is about an order of magnitude lower than those reported for pentacene-based devices.⁷ Polar molecules at the organic semiconductor-dielectric interface have been reported to be responsible for positive V_T shifts in organic devices.¹⁴ The low values for the trapped charge density as well as a negligible ΔV_T are credited to the hydrophobicity and less-polarity of the PMMA surface, which strongly inhibit the adsorption of water molecules and resultant charge trapping activities.

To investigate the bias-stress effect on the OFET, which could be manifested in OFET hysteresis and/or V_T shifts, I-V transfer characteristic was first measured without applying any bias stress. Subsequently, bias stress ($V_G = V_D = -3$ V) was applied for set times and the I-V transfer characteristic was measured after each set time (Fig. 4(a)). In all, the OFET was subjected to 1960 s of dc bias stress. We observe very little shifts (0.08 V) in transfer characteristics—a confirmation of our low trap density estimates and a testament to

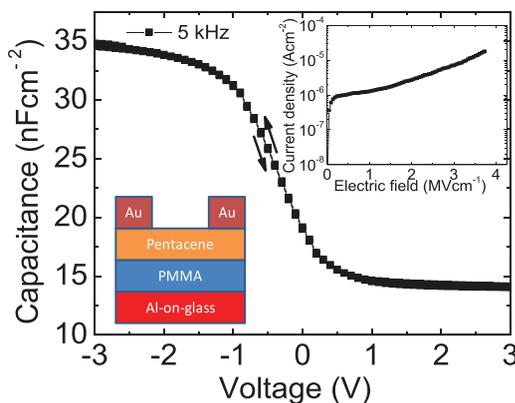


FIG. 2. (Color online) C-V curve of Al/PMMA-PC/Pentacene/Au MIS diode at a frequency of 5 kHz. Inset: Current density versus electric field characteristics of the diode and the device structure of OFETs used for this study.

TABLE I. Summary of electrical characteristics of PMMA (with PC) and PMMA-BTAc OFETs.

	μ (cm 2 /Vs)	V_T (V)	ΔV_T (V) (hysteresis)	S (V/dec)	ΔV_T (V) (bias stress)	V_{on} (V)	V_G (V)
PMMA	0.1	-1	0.11	0.4	0.08	-0.3	< -3
PMMA-BTAc	0.04	-2.7	0.43	1.5	0.18	-0.9	< -8

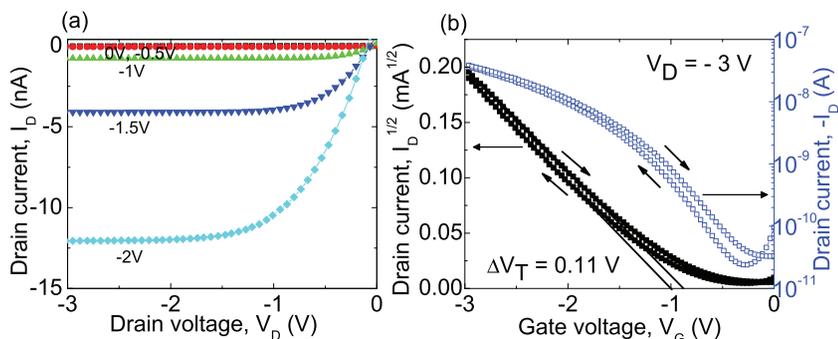


FIG. 3. (Color online) Representative (a) output and (b) transfer characteristics of a pentacene OFET.

the stability and reproducibility of our device. Bias stress investigations with similar time scales on SiO₂-based OFETs have shown V_T shifts higher than 13 V.¹⁵ Considering the critical relationship between MIS V_{FB} and FET V_T ,¹⁶ bias stress measurements carried out on the MIS diode showed no shifts in V_{FB} (inset of Fig. 4(b)) consistent with the OFET observations. Next, we stressed our OFET by repeatedly measuring transfer characteristics in the forward direction at a V_D of -3 V. This was carried out 25 times with a 4 s mea-

surement interval. Shown in Fig. 4(b) are representative plots for the 1st and 25th cycles. Here again, no obvious change in the shape of the transfer curves or any device degradation was seen.

In summary, this study demonstrates that with a proper choice of solvent, thin, non-crosslinked and single PMMA dielectric layers could be used to achieve stable and very low-operating voltage OFETs (below -3 V); hence, opening up the prospects of a possible utilization of less-polar and low- k dielectric polymers in realizing device-quality, stable, and low-voltage transistors that could be employed in the design of practical electronic circuits.

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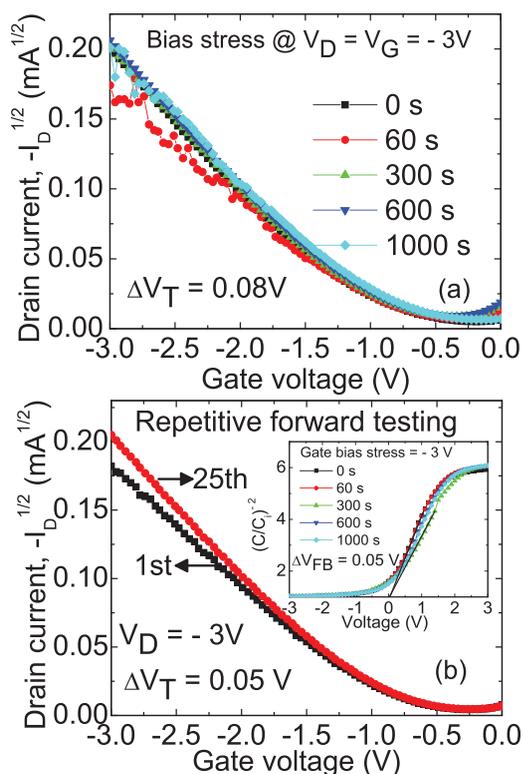


FIG. 4. (Color online) Operational stability of OFET transfer characteristics (a) after bias stress at $V_G = V_D = -3$ V at varying stress times. (b) Repetitive forward testing at $V_D = -3$ V. Inset: operational stability of MIS diode after bias stress at $V_G = -3$ V.

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