



Letter

High performance top contact fused thiophene–diketopyrrolopyrrole copolymer transistors using a photolithographic metal lift-off process



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ABSTRACT

High-performance fused thiophene–diketopyrrolopyrrole copolymer (PTDPPTFT4) top-contact transistors have been fabricated using a top contact metal lift-off process. The source–drain (S/D) top metal contacts were directly formed by i-line photolithographic patterning over the organic semiconductor channel through a lift-off method based on a negative tone photoresist. With a fluorinated imaging material (Orthogonal OSr 2312), devices exhibit field effect mobilities up to $2.56 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and on/off ratio $>10^7$, with channel lengths precisely patterned down to $10 \mu\text{m}$. Devices prepared from the lift-off process through a commercial negative tone photoresist such as AZ nLOF 2020 instead of the fluorinated photoresist exhibited considerable degradation. De-ionized (DI) water, aqueous tetramethyl ammonium hydroxide (TMAH) developer and organic stripper do not apparently degrade the performance of the PTDPPTFT4 transistors upon dip testing. Indeed the observed degradation originates from the incomplete development of the unexposed photoresist in the TMAH developer. This is detrimental to the PTDPPTFT4 channel due to formation of resist residue over the organic semiconductor surface. This residue eventually leads to a significant increase of the S/D contact resistance upon metal contact deposition.

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1. Introduction

Conjugated polymers have been attracting much attention due to many advantages such as low temperature solution processability, excellent mechanical properties and high throughput large area processing capability. Research efforts have been extensively focused on developing high performance new organic materials and it is expected that organic devices will be commercially available and impact the fabrication of modern electronics.

Recently, several high-performance polymer materials with field effect mobilities exceeding $1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, for example, poly(bis-alkylthiophenylthienothiophene) (PBTTT) [1], poly(cyclopentadithiophene–benzothiadiazole) [2,3], isoindigo-based polymers [4,5] and diketopyrrolopyrrole (DPP)-based polymers [6–16], have been reported. Photolithographic processing is believed to be impractical for these OTFT materials since the typical solvent of hydrocarbon based resists, propylene glycol monomethyl ether acetate (PGMEA) and aqueous developers are known to be harmful to the OTFT thin films due to surface swelling and water based medium involved.

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Many organic patterning methods have been developed, such as ink-jet printing [17] and nano-imprint lithography [18]. Photolithography is one of the most precise and high throughput techniques for mass fabrication of organic thin-film transistor (OTFT) devices and circuits. Development of a photolithography compatible high performance organic semiconductor should impact the current industrial manufacturing process and provide new technical possibilities.

In this letter, the use of a donor–acceptor thiophene–DPP organic semiconductor (OSC), namely PTDPTFT4, to fabricate top contact organic transistors [19] is reported. These OTFT top contacts are photolithographically patterned over the OSC layer through a lift-off method. Both hydrocarbon based (AZ nLoF 2020) and fluorinated (Orthogonal Inc OScR 2312) negative tone photoresists are intentionally used for comparison of this lift-off process. AZ nLoF 2020 photoresist is a phenol–formaldehyde (Cresol novolak) based resin with a small portion of melamine resin and an i-line photoacid generator (PAG) that can be dissolved in PGMEA, developed in an aqueous tetramethyl ammonium hydroxide (TMAH) developer, and stripped in organic strippers such as diethylene glycol monomethyl ether (DGME) or N-methyl-2-pyrrolidone (NMP). OScR 2312 is an i-line sensitive semi-perfluoroalkyl substituted highly fluorinated resin that can be dissolved and developed in a family of hydrofluoroethers (HFEs). UV exposure cleaves the semi-perfluoroalkyl side chains and therefore reduces the fluorinated photoresist's solubility in the fluorinated solvent. Stripping of the exposed fluorinated photoresist can be, for example, finished in a mixed miscible solution of HFE and ethanol, where ethanol functions as a stripping agent and HFE behaves as a solvent for carrying the dissolved fluorinated resin.

Compared to the reference device, where its top contact electrodes are patterned through metal evaporation over a shadow mask, the performance of the top-contact lift-off OTFT devices patterned by the hydrocarbon photoresist degrades obviously, while the one patterned by the fluorinated photoresist remains unchanged. In the case of the fluorinated photoresists [20,21], both the developer and the stripper involve mainly fluorinated solvents that do not cause re-dissolving, swelling, or cracking of the ordinary oleophilic or hydrophilic materials. Using this fluorinated photoresist, and hydrofluoroethers as the imaging media to perform top contact lift-off, the PTDPTFT4 transistor achieves a maximum mobility of $2.56 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is compatible to the performance of the reference devices.

The origin of device degradation in the hydrocarbon based lithographic materials system has been further investigated. Investigation of the wet process using AZ nLoF 2020 for top contact lift-off indicates that the TMAH developers and DGME strippers do not obviously affect the overall performance of the PTDPTFT4 devices. The device degradation is indeed originated from the incomplete development of the unexposed photoresist that is detrimental to the PTDPTFT4 channel, due to formation of the resist residue over the OSC surface prior to top contact metal deposition. This residue eventually leads to an increase of the contact resistance of the S/D electrodes.

2. Experimental

Heavily doped Si(100) wafers were used as a gate electrode with a 200 nm thermally grown silicon dioxide layer as the gate dielectric layer. Fig. 1(a) and (b) shows the structure of the common gate top contact PTDPTFT4 OTFT, and molecular structure of PTDPTFT4, respectively. Substrates were cleaned by sonication in semiconductor grade acetone and isopropanol, and then given a 2 min 20 W O_2 plasma treatment. Octyltrichlorosilane (OTS-C8) was used for surface modification of the gate dielectric layer. Prior to OTS-C8 treatment, pre-cleaned Si/SiO₂ samples were baked at 220 °C for 10 min in a nitrogen environment for dehydration. Samples were dipped into an OTS-C8 solution diluted in *p*-xylene (1.3 vol.%) at room temperature for 60 min, followed by an isopropanol rinse. A 5 mg/mL solution of PTDPTFT4 in 1,2,4-trichlorobenzene (TCB) was prepared by stirring at 120 °C for 20 min. OSC films were spin-coated with the spin profile 0 rpm for 10 s, 500 rpm for 5 s, then 1000 rpm for 60 s. Samples were subsequently baked at 190 °C in a nitrogen environment. In the case of the reference device, the patterned Au electrodes were formed by vacuum evaporation of gold through a metal shadow mask (Fig. 2(a)). For lithographically patterned top contacts, the Au electrodes were formed by a lift-off process using a negative tone photoresist. The process schematic diagram is shown in Fig. 2(b). The process flow is briefly described below: The OSC coated sample was removed from the glovebox, then a negative tone photoresist (PR) (either AZ nLoF 2020 from AZ Electronic Materials or OScR 2312 from Orthogonal Inc.) was spin-coated on top of the OSC layer at 1000 rpm in air. After pre-bake, the top-contact patterns were then imaged through a chromium photomask using an i-line ($\lambda = 365 \text{ nm}$) contact aligner (ABM Inc.). Image development was performed under either 2.38% TMAH or a hydrofluoroether developer so as to open up the S/D window for metal deposition. A 40 nm thick Au layer was vacuum deposited at a rate of 0.2–0.4 nm/s to form the top contacts. The PR film was finally lifted off in a mixture of hydrofluoroether and ethanol for OScR 2312 or DGME for AZ nLoF 2020 in order to leave patterned Au electrodes over the OSC layer. The lift-off process flows are summarized in Table 1.

3. Results and discussion

3.1. Performance of top contacts OTFT prepared with lift-off Au S/D contacts

Fig. 2(c) shows a microscopic image of the top contact transistor (10 μm channel length and 1 mm channel width) prepared by the top-contact lift-off method using OScR 2312. All top contact PTDPTFT4 transistors were characterized using a Keithley 4200-SCS semiconductor analyzer in air. Using OScR 2312 for top contact lift-off, devices show a maximum field effect mobility of $2.56 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, V_{TH} of 10.4 V and $I_{\text{on/off}}$ of $>2 \times 10^7$ along the saturation regime, as shown in Fig. 3. As a reference, the typical top contact PTDPTFT4 transistors were also

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