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Micro-scale twistable organic field effect transistors and complementary inverters fabricated by orthogonal photolithography on flexible polyimide substrate



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ABSTRACT

We fabricated micro-scale organic field effect transistors (OFETs) and complementary inverters on a twistable polyimide (PI) substrate by applying orthogonal photolithography. By applying a highly fluorinated photoresist and development solvent, it becomes possible to create organic electronic devices with a micro-scale channel length without damaging the underlying polymer films. The 3 μ m-channel twistable pentacene OFET devices and complementary inverters created using p-type pentacene and n-type copper hexadecafluorophthalocyanine exhibited stable electrical characteristics from flat to twist configurations (angle of up to ~50°). The realization of twistable micro-scale OFETs and inverter devices on a PI substrate may enable the production of functioning organic devices in practical, flexible configurations.

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1. Introduction

Recently, organic electronic devices, including organic field effect transistors (OFETs), non-volatile memories, light-emitting diodes, and solar cells, have received considerable attention based on their excellent advantages, such as a simple device structure, an easy and low-cost fabrication process, printability, flexibility, material variety, and a myriad of potential applications [1–9]. Because of the vast selection of organic materials for electronic devices, a variety of device fabrication processes have been demonstrated, including thermal evaporation, spin-coating, dropcasting, inkjet printing, and roll-to-roll printing [10–15]. However, in spite of these merits, for most organic electronic devices, it remains difficult to apply the conventional

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http://dx.doi.org/10.1016/j.orgel.2014.08.025 1566-1199/© 2014 Elsevier B.V. All rights reserved. photolithographic technology, which is widely used to create micro-scale high-density inorganic devices, because the existing organic solvents for photolithographic processing are not sufficiently selective to dissolve only the photoresist and not the organic layers [16,17]. To solve this problem, fluorous solvents are good candidates for the photolithographic processing of organic electronic devices because highly fluorinated materials generally have orthogonal properties to those of most organic materials, regardless of their polarity. The orthogonality is required to protect the polymer films during the addition of lithographic chemicals [18]. Among the various fluorous solvents that do not dissolve the organic materials, segregated hydrofluoroethers (HFEs) have attracted great attention because of their outstanding properties, such as their nonflammability, lack of ozone-depletion potential, low global warming potential, and low toxicity for humans. For this reason, the use of HFEs as photolithographic solvents has



been studied extensively besides being used as eco-friendly refrigerants and cleaning solvents [18–22].

Because the technological progress of user-friendly devices will be a major focus in future electronics, the flexibility of organic materials and electronic devices has become more important for their application in foldable and wearable electronic devices. To ensure that flexible organic electronic devices are the core elements in future electronics, flexible devices should work stably not only in bending conditions but also in complex twist configurations that occur in practical use [23–25]. As an example, our group has recently reported twistable non-volatile organic resistive memory devices [26].

In this study, we fabricated micro-scale twistable OFET devices and complementary inverters consisting of pentacene and copper hexadecafluorophthalocyanine (F_{16} CuPc) on a flexible polyimide (PI) substrate. The micro-scale device fabrication of the OFETs was made possible by applying orthogonal photolithography using the HFE photolithographic solvents and a compatible highly fluorinated photoresist solution. The 3 µm-channel twistable OFET devices fabricated in this study showed reliable electrical characteristics in the flat, bending (radius of 5 mm), and twist configurations (angle of up to ~50°). Furthermore, the 3 µm-channel complementary inverters had reliable voltage transfer logic inverter operations in both the flat and twist configurations.

2. Experimental section

(a)

(b)

Al gate deposition

on PI substrate

Fig. 1a shows the fabrication process of our micro-scale OFET devices using orthogonal photolithography. To fabricate the OFET devices, first, the PI substrate was cleaned using a standard solvent cleaning process using de-ionized

Ti/Au deposition

(DI) water and 2-propyl-alcohol (IPA) in an ultrasonic bath for 10 min at each cleaning. Next, the substrate was dried in a vacuum oven at 100 °C for 1 h to evaporate the residual solvent and moisture on the PI substrate. The bottom Al gate electrodes (30 nm thickness) were deposited by a thermal evaporator using a shadow mask with a deposition rate of 0.5 Å/s at a pressure of $\sim 10^{-6}$ torr. To prepare the polymeric dielectric layer, 15 wt% of poly (4-vinylphenol) (PVP) and 3 wt% of poly (melamine-co-formaldehyde) as a cross-linking agent were dissolved in a solvent of propylene glycol methyl ether acetate (PGMEA). After the UV-ozone treatment of the Al gate electrode/PI substrate for 10 min to improve the film uniformity and device reliability, the prepared PVP solution was spin-coated onto the Al gate electrode/PI substrate at 3000 rpm for 30 s, followed by soft-baking on a hot plate at 100 °C for 10 min in a N₂-filled glove box. To enable the OFET devices to be electrically measured by probe tips, the portion of the PVP film on the pads of the Al gate electrode was removed with a methanol-soaked swab, followed by hard-baking on the hot plate at 200 °C for 40 min in a N₂-filled glove box [27,28]. The thickness of the spin-coated PVP film was measured at $\sim 1 \ \mu m$ using an Alpha-step profiler. To prepare the fluorinated photoresist solution, 10 wt% of the previously reported semi-perfluoroalkyl resorcinarene (denoted as R_F-Calix-tBoc, shown in Scheme 1a) and 0.5 wt% of an Nnonafluorobutanesulfonyloxy-1,8-naphthalimide photoacid generator (Scheme 1b) were dissolved in a mixed solvent (3-ethoxy-1,1,1,2,3,4,4,5,5,6,6,6-dodecafluoro-2trifluoromethylhexane ((denoted as HFE 7500, shown in Scheme 1c): PGMEA = 4: 1 weight ratio), and then the mixed solution was filtered through a 0.20 µm-sized nylon syringe filter [19]. Afterwards, the fluorinated photoresist solution was spin-coated at 1500 rpm for 60 s onto the

Exposure and development



Lift-off process

PVP spin coating

Fig. 1. (a) The fabrication process of our micro-scale twistable OFET devices on a flexible PI substrate by orthogonal photolithography. (b) Photographic and optical microscopic images of the OFET devices with a schematic illustration of the twisted OFET devices.

PR spin coating

Pentacene deposition

Twist mold

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