



Ionic self-assembled monolayer for low contact resistance in inkjet-printed coplanar structure organic thin-film transistors



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ABSTRACT

To reduce the contact resistance in inkjet-printed organic thin-film transistors (OTFTs), the use of a newly synthesized ionic self-assembled monolayer (SAM) consisting of an anchoring group, a linker group, and an ionic functional group, is investigated. According to the gated transmission line method (TLM) measurements of a series of OTFT devices, where one type has no charge injection layer, another type having a pentafluorobenzenethiol (PFBT) injection layer, and a third type containing a (6-mercaptohexyl)trimethylammonium bromide (MTAB) ionic SAM, the latter exhibits the lowest contact resistance value of $\sim 3.1 \text{ K}\Omega \text{ cm}$. The OTFTs without charge injection layer and with the PFBT SAM have relatively higher contact resistance values of $\sim 6.4 \text{ K}\Omega \text{ cm}$ and $\sim 5.0 \text{ K}\Omega \text{ cm}$, respectively. The reduced contact resistance in the OTFTs with ionic SAMs is attributed to the large charge carrier density induced by the ionic SAM, which allows sufficient tunneling-assisted injection of the carriers from the metal electrode to the polymer semiconductor. These results suggest that the use of appropriate ionic SAM injection layer is an effective way to reduce the contact resistance, hence improving the charge transport characteristics of inkjet-printed OTFTs.

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1. Introduction

The field of organic printed electronics has been intensively investigated for a broad range of applications, such as flexible active-matrix displays, radio-frequency identification, non-volatile memory and bio/chemical sensors. As a result, several working prototypes in the above areas were recently introduced to potential consumers, meaning that the technology is nearing commercialization [1–3]. The components that make the above possible are organic thin-film transistors (OTFTs), which are the basic devices that act as switches in organic printed electronics [1–4].

In this regard, the development of OTFTs is of great importance, especially concerning the optimization of electrical properties so as to allow the manufacture of versatile printed electronic products.

Among the raised issues in the field of OTFTs, one of the most critical problems is the relatively high contact resistance at the organic/metal interface [5–7]. Since surface dipoles are present and usually a mismatch between the Fermi level of the electrode metal and the highest occupied molecular orbital (HOMO) level of the organic semiconductor is observed at the organic/metal interfaces, the contact resistance of the OTFTs is a major parameter that determines the electrical performance. In particular, for OTFTs with short channel lengths, the contact resistance problem is more severe and results in the deterioration of the organic circuit speed and larger power

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consumption of the applications [2,8–10]. In the past few years, to solve this issue, there have been several approaches such as electrode modification, molecular or oxide material doping, and the insertion of a buffer layer between the electrode and the active layer [11–18]. However, the effects of the electrode modification with plasma or UV/O₃ treatment are very limited, and most of the doping methods for the source/drain (S/D) contacts of OTFTs require vacuum-based thermal evaporation of the doping materials, which denies all the advantages of soluble printing process. In addition, selective patterning of the inserted buffer layers on the electrodes is usually accompanied with additional complex process steps, which also negate the benefits of low-cost printing process.

In this study, an ionic self-assembled monolayer (SAM) was synthesized for use as a contact layer, consisting of an anchoring group, a linker group, and an ionic functional group. In particular, the anchoring group of the ionic SAM contains sulfur, which selectively attaches to the metal electrode by forming preferential sulfur-metal coordination bonds (or dative covalent, semi-covalent bonds), and thus allows the ionic SAM to be deposited on the S/D electrodes with a simple dipping process. On the other hand, the ionic functional group is composed of the cation directly connected to the linker group and the anion binding to the counter cation to induce the additional charges in the polymer semiconductor. By applying the ionic SAM on photolithographically patterned bottom S/D contacts, coplanar OTFTs were fabricated and then compared with conventional OTFTs without any SAM treatment and OTFTs with a widely used pentafluorobenzenethiol (PFBT) SAM buffer layer, in terms of contact resistance and electrical performance. The contact resistance measurements were done using a gated transmission line method (TLM) [5–7].

2. Experiments

2.1. Material synthesis

For the synthesis, all chemicals were purchased from Aldrich and used without further purification. Fig. 1 shows the synthetic routes for a (6-mercaptohexyl)trimethylammonium bromide (MTAB) ionic SAM materials [19,20].

2.1.1. Compound 1

1,6-Dibromohexane (27.7 g, 0.10 mol) and potassium acetate (4.68 g, 0.042 mol) were added to an aceto-

nitrile solution (120 mL), which was then stirred at 90 °C for 12 h. The mixture was filtered, then the acetonitrile was evaporated under reduced pressure. A yellow solution was obtained and purified by column chromatography to provide a product solution (14.25 g, 59.6%).

2.1.2. Compound 2

A trimethylamine gas (34.58 g, 0.595 mol) was added to a chilled (−78 °C) mixture solution of compound 1 (14.25 g, 0.060 mol) in dry THF (250 mL). The reaction mixture was left to warm to room temperature and was stirred for 7 days. The white precipitate was filtered, and washed with THF several times and dried under a reduced pressure. Compound 2 was obtained with a 72% yield.

2.1.3. Compound 3 (MTAB)

A mixture of compound 2 (9.0 g, 0.030 mol) in 47% hydrobromic acid (90 mL) and ethanol (200 mL) was stirred for 2 days under N₂ ambient. The solvent was removed under a reduced pressure. The cooled crude mixture was poured into methanol and diethyl ether. Then the white precipitate was obtained by reprecipitation. The final product was dried in vacuum, and compound 3 was obtained with 51% yield. ¹H NMR (CD₃OD, 300 MHz): δ [ppm] 3.37 (t, 2H, CH₂), 3.14 (s, 9H, CH₃), 2.52 (t, 2H, CH₂), 1.79 (m, 2H, CH₂), 1.64 (m, 2H, CH₂), 1.46 (m, 4H, CH₂); Anal. calcd for C₉H₂₂BrNS: C, 42.18; H, 8.65; N, 5.47. Found: C, 42.02; H, 8.70; N, 5.42.

2.2. Device fabrication and electrical characterization

For the fabrication of inkjet-printed OTFTs employed for the contact resistance extraction experiments, molybdenum (Mo) was sputter-deposited onto glass substrates as a first step. The deposited Mo was wet-etched to pattern the gate electrode into the designed shape. Subsequently, a 300 nm thick silicon dioxide (SiO₂, capacitance C_i = 10 nF/cm²) was deposited by plasma enhanced chemical vapor deposition (PECVD) as the gate dielectric. Before the deposition of Au, a (3-Mercaptopropyl)trimethoxysilane (MPS) (Purchased from Aldrich) with 10 mmol concentration in *n*-hexane was applied to enhance adhesion between SiO₂ and Au [21]. The S/D electrodes (Au: thickness ~50 nm) were deposited by e-beam evaporation and patterned again by photolithography. For the surface treatment of the gate dielectric, octadecyltrichlorosilane (ODTS) SAM (Purchased from Aldrich) was used. The substrate was

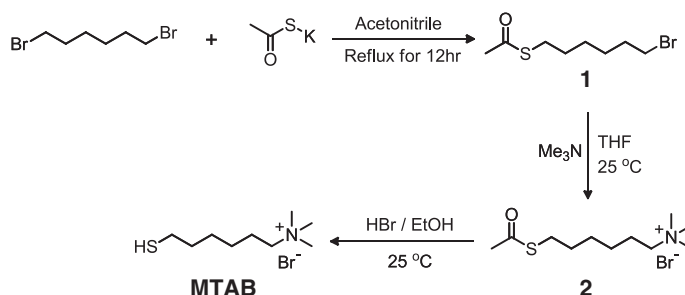


Fig. 1. Synthetic routes of the (6-mercaptohexyl)trimethylammonium bromide (MTAB) ionic SAM compound.

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