Organic thin-film transistors with over 10 cm²/Vs mobility through low-temperature solution coating

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ABSTRACT
Recent studies on organic thin-film transistors (OTFTs) have reported high mobility values, but many of them showed non-ideal current–voltage characteristics that could lead to the overestimation of the mobility values. In this study, the non-ideal transistor behavior was briefly investigated by considering the effect of charge injection, and a method of overcoming the effect was developed. Correspondingly, various charge injection layers were developed, and their effects on the modification of metal contacts, including work function tuning and interfacial doping, were studied. The materials that had been coated formed a good metal-semiconductor interface through fine manipulation in the wetting and dewetting of the selected liquid. With such electrodes, the OTFTs were fabricated at room temperature and exhibited almost ideal transistor behavior in terms of the current–voltage characteristics, featuring high (over 10 cm²/Vs) field-effect mobility.

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1. Introduction
In organic thin-film transistors (OTFTs), surprisingly high mobility has been increasingly reported, from 40 to close to 100 cm²/Vs [1,2]. Yet, the precise extraction of the mobility value for electronic applications is still debatable [3]. It has been reported that the resistance of organic field-effect transistors (OFETs) increases from the Schottky contact between metals and organic semiconductors, and can lead to non-linear conductance in the OTFTs. This not only leads to overestimating the carrier mobility but also causes a low current density in the device. The energetic barriers and/or interface states at the heterojunction contacts considerably consume voltage drop [4], generate Joule heating [5], and decrease the injection efficiency. Especially, a large contact resistance seriously suppresses the performance of thin-film transistors (TFTs) used for the back panel of active displays [6], sensors, memories, and other facile electronics, which require a low driving voltage and a high on–off ratio [7]. Such limitations raise the demand for ohmic-contact injection [8], which has not been achieved in most OTFTs or OFETs. As a result, such transistors exhibit non-ideal current–voltage characteristics and are not suitable for circuit applications in display or sensor technologies.

Since the past several decades, the printing of OTFTs has rapidly developed and has demonstrated a fascinating realization of integrated circuits and flexible wearable electronics. Especially, all-inkjet-printed TFTs have shown promising performances for the textile manufacturing, display circuit, and label-free bio-sensing, as demonstrated by Castro et al. [9], Fukuda et al. [10], and Medina-Sánchez et al. [11]. Moreover, the yielding in the upscaling of manufacturing has been improved through the fine-tuning of printing techniques, as shown by Sowade et al. [12]. The above state-of-the-art advances in the development and production of OTFTs have made it possible to envision a flexible electronic industry based on printed OTFTs in the future.

The non-ideal transistor behavior, which can be understood by considering the effect of charge injection, is briefly discussed in this paper. Various charge injection
layers were developed in this work, and their effect on modifying the contacts was studied. Finally, control of the depositing solution of the charge injection layers was obtained for the fabrication of fine electrodes, and OTFTs were fabricated through the solution process. The resulting OTFT exhibited almost ideal transistor behavior, with high (over 10 cm²/Vs) field-effect mobility.

2. Results and discussion

2.1. Understanding the non-ideal transistor behavior

The non-ideal transistor behaviors in OTFTs can be understood by considering the charge injection process. Figure 1(a) shows the typical structure of OTFTs and Figure 1(b) shows the possible injection process at the metal-organic interface. Actually, injection barriers generally exist at the contacts of OTFTs, caused by the mismatched work functions of the metals with respect to the injection levels in organic semiconductors [13] or the interfacial states with charge transfer and dipole moments [14]. The injection barrier leads to a depletion region [15], where the carriers are depleted and need to diffuse through the region after a number of collisions. Using the common parameters of intrinsic organic semiconductors in the literature, $W_d$ is estimated to be over 1 μm in OTFTs if the effective barrier height is 0.5 eV [16]. This value is much larger than the mean free path ($l_{\text{free}}$) of a carrier in OSCs, which is only in the order of the intermolecular distance (several Å to nm) owing to the structural disorders and localized polarons [17], pointing to the possibility of diffusion-limited injection. Especially, at the metal-organic interface, carriers can recombine with their image charges when thermal energy $kT$ reaches the carrier image binding energy [18,19], and recombination can occur with unipolar carriers. The critical distance for recombination $x_C$ for common OSCs (∼5 nm) [19] is larger than or comparable with $l_{\text{free}}$ (several Å to a few nm) (Figure 1(b)). The recombination current is opposite the thermionic injected current and affects the detailed balance between the two to derive

![Figure 1](image-url)

**Figure 1.** (a) Typical configuration of organic TFTs. (b) Injection processes in the metal-organic semiconductor interface in OTFTs. (c) Non-ideal transistor behaviors in OTFTs with the transfer curve and the differential mobility (obtained by the differential of the square root of $I_d$ against $V_g$), which exhibits an abnormal peak due to the contact effect. (d) Ideal transistor behaviors, where the differential mobility rises as $V_g$ increases. Carriers fill up from the less mobile tail states in the weak accumulation (‘weak acc.’) to the mobile central states in the density of states (DOS) in the strong accumulation (‘strong acc.’).
the net injection current [19]. The surface recombination velocity near the interface is determined by the carrier mobility. The general form of contact resistance \( R_c \) in OTFTs is then presumed to be as shown below [20]:

\[
R_c \propto \mu^{-\gamma} \times (F)^a \exp\left(-B(F)^b\right) \times \exp\left(\frac{\psi_{\text{eff}}}{\varphi_0}\right). \tag{1}
\]

Here, \( \mu \) is the carrier mobility, \( F \) is the electric field near the contact area determined by drain voltage \( V_d \) and gate voltage \( V_g \), \( \psi_{\text{eff}} \) is the effective injection barrier, \( \varphi_0 \) is the thermal energy at certain temperatures, and the other parameters \( (a, b, \gamma \) and \( B \)) are determined by the device structures and materials. From Figure 1(b), a diffusion-limited injection in OTFTs is expected because \( l_{\text{free}} \leq W_d \), and it is probably accompanied by a surface recombination process as \( l_{\text{free}} \leq x_C \).

It can be learned from Equation (1) that the contact resistance is sensitive to \( V_g \) and \( V_d \) if the injection barrier is high [21]. In the case where the contact resistance is sharply reduced as a function of \( V_g \) in transfer scanning [22], it will cause a sudden increase in the current and then lead to a sharp peak of the extracted mobility [23], which is calculated from the saturated regime using the equation below:

\[
\mu_{\text{sat}} = \frac{2L}{W C_i} \left( \frac{\partial \sqrt{I_D}}{\partial V_G} \right)^2. \tag{2}
\]

This scenario is shown in Figure 1(c), where the transfer curve of an OTFT with a large injection barrier (0.6 eV) is simulated by a numerical tool [24]. The carrier mobility of the semiconductor is set to be 1 cm²/Vs, but the non-ideal behaviors lead to peak values much higher than that. In comparison, a TFT with ohmic contact will exhibit transistor behavior, as shown in Figure 1(d). The extracted mobility rises as \( V_g \) increases because the carriers fill up from the less mobile tail states (i.e. weak acc.) to the mobile central states in the density of states (DOS). Various experimental studies confirmed such features, and they roughly follow a power law \( \mu \propto (V_g - V_{\text{th}})^\beta \)

where \( V_{\text{th}} \) is the threshold voltage and \( \beta \) is a characteristic factor determined by the device structures and materials.

### 2.2. Developing aqueous charge injection materials

To achieve ideal transistor behavior capable of supporting circuit applications, the contact injections need to be improved by reducing the injection barrier or narrowing the depletion region, as shown in Figure 1(b). Towards this end, one of the most promising candidate materials for this purpose, molybdenum oxide (MoO₃), which has deep-lying electronic states and is non-toxic, was investigated [25]. In particular, MoO₃ can be solution-processed to allow low-cost and large-area fabrication through cost-effective printing processes. As opposed to the several precursors for fabricating a solution-processed MoO₃ layer that have been reported [26], the synthesis of an aqueous MoO₃ solution that can be fabricated with a low processing temperature, a short reaction time, simple low-cost processing steps, and a well-controllable low surface roughness of the deposited film was developed [27]. The fabrication process of the MoO₃ solution is shown in Figure 2(a). The aqueous transparent solution with MoO₃ in ammonia-water is formed by the chemical reaction between MoO₃, H₂O, and NH₃ (i.e. MoO₃ + 2NH₃ + H₂O → (NH₄)₂MoO₄). The ammonium molybdate (NH₄)₂MoO₄ is soluble in H₂O, and the solution is transparent, colorless, and stable, without undergoing any change in color or transmittance under ambient conditions for 2 months. The aqueous MoO₃ solution was spin-coated onto a pristine Mo surface and annealed at 150°C for 10 min in air. During the annealing process, the ammonium molybdate underwent rapid decomposition into three components: MoO₃, NH₃, and H₂O. NH₃ and H₂O were evaporated into the air and MoO₃ was left in the film.

The topological images of the MoO₃ films with which the Mo substrates were coated were taken from a tapping-mode atomic force microscope (AFM), as shown in Figure 2(b). By controlling the concentration of MoO₃ in the ammonia-water, the density of the nanoparticles increased, and a continuous and smooth MoO₃ film was formed, with a slightly increased roughness (0.557 nm at 4 mg/ml and 0.750 nm at 8 mg/ml). Such a low roughness of the channel region will be ideal for contact modifications. The X-ray photoemission spectroscopy (XPS) image shown in Figure 2(c) reflects the characteristic peaks of the O1s, Mo3s, Mo3p, and Mo3d levels of the MoO₃ film. The core level of Mo3d with the Mo3d5/2 and Mo3/2 peaks is located at 236.3 and 233.2 eV, respectively, which indicates the presence of the Mo6⁺ cation as the characteristic peak of molybdenum. A further test through Kelvin probe indicated that the work function of the MoO₃-coated Mo electrodes ranges from −4.40 to −4.47 eV as the MoO₃ solution concentration increased from 0.1 to 0.8 wt% (Figure 2(d)). The work function was pinned to be around the original value (4.45 eV) because the bare Mo had been readily oxidized and covered by a thin layer of MoO₃ containing the Mo-O-Mo3d(±5) or Mo-O-Mo3d(±6) complexes. The interfacial electronic structure of Mo and poly(3-hexylthiophene) (P3HT) was studied via ultraviolet photoemission spectroscopy (UPS). Figure 2(e) shows that the binding energy of the polymer progressively decreased with a thicker MoO₃ film on the surface: 19.71 eV for P3HT, 19.42 eV at 1 nm MoO₃ on P3HT, 18.58 eV at 2 nm MoO₃ on P3HT, and 18.93 eV at 3 nm MoO₃ on P3HT, respectively. It
Figure 2. (a) Images of the MoO$_3$ powder solution in NH$_3$ solvent and after heating at 80°C for 30 min. (b) AFM images of the MoO$_3$ films when spin-coated at various concentrations on the Mo substrate (the units of rms are 0.001 nm). (c) X-ray photoemission spectra of the MoO$_3$ films with characteristic peaks. (d) Work function of Mo electrode dependence on MoO$_3$ thickness. (e) UPS spectra of the MoO$_3$ films with different thicknesses that were thermally deposited on a P3HT thin film. Redrawn from Ref. [21] with permission from PCCP Owner Societies. Copyright (2015). (f) Schematic diagram of the energy levels in the P3HT polymers with or without MoO$_3$.

also shows that the HOMO states progressively shifted towards the Fermi level, probably indicating that p-doping in P3HT was formed. The valence edge shifted from 1.5 eV for the pristine P3HT film to 0.46 eV in the film with 3 nm MoO$_3$. The doping would promote the charge injection by reducing the depletion thickness at the contacts [16], as indicated by Equation (1).

Besides, another aqueous charge injection material for the V$_2$O$_5$ solution was obtained following a similar strategy, as shown in Figure 3(a)–(c). The aqueous transparent solution of V$_2$O$_5$ in ammonia-water is formed by the chemical reaction among V$_2$O$_5$, H$_2$O, and NH$_3$ (i.e. V$_2$O$_5$ + 6NH$_3$ + 3H$_2$O → 2(NH$_4$)$_3$VO$_4$) [28]. After the film deposition on the Mo electrodes, the topological images indicated that the V$_2$O$_5$ films had a generally uniform quality (Figure 3(d–f)). As the solution concentration went further up to 0.25, 0.5, and 0.8 wt%, the V$_2$O$_5$ film thickness $d$ increased almost linearly to 2.4, 3.7, and 5.8 nm, respectively. As opposed to the MoO$_3$ layers, the work function of the Mo substrate can be significantly changed by depositing the V$_2$O$_5$ insertion layer (Figure 3(g,h)). It was found that the strongly polar V$_2$O$_5$ molecules adsorbed on Mo remarkably changed the work function, which homogeneously increased from 4.45 eV in the case of the pristine Mo film to 4.56 eV (1.2 nm V$_2$O$_5$), 4.65 eV (2.4 nm V$_2$O$_5$), 4.77 eV (3.7 nm V$_2$O$_5$), and 4.85 eV (5.5 nm V$_2$O$_5$). Apparently, the Mo–O–V complexes formed between the nanostructured V$_2$O$_5$ interlayer and the Mo bottom-contact induced the increased surface dipole moment. Also, the gradual change in the work function is attributed to the increased coverage by the additional molecules of V$_2$O$_5$ [29]. The shifting of the work function towards high values is expected to narrow the injection barrier between the electrodes and the deep-lying HOMO levels of the organic semiconductors, and to benefit the contact injection.

2.3. Achieving ideal transistor behaviors in OTFTs by printing

The aforementioned developed materials were applied to fabricated fully printed OTFTs for modifying the interfaces of Au electrodes and organic semiconductors. First of all, electrodes were fabricated through the self-assembly method, by screen-printing a guide layer of hydrophobic CYTOP (500 nm) and then gold nanoparticles successively. The experiment details are the same as those described in [30]. Then, solutions with charge injection materials were deposited onto the Au electrodes, as shown in Figure 4(a). The hydrophilic source/drain electrodes combined with the CYTOP bank
Figure 3. (a–c) Images of the (a) V$_2$O$_5$ powder, (b) V$_2$O$_5$ solution in NH$_3$ solvent, and (3) transparent solution. (d–f) AFM images of the V$_2$O$_5$ films formed via spin-coating using V$_2$O$_5$ solution at various concentrations. (g) Schematic representations of the V$_2$O$_5$ films spin-coated on the Mo electrode. (h) Work function tuning by the V$_2$O$_5$ films spin-coated on the Mo electrodes, and the corresponding film thickness. Redrawn from Ref. [23]. Copyright (2015).

Figure 4. (a) Schematic representations of the depositing charge injection materials. (b) Real-time observation of the water droplet’s evolution near the channel region. (c) Schematic representations of solution dewetting for splitting. (d) The height at which the solution splits onto the electrodes, as a function of the contact angle on the channel region.
layer efficiently guided the aqueous solutions to the electrode surfaces. The real-time observation of the water droplet’s evolution near the channel region clearly illustrated that the solution finally remained on the electrodes (Figure 4(b)). At the beginning, a 0.1-μl droplet of MoO3 solution self-assembled into the electrode and channel area, and then gradually shrunk as the water evaporated. The droplet completely separated and then rapidly dried onto the electrodes. The whole process took around 80 s.

Especially, when the channel is narrow (e.g. a few microns in width), the precise deposition of the charge injection layer solution calls for the consideration of the dynamic process of the wetting and splitting of the solution droplets [31]. The thermal equilibrium is considered only before the liquid layer splits, and a circular curvature is assumed when the liquid splits. The total interface tension of the liquid layer before liquid splitting is denoted as $\gamma_1$ (Figure 4(c), upper panel). When the liquid splits from the top surface to the bottom of the channel, the total surface energy of the liquid layer and the exposed channel on top of the channel is denoted as $\gamma_2$ (the channel width is defined as $L$, Figure 4(c), bottom panel). The liquid layer splits at $h_c$ when $\gamma_1$ is larger than $\gamma_2$.

$$\gamma_1 - \gamma_2 \cong [ L - (\pi - 2)t_c ] \gamma_{sl} + L(\gamma_{sl} - \gamma_{sg}) \geq 0. \quad (3)$$

Here, $L$ refers to the channel length where the surface is hydrophobic. The subscripts $s$, $l$, and $g$ refer to solid, liquid, and gas, respectively. Note that Young’s equation $\gamma_{sg} - \gamma_{sl} = \gamma_{l}\cos\theta$ and $\theta$ is the contact angle of the solution on the hydrophobic channel. Then, the liquid layer splits if it evaporates to reach the critical thickness $t_c$.

$$t_c \leq \frac{L}{\pi - 2}(1 - \cos\theta). \quad (4)$$

The quantitative relations among $\theta$, $t_c$, and $L$ are shown in Figure 4(c). As the channel length is reduced, it is more difficult for the solution to split as reduced $t_c$ and larger contact angles of the hydrophobic area will be needed to facilitate the splitting to form electrodes.

The formation of charge injection layers, including molybdenum oxide (MoO3), vanadium oxide (V2O5), and tungsten oxide (WO3) layers, is expected to decrease the charge injection barrier at the contact interface [32]. Figure 5(a) illustrates the OTFT configuration. The fabrication methods used mainly involved the self-assembled deposition of solutions and regular coating [30]. The flexible PEN substrates were cut into square pieces ($4 \times 4$ cm), and then Perylene-C was deposited through the chemical vacuum evaporation system to reach an 800-nm thickness. This layer was intended to modify the PEN surface and to act as a smoothing layer. Then, the substrates were exposed to UV irradiation (172 nm wavelength) under vacuum for 15–25 s through a photomask, to form the source and drain patterns. Then, the substrate was coated with Au nanoparticle ink using a glass rod to form source and drain electrodes, and the ink was dried out naturally. The charge injection layers (i.e. MoO3, V2O5, and WO3 layers) were then deposited by rod-coating the solution, which self-assembled onto the Au surface, as shown in Figure 4(b). Consequently, to deposit the guide layer, the Cytop solution was used for coating by screen-printing through a screening mask to form banks surrounding 500-nm-deep arrays of semiconductor films. After drying the CYTOP film, C8-BTBT solution (0.4 wt% in anisole) was drop-casted onto the substrates and self-assembled into the banks to form a thin semiconductor film. During the drying, the substrates were held perpendicularly to one another to guarantee the fast and uniform crystallization of the C8-BTBT material. After removing the CYTOP layer by rinsing it with CYTOP solvent 180 and drying it at 40°C for 20 min, the dielectric bilayer with 250 nm Cytop and 600 nm parylene-C was deposited through spin-coating and chemical vapor deposition, respectively. The dielectric capacitance was measured to be 2.8 nF/cm². Finally, top gate electrodes were deposited following the same method as that used in forming the source and drain electrodes, resulting in the devices shown in Figure 5(b).

The transistors showed decent turn-on behavior, and the transfer characteristics are shown in Figure 5(c–e). The OTFTs with V2O5, MoO3, and WO3 exhibited high values of $\mu_{FET}$ (11.6, 13.1, and 10.0 cm²/Vs, respectively). The threshold voltage was generally small (2.0–2.5 V). Moreover, the turn-on characteristics exhibited a quite small threshold slope of 0.3 V/decade. The differential mobility can be extracted using Equation (2), and all the values of $\partial \sqrt{I_D}/\partial V_G$ generally increase in a power law with $V_G$, and almost follow the ideal transistor behavior, as discussed earlier. It has been reported that printed C8-BTBT single crystals exhibit up to 30 cm²/Vs mobility, where the crystals were inkjet-printed through antisolvent for crystallization [33]. Yet, the output characteristics showed a significant signature of non-linear charge injection. Also, it has been reported that the spin-coated C8-BTBT crystals have shown high mobility (more than 40 cm²/Vs) [2], and yet the transfer characteristics show kink features that may be induced by the Schottky contact, and thus, the mobility value does not hold when the gate voltage is large [22]. Compared with that in the previous studies, the mobility value of the presented devices in this study is among the highest values, and the transistor behavior is close to that of the ideal transistors. For comparison, the control OTFTs without the presented technique and structure (i.e. spin-coated or thermally evaporated polycrystalline OTFTs with bare Au
Figure 5. (a) Schematic representations of the OTFT structure. (b) Optical image of OTFT. (c–e) Transfer and output characteristics of OTFTs with charge injection layers, and differentials \( \partial \sqrt{I_D}/\partial V_G \) for calculating the mobility in the saturated regime, which are generally like those of the ideal transistors. The OTFTs with V2O5, MoO3, and WO3 exhibited high \( \mu_{\text{FET}} \) values (11.6, 13.1, and 10.0 cm²/Vs, respectively).

Electrodes) exhibited much field-effect mobility (smaller than 1 cm²/Vs) [34]. Also, the control OTFTs in the same structure but without the charge injection layer exhibited lower field-effect mobility, with average values of 1.8 cm²/Vs, in the authors’ previous studies [21].

The output curves are shown in Figure R2. The curves were fitted through simulations using the following two methods: (1) using the classic equations for TFTs (i.e. \( I_D = (W/L)C_i\mu\left(V_G - V_{th} - V_d/2\right)V_d \)) and (2) using the G-function to extract contact resistance \( R_C \) from the experiment data, as described in Ref**, and then adding it to total resistance

\[
R_{\text{tot}} = R_C + R_{\text{ch}} = R_C + \frac{L}{WC_i\mu\left(V_G - V_{th} - V_d/2\right) V_d}
\]

and so \( I_D = V_d/R_{\text{tot}} \). The results are shown in Figure R3a and b, where the fitted curves without contact resistance are in black and the fitted curves with contact resistance are in blue. It can be seen that the super-linear dependence of the \( I_d - V_d \) curve coming from the contact resistance is small especially in the devices with an MoO3 insertion layer, and that the difference between the measured curves from the fabricated OTFTs and the ideal transistors as simulated is not large. The reason for this can be found by comparing the extracted contact resistance and channel resistance, as shown in Figure R3c and d. As shown in these figures, the contact resistance became smaller than the channel resistance at around \( V_d = -2 \) or \(-3\) V for the OTFTs with MoO3 and WO3, respectively. Therefore, the impact of the contact resistance was not serious, and the measured curves were not significantly different from those of the ideal transistors (Figures 6 and 7).

The surface of the semiconducting layer C8-BTBT was characterized with an AFM and an optical microscope (Figure S1), and it was indicated that the film surface in the channel region was highly crystalline and that there were no noticeable differences between the samples with...
Figure 6. Output characteristics for the OTFTs with (a) V$_2$O$_5$, (b) MoO$_3$, and (c) WO$_3$ insertion layers, respectively.

Figure 7. (a) Comparison of the simulated and experimental output curves for the OTFT with an MoO$_3$ insertion layer. Here, the experiment data (black circles) are compared with the simulated data without contact resistance (gray curve) and with contact resistance (blue curve). (b) Extracted contact resistance and channel resistance by G-function for the OTFT with a WO$_3$ insertion layer. (c, d) Same data for the OTFT with a WO$_3$ insertion layer.

and without charge injection layers, respectively. In addition, the thickness of the organic semiconductor layer was determined to be 100–150 nm using a surface profiler. Such studies showed that the improvement of the devices could not be attributed to the morphological difference between the contacts with and without a charge injection layer, respectively. Instead, it can be said that the crystallization procedures highly affect the crystal morphology. When the substrates were held perpendicularly in this study, the crystals were along a certain direction, showing a better-ordered structure compared with the spin-coated film (Figure S2).

In addition, note that the temperature can significantly affect the device performance. For the temperature effect, it was observed that the OTFTs with C8-BTBT crystals still showed a high performance in the low-temperature measurements, as reported in the authors’ previous work [35]. For the temperature stability, the melting temperature of the C8-BTBT materials, however, was relatively low (i.e., 126°C). This is partially due to the design rules of a low processing temperature and good solubility. Thus, the presented OTFTs work well only at around room temperature and below 100°C. In the future, other semiconducting materials with higher temperature tolerance will be studied for their potential use in the presented fabrication techniques.

3. Conclusions

The non-ideal transistor behavior was investigated by considering the effect of the charge injection. Various
charge injection layers were synthesized, and their effect on the modification of the contacts was studied. The resulting OTFT exhibited almost ideal transistor behavior, with high field-effect mobility (i.e. over 10 cm²/Vs). The study showed the potential of the use of printed OTFTs for electronic applications.

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