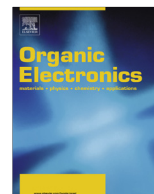
Organic Electronics 15 (2014) 1738–1744

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel



Metal evaporation dependent charge injection in organic transistors

Yong Xu [a,*], Chuan Liu [a], Huabin Sun [b], Francis Balestra [c], Gerard Ghibaudo [c], Will Scheideler [d], Yong-Young Noh [a,*]

[a] Department of Energy and Materials Engineering, Dongguk University, 26 Pil-dong, 3-ga, Jung-gu, Seoul 100-715, Republic of Korea
[b] School of Electrical Science and Engineering, Nanjing University, Nanjing 210093, PR China
[c] IMEP-LAHC, INP-Grenoble, MINATEC, 3 Parvis Louis Neel, BP 257, 38016 Grenoble, France
[d] Electrical Engineering and Computer Sciences, University of California, Berkeley, CA 94720, USA

ARTICLE INFO

Article history:
Received 7 March 2014
Received in revised form 29 April 2014
Accepted 5 May 2014
Available online 17 May 2014

Keywords:
Charge injection
Metal diffusion
Metal evaporation
Organic transistor

ABSTRACT

To illuminate a long-term remaining issue on how contact metallization (metal and speed) affects charge injection, we investigated top-contact pentacene transistors using two categories of metals deposited at various rates. Differing from previous studies such as those devoted to morphological influences by microscopy, in this work we concentrated on their electrical characteristics in particular combining the low-frequency noise which provided a direct quantity of trap density and its evolution with respect to contact metal and deposition rate. It turns out that the transistors with noble metal (Au) suffer from metal-diffusion related charge trapping in the pentacene bulk close to the Au/pentacene interface, and this diffusion-limited injection is greatly tuned from bulk to interface by speeding Au deposition which leads to a Schottky-like injection due to the severe thermal damage to the upper pentacene layer. Applying a conventional contacting metal (Cu), however, Ohmic contacts with much fewer traps are always observed regardless of metallization speed. This is attributed to an ultra-thin interlayer of Cu_xO that guarantees stable Ohmic injection by introducing gap states and protecting the pentacene film so that those transistors appear to be free from Cu metallization. Our results quantitatively show the limiting factors of charge injection for different metals and at various evaporation rates.

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1. Introduction

Organic field-effect transistors (OFETs) have been studied for more than three decades and recently exhibited impressive progresses in both material engineering and fabrication technology [1,2]. However, controlling charge injection remains a big obstacle to overcome. OFETs suffer from significant contact influences (contact resistance $R_C \approx$ kΩ cm) from hetero-junctions created by deposition of metal onto intrinsic organic semiconductor (OSC) or vice versa [3]. The charge injection is essentially governed by the Schottky–Mott limit while it was observed to depend greatly on many other factors such as metal–organic reaction (e.g., Al, In, Ti, Sn with PTCDA) [4] and/or metal oxidation upon exposure to air (e.g., Al) [5] or involving oxygen absorbed in OSC (e.g., Cu with pentacene) [6], metal/organic interdiffusion (e.g., In/PTCDA [7], Al/F_{16}CuPc [5], Au/pentacene [8,9] and Au/DIP [10]) as well as interface charge redistribution [11–13]. These effects play more important roles when depositing metal onto OSC compared to depositing OSC-on-metal, due to enhancement caused by greater metal diffusion [14]. Meanwhile, the

* Corresponding authors. Tel.: +82 2 22604974.
E-mail addresses: xuyong.hn@gmail.com (Y. Xu), yynoh@dongguk.edu (Y.-Y. Noh).

http://dx.doi.org/10.1016/j.orgel.2014.05.006
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molecular stacking geometry that defines the charge transport therein is disordered more significantly by the enhanced reaction and diffusion [5,9]. In reviewing the extensive studies of contact materials, one may notice that they are mostly from the perspectives of chemistry, morphology and electronic structure and seldom from a point of view of practical devices [9,15]. A simple question we may raise is how to fabricate the contacts to attain optimal device performance? Indeed, metal evaporation can substantially change the mechanism of interface formation, e.g., speeding metal deposition (sometimes combined substrate cooling) yields more abrupt interface, due to rapid metal aggregation [16]. Taking into account the interface chemistry (e.g., reactive or non-reactive) [15], the effects of metal evaporation on charge injection in OFETs becomes a complex question which remains unanswered.

Here, we investigate the influences of the contact metal evaporation on the charge injection in top-contact pentacene OFETs using two categories of metals (a noble metal, Au, and a usual contacting metal Cu) which were deposited at various rates. With increasing the evaporation rate for Au and Cu, the corresponding OFETs displayed distinct properties. The Au-contact OFETs showed notably degraded characteristics whereas no obvious change was observed for the Cu-contact OFETs. After a detailed examination of the current–voltage (*I*–*V*) and low-frequency noise (LFN) characteristics we found that, the tunable Au-diffusion at various evaporation rates and the interfacial Cu-oxidation led to rather different mechanisms of charge injection in such pentacene OFETs.

2. Device fabrication and characterizations

Bottom-gate and top-contact pentacene OFETs were made on heavily *p*-doped Si(100) wafers covered with 50 nm SiO_2, see inset of Fig. 1(a). The substrates were cleaned by ultrasonic in acetone and isopropyl alcohol and then spin coated 40 nm PMMA that has been reported to favor pentacene ordered growth [17,18]. The Afterward, a 40 nm-thick pentacene layer was deposited in vacuum ($\approx 10^{-5}$ Pa) at room temperature through a metal mask (0.1 Å/s) for all the substrates. Typical film morphology by AFM is shown as inset in Fig. 1(a) as well. Finally, 40-nm-thick Au or Cu source/drain electrodes were thermally deposited (using tungsten boat) in vacuum ($\approx 5 \times 10^{-4}$ Pa) through a metal mask by various rates (0.1, 0.5, 1.0 and 1.5 Å/s), yielding a uniform channel width $W = 1000$ μm and different channel lengths (L) varying from 50 μm to 350 μm with an interval of 50 μm. The *I*–*V*, capacitance–voltage (*C*–*V*) and LFN characterizations were carried out at room temperature in air by using a semiconductor parameter analyzer HP4156C, a LCR meter Agilent 4284A and a programmable point-probe noise measuring system (3PNMS) [19], respectively.

3. Results and discussions

3.1. Influences on I–V characteristics

Fig. 1(a–d) shows the transfer characteristics with respect to the metal evaporation rate, where the drain current (I_D) is normalized by (L/W) in order to visualize device uniformity and to compare Au and Cu. At higher evaporation rate the Au-contact OFETs exhibit worse characteristics, indicating significant influences of Au metallization on pentacene. Here, it is worth mentioning that the intermediate rate of 1.0 Å/s led to failure of Au's device functionality for the first time, due to a Schottky-like charge injection showed as very poor linearity in the output characteristics (not shown). We tried again using backup substrate and found that the devices still exhibited the worst characteristics as seen in Fig. 1(c). The reproducible issue implies a critical rate around 1.0 Å/s for the tradeoff between Au diffusion and accumulation. Increasing the deposition rate for Au, these two effects compensate each other and their joint impacts reach maxima at a specific rate, and thus degrade the device characteristics very severely and randomly. Detailed discussion will be given below. In contrast to Au, there is nearly no change observed for the Cu-contact pentacene OFETs, indicative of slight influences arising from Cu metallization.

3.2. Mobility and threshold voltage

The low-field mobility (μ_0) extracted by the *Y*-function method [20] in linear regime is shown in Fig. 1(e–h), where a gate dielectric capacitance per unit area ($C_i = 3.5 \times 10^{-8}$ F/cm^2) estimated from the *C*–*V* measurement was used to calculate the mobility. One can readily recognize a constant μ_0 for Cu-contact OFETs, consistent with minor contact influences. Different features are seen from Au-contact counterparts: at a slow rate of 0.1 Å/s, μ_0 is approximately constant and comparable to Cu. Upon a small increase of deposition rate to 0.5 Å/s, μ_0 is lowered for short L and then continues to increase with L. This effect retains for higher rates and is in line with significant impacts of Au metallization. A noteworthy result here is that at a fast rate of 1.5 Å/s, μ_0 is boosted to higher value than that of 0.1 Å/s at long L. Although the μ_0 evaluated by *Y* function has suppressed the contact resistance effects (i.e., its attenuation of transistor's effective mobility) [20], the metallization-induced influences on the charge transport is not eliminated. Hence, such a higher mobility signifies ameliorated access transport. The threshold voltage (V_T) (Fig. 1(i–l)) shows similar features as μ_0: Cu metallization does not affect V_T but fast deposition of Au largely increases $-V_T$, up to −8 V at 1.0 Å/s. A histogram (inset) illustrates their distribution, indicating that V_T considerably varies with Au deposition rate. In contrast, for Cu, irrespective of slow/fast rates, V_T distribution always centers around −2 V with a small dispersion, demonstrating the advantage of Cu against Au to achieve superior process stability. As discussed above, 1.0 Å/s would be the worst rate for Au giving the highest V_T.

3.3. Contact resistance

We then extracted their R_C by using the transfer-length method (TLM), cf. insets in Fig. 2(a and b). A notable difference between these two TLM plots is that the R_C of Au-contact OFETs is obtained from the *Y*-axis intercepts decreasing with the gate voltage (V_G). For the Cu-contact

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