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Ambipolar Carrier Transport in Polycrystalline Pentacene Thin-Film Transistors

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We have investigated the dependence of polycrystalline pentacene thin-film transistors (TFTs) characteristics on work function of source-drain contact metals. The pentacene TFTs using Mg, Al, Ag, and Au source-drain electrodes showed only p-type characteristics and the field-effect hole mobilities were strongly dependent on their work function. On the other hand, the pentacene TFTs using Ca source-drain electrodes showed typical ambipolar characteristics. The field-effect hole mobility of $4.5 \times 10^{-4} \text{ cm}^2/\text{Vs}$ and field-effect electron mobility of $2.7 \times 10^{-5} \text{ cm}^2/\text{Vs}$ were estimated from saturation currents. Appearance of an electron enhancement mode in pentacene TFTs was ascribed to the lowering of barrier for electron injection at source electrodes.

Keywords: mobility; organic semiconductor; pentacene; transistor; work function

INTRODUCTION

Many studies on carrier transport properties of polycrystalline pentacene as p-type organic semiconductor have been reported for organic electronics devices, especially organic thin-film transistors (OTFTs) [1–3]. However, there are few reports that investigate n-type performances in pentacene TFTs, mainly due to the large electron injection barrier from source electrode to the lowest unoccupied molecular orbital (LUMO) of pentacene [4–7]. In this study, we have fabricated pentacene TFTs by depositing the source-drain contacts on the

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polycrystalline pentacene film, in order to realize and investigate n-type performance of pentacene TFTs. We find a clear dependence of p-type and n-type pentacene TFT performance on the metal work function.

EXPERIMENTAL

We fabricated OTETs having a top contact geometry shown in Figure 1(a). The methods of fabricating the gate electrode, the gate insulator and the thin film of polycrystalline pentacene were described in our previous reports [6,8]. The surface image of polycrystalline pentacene on the poly-chloro-*p*-xylylene (diX-C, Daisankasei Co. Ltd.) insulating layer was also reported in our previous paper [9]. A shadow mask with an interdigitated configuration was attached onto the film in the glove box and the substrate with the mask was transferred again in the vacuum chamber to form metal source-drain electrodes. The metals used in this study were Au, Ag, Al, Mg with Ag overlayer and Ca with Al overlayer. The channel length L and width W were 75 mm and 5 mm, respectively. Finally, the fabricated device was placed in the glove box and TFT characteristics were examined using an Agilent 4156C precision semiconductor parameter analyzer. The electric parameters were estimated using a standard analytic theory of MOSFET according to the following equation:

$$I_{D,sat} = \frac{WC_i}{2L} \mu (V_G - V_T)^2$$

where $I_{D,sat}$ is the saturation drain current, C_i is the capacitance per unit area of the insulating layer, V_T is the threshold voltage, V_G is the gate voltage and μ is the field-effect mobility.

The energy level diagrams of pentacene and work function of metals are illustrated in Figure 1(b). The highest occupied molecular

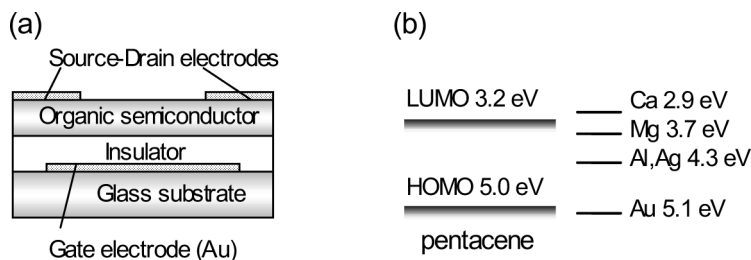


FIGURE 1 (a) Schematic cross-section of the organic thin-film transistor in this study. (b) Energy level diagram of pentacene and metal electrodes.

orbital (HOMO) of pentacene was estimated to be 5.0 eV by photoemission apparatus, AC-2 (Rikenkeiki). Optical energy band-gap (E_g) of pentacene was estimated to be 1.8 eV from the absorption spectra edge, and LUMO of pentacene was calculated to be 3.2 eV from the equation $LUMO = HOMO - E_g$.

RESULTS AND DISCUSSION

Figure 2(a) shows the drain current (I_D) vs. the drain voltage (V_D) with varying the applied gate voltage (V_G) for pentacene TFT using Mg with Ag overlayer source-drain electrodes. On the application of negative bias to the gate electrode, a typical p-type behavior was observed. The on/off drain current ratio ($V_D = -100$) was estimated to be 2.0×10^4 from Figure 2(b). Above the threshold voltage, the field-effect hole mobility can be calculated to be $7.2 \times 10^{-4} \text{ cm}^2/\text{Vs}$ from the slope of the plot of $|I_{D,sat}|^{1/2}$ vs. V_G . On the other hand, upon application of a positive bias to the gate electrode, a typical characteristic in n-type mode was not observed.

Figure 3(a) shows I_D vs. V_D with varying the applied V_G for pentacene TFT with Ca with Al overlayer source-drain electrodes. On the application of negative bias to the gate electrode, a typical p-type behavior was observed. The field-effect hole mobility can be calculated to be $4.5 \times 10^{-4} \text{ cm}^2/\text{Vs}$. From the transfer characteristics in Figure 3(b), the on/off ratio was estimated to be 18 ($V_D = -100 \text{ V}$), 1.4×10^2

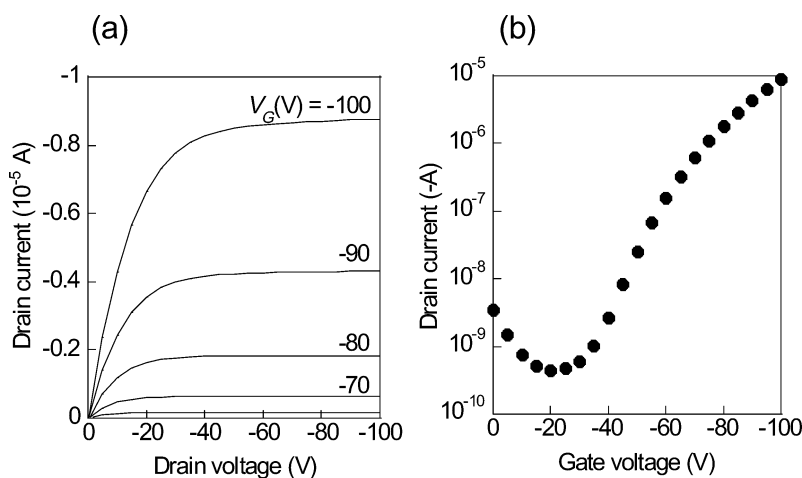


FIGURE 2 (a) Output characteristics and (b) transfer characteristics of p-type TFT using polycrystalline pentacene and Mg source-drain electrodes.

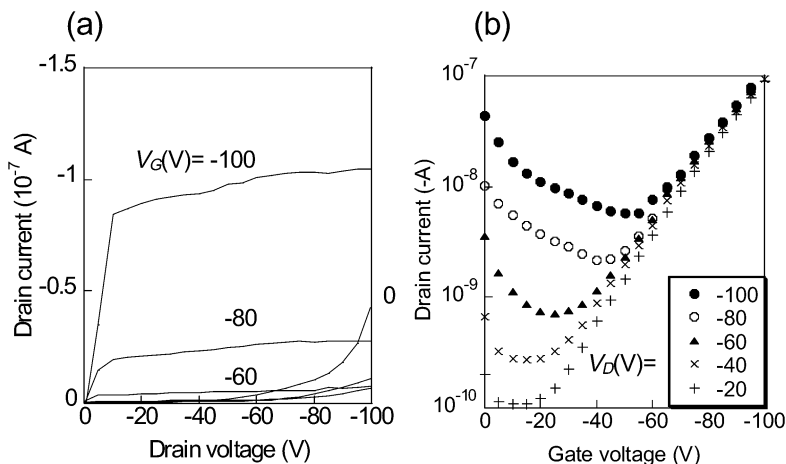


FIGURE 3 (a) Output characteristics and (b) transfer characteristics of p-type TFT using polycrystalline pentacene and Ca source-drain electrodes.

($V_D = -60$ V) and 8.1×10^2 ($V_D = -20$ V). The increase of I_D with high negative V_D is observed for low negative V_G (ranging from -60 to 0 V). This current increase is attributed to the electron injection into pentacene at the Ca drain electrode [10] and lead to the relative low on/off ratio.

Figure 4 shows the dependence of field-effect mobility on the metal work function. The highest performance of p-type pentacene TFT is obtained using metal with the largest work function Au (field-effect hole mobility, $0.56 \text{ cm}^2/\text{Vs}$), intermediate p-type performance is obtained using metals with smaller work function such as Ag ($0.010 \text{ cm}^2/\text{Vs}$), Mg ($7.2 \times 10^{-4} \text{ cm}^2/\text{Vs}$) and Ca ($4.5 \times 10^{-4} \text{ cm}^2/\text{Vs}$). A simulation result reported by Bolognesi *et al.* has shown that a high injection barrier between the source electrode and the organic semiconductor results in a large decrease in field-effect mobility [11]. The observed a few orders of magnitude decrease in the field-effect hole mobilities over the case of the Au source-drain electrodes was also due to the large hole injection barrier. Significantly degraded performance is obtained using Al ($9.8 \times 10^{-5} \text{ cm}^2/\text{Vs}$). This may be due to an unintentional chemical reaction between a thermal evaporated Al and pentacene.

Upon application of a positive bias to the gate electrode, a typical n-type behavior was observed in pentacene TFT with Ca source-drain electrodes as shown in Figure 5(a). The field-effect electron mobility can be calculated to be 2.7×10^{-5} . Appearance of n-type pentacene

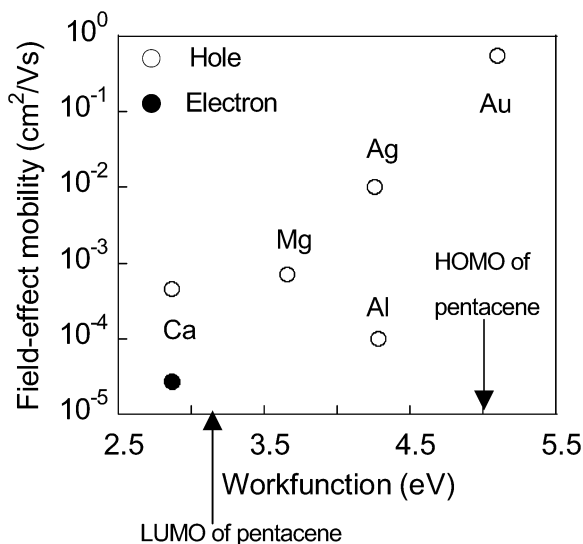


FIGURE 4 Dependence of field-effect mobility in pentacene TFTs on the metal work function.

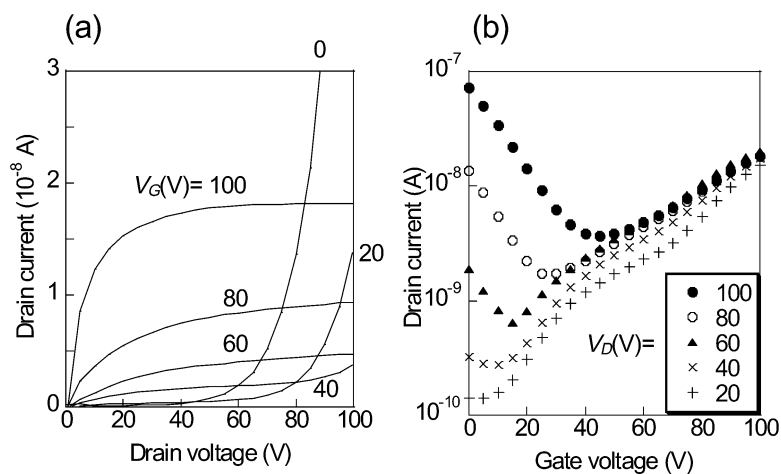


FIGURE 5 (a) Output characteristics and (b) transfer characteristics of n-type TFT using polycrystalline pentacene and Ca source-drain electrodes.

TFT was ascribed to the lowering of barrier for electron injection at source electrodes. From the transfer characteristics in Figure 5(b), the on/off ratio was estimated to be 5 ($V_D = 100$ V), 31 ($V_D = 60$ V) and 1.1×10^2 ($V_D = 20$ V).

CONCLUSION

We have investigated the dependence of polycrystalline pentacene TFTs characteristics on work function of source-drain contact metals. By reducing the electron injection barrier, an n-type characteristic was successfully observed in pentacene TFTs with Ca source-drain electrodes. On the other hand, pentacene TFTs with Mg, Al, Ag, and Au source-drain electrodes exhibited only p-type characteristics.

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