Revealing the effect of electrode materials on the performance of low-voltage pentacene thin film transistor by in-situ surface potential measurement

Q L Chen¹, M Ouyang², Q C Liu¹, H Y Jin¹,³ and W G Xie²

¹ Department of Medical Devices, Guangdong Food and Drug Vocational College, Guangdong 510520, PR China
² Siyuan Laboratory, Department of Physics, Jinan University, Guangzhou, Guangdong 510632, PR China
E-mail: jinhy@gdyzy.edu.cn

Abstract. We investigate the effect of contact potential between Au, CuO₅ and Au-CuO₅, and pentacene thin films by Kevin Probe microscopy. Compared to the Au contact, although the injection barrier is lower for CuO₅ contact, the extraction barrier is higher, and the total contact resistance is only slightly smaller. The better performance of CuO₅ contact device results from the doping effect from the p-type CuO₅. The introduction of additional Au top layer deposited on the thin CuO₅ layer lowers the extraction barrier and facilitates better carrier transfer, which further reduces the contact resistance. Therefore, both the apparent mobility and channel mobility are the best in the device with Au-CuO₅ double layer contact.

1. Introduction

Organic thin film transistors (OTFTs) have received intensive research in recent years because of their potential application in flexible electronics [1, 2]. Gold is the most commonly used electrode material in OTFTs. Owing to the high work function, good conductivity, and environmental stability, high-performance pentacene thin film transistors (TFTs) have been developed in recent years [3, 4]. However, large-scale application of Au electrode is limited by the high cost and scarcity of gold. There have been continuous efforts to replace gold with low-cost materials, for example, Cu-based materials [5-7]. The energy offset at the interface of Cu/pentacene is expected to be higher than that of Au/pentacene. However, Wang et al. [5] showed that the contact resistance of pentacene TFT with Cu electrodes was smaller than that of Au contact device, which was attributed to the narrower trap distribution in Cu contact. Di et al. [6] found that a CuO₅ interfacial layer would form between the Cu and pentacene thin film, which improved the hole injection because of better energy level alignment. Modification of the Cu electrode with 7,7,8,8-tetracyanoquinodimethane (TCNQ) could further reduce the contact resistance. In our recent study, we also demonstrated that a partially oxidized CuO₅ electrode greatly raised the mobility of the pentacene TFT. Furthermore, the introduction of an additional Au top layer deposited on the thin CuO₅ layer results in the increase of apparent mobility from 1.0 cm²/Vs to 2.0 cm²/Vs. Transfer line method has shown that the optimization of the electrode leads to a dramatic decrease of contact resistance [7]. The above findings show that the performance...
of OTFT has been successfully improved by the use of Cu-based materials, however, the underlying mechanism is not systematically revealed. In this study, we investigate the surface potential distribution in the channel of the working pentacene TFTs using Kelvin probe force microscopy (KPFM). The effects of the electrode materials and structural optimization on the performance of the devices are discussed.

2. Experimental methods
An Al2O3/TiOx (ATO) dielectric layer modified by octadecylphosphonic acid (ODPA) was used as gate dielectric for pentacene TFTs [8]. A 30-nm-thick pentacene film was deposited onto the ODPA/ATO substrates at a rate of 0.1 Å/s and pressure of 3 × 10^-4 Pa with the substrate temperature at 60°C. A 40-nm-thick S/D electrode layer was then deposited onto the pentacene layer through a shadow mask at a rate of 0.3 Å/s. For Au electrodes, the deposition pressure was about 5 × 10^-4 Pa. For CuOx electrodes, Cu source is deposited onto the pentacene at a deposition pressure of about 5 × 10^-3 Pa. X-ray photoelectron emission spectroscopy (XPS) has confirmed that Cu can be oxidized to CuOx (mainly Cu2O) at low vacuum condition [7]. For the Au-CuOx complex electrodes, a CuOx layer was deposited first. An Au top layer was then deposited at high vacuum condition. The device performance was measured in ambient conditions using Keithley 4200SCS. The in-situ surface potential profile was measured by integrating a Bruker DI 3100 atomic force microscopy with KPFM mode to the device measurement setup, which is shown in figure 1(a) [9].

![Figure 1](image.png)

**Figure 1.** (a) Schematic setup of in-situ surface potential measurement; Normalized surface potential profile along the channel of Au contact (b), CuOx contact (c) and Au-CuOx contact (d) devices.

3. Results & discussions
To clearly observe the potential distribution induced by the applied VDS, the surface potential profile at
$V_{GS}=V_{DS}=0$ V was subtracted in advance, and the voltage drop along the channel of Au contact device is shown in figure 1(b). At small $V_{DS}$, the linear variation of surface potential across the channel is observed, which identifies that the device is working in the linear region. At higher $V_{DS}$, the surface potential near the drain electrode changes more rapidly. It means that a depletion layer forms and the device is working in the saturation region.

The total resistance of a TFT device is the sum of resistances of the source ($R_S$), drain ($R_D$) and channel ($R_{Ch}$), which can be directly calculated from the potential profiles as shown in figure 2. We find that in the linear operation regime, the contribution from the contact resistance $R_C=R_S+R_D$ is the smallest in the Au-CuO$_x$ contact device ($R_{Ch}/R_C=2.33$, $V_{DS}=-0.3$ V). In the other two devices, the contribution from the contact resistance is similar ($R_{Ch}/R_C \sim 0.3-0.4$, $V_{DS}=-0.3$ V). It’s also expected that in the linear operation regime, the contact resistance $R_C$ is the sum of $R_S$ and $R_D$. The $R_S$ approximately equals to $R_D$ [5]. However, we find that this is correct only for Au-CuO$_x$. In the Au contact device, the ratio of $R_D/R_S$ is 2 – 3. For CuO$_x$ contact, this ratio is as high as 10.

![Figure 2. The calculated resistance of $R_S$, $R_D$, $R_{Ch}$ in the devices with contact of Au (a), CuO$_x$ (b) and Au-CuO$_x$ (c).](image)

To understand the effect of electrode materials on the device performance, we consider the vacuum level alignment of the device at $V_{DS}=V_{GS}=0$ V (figure 3(a)). When a gate bias is applied, the vacuum level will slightly increase initially, but then slowly decrease to its original value, implying that the energy level is pinned. It can be seen from figure 3(a) that there is an energy level offset of -0.27 eV between the Au electrode and pentacene. When the contact is changed to CuO$_x$ (mainly Cu$_2$O, IP=5.3 eV [10]), a clear upward shifting of the energy band is observed. The energy diagrams are constructed in figures 3(b)-3(d) according to the energy level alignment in figure 3(a).

As CuO$_x$ is a p-type semiconductor, the shifting of the energy band can be understood as the hole doping from the electrode. The doping from the electrode is easier to observe from the Au-CuO$_x$ contact. In this case, the CuO$_x$ layer is thinner than the full CuO$_x$ electrode so that the doping effect is smaller. The potential near the electrode drops down, whereas at the center of the channel, the potential increases again. Due to the doping effect, the hole density will increase, and hole traps at the interface of pentacene/dielectric should be passivated, which facilitates better hole transport. As a consequence, the depletion of the channel, which indicated by the rapid increase of $R_{Ch}$ with $V_{DS}$, is more profound in the Au contact device (figure 2). In the other two contacts, especially in the case of CuO$_x$ contact, the depletion region is restricted near the vicinity of the drain contact (figures 1(c) & 1(d)).

We then consider $R_S$ in the linear operation region at -0.3 V, Where $R_S$ is not affected by the depletion of the carriers. We find that $R_{S,\text{Au}}$ (0.5M) > $R_{S,\text{CuO}_x}$ (0.04 M) > $R_{S,\text{Au/CuO}_x}$ (0.03 M). Considering the energy level alignment in figure 3(b), the large contact resistance in the case of Au contact should be partially caused by the large injection barrier of 0.27eV. In addition, studies have revealed that the Au atoms will diffuse into the pentacene layer, which will cause interfacial defects and raise the contact resistance [11]. In figure 3(c), holes at CuO$_x$/pentacene interface do not feel the barrier when injected from the electrode to the pentacene thin film. The resistance of the source is thus caused by the high bulk resistance of CuO$_x$. 

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The resistance of the drain contact is determined by the extraction barrier and the depletion extent. For Au contact in figure 3(b), the extraction of the holes from the channel should be easier than the injection of the holes, so it cannot account for the large $R_D/R_S$ ratio. Considering the trend of $R_D$ and $R_{Ch}$ with the $V_{DS}$ in figure 2(a), we find that the $R_D$ comes to a saturated value at low bias. At the same time, the channel depletion becomes obvious. This indicates that the drain contact is depleted, which leads to a relatively high $R_D/R_S$ ratio. The easy depletion of the contact should be caused by the traps induced by the pentacene/dielectric interface and the diffusion of Au atoms into the pentacene thin film. In the case of CuO$_x$/pentacene contact, the large $R_D/R_S$ ratio at low $V_{DS}$ should come from the large barrier of 0.46 eV for hole when it injects from the pentacene into the CuO$_x$ (figure 3(c)). Such a high barrier seriously limits the carrier extraction, leading to high $R_D$. The $R_D$ will decrease dramatically when an increasing $V_{DS}$ bend down the barrier. It comes to a minimum resistance at $V_{DS} = 0.6$ V, after which, the depletion effect leads to increased resistance again (figure 2(b)). Thinning of the CuO$_x$ layer may lead to increased tunneling possibility of the carriers; however, the relative poor resistance of CuO$_x$ itself will also limit the transfer of holes as well (figure 3(b)). When the CuO$_x$...
layer was thinned, and a top Au layer was used to encapsulate the electrode, the problem solved. The holes can tunnel easier to the Au layer and be transferred out (figure 3(d)). This is why we observed a dramatic decrease of $R_D/R_S$ ratio in the contact of Au-CuOx.

From the above discussion, we can see that the contact effect in the three types of device is different. With the Au-CuOx contact, the contact resistance is reduced to a minimum value. To ensure that the device still keeps the best performance after the removal of the contact effect, we evaluate the real device performance by calculating the mobility of the channel from the potential profiles near the source electrode. The channel mobility at a $V_{DS}$ can be extracted from the potential profile where it varied linearly using the following equation:

$$I_{DS} = eWn(x)\mu(V'_{G}, E)E(x)$$  \hspace{1cm} (1)

where $W$ is channel length, and $e$ is the elemental charge. $E(x)$ is the local electrical field that can be determined by $-\frac{\partial V}{\partial x}$ from the measured surface potential profile. The interfacial carrier density $n(x)$ is given by $C_{ox}(V_{GS}-V_{t})/e$.

Figure 4(a) shows that the saturated electrical field in the channel of CuOx contact device is the smallest. The small electrical field is due to the localization of the potential near the drain contact, which leads to the low voltage drop in the channel. In the Au-CuOx device, the contact effect is the smallest. Most of the applied voltage will drop at the channel, which strongly raises the channel electrical field.

![Figure 4](image_url)

**Figure 4.** Electrical field (a) and mobility (b) of the channel calculated from the linear region in the surface potential profile

As shown in figure 4(b), the carrier mobility is almost the same under different $V_{DS}$. The mobility of the Au-CuOx device is still the highest. It is partially caused by the high electrical field in the channel, which enhances hoping transport of carriers. Moreover, although the electrical field of the CuOx contact device is smaller than that of the Au contact device, its mobility is higher. This indicates that doping from the electrodes, which pre-passivates the interface traps, also contributes to the high carrier mobility by reducing the trapping and detrapping probability of carriers. The results confirm that the optimization of the contact materials improves the device performance.

4. Conclusions

In this work, the effect of the electrode materials on the performance of pentacene TFT devices is revealed. Although the CuOx electrode is not so efficient for holes extraction, it reduces the injection barrier and induces hole doping to the pentacene channel, which favor the hole transport easier. With the additional Au top layer covered, the CuOx interlayer reduces the extraction barrier. As a consequence, the contact resistance of pentacene TFT is dramatically reduced by the use of Au-CuOx double layer electrode. Both the apparent mobility and channel mobility of the OTFT are greatly
improved over that of the Au or CuO x contact devices. The findings show that the OTFT performance can be optimized by carefully choosing the materials types and electrode structures.

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