

High performance of pentacene field effect transistors using graphene electrodes and substrate treatments

Sangchul Lee¹, Gunho Jo², Seok-Ju Kang², Gunuk Wang², Minhyeok Choi², Woojin Park², Jongwon Yoon², Taehyeon Kwon¹, Dong-Yu Kim^{1,2}, Yung Ho Kahng³, and Takhee Lee^{1,2,4,*}

¹Department of Nanobio Materials and Electronics, ²School of Materials Science and Engineering, ³Research Institute for Solar and Sustainable Energies, Gwangju Institute of Science and Technology, Gwangju 500-712, Korea

⁴Department of Physics and Astronomy, Seoul National University, Gwanak-gu, Seoul, 151-747 Korea
Phone: +82-62-715-2313 E-mail: tlee@gist.ac.kr

1. Introduction

Organic field effect transistors (OFETs) have received great interest due to their cost-efficient fabrication, limitless material variety, and the wide variety of potential applications [1]. For improving performance of OFETs, choosing appropriate contact electrode material and substrate treatments are significantly important [2]. Recently, graphene have attracted a great deal of attention as an alternative electrode material. In addition, self-assembled-monolayer treatment for dielectrics, has been carried out.

In this study, we report on the highly improved performance of pentacene OFETs with graphene electrodes and surface treatments [3,4]. The results revealed that performance improvement was induced from the formation of favorable interface which results in the reduction of the resistances. Through detailed characterizations, the output and transfer currents were increased and the total resistance was decreased. Moreover, we show significant improvement of OFET performances such as higher mobility of $\sim 1.2 \text{ cm}^2/\text{V}\cdot\text{s}$. Our findings will foster establishing graphene films as an efficient electrode material with surface treatments for improving pentacene OFETs and other types of organic electronic devices.

2. General Instructions

The grown MLG (multilayer graphene) film was detached from the growth substrate by using the catalyst-etching and scooping-up technique with aqueous iron chloride (FeCl_3) solution ($\sim 1 \text{ M}$) as the etchant and transferred onto a SiO_2 of 300-nm-thick layer on a heavily doped Si wafer. Then, Ni of 50-nm-thick was evaporated through a shadow mask, and we exposed oxygen plasma to the substrate at 200 mTorr and 50 W to remove the MLG film from the unprotected regions. Then, the Ni mask patterns were then etched away in FeCl_3 solution. The patterned graphene electrodes were coated with PMMA. Subsequently, the PMMA-coated graphene electrodes were detached from the SiO_2 substrate by etching the SiO_2 layer with buffered oxide etchant. Then, we transferred detached graphene electrodes to octadecyltrichlorosilane (OTS) treated substrate. After removing the PMMA by acetone, finally, the pentacene layer ($\sim 60 \text{ nm}$ thick) was deposited using a thermal evaporator under vacuum through a shadow mask onto the patterned MLG electrodes. The fabricated OFETs have 1000 μm width and channel lengths of 50, 100, 200, and 300 μm .

In this fabrication method, to improve the performances of such devices, we applied two research efforts. First, we identify the optimal electrode material for efficient charge injection to the active layers. Second, we optimize the grain size and organization of the active organic semiconductor channel for efficient charge transport. For example, to improve the grain size and molecular arrangement of the organic semiconductor channel, techniques such as high-temperature deposition of organic materials and dielectric-surface treatments with self-assembled monolayers of materials such as OTS have shown promising results.

Using such devices, we can calculate contact resistance (R_C). The R_C can be obtained by characterizing the device characteristics as a function of the channel length. In particular, the I_D values decreased as the channel length increased because the channel resistance increased with channel length. We obtained the R_C as the y-intercept of the extrapolation fit of R_{tot} versus the channel length data. The obtained R_C was determined from 0.56 to 1.89 $\text{M}\Omega$ for GR-pentacene OFETs. However, using the same characterization method, a larger R_C value (0.85 $\sim$ 3.34 $\text{M}\Omega$) was obtained for Au-pentacene OFETs at V_G from -10 to -50 V.

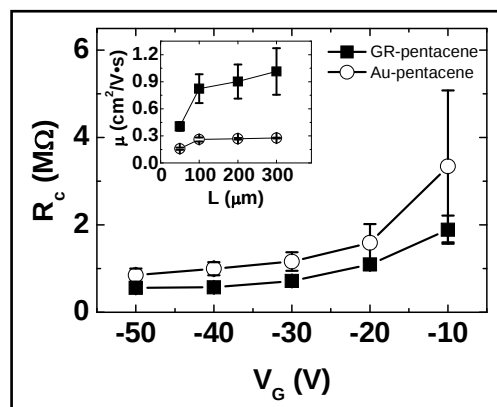


Fig. 1 R_C versus V_G of GR-pentacene and Au-pentacene OFETs. The error bars were obtained from measuring the standard deviation of ~ 7 devices for each data point. The inset compares the saturation mobility (μ_{sat}) of both types of OFETs with different channel lengths at a fixed V_G of -50 V.

As summarized in Figure 1, the R_C decreased with V_G becoming more negative for both GR-pentacene and Au-pentacene OFETs. Such trend of a decrease in R_C with V_G being more negative is correlated to the higher density of charge in the conducting channel in the large channel-length regime. More importantly, the saturation region mobility for GR-pentacene OFETs ($0.40 \sim 1.01 \text{ cm}^2/\text{V}\cdot\text{s}$) were observed to be higher than those for Au-pentacene OFETs ($0.16 \sim 0.28 \text{ cm}^2/\text{V}\cdot\text{s}$), as shown in the inset of Figure 1.

The R_C between the electrode and the organic channel is primarily caused by the charge injection barrier formed at the interface. Therefore we directly measured the effective barrier height (Φ) via temperature-variable I_D - V_D characterization.

The devices followed typical thermionic emission. So we estimated barrier height using this method was $\sim 0.24 \text{ eV}$ for GR-pentacene OFETs and was significantly lower than that ($\sim 0.45 \text{ eV}$) for Au-pentacene OFETs. Therefore, it can be concluded that GR-pentacene has better contact form than Au-pentacene due to the favorable orientation of the interfacial dipoles at the GR-pentacene interface, which resulted in lowering of the hole injection barrier. Accordingly, the GR-pentacene interface forms a small and favorably-oriented interface dipole layer, unlike the Au-pentacene interface which forms a significant interface dipole layer and results in a high carrier injection barrier. In addition, graphene is expected to induce strong interaction with pentacene and to form excellent interface contact because it has a similar molecular structure to organic materials.

We also measured the effect of interfacial dipoles by the Kelvin probe measurement. The work functions of Au and graphene films changed to 4.46 and 4.71 eV from 4.90 and 4.62 eV, respectively. Therefore, the Kelvin probe measurement results indicate that the interfacial dipole barriers of Au-pentacene and GR-pentacene are -0.44 and 0.09 eV , respectively. After using these data to align energy levels at the Au-pentacene and GR-pentacene interfaces the expected hole injection barrier height is obtained as $\sim 0.54 \text{ eV}$ for Au-pentacene and $\sim 0.29 \text{ eV}$ for GR-pentacene. Therefore, the interfacial dipole layer functions unfavorably (that is, it increases the charge injection barrier) for the Au-pentacene interface, whereas it functions favorably for the GR-pentacene interface.

Table I. Summary of Au-pentacene and GR-pentacene OFETs

	Work function	Ionization energy	Barrier height(Φ)	Mobility ($\text{cm}^2/\text{V}\cdot\text{s}$)
Au	5.10		0.45	$0.05 \sim 0.26$
Graphene	4.42 4.7-4.9		0.24	$0.41 \sim 1.01$
Pentacene		5.0		

3. Conclusions

In conclusion, we demonstrated that the performance of pentacene OFETs could be improved significantly by combining the use of graphene electrodes and dielectric surface treatments. We showed that as the pentacene film morphology was improved with various dielectric surface treatments, the electrical performance of the graphene-electrode pentacene OFETs was also improved accordingly in terms of low threshold voltage, low sub-threshold slope, large on/off ratio, and high field-effect mobility. And, through the detailed characterizations of the graphene and pentacene interface, key parameters such as the contact resistance and the charge injection barrier height were determined. Such results suggest that the use of various surface-treatment techniques brings synergetic improvements for high-performance OFETs with graphene electrodes and therefore may foster the wide application of graphene electrodes in other organic electronic devices.

Acknowledgements

This work was supported by the National Research Laboratory Program, a National Core Research Center grant, and the World Class University program from the Korean Ministry of Education, Science and Technology. This work was also partly supported by the core technology development program of the Research Institute for Solar and Sustainable Energies/GIST.

References

- [1] A. K. Geim, and K. S. Novoselov, Nature materials, **6**, (2007) 183
- [2] C. M. Aguirre et al., Nano Letter., **9**, (2009) 1457
- [3] S. Lee et al., Advanced Materials, **23**, (2011), 100
- [4] S. Lee et al., Applied Physics Lettet, (2011), in revision

Appendix

Secretariat of SSDM

c/o Inter Group Corp.

E-mail: ssdm_secretariat@intergroup.co.jp

URL: <http://www.ssdm.jp>