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Wettability Improvement by Silica Nanoparticle Addition in Solution-Processed TIPS-Pentacene Field-Effect Transistors

Saori Yamazaki[1,2], Takashi Hamada[2,*], Sakae Tokai[1], Masashi Yoshikawa[2], Takashi Nagase[2,3], Takashi Kobayashi[2,3], Yoshiki Michiwaki[4], Seiji Watase[5], Mitsuru Watanabe[5], Kimihiro Matsukawa[5], and Hiroyoshi Naito[2,3]

[1] Citizen Holdings CO., LTD., 840 Shimotomi, Tokorozawa, Saitama 359-8511, Japan
E-mail: yamazakisa@citizen.co.jp
[2] Department of Physics and Electronics, Osaka Prefecture University,
1-1 Gakuen-cho, Naka-ku, Sakai 599-8531, Japan E-mail: naito@pe.osakafu-u.ac.jp
[3] The Research Institute for Molecular Electronic Devises, Osaka Prefecture University,
1-1 Gakuen-cho, Naka-ku, Sakai 599-8531, Japan
[4] Fuso Chemical CO., LTD., 1-5 Osadano-cho, Fukuchiyama, Kyoto 620-0853, Japan
[5] Osaka Municipal Technical Research Institute, 1-6-50 Morinomiya, Joto-ku, Osaka 536-8533, Japan

1. Introduction

Organic field-effect transistors (OFETs) have received considerable attention due to their possible use in large-area, flexible, and low-cost electronics such as organic light-emitting displays, electronic papers, and radio frequency identification tags. For practical application, solution-processable OFETs using inexpensive fabrication processes including spin-coating and ink-jet printing have been recognized as the most promising candidate and soluble organic semiconductors with high charge mobilities have attracted increasing interest in recent years.

Field-effect mobility of soluble organic semiconductors is known to depend on the surface energy of gate dielectrics and several orders-of-magnitude increase of mobility by hydrophobic treatment of gate dielectrics has been reported. Lowering the surface energy of gate dielectrics is therefore essential for improving the performances of solution-processable OFETs. However, the fabrication of soluble organic semiconductors on hydrophobic substrates generally suffers from the dewetting of organic solution, which prevents the deposition of uniform semiconductor films using spin-coating process and the direct pattering of semiconductor layers using printing processes. To prevent the spreading of organic solution from channel regions in ink-jet printing process, the fabrication of bank structures has been employed [1].

In this paper, we report the improvement of the wettability of soluble organic semiconductor of 6,13-bis(triisopropyl-sililethynyl)pentacene (TIPS-pentacene) [1-3] on hydrophobic gate dielectrics by the addition of silica nanoparticles (SNPs). The effects of SNPs addition are investigated through the fabrication of OFETs on hydrphoic polymer gate dielectrics of poly(methyl silsesquioxane)s (PMSQs) [4-6] using spin-coating process. In addition, the direct pattering of TIPS-pentacene using ink-jet printing process is demonstrated.

*Present address: The Institute of Industrial Science, the University of Tokyo, 4-6-1 Komaba, Meguro-Ku, Tokyo 153-8505, Japan

2. Experiments

The OFETs with PMSQ gate dielectrics were fabricated onto glass substrates with an indium titanium oxide (ITO) layer, which was used as a gate electrode. The PMSQ dielectric layers were prepared by spin-coating, followed by curing at 200 °C for 1 h in ambient air through cross-linking polycondensation, which allows the subsequent deposition of soluble organic semiconductors. The SNPs used in this study are sol-gel synthesized colloidal particles having the organic surfactants of phenyl groups. The average diameter of SNPs is about 15 nm. The toluene solutions of organic semiconductors were prepared by adding SNP solution with different concentrations to the 2 wt % solution of TIPS-pentacene. The organic semiconductor layers of top-contact and bottom-contact devices were deposited onto the PMSQ dielectrics by spin-coating and ink-jet printing, respectively. In the ink-jet printing process, the organic solvent having higher boiling point of 1,3,5-trimetylbenzene (TMB) (165 °C) was used to enhance the crystallinity of TIPS-pentacene films. The interdigitated source-drain Au electrodes of top-contact and bottom-contact devices were fabricated by vacuum evaporation using a shadow mask and photolithography, respectively. The channel length and width were 200 μm and 48 mm, respectively. For comparison, the top-contact devices with Si/SiO_2 substrates were also fabricated. The FET measurement of fabricated devices was performed in ambient air using Keithley 238 source meters.

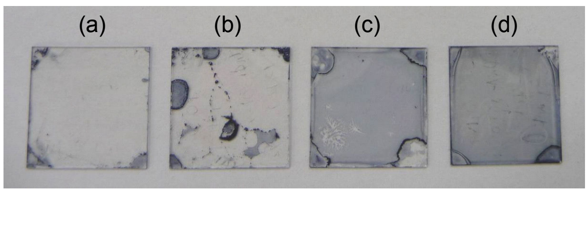

Fig. 1. Photographs of spin-coated films on PMSQ dielectrics from TIPS-pentacene solutions containing (a) 0, (b) 0.01, (c) 0.1, and (d) 1 wt% SNPs.

3. Results and discussion

Figure 1 shows the photographs of spin-coated films on PMSQ dielectrics from TIPS-pentacene solutions containing 0, 0.01, 0.1, and 1 wt% SNPs. When the concentration of SNPs is less than 0.01 wt%, the TIPS-pentacene solution can not be deposited on hydrophobic PMSQ surfaces because of dewetting.

Figures 2(a) and 2(b) show the transfer characteristics of top-contact OFETs fabricated from the TIPS-pentacene solution for the various concentrations of SNPs on SiO_2 and PMSQ gate dielectrics, respectively. We measure the transfer characteristics ofeach device with respect to the gate voltage V_G sweep from +20 to -40 V and then from +40 to +20 V to investigate hysteresis characteristics. The mobility enhancement observed in the devices on PMSQ dielectrics is attributed to the improvement of structural ordering of TIPS-pentacene molecules on hydrophobic low-energy surface. The suppression of the hysteresis in the devices on PMSQ dielectrics is likely to be due to the low OH density of PMSQ surfaces.

It also can be seen that the amount of ΔV_{th} in the devices fabricated on SiO_2 dielectrics decreases with concentration of the SNPs, indicating that the effective density of OH trapping sites at SiO_2 surfaces is reduced by the SNP addition. These results suggest that SNPs sink faster than TIPS-pentacene molecules to dielectric surfaces due to the difference in specific gravity, and the SNPs stay at dielectric surfaces act as anchors of TIPS-pentacene by an interaction between the phenyl groups of the SNPs and TIPS-pentacene molecules. Hence, the TIPS-pentacene : SNP solution can be spin-coated onto hydrophobic surfaces.

We find that the improvement of the wettablity by the SNP addition is also effective in the fabrication of OFETs by ink-jet printing. Figures 3(a) and 3(b) show the photographs of ink-jet printed square patterns on PMSQ dielectrics from TIPS-pentacene solutions containing 0 and 0.1 wt% SNPs, respectively. It can be seen that ink-jet printing using the TIPS-pentacene solution with SNPs forms well-defined patterns although ink-jet printed patterns using the solution without SNPs significantly suffer from dewetting. The polarized optical microscope image and the transfer characteristics of the fabricated bottom-contact OFET devices are shown in Figs. 3(c) and 3(d), respectively. The crystallinity of TIPS pentacene film is significantly improved by ink-jet printing using the high boiling solvent of TMB. The ink-jet printed OFET exhibits the high mobility of 2.57×10^{-2} cm^2/Vs and the high on/off ratio over 10^4.

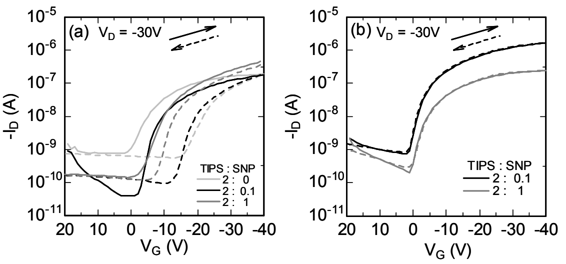

Fig. 2. Electrical characteristics of TIPS-pentacene : SNP FETs. Transfer characteristics of the devices fabricated on (a) SiO_2 and (b) PMSQ gate dielectrics with different SNP concentrations.

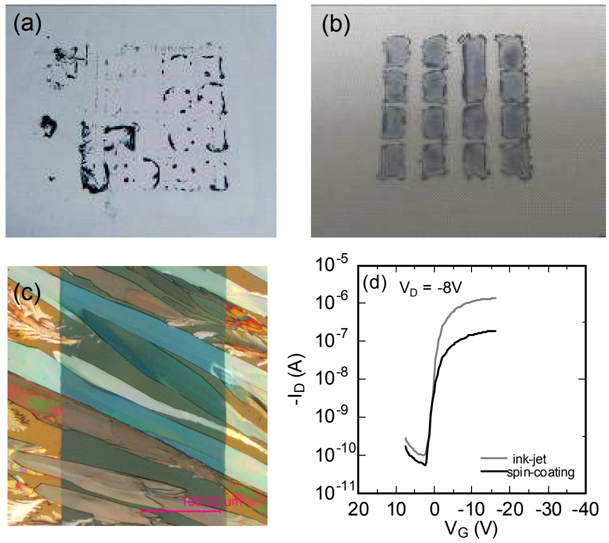

Fig. 3. Photographs of ink-jet printed square patterns on PMSQ dielectrics using TIPS-pentacene solutions containing (a) 0 wt% and (b) 0.1 wt% SNPs. (c) Polarized optical microscope images and (d) transfer characteristics of bottom-contact OFETs with 0.1 wt% SNPs fabricated on PMSQ dielectric by ink-jet printing. The thickness of PMSQ dielectric was about 150 nm.

4. Conclusions

We have found that the wettability of the soluble organic semiconductors of TIPS-pentacene on hydrophobic PMSQ gate dielectrics is significantly improved by the small amount of the SNP addition. It has been also found that the application of this approach to ink-jet printing process allows the direct patterning of soluble organic semiconductors on polymer gate dielectrics. The observed improvement of the wettability by the SNP addition would be attributed to the anchoring of TIPS-pentacene molecules to phenyl-modified SNPs during coating and printing process.

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