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The Effect of Interfacial Roughness on the Electrical Properties of Organic Thin Film Transistors with Anisotropic Dielectric Layer

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We investigated interfacial roughness effect of pentacene-based organic thin film transistors on an anisotropic insulator fabricated by obliquely evaporated silicon dioxide. It was observed that the obliquely evaporated gate dielectric layer affected molecular ordering of evaporated pentacene molecules. As the evaporation angle of SiO_2 increased, the anisotropic interaction at the dielectric interface and the ordering of organic semiconductor molecules increased. We obtained the anisotropic ratio of the field-effect mobility for carriers transported parallel and perpendicular to the evaporation direction was 2.9.

Keywords: anisotropic interfacial effect; liquid crystal alignment; molecular ordering; obliquely evaporated SiO_2; organic thin film transistor; pentacene

INTRODUCTION

Organic thin film transistors (OTFTs) are attracting much attention in flexible electronic applications such as flexible display, smart card, and radio frequency identification tag. Recently, the field effect

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mobility of OTFTs approached or exceeded that of hydrogenated amorphous silicon TFTs [1,2]. In such a rapid development of OTFTs' performances, much improvement in interfacial properties had an important role. The degree of molecular ordering and crystalline orientation of the organic molecules was proven to be significant factors in determining the performance of OTFTs [3,4]. In order to enhance molecular ordering and crystalline orientation, there were several efforts to modify interfacial anisotropic properties between the pentacene molecules and the insulator surface. Considering that pentacene is a rigid rodlike molecule similar to the core structure of liquid crystal, it is expected that similar surface-induced order can be also imposed on pentacene [5] by surface treatments such as mechanical rubbing [6] or irradiation of polarized UV light [7] on organic insulators.

In this paper, we used an obliquely evaporated silicon dioxide layer as gate insulator in order to align pentacene molecules. As the evaporation angle of SiO_2increased, the anisotropic interaction at the dielectric interface and molecular ordering of evaporated pentacenes increased. However, in highly obliquely evaporated dielectric surface, it was observed that growth of the pentacene molecules was highly limited due to increased surface roughness. The molecular ordering effect and the grain size effect depending on the surface anisotropy and the surface roughness were discussed with the results of the field-induced mobility and the surface morphology.

EXPERIMENTAL

To investigate the anisotropic interfacial effects on the pentacene-based OTFTs, a bottom-gate device was fabricated as show in Figure 1(a). The anisotropic insulator interface was fabricated by oblique evaporation of silicon dioxide with an e-beam evaporator at angles of 0°, 10°, 20°, and 40° to normal on the thermally grown oxidized surface, as shown Figure 1(b). Then, the 700 Å pentacene film was deposited at a deposition rate of 0.5 Å/s. This film was grown by thermal sublimation at a pressure of 10^{-6} torr and the substrate was held at room temperature during pentacene evaporation. Source and drain contacts were formed by evaporation of a 400 Å gold layer at a deposition rate of 1 Å/s through a shadow mask. The channel width and length were 300 μm and 50 μm, respectively. The channels of the transistor were aligned parallel and perpendicular to evaporation direction in order to investigate anisotropic electrical properties. We denote the samples which direction of current flow were parallel and perpendicular to the direction of the evaporation of SiO_2 by 'sample A' and 'sample B', respectively, in this article. For comparison, conventional OTFT with an isotropic silicon

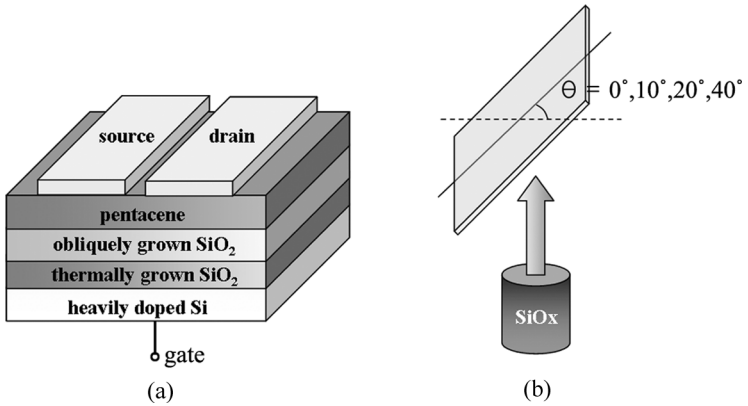

FIGURE 1 (a) Structure of pentacene-based TFT with an anisotropic inorganic dielectric layer and (b) schematic of oblique evaporation of SiO_2 by e-beam method.

dioxide surface was prepared by thermally grown oxidation with about 1300 Å thick on the heavily doped silicon wafer. The I-V characteristics of the OTFTs were obtained by using a semiconductor analyzer in ambient condition. In our analysis, the carrier mobility of the OTFTs were calculated in the saturation regime as follows,

$$I_D = -\frac{1}{2} C_i \mu \frac{W}{L} (V_{GS} - V_{TH})^2 \text{ for } V_{GS} < V_{TH} \text{ and } V_{DS} < (V_{GS} - V_{TH}), \quad (1)$$

$$C_i = \frac{\varepsilon_0 \cdot \varepsilon_i}{d_i} \quad (2)$$

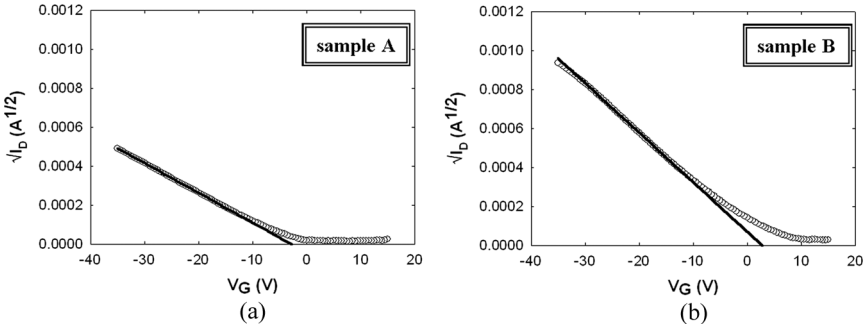

FIGURE 2 $\sqrt{I_D}$-V_{GS} characteristics of OTFTs where the oblique incidences of the SiO_2 evaporation at 20° are (a) perpendicular and (b) parallel to the channel direction.

Where C_i, V_{TH}, and μ were total insulator capacitance, threshold voltage, and carrier mobility, respectively. The surface morphologies of insulator and evaporated pentacene layer were measured by atomic forced microscope (AFM) in the non-contact mode.

RESULTS AND DISCUSSION

In order to induce interfacial anisotropy, we fabricated OTFTs with obliquely evaporated SiO_2 at 20° and measured electrical properties. Figure 2(a) shows a plot of the $\sqrt{I_D}$-V_{GS} characteristic for the 'sample A'. When gate voltage increased until -35 V, the drain current was measured at -0.24 μA. In case of 'sample B', the drain current was obtained three times higher than that of 'sample A' at the same gate-source voltage. The mobility of the 'sample B', calculated from Eq. (1), was higher than that of 'sample A', with the values of 5.13×10^{-3} cm^2/Vs and 1.49×10^{-2} cm^2/Vs, for 'sample B' and 'sample A', respectively. Such mobility difference depending on the evaporation direction was due to anisotropic charge transport induced by interfacial anisotropy. The threshold voltage had no difference in the evaporation direction, which meant that the electrical anisotropy was not caused by difference of defect structure at the interface between 'sample A' and 'sample B'.

In organic semiconducting material, charges transport by $\pi-\pi$ interactions between stacked molecules through the hopping mechanism. During the hopping process, charges propagate along the stacking axis of molecules through their overlapping π-obitals [5]. Accordingly, mobility difference shows that 'sample A' has more π-orbital overlaps than 'sample B', which means that the anisotropic interface affected molecular ordering of pentacene molecules.

However, when we evaporated SiO_2 at a smaller angle of 10°, there was no electrical anisotropy, as shown in Figure 3, which represented that molecular ordering of pentacene could not be achieved at too weak surface anisotropy condition. From the previous results, it was expected that the electrical anisotropy of the OTFT device on obliquely evaporated SiO_2 interface at an angle of 40° would be the most largest one among our test conditions, but both the mobility and the mobility anisotropy were lower than those of the OTFT on anisotropic SiO_2 surface fabricated by 20° oblique evaporation.

Such phenomenon could be explained by increase of the surface roughness accompanied by the increase of the surface anisotropy in our obliquely evaporated SiO_2 surface. Figure 4 is the AFM images of the topology of obliquely evaporated SiO_2. The RMS values of obliquely evaporated SiO_2 at 0°,10°, and 40° were 0.298 nm, 0.459 nm,

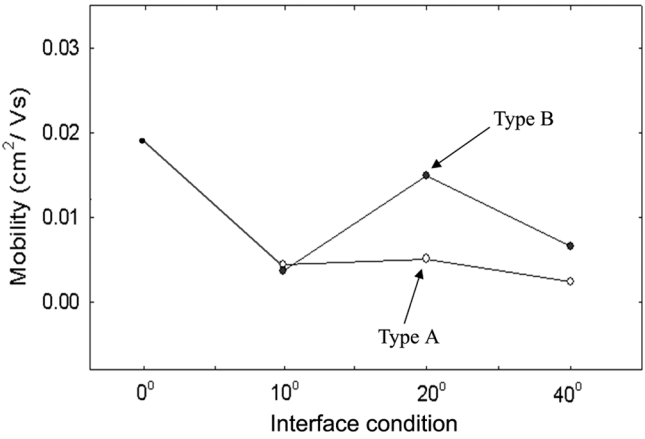

FIGURE 3 Variation of field effect mobilities of pentacene-based OFET devices fabricated by on obliquely evaporated SiO_2 at various angle.

and 0.667 nm, respectively. It showed that steep evaporation angle induced surface roughness. Rough surface of insulator resulted in reduction of diffusion length and energy barrier for nucleation, causing grain size to be decreased. The AFM images of pentacene surface on thermally grown SiO_2 (RMS roughness = 0.1 nm) and obliquely evaporated SiO_2 are shown in Figure 5. We observed the typical herringbone-like growth of pentacene crystals, in the pentacene surface evaporated on the thermally grown SiO_2. But, on obliquely evaporated SiO_2, small grain size was observed. In OTFTs with small grain sizes, the mobility significantly decreased because the grain boundaries

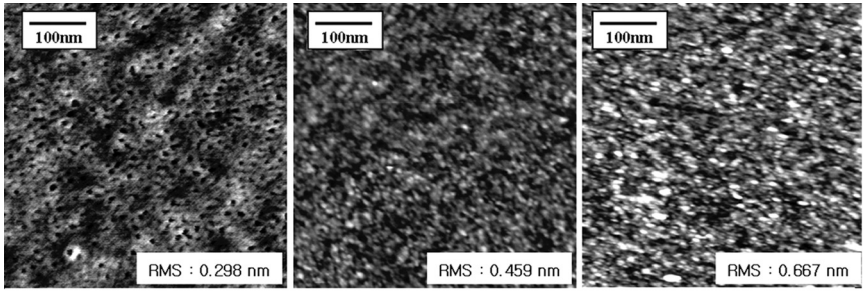

FIGURE 4 AFM images of obliquely evaporated SiO_2 surfaces fabricated by evaporation angle, Θ, of (a) 0°, (b) 10°, and (c) 40°.

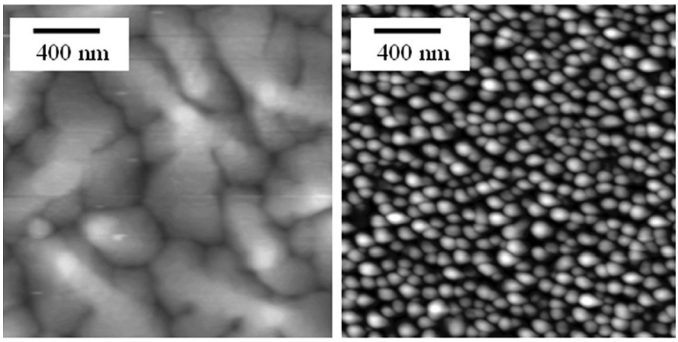

FIGURE 5 AFM images of pentacene surface on (a) thermally grown SiO_2 and (b) obliquely evaporated $SiO_2(\Theta = 20^\circ)$.

acted as trap sites in charge transport, limiting the carrier mobility [3, 8]. Accordingly, though OTFT fabricated by obliquely evaporated SiO_2 at steep angle had high anisotropic surface interaction during pentacene growth, the field-effect mobility was decreased by the grain boundary effect. Notice that the surface roughness of SiO_2 evaporated at an angle of 20° was higher than that of SiO_2 evaporated at an angle of 10° but both the mobility and the mobility anisotropy of the OTFT device on the former rougher interface was larger than that on the later one. Therefore, to obtain high field effect mobility and electrical anisotropy, it is required to optimize evaporate condition such as evaporate angle and deposition rate.

CONCLUSIONS

We studied the enhancement of molecular ordering and crystal orientation on the obliquely evaporated SiO_2 as gate insulator. The sample which was evaporated at an angle of 20° had the highest mobility than other samples and the mobility anisotropic ratio between perpendicular and parallel evaporation direction was 2.9. As evaporation angle became steeper, grain boundary effect induced by surface roughness was also more increased. So, the carrier mobility and electric anisotropy decreased, again. Therefore, to enhance field-effect mobility by molecular ordering on an anisotropic interface, anisotropic surface should be prepared without increasing surface roughness, which resulted in the existence of optimum evaporation angle range in our anisotropic surface fabricated by oblique SiO_2 evaporation.

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