Surface Modification Effects on the FET Property of Pentacene Thin Film

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1. Introduction
Organic molecular solids have been intensively investigated due to their versatility. Especially, the field-effect transistor (FET) structure has been widely utilized to evaluate electrical properties (charge carrier mobility, charge carrier density, etc.) of various organic materials [1]. However, since charge carrier mobilities of organic molecular devices tended to be compared with those of silicon devices, these materials have not been regarded as a promising candidate for electronic applications. Recently, relatively high values of the charge carrier mobility (0.5-1.5 cm²/V·s) for a pentacene thin film device were reported [2-4]. Organic thin films have been reevaluated as practical materials for electronic applications since then. Nowadays, with rapid spread of mobile electronic devices, thin, lightweight and flexible materials are earnestly requested in the electronic industry. Organic materials would sufficiently fulfill these requests and be suitable for such mobile electronic devices.

For improving the performance of pentacene FETs, it is necessary to find out the factor controlling the carrier transport property of devices. Differing from the single-crystal FET, FETs of thermally evaporated pentacene thin film contain several crystalline domains between drain-source electrodes. Charge carriers must hop from one domain to another between the electrodes. Namely, the charge carrier mobility is determined by how many boundaries, which carrier must hop, exist between the electrodes. Therefore, the size of crystalline domains strongly affected the carrier transport property, as previously reported for pentacene FET by J. H. Schönh et al. [5]. Moreover, the orientation of the direction of the pentacene crystal axes to the substrate plane would be also important. In this report, we will notice the relation between the charge transport property and the crystal growth of pentacene at different substrate temperature (T_sub) during evaporation. We will also refer to the effect of the surface modification on the crystal growth of pentacene.

2. Experimental
The pentacene FET device was fabricated as following procedure. Heavily doped Si with thermally grown SiO₂ (200nm) was employed as a substrate and a gate electrode. Substrates were carefully UV-ozone cleaned before the fabrication of FET devices. Capacitance of the SiO₂ layer was calculated by the value from a reference [6]. Pentacene was train-sublimated 5 times for purification. A pentacene thin film (50 nm) was deposited onto the substrate, and gold drain-source electrodes were deposited on the pentacene thin film surface by using thermal evaporation method at a pressure of ca. 10⁻³ Torr (deposition rate = 0.1-0.2 nm/s). The channel length (L) and width (W) of the drain-source electrode set were 100 µm and 4 mm, respectively. Electrical measurements were carried out by using a source meter (Keithley 2420) and a voltage source (Kepco ABC125-1DM).

3. Results and discussions
Fig.1 shows XRD spectra of deposited pentacene films at different T_sub. The XRD spectrum of the pentacene film deposited at T_sub = 343 K is definitely composed of two crystalline phases. The peaks at 6.02° and 5.67° correspond to the thin film phase (vertical periodicity 15.5 Å) and the bulk phase (vertical periodicity 14.5 Å), respectively. In the spectra of thin films deposited at T_sub = 318 K and R.T., the only peak attributed to the thin film phase was observed. On the other hand, the XRD spectrum of the film deposited at T_sub = 87 K has no peaks, indicating that this film is completely composed of amorphous phase. The FET mobility of each film deposited at different T_sub is shown.

Fig.1. XRD spectra of pentacene thin films deposited at different T_sub
in Table I. Obviously, FET mobilities are decreased with increase of $T_{\text{sub}}$ (the content of bulk phase is also increased) except the mobility of the film of $T_{\text{sub}} = 87$ K. The film deposited at $T_{\text{sub}} = 87$ K did not work as FET. AFM images show that the average domain size in each film is increased with increase of $T_{\text{sub}}$. It has generally known that larger crystal domain gives higher mobility [5]. However, mobilities decreased by the increase of the average domain size of pentacene crystal in this study. We have considered that this is due to the poor orientation of pentacene crystal axes in the film. In order to obtain higher mobility, both the average domain size and the wettability should be improved.

We have investigated the modification of the SiO$_2$ surface to improve the wettability to pentacene. Polydimethylmethacrylate (PMMA) and n-Octadecyltri-chlorosilane (OTS) were employed as coating agent of SiO$_2$. Fig.3 shows XRD spectra of pentacene films deposited onto an OTS-treated or PMMA-coated substrate. In spite of the deposition at $T_{\text{sub}} = 343$ K, the peak attributed to the bulk phase is not observed in the XRD spectrum of the film deposited on the PMMA coated substrate. The AFM image of this film (Fig.4 (b)) shows the dendric growth of large crystal domain. As a result, mobility improvement (0.28 cm$^2$/V-s shown in Table II) could be obtained in this device. On the other hand, the film deposited on the OTS-treated one still contains bulky grains in Fig.4 (a). The obtained mobility (0.08 cm$^2$/V-s) of this film was much lower than that of the deposited film on PMMA coated substrate.

Consequently, we have found that both the domain size and the orientation of crystal axes strongly affect FET mobilities and that the wettability of the substrate to pentacene is dominant to these two factors. In this study, we have shown that PMMA coating of SiO$_2$ substrate gives high improvement of wettability of substrates to pentacene. This indicates that insulator layer design is fairly important to obtain a high perfomance of organic FET.

References