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Structure and Performance of Thin-film Transistors of Pentacene Formed by Dispersion of Crystals

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1. Introduction

Among the organic semiconductor transistors, pentacene is often studied because of its large carrier mobility and on/off ratio. Due to the poor solubility of pentacene in common organic solvents, most of the studies are performed using vacuum deposited films. Solution processing is a promising method for low-cost fabrication of thin films for large area electronics. There have been several reports on solution processing by precursors [1)] and substituted pentacene [2)].

We have reported a different approach to solution processing of thin films using the solutions of unsubstituted pentacene at elevated temperatures [3)]. The solution-processed thin-film transistors exhibit large carrier mobility and the films are assumed to be polycrystalline, though the grain boundaries are not clearly identified.

Recently, we have demonstrated a second approach to forming thin films of pentacene by transferring pre-grown crystals using a liquid process [4)]. This dispersion process produces single or poly-crystalline thin films with distinct particulate grains. In order to study the effect of grain boundary on the transport properties, several kinds of dispersion-processed films are fabricated by changing the crystal size. This fabrication process is unique since it is a "built-in system" in contrast to the "build-up system" of conventional thin film process.

2. Experimental details

Formation of crystals and dispersion

Crystalline particles of pentacene are grown in liquid phase from the solution. Crystals are prepared by cooling from a hot pentacene solution of 1,2,4-trichlorobenzene (1 g/l) at 200℃, and the crystal size can be varied by adjusting the cooling rate of the solution. Large rectangular or prismatic platelet crystals of size up to mm in size are grown by slow cooling (1℃/min) whereas small needle plate crystals with a width of a few microns are grown by rapid cooling (10℃/min). Small crystals of pentacene are prepared by dropping the hot solution into isopropanol, a poor solvent, which solidifies them. From these preparations, several kinds of crystals with sizes ranging from sub-micron to 50 microns are grown. After the growth of particulate crystals, pentacene crystals are concentrated by decantation of the mixture and are diluted with a dispersion media of isopropanol to form inks having a solid concentration of 1 wt%.

Thin film and thin-film transistor

Thin films are prepared by drop-casting the ink on the substrate and vaporizing the dispersion media at room temperature under a nitrogen atmosphere.Film structures are studied by polarized microscopy, scanning electron microscopy, wide-angle, and grazing incidence X-ray diffractions.

Thin-film transistors are fabricated by forming the films on SiO2/Si substrates with patterned Ti under-layered with Au as source and drain electrodes. The SiO2 surface is treated with hexamethylenedisilazane. The carrier mobility in a saturation regime is evaluated from the transfer characteristics.

3. Results and discussion

In contrast to the process of unsubstituted pentacene using a hot solution, this dispersion process has several process merits since it can be performed at ambient conditions using relatively stable inks of dispersion in air. The grown crystals are thin platelet and single domain crystals as observed by polarized microscopy. Thin films are formed with assembled platelet shaped crystals, and most of which are aligned with the plate plane facing the substrate surface. This is confirmed by optical and electron microscopy as shown in Figure 1 (a)- (d) . Molecular orientation of the films is confirmed by measuring the wide-angle X-ray diffraction pattern, which shows that the pentacene molecules are oriented regularly in the films with long molecular axis perpendicular to the substrate plane, which is the same with thin films by conventional process.

The carrier mobility is plotted with the threshold voltage as shown in Figure 2. Single domain thin films assembled with large crystal grain sizes exceeding the channel length exhibit an average carrier mobility of 1cm^2/Vs, which is comparable to single crystal transistors [5)]. Surprisingly, transistors of assembled small crystals with diameter less than the channel length performed well. The carrier mobility of the transistor deceases with the decreasing the crystal size, thus a narrower distribution of the carrier mobility and threshold voltage are observed. It is assumed that the defects such as grain boundaries and vacancies in the films are uniformly distributed.

The influence of grain boundaries can be identified from the relation between the channel length and carrier mobility. The carrier mobility slightly increases of with decreasing channel length in films with small crystals, while is independent of the channel length in films assembled with large and medium crystals. We believe that the effect of grain boundaries is less in short channel transistors, and that the electrode contact resistance of the transistors is relatively small.

In order to clarify this, the electrode contact resistance is studied by the gradual transfer line method in a linear regime [6]. The assumed contact resistance up to 100 kohms is very small compared to the channel resistance of several Mohms.

The transport barrier between grain boundaries of the films is studied from the temperature dependence and determined to be below 100 meV. A plausible explanation for the small barrier heights and contact resistances is that the crystals form large contact planes with a smooth surface [7].

Further studies on the grain structure in the films and the quality of the assembled crystals applying other small molecules are in progress.

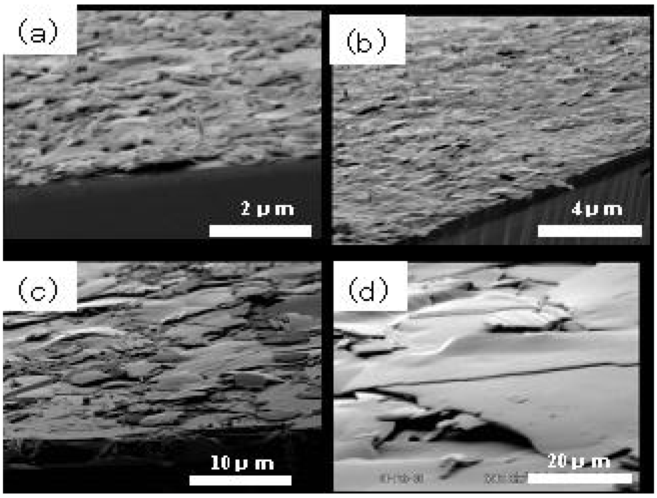

Fig. 1 Scanning electron micrographs of thin films formed by dispersion coating with changing crystal size of 0.3 (a), 0.5 (b), (c), and 50 μ m (d).

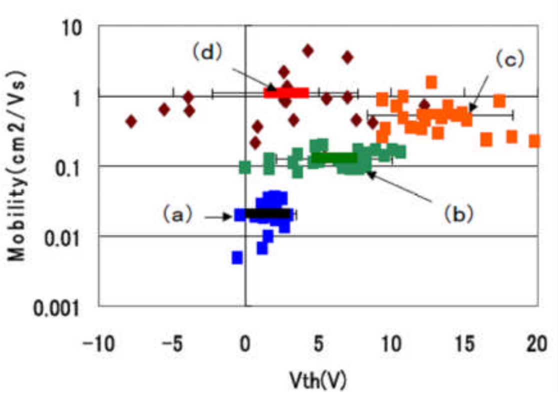

Figure 2 Plots of carrier mobility and threshold voltage (Vth), transistors are formed with film (a) to (d).

4. Conclusions

Thin films of pentacene have been fabricated by transferring a dispersion of pre-grown crystals in liquid media. Transistors are formed by changing the crystal size and show a good performance. The interfacial properties including both the contact resistance to the electrodes and the transport barrier height are found to be small.

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