Pressure Control Organic Vapor Deposition Methods for Fabricating Organic Thin-Film Transistors

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In this letter, we report on the development progress of a pressure control organic vapor deposition (PCOVD) technology used to design and build a large area deposition system. We also investigate the growth characteristics of a pentacene thin film by PCOVD. Using the PCOVD method, the mobility and on/off current ratio of an organic thin-film transistor (OTFT) on a plastic substrate are 0.1 cm²/Vs and 10⁶, respectively. The developed OTFT can be applied to a flexible display on a plastic substrate.

Keywords: PCOVD, pentacene, OTFT, plastic substrate.

I. Introduction

In recent years, there has been a growing interest in the field of organic thin-film transistors (OTFTs) on plastic for use in organic electronics, such as smart cards, low-cost identification (ID) tags, and flexible displays [1]. OTFT offers such advantages as mechanical flexibility and weight reduction compared to traditional field-effect transistors [2]. Pentacene consists of five linear benzene rings and has demonstrated the highest electron and hole mobility of organic small molecules. The material exhibits a strong tendency to form highly ordered films, which depend on the growth conditions and substrate. OTFTs have been fabricated using solution precipitation [3]-[5], organic molecular beam deposition [6], vacuum thermal evaporation (VTE) [7], [8], and organic vapor phase deposition [9], all having comparable performances.

Pressure control organic vapor deposition (PCOVD) is a new technology for the large area deposition of small molecular organic semiconductor devices and can be applied to the OLED deposition [10]. In PCOVD, organic source materials are sublimed into a stream of hot inert carrier gas. The molecules undergo a convective transport through a coldwall chamber to a cooled substrate on which they condense to form a film. The biggest difference between VTE and PCOVD is as follows. In the case of PCOVD, the properties of organic thin film can be adjusted by changing the deposition pressure.

In this letter, we study the growth characteristics of pentacene thin film using PCOVD, which is an innovative deposition technique.

II. PCOVD system

Figure 1 shows a schematic diagram of a PCOVD system employing a cold-wall chamber and the showerhead concept of a gas distributor capable of depositing highly uniform layers [11]. Organic source materials are heated in physically separated source containers and transported by a hot inert carrier gas into the cold-wall chamber. Individual valves switch the source and dilution flow to enable a rapid on/off control of the respective deposition, which offers high precision control of the layer interfaces as well as a minimization of material waste. The organic molecules are introduced uniformly across the entire substrate surface through a heated showerhead injector and condensed onto the substrate to form the desired film.

The deposition rates and layer composition, such as doping, are adjusted by means of the mass flow rate of the carrier and the dilution gas at a constant source temperature, using individual electronic mass flow controllers for each source.

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Fig. 1. Schematic diagram of PCOVD system.



Fig. 2. SEMs of pentacene thin films deposited by PCOVD on polymer gate dielectric at (a) 20 min, (b) 30 min, (c) 60 min, and (d) 120 min (scale bar is 3 μm in size).

This offers a high degree of precision and reproducibility combined with flexibility in multilayer deposition sequences as well as an accurate composition of multicomponent layers.

Gas phase transport allows the remote placement of multiple organic sources with respect to the deposition chamber, thus preventing cross contamination between source materials.

Consequently, PCOVD offers the potential for low maintenance cycles, high material yield, high reproducibility, well-defined doping with multiple dopants, and high throughput, which are key factors for industrial mass production at a low cost of ownership.

III. Results

Pentacene, which was obtained from Aldrich Chemical Co., was used without additional purification. The source material was maintained at 300°C during pentacene thin film deposition. The pressure in the deposition chamber was varied from 1 torr to 30 torr, and deposition temperature was varied from 0°C to 80°C. Compared with thermal evaporation, this method minimizes material waste and achieves a uniform deposition of pentacene thin film on a five-inch wafer.

Figure 2 shows scanning electron micrographs (SEM) of a pentacene thin film on a polymer gate dielectric at various deposition times. The deposition pressure was 1 torr. The pentacene thin films formed a continuous thin film.



Fig. 3. SEMs of pentacene thin films deposited by PCOVD on polymer gate dielectric at (a) 1 torr, (b) 5 torr, (c) 10 torr, and (d) 30 torr (scale bar is 3 μm in size).



Fig. 4. SEMs of pentacene thin films deposited by PCOVD on polymer gate dielectric at (a) 0°C, (b) 40°C, (c) 60°C, and (d) 80°C (scale bar is 3 μm in size).

Additionally, the grain size with increasing deposition time was observed clearly.

Figure 3 shows the morphology of a pentacene thin film on a polymer gate dielectric at various deposition pressures. The deposition time was 30 minutes. Below 10 torr, the pentacene thin film formed a continuous thin film. However, above 10 torr, the pentacene thin film formed needle-shaped islands. In X-ray diffraction analysis, the peaks arise from the well-known bulk and thin film phases of pentacene. The thin film phase has an interplanar spacing of 15.4 Å, while the bulk phase has an interplanar spacing of 14.5 Å. Pentacene thin film phase at 1 torr. However, above 10 torr, the increase of deposition pressure leads to an amorphous phase.

Figure 4 shows the morphology of a pentacene thin film on a polymer gate dielectric at various deposition temperatures. The deposition time and pressure were 30 minutes and 1 torr. At 80°C, the pentacene thin film does not form a continuous thin film. The grain size of the pentacene thin film increases as the deposition temperature increases.

The pentacene TFTs were fabricated into a bottom-contact structure. The deposition time, pressure, and temperature of the pentacene thin film were respectively 30 minutes, 1 torr, and 0°C. Cr/Al/Cr and Au/Ti were used for the gate and source/drain metals, respectively, and were deposited by sputtering and e-beam evaporation methods, respectively. For an OTFT with an organic gate insulator, a polymer gate dielectric was deposited onto the patterned gate metal by spin



Fig. 5. OMs of pentacene thin film by deposited (a) PCOVD and (b) VTE after patterning of active layer.



Fig. 6. (a) Output characteristics of pentacene TFT after patterning of active layer and (b) image of OTFT array on plastic substrate.

coating from a solution. The gates were planarized by the spincoated polymer gate dielectric at a thickness of about 300 nm.

After pentacene deposition, CVD-parylene and ALD-Al₂O₃ thin films were used as a passivation layer [12]. Parylene and ALD-Al₂O₃ thin films were very well suited to this process. The material was deposited onto the pentacene surface at room temperature and polymerized during the deposition process into a solvent-resistant, chemically-inert film. The passivation layer protected pentacene against any attack from the solvents used for the photolithographic process.

Figure 5 shows optical micrographs (OMs) of a pentacene thin film deposited by PCOVD and VTE after patterning of the active layer. In PCOVD, the pentacene thin film was not peeling after active patterning. In VTE, however, the pentacene thin film was peeling after active patterning. This shows that the adhesion of the pentacene thin film deposited by PCOVD is superior to the adhesion of the pentacene thin film deposited by VTE.

Figure 6(a) shows the output characteristics of a pentacene TFT after patterning of the active layer, and Fig. 6(b) shows an image of an OTFT array on a plastic substrate. We observed that the mobility did not change after patterning of the active layer.

IV. Conclusion

We grew a pentacene thin film using a new deposition technology, PCOVD. Deposition of a pentacene thin film using the PCOVD process is a promising deposition technique. A bottom contact OTFT with the pentacene thin film exhibited a field-effect mobility of $0.1 \text{ cm}^2/\text{Vs}$ and an on/off current ratio of about 10^6 .

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