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Feasible organic thin-film deposition architecture for large-area organic electronics by roller vacuum thermal evaporation

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Pentacene thin film deposition with high uniformity of 2.7% on flexible polyethylene terephthalate (PET) substrate is achievable over area of 300 mm × 500 mm using a simple roller vacuum thermal evaporation system with point sources. Thickness numerical simulations, agree well with measurements, indicate that the multi-dimensional movement of the cylindrical sample holder guarantees film uniformity over a larger area. High device performance with excellent uniformity and reproducibility is demonstrated by pentacene-based thin film transistor arrays on large PET substrates with average saturation mobilities of 0.42 cm²/V s. The system provides a feasible thin film deposition architecture for large-scale organic electronics. © 2011 American Institute of Physics. [doi:10.1063/1.3657411]

Organic semiconductors and devices have attracted a lot of research attention during the past decade because of their inherent advantages of low costs, lightweight, near-ambient fabrication temperatures, compatibility with flexible substrates, and potential for large-scale mass production. The successful application of the organic materials depends on capturing their low-cost potential through the innovative fabrication architecture for devices on inexpensive, large-area substrates. Many processes have been studied to achieve these potentials, including vacuum thermal evaporation (VTE), organic vapor phase deposition (OVPD), direct printing through use of contact with stamps, or via ink-jets and other solution-based methods. The solvents used in solution-based process can, and often do, smear previously applied layers, thereby limiting the freedom of device design. VTE is nevertheless a widely used “dry processing” in small molecule organic electronics manufacturing, particularly for multilayered electronic devices, because of the relative simplicity of the process and high quality of interface control. However, conventional VTE can be wasteful of materials and, especially difficult to maintain uniform film deposition on a large area. To avoid many of the shortcomings of conventional VTE, an OVPD source has been proposed and in-depth studied in recent years which has made significant progress in large area uniform deposition. However, this technique still leaves much margin considering the expenses, wafer size limitation and the compatibility with the electrode formation.

In this work, we propose a roller-VTE technique to deposit high uniform films over large-area while keeping the normal point source designs as in conventional VTE systems. This technique can deposit both organic and metal films or improve the uniformity in large deposition area using OVPD source or point source arrays. A pentacene film with average thickness of 85 ± 2 nm and pentacene-based organic thin film transistor (OTFT) arrays with average saturation mobility up to 0.88 ± 0.12 cm²/V s over an area of 300 mm × 500 mm have been demonstrated in our recent work. Here, numerical simulations on both effusion and deposition subsystems are presented to compare with the experimental results and to prove the availability of roller-VTE systems on even much larger uniform area which provides a feasible technique route for organic thin film deposition on large area in both industrial production and laboratory manufacture. High device performance with excellent uniformity and reproducibility using this system is also demonstrated by pentacene-based thin film transistor arrays on large flexible polyethylene terephthalate (PET) substrates.

Figure 1 shows a photograph and a schematic drawing of the roller-VTE system. At present design, the roller, a cylinder with 160 mm in outer diameter and 300 mm in length, can reciprocally travel along the horizontal axis (Z) at a horizontal speed $V_H$ from 1 to 15 mm/s and self-rotate simultaneously around the Z axis at angle speed $\omega$ from 0.3 to 2.7 rad/s during film deposition. The maximum horizontal displacement along the Z axis is 500 mm to avoid the edge effect. The length of the deposition window on the Z axis is about 126 mm, controlled by a shutter between the sources and the roller. The open crucibles for organic semiconductors and

![Figure 1](https://via.placeholder.com/150)
thermolecular evaporation boats for metal materials are situated at the bottom of the deposition chamber, as schematically shown in Fig. 1(b). Quartz oscillators are positioned over the cylinders to monitor the vapor flux distribution during real-time monitoring of the film growth rate.

The vapor flux distribution for the organic source was calibrated on a flexible PET sheet which was mounted onto the roller surface and held in place during vapor flux calibration. A static deposition rate ($R_S$) was defined as the values monitored by the quartz oscillator. The film thickness distribution on the whole roller surface which was in a multi-dimensional moving state during film deposition was calibrated by discrete film thicknesses on many $15 \times 15$ mm SiO$_2$ wafers which were homogeneously attached onto the roller surface. Film thicknesses were measured post-growth and using a surface profiler (Dektak-150, Veeco Co.) and an atomic force microscope (AFM) by tapping mode (Nanoscope III, Veeco Co.). Device performance using the roller-VTE system was characterized by top-contact bottom-gate pentacene-based OTFTs arrays on PET foils with indium tin oxide (ITO) gates. The OTFTs were fabricated using the following processes. First, polystyrene (PS) (Alfa Aesar, molecular weight = 100 000) was spin-coated onto the PET–ITO foils with final thickness of 450 ± 20 nm. Then, pentacene films with thicknesses of about 36 nm were deposited onto these PS-treated substrates at room temperature under $R_S = 0.28$ nm/s, $V_H = 10$ mm/s, and $\omega = 1.8$ rad/s. At last, 30-nm-thick Au or Cu source-drain electrodes were deposited onto the pentacene layer through a flexible metal shadow-mask by this roller-VTE system. Limited by the size of the spin-coating machine, the PET–ITO foil size was chosen to be $100 \times 100$ mm.

Apart from experimental characterizations, systematic numerical simulations for the roller-VTE system were carried out to compare with experimental results. For the effusion subsystem simulation, we used an empirical formula of $\cos^n(\theta)$, which was commonly used in physical vapor deposition process, to describe the open crucible effusion flux. For the deposition subsystem simulation, the film thickness on each grid point on the roller surface can be determined by the vapor flux distribution and the trajectory of the point. The former is described by the effusion subsystem, while the latter is determined by the roller geometry and the movement parameters. Three idealized conditions were assumed to simplify the thickness simulation. These were regarding evaporation source as point source, straight line trajectories for effusion molecules, and no desorption on the substrate surface.

A photograph of the beam spot of the open crucible and the fitting deposition rate distribution are shown in Figs. 2(a) and 2(b), respectively. The normalized vapor flux distribution versus effusion angle is shown in Fig. 2(c). The corresponding maximum static deposition rate was $R_S = 0.56$ nm/s. Note that the $\cos^n(\theta)$ empirical formula gave a good description of the data trends of the vapor flux distribution, although the normalized experimental data distributed loosely around the fitting curve which was caused by the accuracy limitation of the measurements.

For any point on the roller surface, the point coordinates at a given time can be determined by the roller geometry and movement parameters. For an instance, Fig. 3(a) shows the trajectory of the lowest center point P on the roller surface with the initial position of $(0, 80, 250)$ under $V_H = 10$ mm/s, and $\omega = 1.8$ rad/s during deposition time of 100 s. Using the preceding deposition rate distribution as indicated in Fig. 2(b) or the inset of Fig. 3(a), we got the real-time deposition rate on point P at any given time and then the corresponding film thickness (i.e., time integral of the deposition rate), shown as Fig. 3(b). Figure 3(c) shows both the experimental and simulation results of the film thicknesses on point P and the relative standard deviation (RSD) over $300 \times 500$ mm area. Note that the thickness simulation results are in good agreement with the experimental results in short vaporization time and
have a little deviation in long evaporation time. The deviation may be caused by pentacene consumption in the crucible which has not been considered in our present model. The RSDs in experiments are excellent and less than 2.7% in long evaporation time. Considering all the standard deviation of the measurements were less than 2.5 nm, the deviation of RSD 

\[ RSD \]

in experiments are excellent and less than 2.7% in long evaporation time. The deviation of the OTFT arrays. The saturation mobility is 0.42 ± 0.02 cm²/V s and the on/off ratio is close to 2 × 10⁵.

FIG. 4. (Color online) Pentacene-based OTFT arrays and device performance. (a) A photograph of the OTFT arrays. (b) Schematic cross-sectional structure of the OTFT. (c) Typical output curves of the OTFTs prepared on PET substrates by the roller-VTE system. (d) Superimposed transfer curves of the OTFT arrays. The saturation mobility is 0.42 ± 0.02 cm²/V s with the drain-source voltage V_DS and the gate-source voltage V_GS both at −60 V.

Figures 4(c) and 4(d) show typical output curves and superimposed transfer curves of tens of pentacene-based OTFTs with Cu electrodes which are randomly selected from the OTFT arrays, respectively. The average saturation mobility was 0.42 ± 0.02 cm²/V s with an RSD of 5%. We have carried out a lot of repeated experiments. The results show that this roller-VTE system achieves high uniformity device performance over area of 300 mm × 500 mm and has good reproducibility for electric characteristics.

In conclusion, a roller-VTE system has been developed and optimized for deposition of uniform pentacene and Cu thin films over 300 mm × 500 mm area. It is demonstrated that pentacene-based OTFTs on flexible PET substrate fabricated by the roller-VTE system have average saturation mobility of 0.42 ± 0.02 cm²/V s with the RSD of 5%. The thickness simulations, agree well with measurements, indicate that the multi-dimensional moving of the roller guarantees the uniformity of the film thickness and that the system can be easily extended for a larger area. The roller-VTE system provides a possible tool for depositing uniform organic and metal films on large flexible substrates and may be a promising candidate for industrial production.

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9In addition to the cos²(θ) empirical formula, we also studied the vapor flux formula of circular section tube proposed by Clausung (W. Steckelmaeher, *Vacuum* **16**, 59 (1966)) and a quartic polynomial P₄ (z, e) to fit the beam spot, respectively. The fitting results were similar to the cos²(θ) formula.