Supporting Information

Gas Blow Coating: a Deposition Technique to Control the Crystal Morphology in Thin Films of Organic Semiconductors

Jincheng Tong*, Amadou Doumbia, Adriana Alieva, Michael L. Turner* and Cinzia Casiraghi*

J. C. Tong, A. Doumbia, A. Alieva, Prof. M. L. Turner and Prof. C. Casiraghi
School of chemistry
University of Manchester
M13 9PL
United Kingdom
E-mail: Jincheng.tong@manchester.ac.uk, Michael.Turner@manchester.ac.uk, cinzia.casiraghi@manchester.ac.uk

Table S1 Comparation of state-of art works

S1 Experimental setup

S2 Characterization of the deposited films

S3 Characterization of the devices
Table S1 compare our results to state-of-art works reporting OFETs of TIPS-pentacene and C8-BTBT.

Table S1. Typical OFETs based on TIPS-pentacene and C8-BTBT thin films deposited by different coating methods in literature. Details on crystals morphology, dielectric used (Silicon Oxide-SiO$_2$, hexamethyldisilazane-HMDS, polytitanosiloxanes-PTS, benzocyclobutene-BCB, dodecyltrichlorosilane-DTS, 4,40-(hexafluoroisopropylidene)diphthalic anhydride-HAD, octadecyltrichlorosilane-OTS, Polyvinylpyrrolidone-PVP, poly(methyl methacrylate)-PMMA), and device characteristics.

| Semiconductors | Coating Methods | Crystals morphology | Dielectric | Average mobility ($cm^2V^{-1}S^{-1}$) | Maximum mobility ($cm^2V^{-1}S^{-1}$) | Operating Voltage (V) | Ref. |
|----------------|-----------------|---------------------|------------|----------------------------------|----------------------------------|-----------------------|------|
| TIPS-pentacene | Spin coating    | small crystals      | SiO$_2$    | 0.01                             | NA                               | 40                    | S1   |
|                | Small angle drop casting | big crystals      | SiO$_2$/HMDS | 0.3±0.08                         | 0.48                              | 40                    | S2   |
|                | Dip coating     | oriented crystals  | SiO$_2$    | NA                               | 0.2                               | 10                    | S3   |
|                | Inkjet printing | big crystals       | SiO$_2$    | 0.018                            | 0.12                              | 40                    | S4   |
|                | Double shot Inkjet printing | big single crystals | SiO$_2$ | 0.042 | NA | 40 | S5 |
|                | solution shearing | aligned single crystals | SiO$_2$/PTS | 1.49 | 4.6 | NA | S6 |
|                | Droplet pinning growth | aligned single crystals | SiO$_2$/BCB | 2.4±0.6 | 3.8 | NA | S7 |
|                | spray printing  | big single crystals | SiO$_2$    | NA                               | 0.4                               | 35                    | S8   |
|                | Gas blow coating | aligned single crystals | SiO$_2$ | 0.07±0.04 | 0.15 | 40 | our work |
| C8-BTBT        | Vacuum deposition | islands of crystals | SiO$_2$    | 0.30–0.74                        | NA                               | 60                    | S9   |
|                | Spin coating    | NA                  | SiO$_2$    | 0.46–1.8                         | NA                               | 60                    | S10  |
|                | small angle drop casting | big crystals | SiO$_2$/DTS | 3.5-5 | NA | NA | S11 |


| Blend                  | Method                                      | single crystals | Parylene C | SiO₂ | 0.26 | 1.6 | 40  | S12 |
|-----------------------|---------------------------------------------|-----------------|------------|------|------|-----|-----|-----|
| Bar-assisted          | Meniscus shearing                           | spherulitic     | SiO₂       | 0.26 | 1.6  | 40  | S13 |
| Off-center spin       | NA                                          | single crystals | SiO₂/OTS   | 6.9±2.6 | 11.2 | 40  | S15 |
| Solution-epitaxy      | spin coating                                | single crystals | SiO₂       | 4.8  | 13   | 20  | S16 |
| Doctor blade coating  | aligned single crystals                     | SiO₂/PTS       | 4.8        | 13   | 20   | 20  | S17 |
| polymer assisted      | solvent vapor annealing                      | one dimensional | SiO₂/PMMA  | 1.1±0.78 | 3.8  | 40  | S18 |
| Gas blow coating      | aligned single crystals                     | SiO₂           | 0.7±0.31   | 1.4  | 40   | 40  | our work |

**S1 Experimental setup**

![Figure S1. Picture of the gas blow coating system showing the various components.](image-url)
**Figure S1** shows the various components of the developed gas blow coating system. A N₂ cylinder is used to supply the gas. A breather check valve is applied to control the start or finish of the gas blow coating process. The coating system is based on a doctor blade coater (Kpaint applicator purchased from RK Printcoat Instruments), in which the speed can be controlled by the speed control panel. The gas knife is purchased from EXAIR and it is assembled into the coating panel for the deposition of thin films with both pressure and moving speed controlled. And in this set up, the distance (5 mm) and the angle (57°) of the air knife to the substrate are kept constant.

**S2 Characterization of the deposited films**

**S2.1. Gas blow coating of TIPS-Pentacene**

The gas blowing deposition parameters of TIPS-Pentacene in o-xylene have been studied in detail, using glass as substrate. The most important parameters are: the gas knife moving speed, the gas supply pressure and the solution concentration. In this study, only one parameter is changed, while the other parameters are kept constant.
Figure S2. (a-g) Pictures of the TIPS-Pentacene films deposited by using different knife speed on glass slides (length of 7.5 cm and width of 2.5 cm). (h-m) Optical pictures showing the different morphology of the films at different moving speeds, from 2.1 cm/s to 4.4 cm/s (the red arrow indicates the gas blow coating direction and the blue arrows show the disconnected part of the film).

We first study the effect of the moving speed for fixed pressure (0.3 bar) and concentration (8 mg/mL). Figure S2 shows optical pictures of the films deposited by changing the moving speed of the gas knife from 4.4 cm/s to 1.7 cm/s. Films deposited at speed between 4.4 cm/s and 3.8 cm/s are composed by all well aligned crystals, starting from the edge to the middle of the film, in the vertical direction to the gas flow coating direction (red arrow in Figure S2h). With the decrease of the speed, the length of the deposited films increases (Figure S2a-2g). When the speed further goes below 3.3 cm/s, the films show some disconnected crystals (blue arrows in Figure S2 k-m). This effect is due to the reduced amount of solution, as the film is not thick enough to supply sufficient solute for the crystal to growth into a whole film.
Figure S3. (a-d) Pictures of the TIPS-Pentacene films deposited on glass by using different gas supply pressure. (e-h) Optical pictures at higher magnification showing the different morphology of the films at different gas supply pressure (the red arrow indicates the gas blow coating direction and the blue arrow shows the disconnected part of the film).

We therefore fixed the speed at 3.5 cm/s. The effect of the gas supply pressure was studied using this speed and concentration of 8 mg/mL. Figure S3 shows that when the pressure increases, the films length increases. When the pressure is set as 0.1 bar, the wet film cannot spread well, resulting in a thick fish skin-like film. Relatively well aligned crystals appear as the pressure is increased to 0.3 bar, but further increase of the pressure makes the crystals of the film to be disconnected (Figure S3g and 3h). This indicates that high pressure forces the solution to spread further on the substrate, thus decreasing the thickness of the wet film, which in turn will create many nucleation sites for small crystals to grow.
Figure S4. Microscope pictures of the films deposited by using solution with 8 mg/mL TIPS-Pentacene on (a) glass and (b) silicon wafer with a moving speed of 3.5 cm/s and gas pressure of 0.3 bar (the red arrows show the gas blow coating direction and the blue arrows show the disruption appear in the middle of the film).

Figure S5. Microscope pictures of the films deposited by using solution with 20 mg/mL TIPS-Pentacene on (a) glass and (b) silicon wafer with moving speed of 0.6 cm/s and gas pressure of 1.0 bar; (c) AFM image of the film deposited on silicon.

Figure S4 shows that a disruption line appears in the middle of the obtained film, as the result of the crystals growing from both sides of the contacts (see main text). By using a high concentration (20 mg/mL), a relatively low moving speed (0.6 cm/s), and a relatively high gas pressure (1.0 bar), uniform films of TIPS-Pentacene on both glass (Figure S5a) and silicon (Figure S5b) are obtained. AFM (Figure S5c) shows the film to consist of small crystals, further confirming the ability of the gas pressure in controlling the thickness of the wet film and the fast evaporation of the solvent, thus creating large quantities of nuclei in the whole film.
Figure S6. (a-d) Pictures of the TIPS-Pentacene films deposited by gas blow coating using different concentration of TIPS-Pentacene on glass with moving speed of 3.5 cm/s and gas supply pressure of 0.3 bar. (e-h) Optical pictures taken with the microscope showing the different morphology of the films at different concentration (the red arrow indicates the gas blow coating direction and the blue arrow show the disconnected part of the film).

We now change the concentration. Figure S6a-d show that with the decreasing of the concentration from 8 mg/mL to 2 mg/mL, the total film length and width are similar, indicating that the wet film obtained by gas blow coating are not affect much by changing the concentration. Correspondingly, Figure S6e-g show that when the concentration is higher than 4 mg/mL, thin films with aligned crystals on glass are obtained. However, at concentration of 2 mg/mL, the crystals are not well aligned and many disruption lines appear in the middle of the film as a result of the insufficient solute to support continuous growth of the aligned crystals on the whole substrate. Figure S6h shows a represented view of two disruption lines in the middle of the film. To see more details and for further characterization of the crystals, the films were deposited with the same parameters on silicon wafer, Figure S7.
Figure S7. Optical microscopy images of the films of (a) 2 mg/mL, (b) 4 mg/mL, (c) 6 mg/mL and (d) 8 mg/mL of TIPS-Pentacene on silicon wafer by gas blow coating with moving speed of 3.5 cm/s and gas supply pressure of 0.3 bar (the red arrow indicates the gas blow coating direction and the red circles show the isolated nuclei, dendritic branches or defects in the middle of the films).
**Figure S8.** Thickness distribution of the crystals deposited at 2, 4, 6, 8 mg/mL TIPS-Pentacene solution by gas blow coating.

**Figure S7** shows that when the concentration is not high enough (2-6 mg/mL), random nucleation sites or big branches or disconnected crystals appear in the films. But concentration of 8 mg/mL allows achieving a thin film with long and well aligned crystals. Accordingly, the average thickness of the TIPS-Pentacene films obtained using different concentrations are: 43.6 ± 9.1 nm (at 2 mg/mL), 66.0 ± 6.9 nm (at 4 mg/mL), 98.6 ± 19.5 nm (at 6 mg/mL), and 121.8 ± 17.5 nm (at 8 mg/mL). Thus, the thickness increases linearly with the concentration, as one should expect (**Figure S8**).
S2.2. Gas blow coating of C8-BTBT films

Figure S9. Optical microscope images of C8-BTBT film deposited from (a) 2 mg/mL, (b) 6 mg/mL and (c) 8 mg/mL solution by gas blow coating (insert are the corresponding AFM images). (d) AFM image of a selected area from panel b (the red arrows show the defects in the layers) and (e) two profile of the selected lines in d showing the deposition of a single layer of C8-BTBT.

Optimal deposition of C8-BTBT is found to be obtained with similar parameters used for TIPS-Pentacene (i.e. knife speed of 3.75 cm/s and gas pressure of 0.18 bar), probably due to the similar nature of the molecules and use of the same solvent. Here, we will only show the effect of the concentration on the deposited films. When the concentration of C8-BTBT is 2 mg/mL, large branches in the crystalline thin films are observed normal to the blow coating direction, resulting in incomplete surface coverage (Figure S9a). By increasing the concentration to 4 mg/mL and 6 mg/mL, the branches are reduced and the crystals fully cover the substrate.
(Figure 3a and Figure S9b). This indicates that enough solute is required to grow well-aligned facet single crystals. Further increasing the C8-BTBT concentration introduces excess nucleation and crystals are observed on top of the aligned crystals (Figure S9c).

**S2.3. Gas blow coating of P3HT and DPPTTT films**

The gas blow coating of P3HT on glass has been studied as a function of the deposition parameters, as done for TIPS-Pentacene and C8-BTBT (Sections S2.1-2.2).

![Figure S10](image)

**Figure S10.** Pictures of the P3HT films deposited on glass by using different gas supply pressure.

The effect of the gas supply pressure is first studied for a fixed speed of 0.6 cm/s and concentration of 10 mg/mL. **Figure S10** shows that when the gas pressure is lower than 0.15 bar, a wet film is formed, which turns into a dense film upon solvent evaporation. However, no aligned crystals are formed as polymers are not easy to organize during fast solvent evaporation process. When the pressure exceeds 0.3 bar, the gas can force the solution to move ahead to cover the whole length of the substrate. When the pressure increases from 0.3 to 1.35 bar, films are formed, but they are characterized by dark-red lines, which are thicker than the rest of the film. This means that the gas pressure is not able to produce enough shearing force to spread forward the whole droplet in a uniform way. When the pressure is increased to 1.8 bar, a narrower uniform film can be achieved. No darker lines are observed. A further increase in the
gas pressure results in stripes, indicating that the pressure is too high and produces a driving force that spreads the solution perpendicular to the moving direction.

Figure S11. Pictures of the P3HT films deposited by using different moving speed of the gas knife on glass.

We then study the effect of the moving speed of gas knife on the obtained film at pressure of 1.8 bar. Figure S11 shows that with increasing moving speed of the gas knife, darker lines appear. These are signatures of not enough high shearing force to move forward the whole droplet while spreading it uniformly on the substrate. Figure S12 shows that a uniform thin film of P3HT can be easily deposited on the silicon wafer with moving speed of 0.6 cm/s and gas supply pressure of 1.8 bar.

Figure S12. Optical microscope image the P3HT film deposited by gas blow coating with moving speed of 0.6 cm/s and gas supply pressure of 1.8 bar on silicon wafer.
Figure S13. Pictures of the DPPTTT films deposited by using different moving speed of the gas knife on glass (the blue line shows the coating direction).

According to the optimal results for P3HT film, a low moving speed should be used for the deposition of uniform film using semiconducting polymers. Therefore, we decided to use the lowest speed (0.6 cm/s) in our set up and optimize the gas supply pressure for deposition of DPPTTT films. Figure S13 shows that with increasing the gas supply pressure, the width of the film become quite narrow when the pressure is higher than 1.25 bar. When the pressure is further increased to 1.6 bar, the film is almost transparent without any disruption lines. Figure S14 shows an optical picture of a thin film of DPPTTT, deposited on silicon using a moving speed of 0.6 cm/s and gas supply pressure of 1.6 bar. This figure shows that the film is relatively uniform on a large area.

Figure S14. Optical microscope image the DPPTTT film deposited by gas blow coating with moving speed of 0.6 cm/s and gas supply pressure of 1.6 bar on silicon wafer.
S3 Characterization of the devices

Tables S2 summarizes the results obtained for OFET made of TIPS-pentacene deposited under optimized conditions by gas blowing. We noticed that the highest mobility is achieved by using the lowest channel length (20 µm), which is ascribed the fact that some defects or disruptions in the crystals have a lower chance to appear in such a short channel. However, only one device works for this channel length as a result of the difficulty in deposition of the gold electrodes that do not connect with each other in such small channel length.

Tables S3 summarizes the results obtained for devices based on C8-BTBT aligned crystals. This table shows that compared with the devices with channel length larger than 60 µm, the devices with smaller channel length of 20 µm and 40 µm show lower charge mobility and higher $V_{th}$, which is ascribed to parasitic contact effects.\[^{[S19]}\] However, when the channel length is larger than 60 µm, this effect is minimal and the mobility is almost constant.

Tables S4 and Table S5 summarize the results obtained for the devices based on P3HT and DPPTTT thin films, respectively. With the changing of the channel lengths, the charge mobility and $V_{th}$ are almost constant, confirming that the films are uniform at least over length of hundreds of µm.

Table S2. OFET electrical characteristics of TIPS-pentacene deposited by gas blow coating.

| Channel Length/µm | Number of Devices | Mobility/cm²V⁻¹S⁻¹ | Max Mobility/cm²V⁻¹S⁻¹ | $V_{th}$/V | On/off ratio |
|-------------------|-------------------|----------------------|--------------------------|------------|--------------|
| 20                | 1                 | 0.15                 | 0.15                     | -6.7       | 7.8*10⁴      |
| 40                | 5                 | 0.06±0.02            | 0.07                     | -6.7±6.2   | 10⁴–10⁷      |
| 60                | 6                 | 0.07±0.04            | 0.13                     | -5.1±3.2   | 10⁴–10⁷      |
| 80                | 5                 | 0.07±0.02            | 0.09                     | -7.2±2.5   | 10⁴–10⁷      |
| 100               | 6                 | 0.05±0.03            | 0.09                     | -4.9±2.0   | 10⁴–10⁷      |
Table S3. OFET electrical characteristics of C8-BTBT thin films deposited by gas blow coating.

| Channel Length/µm | Number of Devices | Mobility/cm²V⁻¹S⁻¹ | Max Mobility/cm²V⁻¹S⁻¹ | V_th/V | On/off ratio |
|-------------------|-------------------|----------------------|--------------------------|--------|--------------|
| 20                | 4                 | 0.4±0.19             | 0.67                     | 15.2±3.6 | 10⁶-10⁷      |
| 40                | 5                 | 0.3±0.11             | 0.42                     | 7.9±3.2  | 10⁶-10⁷      |
| 60                | 7                 | 0.6±0.37             | 1.40                     | 1.4±4.5  | 10⁶-10⁷      |
| 80                | 7                 | 0.7±0.24             | 0.95                     | 0.3±5.4  | 10⁶-10⁷      |
| 100               | 5                 | 0.7±0.31             | 1.19                     | -4.6±0.9 | 10⁶-10⁷      |

Table S4. OFET electrical characteristics of P3HT thin films deposited by gas blow coating.

| Channel Length/µm | Number of Devices | Mobility/cm²V⁻¹S⁻¹ | Max Mobility/cm²V⁻¹S⁻¹ | V_th/V | On/off ratio |
|-------------------|-------------------|----------------------|--------------------------|--------|--------------|
| 20                | 5                 | 1.48E-03±3.92E-04    | 0.02                     | 3.4±4.7 | 10³-10⁴      |
| 40                | 5                 | 1.04E-03±3.40E-04    | 0.0015                   | -3.4±8.4 | 10³-10⁴      |
| 60                | 5                 | 9.5E-04±2.59E-04     | 0.0013                   | -6.4±5.7 | 10³-10⁴      |
| 80                | 5                 | 6.7E-04±3.66E-04     | 0.0011                   | -3.1±2.0 | 10³-10⁴      |
| 100               | 5                 | 8.06E-04±2.19E-04    | 0.0010                   | -4.2±3.6 | 10³-10⁴      |

Table S5. OFET electrical characteristics of DPPTTT thin films deposited by gas blow coating.

| Channel Length/µm | Number of Devices | Mobility/cm²V⁻¹S⁻¹ | Max Mobility/cm²V⁻¹S⁻¹ | V_th/V | On/off ratio |
|-------------------|-------------------|----------------------|--------------------------|--------|--------------|
| 20                | 12                | 0.31±0.10            | 0.52                     | 6.8±5.5 | 10⁴-10⁷      |
| 40                | 12                | 0.34±0.16            | 0.65                     | 6.6±3.1 | 10⁴-10⁷      |
| 60                | 11                | 0.43±0.26            | 0.88                     | 5.2±2.1 | 10⁴-10⁷      |
| 80                | 13                | 0.35±0.22            | 1.0                      | 4.1±2.8 | 10⁴-10⁷      |
| 100               | 12                | 0.41±0.41            | 1.5                      | 4.8±3.3 | 10⁴-10⁷      |
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