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Abstract: The printing of organic semiconductors (OSCs) by means of meniscus guided coating (MGC) has great potential for the fabrication of high-performance, uniform, and large area flexible electronics. Furthermore, surface energy patterning allows low-cost, large area manufacturing, and is a requirement for device isolation to achieve accurate drive and to minimize power consumption. We investigated the morphological and electrical characteristics of surface energy patterned OSCs coated using the MGC method. A unique phenomenon, contact line curvature-induced molecular misorientation effect, which occurs in the patterning of OSCs with the MGC method, is demonstrated. This effect is highly dependent on the pattern width which decided contact line curvature. It influences the charge transport properties and hence the field-effect mobility of organic thin-film transistors (OTFTs). OTFTs in widths ranging from 50 to 500 μm were fabricated. The highest misorientation angle was induced in the 50 μm pattern width and the corresponding OTFTs exhibited the highest mobility.

Cover

1 **Contact line curvature-induced molecular misorientation of a**
2 **surface energy patterned organic semiconductor in meniscus-**
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4

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21 **Abstract**

22 The printing of organic semiconductors (OSCs) by means of meniscus guided coating (MGC) has great
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32 width and the corresponding OTFTs exhibited the highest mobility.

33 **Keywords**

34 Molecular misorientation, surface energy patterning, organic semiconductor, meniscus-guided coating

35

36 **Introduction**

37 Great expectations for next-generation electronics have prompted active research on solution-processed
38 organic thin-film transistors (OTFTs) which can be front-running candidates for low-cost, large-area
39 device manufacturing, and flexible active-matrix displays and sensors¹⁻³. High electrical performance
40 beyond that of amorphous silicon TFTs has been achieved in solution-processed OTFTs through the
41 tuning of small molecule organic semiconductors (OSCs)⁴⁻⁷. One of the best tuning methods is to control
42 the solution coating. In this regard, many solution coating techniques like inkjet printing, shearing, spin-
43 coating, spray coating, dip-coating, blade-coating, and slot-die coating have been developed⁸⁻¹⁴. Among
44 these methods, meniscus guided coating (MGC) methods, as in blading, shearing, or slit-die coating,
45 which deposit solution directionally on the target substrate by the virtue of capillary force between the
46 substrate and the coating head, have been extensively studied because of their special advantages¹⁵. They
47 can form uniform thin films with easily controlled film thickness in large-area fabrication compared to
48 other coating methods. Moreover, they induce molecular alignments, and in some cases molecular
49 packing due to their directionality. As a result, the charge transport properties of small molecule OSCs are
50 enhanced and high field-effect mobility can be achieved⁹.

51 Patterning of OSC thin films plays a significant role in the improvement of the electrical characteristics of
52 OTFTs. Confinement of the OSCs through patterning suppresses leakage currents and crosstalk effect in
53 integrated circuits, thus yielding more accurate TFT drive and lower power consumption¹⁶⁻¹⁸. These
54 positive attributes have driven the development of various patterning methods for selective deposition of
55 semiconductor thin films. Photolithography, template guided patterning, stamping, and chemical
56 patterning have been introduced to pattern soluble OSCs¹⁹⁻²¹. In particular, chemical patterning that
57 induces hydrophobic and hydrophilic region selectively through self-assembled monolayer formation or
58 surface terminal group control has been widely studied due to its merits^{16-18,21}. It is a facile method that
59 does not require conventional photolithography and is not area constrained. This means that fewer
60 process steps, lower manufacturing costs, and large-area fabrication can be achieved.

61 Nevertheless, many studies of MGC methods have been focused on improving unpatterned thin-film
62 characteristics and TFT electrical performance by controlling coating velocity or solvents^{6,22,23}. Although
63 some reports have studied patterning methods and have fabricated patterned OTFTs, the morphological
64 and electrical characteristics of MGC methods have not been fully understood. Furthermore, a unique

65 behavior of patterned small molecule OSCs that does not occur in an unpatterned layer when using MGC
66 methods should be addressed.

67 In this study, we investigated the morphological and electrical characteristics of the surface energy
68 patterned bis (triisopropylsilylethynyl) (TIPS)-pentacene deposited with blade coating, one of the
69 representative MGC methods. We observed the molecular misorientation that affected the charge
70 transport properties between source and drain electrodes was induced in the mesoscale patterned layer by
71 the contact line curvature of the blade coating process. In addition, the pattern width highly affected the
72 contact line curvature-induced molecular misorientation. To demonstrate the electrical result of this effect,
73 the OTFTs were fabricated with TIPS-pentacene thin films of various pattern widths. TIPS-pentacene thin
74 films patterned with a 50 μm width showed the lowest misorientation angle of 7.75° , while those
75 patterned with a 300 μm width exhibited the highest misorientation angle of 18° . Since the molecular
76 misorientation affected their charge transport, OTFTs patterned with 300 μm width showed the lowest
77 field-effect mobility of $0.216 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. In contrast, the 50 μm wide patterned OTFTs showed a
78 relatively high field-effect mobility of $0.317 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, a more effective charge transport value.

80 **Materials and methods**

81 Figure 1(a) shows a schematic of the OSC patterning process. Indium-tin oxide (ITO)-deposited glass
82 was prepared for the substrate and gate electrodes of the TFTs. Cross-linked poly(4-vinyl phenol) (c-PVP),
83 mixed with methylated poly(melamine-co-formaldehyde) at a mass ratio of 1:1.25 and dissolved in
84 propylene glycol methyl ether acetate at 10 wt. % was spin coated on the ITO gates to form a polymer
85 insulator. The substrate was spun at 3,000 revolutions per minute for 30 s and annealed at 200°C for 1 h
86 on a hot plate for cross-linking. For chemical patterning of the TIPS-pentacene, hydrophobic regions were
87 generated through polydimethylsiloxane (PDMS) attachment on c-PVP and heated for 10 min at 120°C .
88 In this step, the methyl group, which is the surface terminal group of PDMS, was transferred onto the c-
89 PVP insulator¹⁸. To remove the methyl groups and generate the hydrophilic regions selectively, a shadow
90 metal mask covered c-PVP surface was exposed to ultraviolet-ozone (UVO) for 10 min. The detailed
91 principles and the process of chemical patterning used in this study were described in our previous study¹⁸.
92 Figure 1(b) describes the patterning process of TIPS-pentacene solution. The widths of the hydrophilic
93 wetting regions were controlled in a mesoscale of 50 – 500 μm . Subsequently, a 2 wt. % TIPS-pentacene
94 solution dissolved in anisole was blade coated onto the c-PVP at 60°C ²⁴. The TIPS-pentacene solution
95 was captured between the blade and the substrate by capillary force and coated on the c-PVP insulator
96 surface. It was coated directionally at various speeds of 0.1, 0.3, and 0.5 mm/s and spontaneously
97 deposited on the hydrophilic c-PVP region. An image of the patterned TIPS-pentacene is shown in Figure
98 1(c). Red arrows indicate the coating direction. Finally, 50 nm of Au was thermally deposited at a rate of
99 0.1 nm/s to form the source and drain electrodes. Channel length was 100 μm and the channel width
100 depended on the patterned width of TIPS-pentacene thin films. The completed OTFTs were measured at
101 room temperature under ambient pressure in a dark environment.

103 **Results and discussion**

104 To describe blade coated TIPS-pentacene structure and define the contact line curvature-induced
105 molecular misorientation angle θ_1 , a 3D atomic force microscope (AFM) image of TIPS-pentacene was
106 recorded as shown in Fig. 2. TIPS-pentacene was blade coated at the speed of 0.3 mm/s with a substrate
107 temperature of 60°C , and a blade gap of 200 μm . For organic semiconductors, π - π stacking structure of
108 molecules facilitates charge transport^{9,25}. Theoretically, the π - π stacking direction of solution-processed
109 small molecules follows the direction of solution evaporation which is the same as the coating direction
110 for MGC methods^{9,26}. MGC methods form a well aligned and continuous molecular stacking of TIPS-
111 pentacene parallel to the coating direction due to their intrinsic directionality. Needle-shaped crystalline
112 domains with 3 μm width and 30 nm thickness were observed in this study as shown in Fig. 2. The long
113 axis of the needle-shaped TIPS-pentacene crystalline domains was parallel to [210] as well as the crystal
114 growth direction (blue arrow). Source and drain electrodes are usually designed to be parallel to the
115 coating direction of OSC solutions for efficient field-effect charge transport., If molecular misorientation

116 is induced, however, the unintended angular mismatch between the coating direction (red arrow) and
117 crystal growth direction results in a disturbance of charge transport and a decrease in mobility. In Figure 2,
118 θ_1 is the contact line curvature-induced misorientation angle which is the angle between coating direction
119 and crystal growth direction.

120 The morphological characteristics of surface energy patterned TIPS-pentacene thin films were observed.
121 Figure 3 display schematic and the corresponding cross-polarized optical microscope (CPOM) images of
122 patterned TIPS-pentacene at varying pattern widths. Red arrows in the schematic and CPOM images
123 indicate the coating direction, and the yellow arrow in the CPOM images indicates the crystal growth
124 direction, which is the [210] direction of the TIPS-pentacene molecules. Regions 1 and 2 are the edge and
125 middle sections of the patterned TIPS-pentacene thin films. The scale bars in the CPOM images are 50
126 μm . The TIPS-pentacene solution was coated at different speeds of 0.1, 0.3, and 0.5 mm/s. As shown in
127 Figure 3, the crystal texture and thickness vary with increasing coating speed, from long, wide and thick
128 crystallites to short, narrow and thin crystallites. The molecular misorientation was observed in both
129 regions 1 and 2 regardless of the coating speeds and widths.

130 To convey the effect of contact line curvature-induced misorientation on the pattern width, the contact
131 line curvature-induced misorientation angles at various pattern widths are shown in Fig. 4(a). In this study,
132 contact line curvature-induced misorientation angles and OTFTs were studied at a coating speed of 0.3
133 mm/s because, at that speed, the highest crystallinity and best alignment of TIPS-pentacene were obtained,
134 and as a result, the highest mobility of the OTFTs was observed²⁷. The misorientation angle of
135 unpatterned layers occurring due to fluid-flow was $9.87 \pm 2.13^\circ$. When the TIPS-pentacene was deposited
136 on the confined hydrophilic wetting region of 500 μm width, the misorientation angle increased to
137 $15 \pm 3.61^\circ$. It increased more to $18 \pm 3.24^\circ$ at the pattern width of 300 μm . By contrast, it began to decrease
138 with a decrease in the pattern width from 300 μm . The misorientation angles of $13.63 \pm 1.08^\circ$ and
139 $7.75 \pm 1.14^\circ$ were observed at the widths of 100 and 50 μm , respectively. Interestingly, the misorientation
140 angle of patterned TIPS-pentacene at 50 μm width was smaller than that of the unpatterned layer. These
141 results might have originated from the different contact line curvature of varying pattern widths.

142 Figure 4(b) shows several of the forces acting in a blade coating solution. First, capillary force helps in
143 capturing the solution between substrate and blade head, and the surface tension makes a meniscus line.
144 In addition, there is attraction between solution and wetting surface due to the hydroxyl group on the c-
145 PVP surface which generates high surface energy, so a solution in contact with the wetting surface
146 remains on the wetting regions when the blade moves. There are dewetting regions outside of the wetting
147 regions and the methyl group of the dewetting region forms a repulsive force between the hydrophobic
148 surface and TIPS-pentacene solution (Figure 4(c)). The complexity of the forces and the different
149 influences of these forces at varying pattern widths result in different contact line curvature-induced
150 molecular misorientations while blade coating. When the pattern is narrow, as in the 50 μm width, the
151 solution on the wetting region is too small compared with the main solution captured between the blade
152 and the surface, so the surface tension is dominant resulting in a high θ_2 which is the angle induce the
153 molecular misorientation in patterned layer. In addition, fewer nucleates in the wider patterns than in the
154 unpatterned layer may induce well-oriented molecular stacking⁶. In summary, the patterned TIPS-
155 pentacene on the 50 μm width showed the lowest misorientation angle θ_1 . When the pattern width is large,
156 the amount of solution on the wetting region and the molecules sited at the center of the wetting region
157 are predominantly affected by the attractive force but scarcely affected by the repulsive force. This results
158 in a high contact line curvature, a small θ_2 , and a large θ_1 . As the pattern width increases beyond 300,
159 the repulsive forces cannot dominate most molecules except those sited at the edge due to the wide
160 hydrophilic region, so the contact line curvature decreases again and results in a large θ_2 and a small θ_1 as
161 shown in Figure 4(a).

162 We fabricated OTFTs to demonstrate the impact of contact line curvature-induced molecular
163 misorientation on the OTFT electrical performance. Figure 5 (a)-(e) shows the transfer curves of the 50,
164 100, 300, and 500 μm patterned OTFTs, and the unpatterned OTFTs coated at the speed of 0.3 mm/s. The
165 corresponding field-effect mobilities were 0.317, 0.240, 0.216, 0.267 and 0.276 $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$. As the pattern
166 width increased up to 300 μm , the mobility decreased from 0.317 to 0.216 $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$. In contrast, the
167 mobility increased again from 0.216 to 0.276 $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ as the pattern width increased from 300 μm . It
168 exhibited a tendency opposite to the misorientation angle graph (Figure 4(a)) as shown in Figure 6. The

169 contact line curvature-induced misorientation angle affected the charge transport properties and therefore
170 the mobility of OTFTs.

171

172 **Conclusions**

173 We investigated the morphological characteristics and the molecular misorientation effect of surface
174 energy patterned TIPS-pentacene deposited by means of blade coating. We demonstrated that contact line
175 curvature-induced molecular misorientation occurs only in patterned OSCs coated using an MGC method
176 and is highly dependent on the pattern width. The lowest molecular misorientation angle of 7.7° occurred
177 in a $50\ \mu\text{m}$ pattern width. This was due to the dominance of surface tension and the generation of fewer
178 nuclei resulting in the highest mobility of $0.317\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. The highest misorientation angle was induced
179 in a $300\ \mu\text{m}$ pattern width of due to the dominance of repulsive and attractive forces compared to other
180 forces. The corresponding OTFTs exhibited the lowest mobility of $0.216\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. We expect this work
181 will provide an optimized pattern width for small-molecule OSCs to achieve the most effective charge
182 transport properties and to maximize the field-effect mobility of OTFTs.

183

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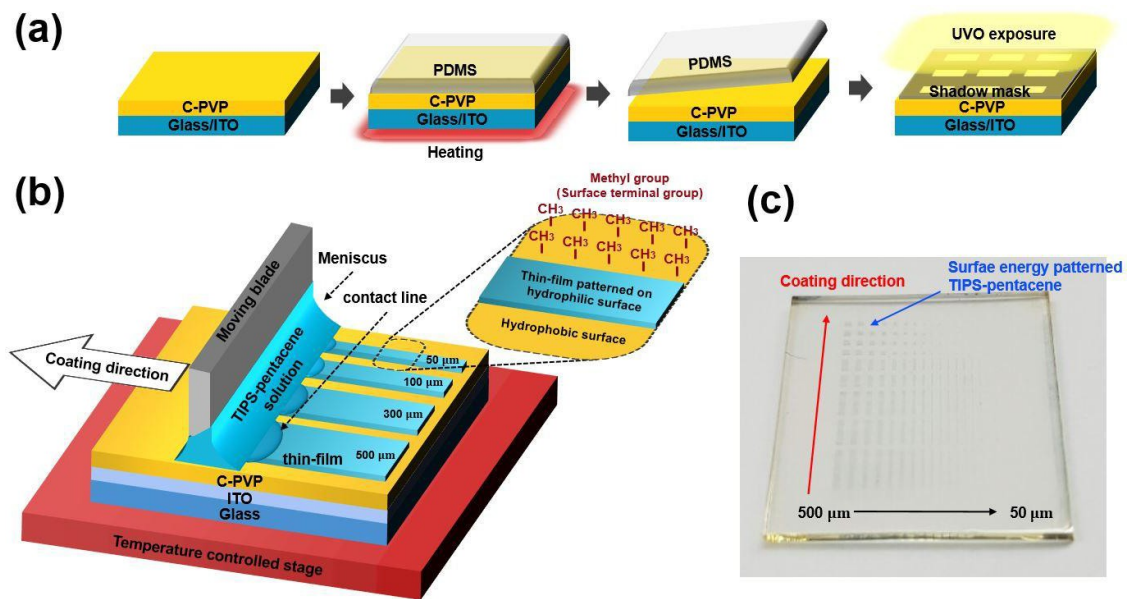
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Figure 1. (a) Schematic illustration of the surface energy patterning process. (b) Schematic illustration of the blade coating of TIPS-pentacene on pre-patterned C-PVP surface. (c) Photographic image of surface energy patterned TIPS-pentacene on c-PVP insulator. Red arrow indicates blade coating direction.

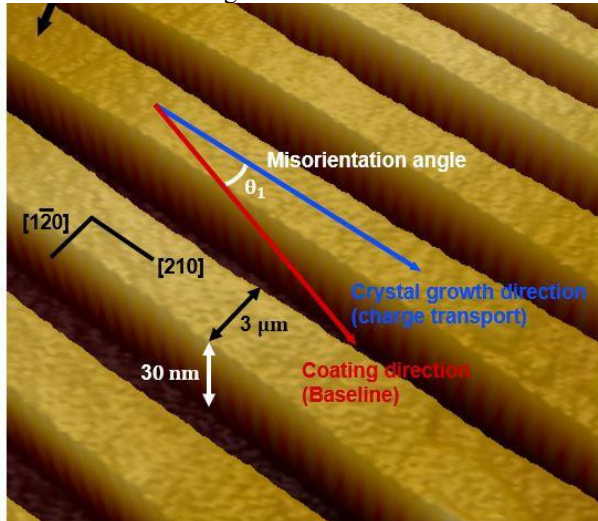


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Figure 2. 3D AFM image of TIPS-pentacene crystalline domain and the schematic defining the

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misorientation angle.



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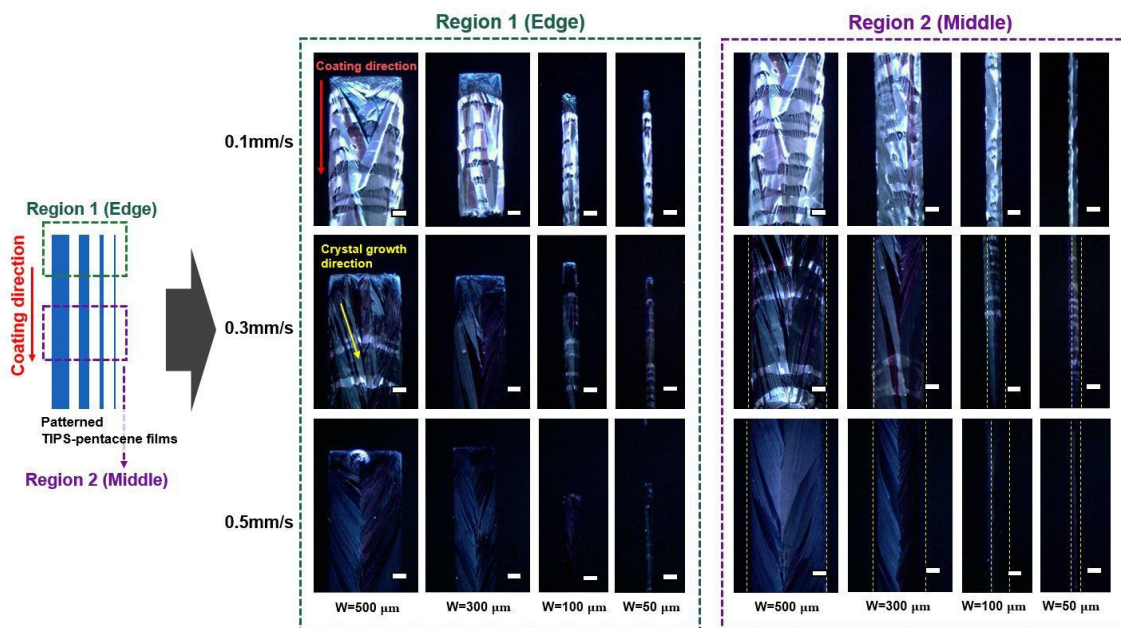
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329 Figure 3. (a) Schematic illustration of patterned TIPS-pentacene thin films and (b) corresponding CPOM
330 images for various coating speeds and various widths.



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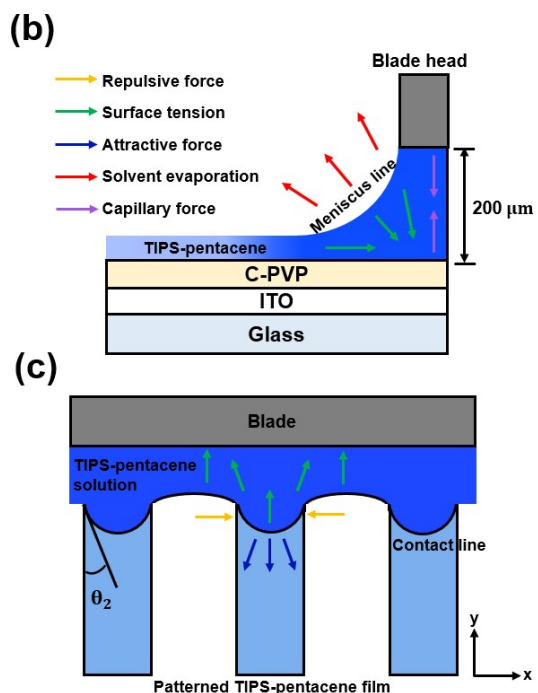
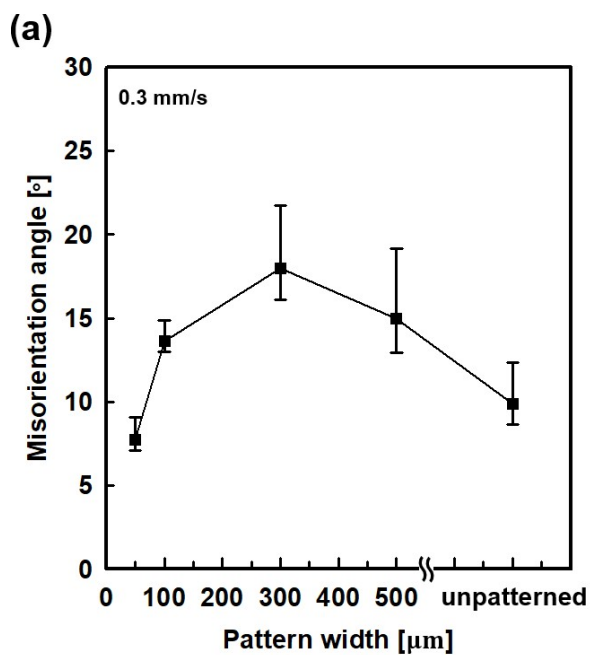
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353 Figure 4. (a) Molecular misorientation angles of patterned TIPS-pentacene for various pattern width.

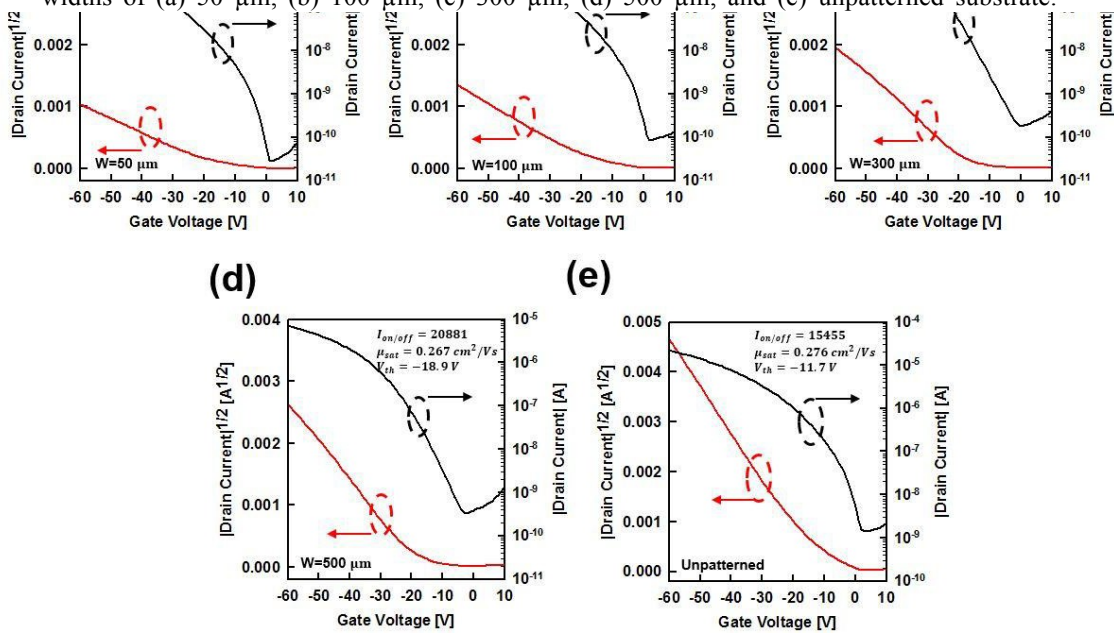
354 Schematic illustrations of (b) side view and (c) top view of meniscus-guided coating of TIPS-pentacene.

355 Red and yellow arrows indicate coating and crystal growth direction, respectively.



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377 Figure 5. Representative transfer characteristic curves of the OTFTs coated at 0.3 mm/s on the pattern
 378 widths of (a) 50 μm , (b) 100 μm , (c) 300 μm , (d) 500 μm , and (e) unpatterned substrate.



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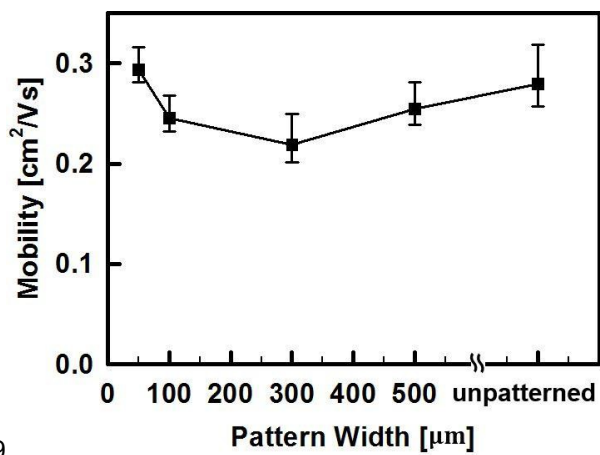
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387 Figure 6. Average field-effect mobilities of OTFTs fabricated observed from surface energy patterned
 388 TIPS-pentacene thin films for various pattern widths.



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