

2D and 3D Crystal Formation of 2,6-Bis[4-ethylphenyl]anthracene with Isotropic High Charge-Carrier Mobility

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Organic semiconductor materials with high charge-carrier mobility and high luminescence are promising for fabricating high-performance organic light-emitting transistors and electrically pumped organic laser diodes. Some derivatives with an anthracene core show both characteristics; however, independently controlling the size and the thickness of single-crystal films is challenging. Here, 2D and 3D crystal formation is demonstrated by using an air-stable organic semiconductor, 2,6-bis[4-ethylphenyl]anthracene (BEPAnt). The shape of the single-crystal films can be changed from about 0.10 mm in diameter and 400 nm in thickness to 1.0 mm in diameter and 50 nm in thickness by controlling the growth conditions using saturation–supersaturation vapor pressure curves. The in-plane growth rate increases by ten times while the out-of-plane growth rate decreases to about 1/8 as the growth temperature increases from 210 to 250 °C. Single-crystal organic field-effect transistors based on BEPAnt show isotropic mobilities as high as $7.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

1. Introduction

Organic semiconductor materials with high mobility and strong fluorescence emission have attracted much interest for fabricating high-performance organic light-emitting transistors (OLETs)^[1,2] and organic laser diodes (OLDs) by current injection.^[3–5] Especially, single crystals are preferred for these devices because the absence of grain boundaries or molecular disorders results in high mobility.^[6–9] Meanwhile, it is challenging for organic single crystals to satisfy both high mobility and high emission simultaneously, because the requirements of those two characteristics are completely conflicting in many organic semiconductors.^[10] Interestingly, anthracene derivatives belong to a special type of organic semiconductors, in which both characteristics can be achieved.^[11–14] However, the crux limiting

the application of anthracene derivatives is the difficulty to grow large and thin single crystals, which are highly desired in OLETs and OLDs. For example, Liu et al. demonstrated that 2,6-diphenylanthracene (DPA)^[15] based single-crystal organic field-effect transistor (SC-OFET) showed mobilities as high as $34 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and photoluminescence quantum yield of 41.2%.^[16] Unfortunately, since the size of the DPA crystals was very small (around 10–30 μm in diameter), it seems to be very difficult to fabricate single-crystal OLETs and OLDs using DPA. Generally, it is extremely challenging to independently control the lateral dimensions and the thickness of an organic single-crystal film by physical vapor transport (PVT). For PVT method, when the strength of the interaction between the surface and

molecules is appropriately controlled, the molecules will diffuse across the surface in the initial process of the deposition. Then, a sufficient number of molecules are packed together by intermolecular forces to form stable nuclei at intervals corresponding to the diffusion length. Finally, crystal growth occurs at these nuclei and this acts as a template for forming single-crystal films. Therefore, we can potentially control the lateral dimensions and the thickness by changing the side chains to anthracene derivatives. Actually, our group introduced different heteroatoms into the peripheral side chains to anthracene derivatives,^[17] and obtained large single crystals of 2,6-bis[4-methoxyphenyl]anthracene (BOPAnt) by PVT. The material exhibited isotropic charge transport with high mobility of $16.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ as well as highly responsible phototransistors.^[18,19] However, the growth mechanism was not clarified in the previous work.

In this paper, a similar compound 2,6-bis[4-ethylphenyl]anthracene (BEPAnt) was selected to demonstrate the growth mechanism of PVT, which could be used as a guideline for single-crystal growth of organic semiconductors for organic electronics. Here, the lateral dimensions and the thickness of the BEPAnt crystals can be controlled independently by considering the PVT growth mechanism. More interestingly, 2D and 3D crystal formation of BEPAnt was demonstrated by PVT. By changing the growth conditions using the saturation–supersaturation vapor pressure curves. The dimensions of single-crystals can be changed from about 0.10 mm in diameter and 400 nm in thickness to 1.0 mm in diameter and 50 nm in thickness. The in-plane growth rate became ten times faster

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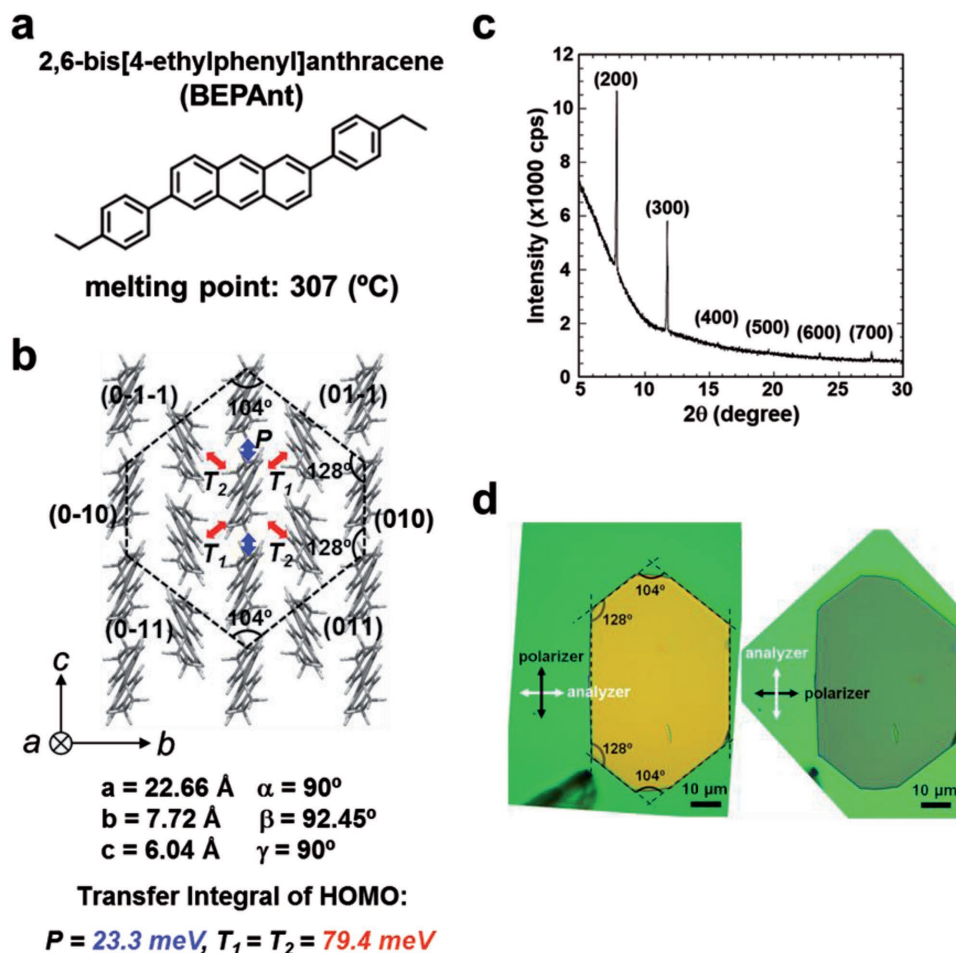


Figure 1. a) Molecular structure used in this experiment, 2,6-bis[4-ethylphenyl]anthracene (BEPAnt). b) Top view of the lamellar herringbone packing structure with lattice parameters and transfer integral values. c) X-ray diffraction result. d) Crossed Nicol polarized microscopy images of the same thin film.

while the out-of-plane growth rate reduced to about 1/8 as the growth temperature increased from 210 to 250 °C. Finally, the fabricated SC-OFETs showed an isotropic mobility as high as $7.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Based on the results, it can be concluded that the proposed growth mechanism provides a more effective way for growing large and thin single-crystal films of anthracene derivatives by PVT, which can further promote its applications in OLETs and OLDs.

2. Results and Discussion

2.1. Analysis of BEPAnt Single-Crystal Film

Figure 1a illustrates the molecular structure of BEPAnt. In order to obtain information about the maximum sublimation temperature, we measured the melting point of BEPAnt to be 307 °C using differential scanning calorimetry (DSC). In order to improve the material purity, we purified the BEPAnt three times before it was used for crystal growth. Figure 1b shows the top view of the crystal structure of BEPAnt in the b - c plane, which is composed of aligned herringbones with structural

parameters of: $a = 22.66 \text{ Å}$, $b = 7.72 \text{ Å}$, $c = 6.04 \text{ Å}$, $\alpha = \gamma = 90^\circ$, and $\beta = 92.45^\circ$. The transfer integral values of HOMO were $P = 23.3 \text{ meV}$ and $T_1 = T_2 = 79.4 \text{ meV}$, as derived by our previous work.^[17] The detailed information about the design, synthesis, and polycrystalline thin-film transistor (TFT) characteristics of this material can be referred to our previous work.^[17] Figure 1c shows the X-ray diffraction (XRD) result of the BEPAnt single-crystal film on an octyltrichlorosilane (OTS)-modified SiO_2/Si substrate. The simulated powder X-ray diffraction pattern of BEPAnt based on the single crystal data is shown in Figure S1 (Supporting Information) for comparative purpose. The peaks at $2\theta = 7.82^\circ$, 11.66° , 15.59° , 19.53° , 23.46° , and 27.50° were attributed to the (200), (300), (400), (500), (600), and (700) reflections of the BEPAnt, respectively. A lattice plane distance calculated from the (200) reflection was 22.61 Å , which corresponds to the crystallographic a -axis lattice parameter (Figure 1b), indicating that the BEPAnt molecules stood up perpendicular to the substrate surface. Figure 1d presents two polarized optical microscopy images of the same BEPAnt single-crystal film. In images recorded through crossed Nicol prisms, the film color changed from bright to dark, as it was rotated from 0° to 90° . Furthermore, the film exhibits

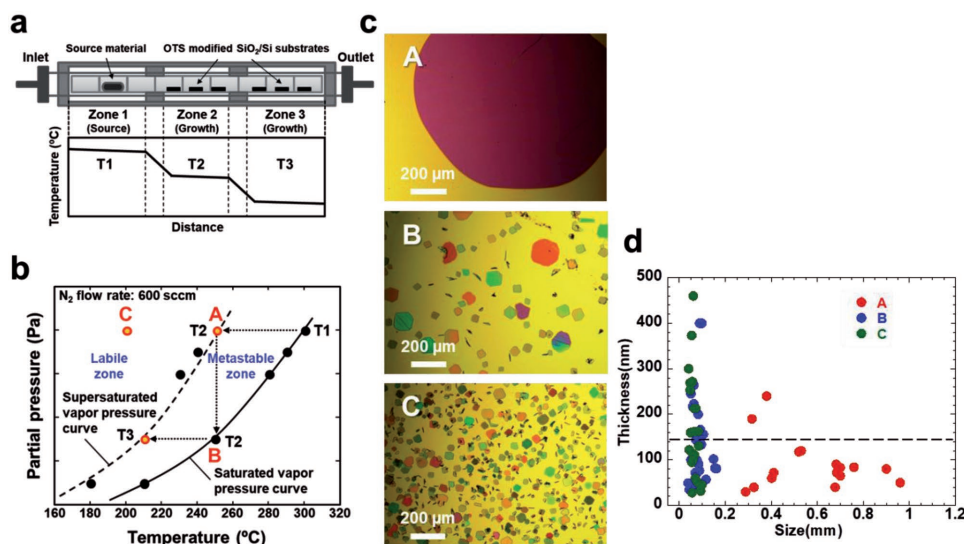


Figure 2. a) Schematic illustration of our furnace with three temperature control zones. Zone 1 is for sublimating source material, and zone 2 and zone 3 are for growing single-crystal films. The temperature distribution is also schematically inserted. b) Experimentally obtained saturated-supersaturated vapor pressure curve of BEPant. c) Crossed Nicol polarized microscopy images of single-crystal films obtained from point A (Zone 2: T1 = 300, T2 = 250, and T3 = 210 °C), B (Zone 3: T1 = 300, T2 = 250, and T3 = 210 °C), and C (Zone 2: T1 = 300 and T2 = 200 °C), respectively. Growth duration was 50 min. The density of single-crystal films for point A, B, and C was 5–15, 160–250, and around 1000 mm⁻², respectively. d) Relationship between thickness and size of single-crystal films obtained from point A, B, and C. The size is defined as $R = 2(S/\pi)^{1/2}$; R and S are the diameter and the area of each single-crystal film.

similar facet angles of 104° and 128°, indicating that facet growth occurred. From these results, we can conclude that the film is single crystal. By comparing Figure 1b with Figure 1d, two characteristic angles of 104° and 128° can be obtained between the {011} and {01-1} facets, and between the {010} and {011} or {01-1} facets, respectively.

2.2. Growth Method and 2D and 3D Crystal Formation

Figure 2a schematically depicts our PVT furnace with three temperature zones. We set the temperature of zone 1 as T1, and filled zone 1 with saturated BEPant vapor. The temperature of zone 2 was set as T2 (<T1), and as the BEPant vapor was carried from zone 1 to zone 2 by N₂ gas, the degree of supersaturation became higher in zone 2. If the state of the vapor in zone 2 exceeded the threshold value of supersaturation by decreasing T2, nuclei started to form. We defined the threshold values as “supersaturated vapor pressure curve” in Figure 2b. Since nucleation generally requires higher supersaturated vapor pressure than crystal growth, only crystal growth can take place in the zone between the saturated vapor pressure curve and the supersaturated vapor pressure curve. This zone is called the “metastable zone.” Below the supersaturated vapor pressure curve (labile zone), spontaneous crystallization can occur, which means that the continuously supplied molecules are used for growing existing crystals and forming new nuclei. This phenomenon is similar to that seen in the solution growth^[20,21] because the growth proceeds under the quasi-equilibrium state. In our equipment, we had two crystal growth zones, where zone 2 is filled with the saturated BEPant vapor at T2 and zone 3 is set at an even lower temperature of T3 (<T2). We could also obtain single crystals if the vapor

state in zone 3 changed from the metastable zone to the labile zone. Figure 2b shows the experimentally obtained saturated-supersaturated vapor pressure diagram for BEPant in our growth system. First, we set T1 at 300 °C (<307 °C: BEPant melting point) and tried to grow single crystals by changing T2 from 270 to 250 °C at 10 °C decrement. At T2 = 270 °C, we could not find single crystals on any SiO₂/n-Si substrates. At T2 = 260 °C, we obtained single crystals only on the substrate arranged at the right hand side of zone 2 (Figure 2a). In the case of T2 = 250 °C, we could obtain single crystals on all of the substrates. By these results, it was found that there was a temperature distribution in zone 2 (and also zone 3) in our growth equipment. However, fortunately, we could verify that the supersaturated vapor pressure curve exists between 250 and 260 °C. Next, we fixed T1 (300 °C) and T2 (250 °C), and in the same way, obtained T3 of 210 °C on the supersaturated vapor pressure curve. Figure 2c shows crossed Nicol polarized microscopy images of substrates grown under the condition at point A, B, and C, respectively, in Figure 2b. Points A and B lie on the supersaturated vapor pressure curve, and point C lies in the labile zone. At point A and point B, T1 = 300 °C, T2 = 250 °C, and T3 = 210 °C, while sample A was grown in zone 2 and sample B was grown in zone 3. At point C, T1 = 300 °C and T2 = 200 °C, sample C was grown in zone 2. It should be noted that a very large single-crystal film with a diameter of about 1.0 mm was obtained at point A. While, in the sample grown at point C, there were many small single crystals because the spontaneous nucleation occurred in the labile zone. Note that many single crystals can also be seen in the sample grown at point B. The size of the crystals ranges from several 10s to 100s μm with a population density of 160–250 mm⁻². The population density is much lower in the sample grown at the point A, which is only 5–15 mm⁻².

Since both points A and point B lie on the supersaturated vapor pressure curve, it was thought that spontaneous crystallization could not occur. Thus, the population density of the nuclei should be lowest under both conditions. First, to explain the differences in the population density and the crystal size between points A and point B, we need to look at the migration distance of the BEPant molecules on the OTS-modified SiO₂ surface. A typical migration distance of BEPant is about 60–80 and 260–450 μm at the growth temperature of 210 and 250 $^{\circ}\text{C}$, respectively, as estimated by the population density. Second, in order to explain the difference of the crystal size, we observed carefully the microscopy image of the sample grown at point B (Figure 2c), and found cracks in some of the crystals. Third, we investigated the thickness of each single-crystal film. Figure 2d shows the relationship between the thickness and the crystal size for samples grown at points A, B, and C. The crystal size was calculated by $R = 2(S/\pi)^{1/2}$, where R and S represent the equivalent diameter and the area of each single-crystal film, respectively. Interestingly, almost all of single-crystal films grown at point A were large (the diameter was larger than 0.30 mm) and thin (the thickness was less than 130 nm). While, those grown at points B and point C were small (the diameter was around 0.050 mm) and thick (the thickness ranged from 40 to 460 nm).

2.3. Growth Mechanism

Figure 3 summarizes the crystal analysis results of BEPant single-crystal films obtained (a) at point A (250 $^{\circ}\text{C}$) and (b) at point B (210 $^{\circ}\text{C}$) in Figure 2c, respectively. We show the optical microscopy images of the single-crystal film, the schematic 3D view, and the schematic in-plane growth rates of each facet, $\{001\}$, $\{011\}$, and $\{010\}$. At the growth temperature of 250 $^{\circ}\text{C}$, growth rates of all facet directions become similar. Therefore, the shape of the crystal resembles a circle. We calculated the growth rate of out-of-plane and in-plane directions by using the thickness of 50 nm, the diameter of 1.0 mm, and the growth time of 50 min. The results were 1.0 nm min⁻¹ and 10 $\mu\text{m min}^{-1}$ for out-of-plane and in-plane direction, respectively. It should be noted that the ratio of the growth rates is about 1/10 000, that is, almost all of the supplied BEPant molecules were expended to expand the (100) plane laterally. While, at the growth temperature of 210 $^{\circ}\text{C}$, the crystal shape changes from 2D to 3D. In the same way, we calculated the growth rates of out-of-plane and in-plane directions by using the thickness of 400 nm, the diameter of 0.10 mm, and the growth time of 50 min. The results were 8 nm min⁻¹ and 1.0 $\mu\text{m min}^{-1}$ for out-of-plane and in-plane direction, respectively, and the ratio of the growth rates was 1/125. Interestingly, the in-plane growth

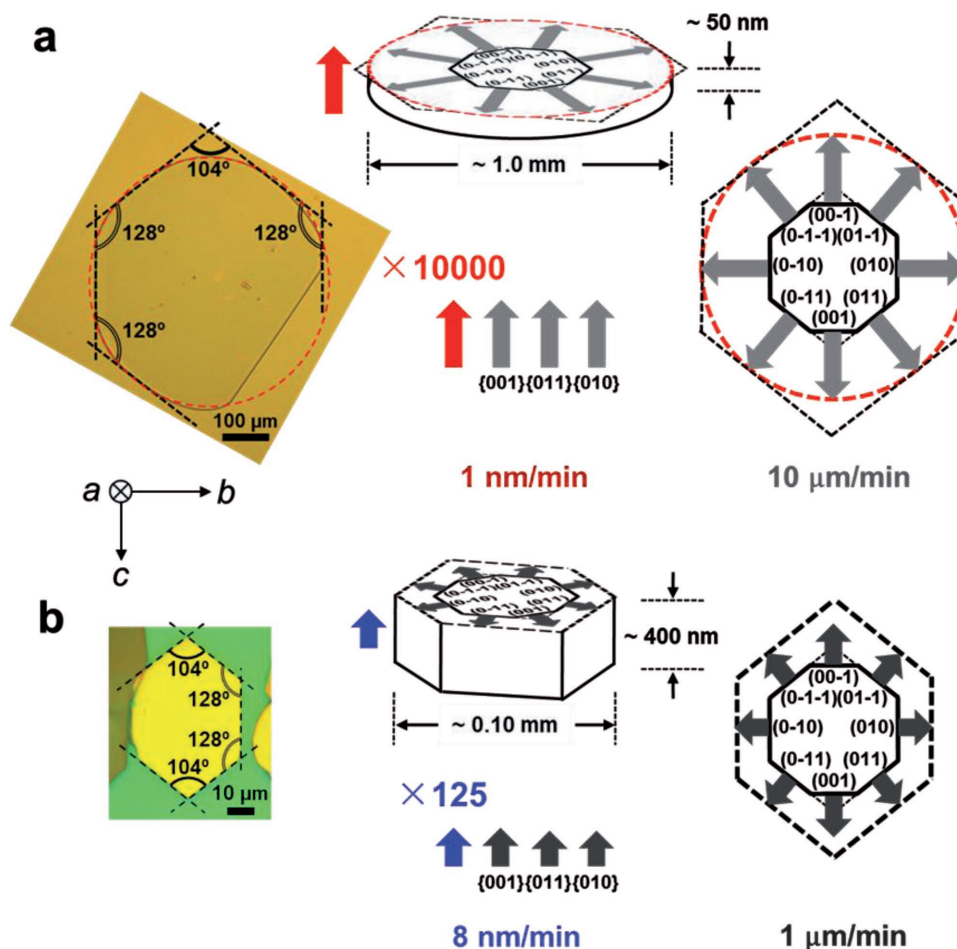


Figure 3. Crystal analysis results of BEPant single-crystal films a) at point A and b) at point B in Figure 2c. The optical microscopy images of the single-crystal film, the schematic bird's eye view, and the schematic growth rates of each facet, $\{001\}$, $\{011\}$, and $\{010\}$ are shown.

rate became ten times faster while the out-of-plane growth rate reduced to about 1/8 as the growth temperature increased from 210 to 250 °C.

We could obtain BEPAnt single-crystal films with lateral dimensions of millimeters and a thickness of only tens of nanometers at the growth temperature of 250 °C. There are two main reasons. One is that the BEPAnt molecule has a long migration distance (260–450 μm at 250 °C) on the OTS-modified SiO_2 surface. The other is that the BEPAnt molecule also has a long migration distance on the (100) surface, and uptake rate of BEPAnt molecules for each facet drastically increases as the growth temperature increases. Generally, in order to increase the migration distance on the surface, we can use a condition favoring a lower growth rate. In practice, we would need to decrease the N_2 flow rate. However, since lowering the flow rate changes the absolute amount of molecules in the vapor formed in the PVT system, we will have to obtain a different saturated-supersaturated vapor pressure curve experimentally.

2.4. Isotropic Hole Mobility of BEPAnt SC-OFET

As described above, we could grow large and thin single-crystal films of BEPAnt in a reproducible manner. Subsequently, we tried to investigate the dependence of the mobility on the crystal orientation by fabricating SC-OFETs. **Figure 4a** shows the optical microscopy images of the fabricated device 1 with a BEPAnt thickness of 60 nm. According to two specific angles, we could determine the crystal orientation. We obtained 12 SC-OFETs with a top-contact and bottom-gate configuration. However, some of them were broken during measurement and we finally obtained eight SC-OFET results. **Figure 4b** shows the measured mobilities from different electrodes. The mobility

values were calculated from the transfer curves in the saturated region at a source–drain voltage (V_{sd}) of -60 V. It should be noted that the mobility clearly showed the isotropic nature, and the average mobility value was around $4.3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. We fabricated device 2 with the thinnest BEPAnt which had a thickness of 40 nm. **Figure 4c,d** shows the optical microscopy image of the fabricated device 2, and the characteristics of SC-OFET2 (transfer curve and output curve). The calculated mobility value was $7.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in the saturated region at $V_{\text{sd}} = -60$ V.

Since the mobility values are strongly affected by the thickness,^[22] we investigated the dependence of mobility on the thickness for the fabricated BEPAnt devices. **Figure S2** (Supporting Information) shows the results from a schematic SC-OFET structure in the inset with the top-contact and bottom-gate configuration. For this type of OFET, the total resistivity (R) is expressed by Equation (1)

$$R = 2R_c + 2R_s + R_{\text{ch}} \quad (1)$$

Here, R_c , R_s , and R_{ch} are the contact resistance, the series resistance, and the channel resistance, respectively. When the single-crystal film is very thin ($R_c \gg R_s$), we only consider the contact resistance and channel resistance. However, when the thickness increases and R_s is not negligible ($R_c \leq R_s$), the mobility decreases in inverse proportion to $1/R$. Therefore, the behavior of mobility versus thickness as seen in **Figure S2** (Supporting Information) seems to be correct. For BEPAnt SC-FETs, the mobility starts to decrease with the thickness around 100 nm. Based on this model, the mobility values for the devices with a thickness of 60 and 80 nm are a little bit small. We identified poor contact characteristics in the output curves, which may be a cause for the lower mobility values. As seen in **Figure S2** (Supporting Information), the mobility values for the devices with a thickness of 145 nm are very small. However, the mobility

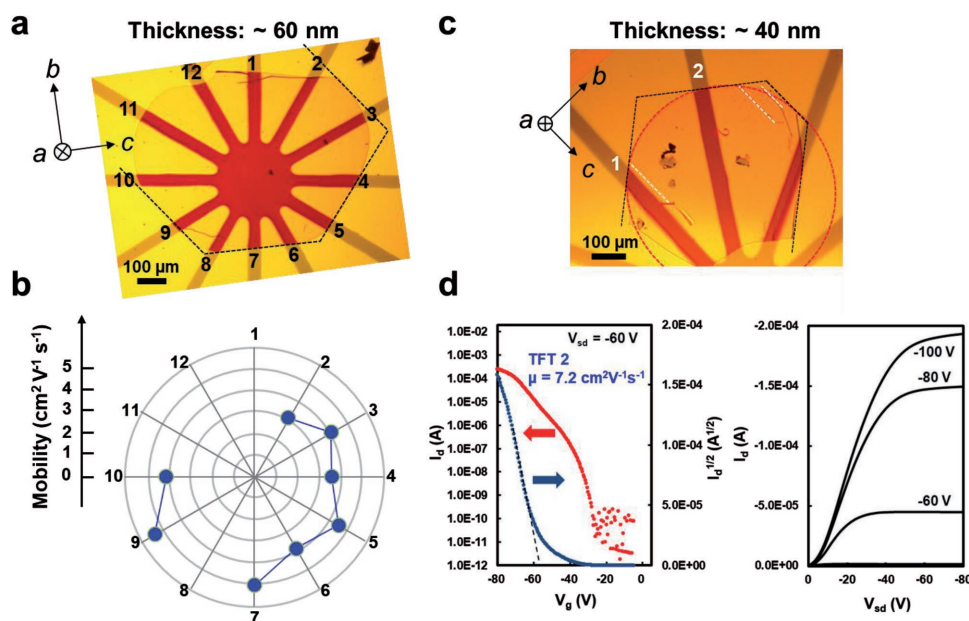


Figure 4. SC-OFET characteristics for BEPAnt single-crystal films with different thickness. a) Optical microscopy image of device 1. The thickness was about 60 nm. b) Isotropic transfer characteristics. c) Optical microscopy image of device 2. The thickness was about 40 nm. d) Transfer curve at $V_{\text{sd}} = -60$ V and output curve for SC-OFET2.

was almost perfectly isotropic, and the contact characteristics were excellent as indicated in the output curve. Figure S3 (Supporting Information) shows the dependence of mobility on the crystal orientation for this device calculated in the saturated region at V_{sd} of -100 V. Unfortunately, we used a very old OTS-modified substrate which seems to produce very low mobility values. However, the isotropic transfer characteristics were certainly achieved and the highest mobility of $7.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ was obtained for the sample with a thickness of 40 nm .

According to the well-known Marcus theory, hopping mobility (μ) is proportional to the forth power of the transfer integral and the square of the distance between neighboring molecules. Here, we calculated the ratio of $P/T1$ ($= T2$) using our materials (Figure 1b)

$$\mu_P/\mu_{T1(T2)} = (23.3/79.4)^4 \times (6.04 \times 10^{-8}/4.90 \times 10^{-8})^2 \times 100 = 1.12 (\%) \quad (2)$$

This result indicates that the mobility along the c -direction can be negligible compared with that along the $(b \pm c)$ -directions, suggesting that this material had the isotropic carrier transfer characteristics. Previously, we investigated a potentially promising organic material, 2,7-di-*tert*-butyl[1]-benzothieno[3,2-*b*]benzothiophene (ditBu-BTBT), for obtaining isotropic transfer characteristics. As a reference point, ditBu-BTBT gave a transfer integral value of P and $T1$ ($= T2$) of 60 and 53 meV , respectively,^[22] and a ratio of

$$\mu_P/\mu_{T1(T2)} = (60/53)^4 \times (6.04 \times 10^{-8}/6.08 \times 10^{-8})^2 \times 100 = 61.7 (\%) \quad (3)$$

For ditBu-BTBT, the difference between the mobility along each direction was relatively small. Therefore, there seems to be an organic material with two types of ratios of $\mu_P/\mu_{T1(T2)}$ that shows isotropic transfer characteristics. This finding becomes one of the reference indexes that are critical to determine the isotropy of any new synthetic organic compounds with isotropic transfer characteristics.

3. Conclusion

We have demonstrated 2D and 3D crystal formation of BEPAnt by PVT. We could change the shape of single-crystal films from about 1.0 mm in diameter and 50 nm in thickness to 0.10 mm in diameter and 400 nm in thickness by changing the growth conditions using the saturation-supersaturation vapor pressure curves. The in-plane growth rate became ten times faster while the out-of-plane growth rate reduced to about $1/8$ as the growth temperature increased from 210 to 250°C . By using these large and thin single-crystal films, the isotropic transfer characteristics and the highest mobility of $7.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ were achieved for the sample with a thickness of 40 nm . We expect that this material will be very useful to fabricate OLETs and OLET-structure-based OLDs because we do not need to think about the electrode direction. Furthermore, using an appropriate organic single-crystal material, single-crystal heterostructures and p-n junction structures are expected to be grown, enabling high-quality and high-functionality electronic and optoelectronic devices to be fabricated.

4. Experimental Section

Substrate Treatment: Heavily doped n-type silicon wafers with 250 nm thermally grown SiO_2 were cleaned in an ultrasonic bath with acetone, deionized water, and isopropanol in sequence, then dried by nitrogen gas and treated with UV-ozone plasma for 15 min . They were transferred into a nitrogen glovebox and dipped into solution of OTS (0.5 g , 0.025 g mL^{-1}) in toluene (20 mL) at 60°C for 20 min to form a self-assembled monolayer (SAM). The OTS-modified SiO_2/Si substrates were rinsed with fresh toluene for three times and dried by nitrogen gas. After treatment, these substrates were kept in the nitrogen glovebox.

Crystal Growth: The single crystals were grown by the PVT with three temperature control zones at atmospheric pressure in high-purity N_2 at 600 sccm . BEPAnt was synthesized by laboratory, and the synthesized procedure was described in elsewhere.^[12] The raw BEPAnt was purified three times by another sublimation furnace before single-crystal growth.

Crystal Analysis: The single-crystal data of BEPAnt were collected using Rigaku R-Axis RAPID diffractometer with a graphite monochromated $\text{Cu K}\alpha$ radiation ($\lambda = 1.54187 \text{ \AA}$) by ω scans at room temperature. The crystal structure was solved by a direct method and refined with SHELX-97 software package.

A Bruker D8 Advance diffractometer was used to obtain in-plane diffraction patterns from thin films of the single crystals grown on OTS-modified SiO_2/Si substrate. 2θ scan was carried out from 2° to 50° using $\text{Cu K}\alpha$ radiation ($1.5418 \text{ \AA}$) at a power of 40 kV and 40 mA , with slit width of 1.000 mm , a step size of 0.02° and a step time of 0.1 s . The thickness of crystals was measured by Bruker DEKTAK XT Profilometer.

Device Fabrication and Characterization: Top-contact bottom-gate SC-OFTs were fabricated by using the single crystals grown on OTS-modified SiO_2/Si substrate. The n -Si layer was used as the gate electrode, and the SiO_2 layer served as the gate dielectric. After lamination of shadow mask, gold source and drain electrodes ($\approx 50 \text{ nm}$) were deposited at $0.3\text{--}3 \text{ \AA s}^{-1}$ under a pressure of $4 \times 10^{-4} \text{ Pa}$ in the vacuum deposition chamber. The fun-shaped shadow masks with channel length of about $50 \text{ }\mu\text{m}$ were used in this work. The morphology of single crystals was inspected under a polarized optical microscope (Shanghai BIMU Company XP-213) and the accurate size of channels was measured in the microscopic photograph. The channel width (W) is decided by the average value of the widths of single-crystal thin film bordering upon the source/drain electrodes. Transfer ($I_d\text{--}V_g$) and output ($I_d\text{--}V_d$) characteristics of BEPAnt SC-OFTs were measured by Keysight B2500A combined probe station in ambient air. All devices were tested in saturation regimes and the key parameters, such as field-effect mobility, threshold voltage, and I_{on}/I_{off} ratio, were extracted using the following equation

$$I_d = \frac{W}{2L} C_i \mu (V_g - V_{th})^2 \quad (4)$$

where I_d , W , L , C_i , V_g , and V_{th} represent the drain-source current, channel width, channel length, the capacitance per unit area of the gate dielectric layer, gate voltage, and threshold voltage, respectively. The field-effect mobility was determined from the slope of plots of $(I_d)^{1/2}$ versus V_g .

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D crystals, 3D crystals, isotropic charge-carrier mobility, single-crystal organic field-effect transistor (SC-OFET)

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