



# High performance of rubrene thin film transistor by weak epitaxy growth method



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## ABSTRACT

Rubrene single-crystal transistors have achieved one of the highest carrier mobilities in organic semiconductors. However its thin film transistor usually shows inferior performance due to the poor film quality. Therefore how to obtain large-area and high quality rubrene thin film has become a prominent challenge. This work utilized weak epitaxy growth method with new inducing layer 1,3-di(terphenyl) benzene (m-7P), and larger-area highly ordered terrace rubrene film was obtained. Based on this high quality film, the hole mobility of rubrene polycrystalline thin film transistor has been enhanced to  $11.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  with VOPc as buffer layer between semiconductor layer and electrodes. This high device performance was attributed to the flat inducing layer and the single orientation of rubrene domains on m-7P layer, which may reduce grain boundaries and improve the film quality. This easy process to prepare large-area high performance rubrene device supplies a good opportunity for large-area electronic device manufacture.

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## 1. Introduction

In the past few decades, organic semiconductors have received considerable attentions because of their advantages such as easy preparation process for large-area, low-cost manufacture, which make them promising to fabricate flexible, low-cost and lightweight electronic components [1–6]. Recently, organic thin film devices showed rapid development, which injected more vitality for their application due to their much better device performance than amorphous silicon [7]. To achieve better device performance, the improvement of organic film quality usually plays a vital role [8–10]. Generally, different substrates were used to grow active materials aiming at the

improvement of film morphology and electronic structure of interface, and the increase of mobility in transistors is usually observed [11–14].

Rubrene (5,6,11,12-tetraphenylnaphthacene), a tetraphenyl derivative of tetracene as a typical organic semiconductor has been investigated since the 1920s [15]. In recent few years the carrier mobility of its single-crystal transistor has reached to about  $40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  which is one of the highest values in organic semiconductors [16]. Additionally, the anisotropy of carrier mobility in the rubrene single-crystal transistor has also been reported that  $\sim 20 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and  $\sim 8 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for carrier transport along *b* axis and *a* axis, respectively [17]. This excellent transistor performance has made it become the most promising candidate material for fabricating thin film devices. However the vacuum-sublimated and solution-processed rubrene thin film transistors' performances were

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not up to expectation since their carrier mobilities were usually several orders of magnitude lower than the value of single-crystal device [18,19]. This diversity usually originates from the poor thin film quality such as amorphous spherulites and nanowires formed on the SiO<sub>2</sub> substrate [20]. Therefore, how to obtain large-area and high quality rubrene thin film has become a prominent challenge. Many research groups have focused on this motif through improving thin film preparing process including decorating substrate, annealing process and epitaxy growth to ameliorate thin film quality then to further enhance device performance [21–23]. The optimal mobility has achieved  $\sim 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for rubrene crystal film grown on h-BN crystals [24]. However, the single-crystal substrate may still be their shortcoming for further applications.

Recently, weak epitaxy growth (WEG) method has been proved to be an efficient way to obtain high quality rubrene films with preferable device performance [25]. This easy process for the preparation of large-area and high-quality rubrene films supplied a good opportunity to obtain high performance rubrene devices. However, in the *para*-sexiphenyl (*p*-6P)/rubrene system, thick *p*-6P film was needed to epitaxially grow rubrene polycrystalline thin film, due to its crystalline phase, not the thin-film phase [26,27]. This thick *p*-6P film caused a large surface roughness, which was infaust for the carrier to transport in the rubrene film adjacent to the *p*-6P layer. Consequently, the inducing layer with crystalline phase and two dimensional (2D) layer-by-layer growth mode would be more suitable to epitaxially grow rubrene thin film. While bend shape molecule is a good candidate according to our previous experiences, like 2,5-bis(4-1,1':4,1''-terphenyl)-thiophene (3PT) which has been proved to have characteristics of crystal and 2D layer-by-layer growth mode for monolayer of 3PT, but it also showed fissures [28]. Here, we selected a new bend-shape molecule 1,3-di(terphenyl) benzene (*m*-7P) as the inducing layer in WEG process. This *m*-7P can form continuous flat monolayer to epitaxially grow rubrene film. By the optimal conditions of thin film growth, larger-area highly ordered terrace rubrene film was obtained, and the transistor performance based on this high quality rubrene film has been further enhanced achieving a higher carrier mobility for the polycrystalline rubrene films. Compared with the rubrene film grown on SiO<sub>2</sub> substrate, both the film crystallinity and device mobility showed dramatic improvement.

## 2. Experimental section

### 2.1. Synthesis of 1,3-di(terphenyl) benzene (*m*-7P)

The detailed synthetic procedures can be found in the [Supporting Information](#).

### 2.2. Fabrication of organic thin films and OTFT

The rubrene material was purchased from Aldrich Company (USA) which was purified twice via thermal gradient sublimation prior to sample preparation. A heavily doped *n*-type silicon wafer with a 300 nm thermal oxidation

SiO<sub>2</sub> layer was used as the substrate. Then *m*-7P monolayer as the inducing layer in WEG process was deposited at 175 °C in a vacuum chamber when the air pressure down to  $10^{-4}$  Pa. Subsequently 20 nm rubrene film was deposited on *m*-7P monolayer at 96 °C, and then 10 nm VOPc was deposited at the same temperature as buffer layer on the rubrene film. Also 20 nm rubrene film was directly deposited on SiO<sub>2</sub> substrate at 96 °C with the pressure down to  $10^{-4}$  Pa. After depositing the organic semiconductor films, the sample was quickly transferred to another vacuum chamber to evaporate 50 nm Au with the pressure about  $10^{-4}$  Pa as source and drain electrodes with a mask shadow to complete the top contact transistor configuration. The width and length of the channels were 3000 and 100  $\mu\text{m}$  respectively, and the unit capacitance of the insulator SiO<sub>2</sub> was  $10 \text{ nF cm}^{-2}$ .

### 2.3. Characterization of organic films and OTFT

The AFM images were obtained by an SPI3800N (Seiko Instruments Inc.) with tapping mode. The out-of-plane X-ray diffraction pattern of thin film was taken from D8 Discover thin film diffractometer with Cu K $\alpha$  radiation ( $\lambda = 1.54056 \text{ \AA}$ ). And the in-plane GIXRD diffraction pattern of thin film was obtained at Shanghai Synchrotron Radiation Facility (SSRF) on beamline BL14B1 with  $\lambda = 1.24 \text{ \AA}$ . The selected area electron diffraction (SAED) was imaged with a JEOL JEM-1011 transmission electron microscope operated at 100 kV. For SAED measurement, organic films were deposited on the SiO<sub>2</sub>/Si substrate first; subsequently, a carbon thin film used as support layer was deposited on the films. And then the films were separated from the SiO<sub>2</sub> surface by floating from 10% HF solution and then were transferred to a copper grid for measurement. Au was directly deposited on the copper grid for demarcating if necessary. Dark field was used for experiments to provide weaker-intensity beam and high contrast. All current-voltage tests of transistors were performed via two Keithley 236 source-measurement units under ambient circumstance at room temperature. The field-effect mobilities were extracted from the saturated region of the transfer characteristic curve.

## 3. Results and discussion

### 3.1. Film topography and structure

Fig. 1a shows the molecular structure of *m*-7P and the AFM image of *m*-7P monolayer topography deposited on SiO<sub>2</sub> substrate at 175 °C. This *m*-7P film has formed compact islands and adjacent islands coalesced well covering SiO<sub>2</sub> substrate with little second layer islands showing up. This indicates *m*-7P has presented layer-by-layer growth behavior which could be appropriate to be the inducing layer in WEG process. It can supply a flat substrate for subsequent organic semiconductor growth. We further investigated the crystalline structure by X-ray diffraction measurement, and the out-of-plane X-ray diffraction pattern of serial *m*-7P films including monolayer,

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