

Letter

High-performance organic transistors based on solution-processed rubrene crystals directly grown on a polymeric dielectric

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ABSTRACT

High-quality rubrene crystals were directly grown on a polymeric gate dielectric-coated ITO/glass substrate by a simple solution process. Organic field-effect transistors were fabricated based on these rubrene crystals, and their electrical performance was investigated. Atomic force micrographs of the rubrene crystal reveal that its surface has a conventional terrace structure, confirming the well-ordered rubrene molecules. The organic transistor with the rubrene crystal shows the highest charge carrier hole mobility of $3.74 \text{ cm}^2/\text{V}\cdot\text{s}$ that can be attributed to the strong π - π overlap between the adjacent rubrene molecules. The device shows a stable static and dynamic electrical stress response under ambient conditions. For evaluating the applicability of the transistor in a logic circuit, a simple load-type inverter is fabricated by combining the rubrene transistor and a $10 \text{ M}\Omega$ load-resistor, to achieve a clear dynamic switching response up to 100 Hz .

1. Introduction

Organic semiconducting materials, including both conjugated polymers and small molecules, have been widely exploited for organic electronics such as organic field-effect transistors (OFETs) [1–6], organic photovoltaics [7–10], and organic light-emitting diodes [11,12] because of various advantages such as their low-cost, low-temperature processability, facile molecular structure engineering, and mechanical flexibility [13–15]. Although significant progress has been made in the development of polymer semiconductors recently owing to their applicability in large-area electronic devices, semiconductors based on small molecules present several advantages over their polymeric counterparts, such as well-defined structures for straightforward chemical structure engineering, easy purification, and good reproducibility (small batch-to-batch variation), leading to stable device performance. Therefore, efforts for the development of organic electronic devices, which exploit small molecules as an active layer, are essential. Among a variety of organic small-molecule semiconductors, rubrene (5, 6, 11, 12-tetraphenylanthracene, Fig. 1(a)), a tetraphenyl derivative of tetracene, is one of the most attractive for organic electronic devices. High intrinsic field-induced mobility up to $20 \text{ cm}^2/\text{V}\cdot\text{s}$ has been achieved for the “air-gap” transistor based on ultra-pure single crystalline rubrene prepared from the vapor phase [16,17]. The high mobility of rubrene is

attributed to the π - π stacking of the tetracene backbone of the adjacent molecules in the crystal structure, which facilitate facile charge carrier transport between the rubrene backbone [16,18].

Despite the high operational performance of the rubrene-based transistors, several challenges in terms of the fabrication process persist. The high-quality rubrene single crystal enabling facial π - π stacking is usually formed by the physical vapor transporting (PVT) technique, which requires expensive equipment [17,19–22]. In addition, the PVT method requires sophisticated control and a relatively high temperature ($> 200 \text{ }^\circ\text{C}$), which limit the use of conventional organic flexible substrates and gate dielectrics. At such high temperatures, direct crystal growth over organic substrates and dielectrics is challenging, and this limits the implementation of this technique for realizing all-organic transistors.

To overcome these disadvantages, several studies have been reported. A rubrene thin film prepared by thermal evaporation was reported by Lee et al. To increase its crystallinity in the thin film, an “abrupt heating method” was developed, and the fabricated OFETs showed a field-effect mobility of $1.21 \text{ cm}^2/\text{V}\cdot\text{s}$ [23]. Fusilli et al. recently discovered that an additional organic underlayer can improve the crystallinity of rubrene thin films via post-deposition annealing [24]. Furthermore, a polymer-assisted solution process had been reported, where the polymer was used as a binder to form a highly

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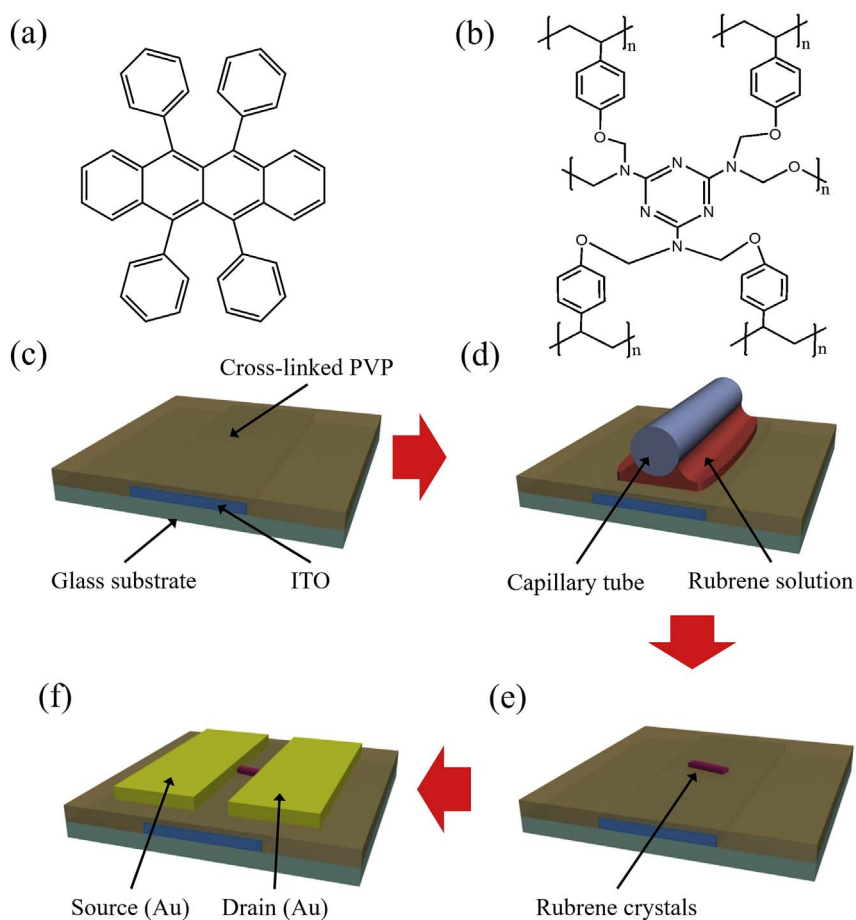


Fig. 1. Molecular structure of (a) rubrene and (b) cross-linked poly(4-vinylphenol). Illustration of fabrication of the device based on rubrene crystals: (c) Cross-linked PVP thin film was formed on an ITO-patterned substrate, (d) dropping a few mL of the rubrene solution onto the capillary tube placed on the cross-linked PVP film, (e) the rubrene crystal is formed after the complete evaporation of the solvent, and (f) device fabrication is finalized by the deposition of gold as source/drain electrodes. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

crystalline rubrene polymorph [25,26]. Methods based on vitreous solution-processed rubrene hypereutectics [27] and epitaxial growth [28] have been also developed for the fabrication of rubrene-based OFETs. However, the aforementioned methods still require rigorous conditions and complicated processes to form high-quality rubrene films or crystals. As rubrene is highly soluble in typical organic solvents such as chloroform, toluene, and *o*-dichlorobenzene, a simple solution-based process could be an effective alternative for the facile and low-cost growth of rubrene crystals. However, until now, reports of OFETs based on rubrene crystals grown through a simple solution process are rare. In particular, ‘direct growth’ of the rubrene crystal over ‘polymeric gate dielectric’ through a solution process is rarely developed yet even though it is highly important for low-cost all organic electronics.

In this study, we report high-performance *p*-type OFETs based on rubrene crystals directly grown over substrates coated with polymeric gate dielectric, cross-linked poly(4-vinylphenol) (Fig. 1(b)) through a simple solution process. The device shows typical dominant hole carrier transport behavior. The highest charge carrier mobility of $3.74 \text{ cm}^2/\text{Vs}$ was obtained under ambient atmosphere, which is comparable to that of other high-performance *p*-type OFETs. We also report an inverter, composed of the rubrene OFET and a load resistor, which shows a distinct inversion of the input/output bias and a dynamic response up to 100 Hz. Moreover, the rubrene-based device fabricated by our simple solution process is highly stable over ~ 500 cycles of ON/OFF switching, and shows no significant degradation over 20 days under ambient conditions.

2. Experimental

For the device fabrication, indium tin oxide (ITO)-coated glass was used as the gate electrode/substrate; the ITO film was patterned by

conventional photolithography to reduce the gate leakage of the device by decreasing the overlap area between the gate and source/drain electrodes. The patterned ITO glass was cleaned using the standard cleaning process for electronic application: ultrasonic treatment in detergent, deionized water, acetone, and 2-propanol for 15 min, respectively. For the formation of the polymeric gate dielectric, a solution consisting of 10 wt% poly(4-vinylphenol) (PVP) ($M_w = 20,000 \text{ g/mol}$, Aldrich), 5 wt% poly(melamine-co-formaldehyde) ($M_n = 511 \text{ g/mol}$, Aldrich) as the cross-linking agent, and 85 wt% propylene glycol monomethyl ether acetate (PGMEA) as the solvent was stirred for at least 1 h at room temperature. This solution was spin-coated on the cleaned ITO glass, baked at 90°C for 1 min on a hotplate, followed by baking at 175°C for 1 h in a vacuum oven, inducing the formation of a very dense polymer chain structure by the chemical cross-linking between PVP and poly(melamine-co-formaldehyde). The expected chemical structure of the chemically cross-linked gate dielectric (capacitance (C_i) = 57.8 pF/mm^2 , thickness (t) = 500 nm) is shown in Fig. 1(b). For the direct growth of a rubrene crystal over the gate dielectric, we used a capillary tube-assisted solution process developed by our group [29]. The rubrene solution (0.01 wt%) in 1, 2, 4-trichlorobenzene (TCB) was dropped on a capillary tube placed on the gate dielectric layer and substrate with the capillary tube, and the solution was heated at 175°C on a hotplate for 60 min under ambient conditions to cause solvent evaporation from the edge of the solution droplet, resulting in the formation of the rubrene crystal. Finally, gold was thermally deposited on top of the crystal at an evaporation rate of 0.3 \AA/s under a high vacuum of 10^{-6} Torr to fabricate the source/drain electrodes. The entire device fabrication process is depicted schematically in Fig. 1(c)–1(f).

The rubrene crystal formed using our solution method was investigated by polarized optical microscopy (OLYMPUS BX51) and

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