



# Crystallization of sexiphenyl induced by polyurethane containing terphenyl groups affording high-mobility organic thin-film transistor



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## ABSTRACT

Novel polyurethane containing terphenyl groups were designed and synthesized as gate insulators to induce the crystallization of p-sexiphenyl(p-6P) for organic thin-film transistors (OTFTs). Different sizes and shapes of p-6P grains were measured by atomic force microscopy (AFM), and results showed that the large size of p-6P grain can improve the performance of OTFTs. About 900 nm thick films can be easily fabricated by spin-coating under ambient conditions, followed by curing at UV irradiation for 10 min. OTFTs with this film as gate insulator were found to have good processability, a high charge-carrier mobility of  $1.1 \text{ cm}^2/\text{Vs}$ , a threshold voltage of  $-25 \text{ V}$ , and an on/off current ratio  $>10^5$ . The result indicated that this material is a promising candidate for the exploration of devices using OTFTs.

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## 1. Introduction

Organic thin-film transistors (OTFTs) have received considerable interest in recent years because of their attractive characteristics, such as simple fabrication process, low cost, and mechanical flexibility [1–3]. The charge transport of OTFTs is confined to an extremely thin semiconductor region (about 2–6 monolayers, depending on the semiconductor) between the surface of the semiconductor/dielectric [4]. Recently many research results have indicated that the quality of the semiconductor/dielectric interface has a critical role in enhancing OTFT performance for a given semiconducting material [5–7]. To obtain high performance OTFTs, great efforts have been made to modify the dielectric surface and search for novel dielectric

materials [8–10]. Nunes et al. [14] have reported improved pentacene OTFT performance on various styrenic polymer dielectrics. Choongik et al. [4d] have reported on pentacene films with varying thicknesses, grown in parallel with various untreated/polymer-coated  $\text{SiO}_2$  gate dielectrics, however, both rarely involved novel polymer materials as insulator instead of commercial polymer. Despite these important aforementioned findings, the kind of insulator surface is more suitable for the growth of semiconductors remains poorly understood.

In our previous research, a series of novel polyurethane containing terphenyl groups as side chain were synthesized by click chemistry, and their properties were characterized. The polyurethane film spin-coating on an ITO substrate, was directly crosslinked by UV irradiation and obtained the anti-solvent surfaces. Polyurethane with different contents of terphenyl groups were prepared to understand the influence of the structure of the insulator

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surface to the crystallization of sexiphenyl(p-6P) and the performance of OTFTs. The p-6P OTFTs fabricated on the ITO substrates with these new polyurethane films (i.e.,  $k = 3.9\text{--}4.1$ ), exhibited low layer thickness of about 900 nm, the on/off current ratio about  $10^5$  and high mobility of up to  $1.1\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ , which is higher than that of the most polymer insulators [11–13]. These results demonstrated that these polyurethane materials meet the important requirements for application as gate dielectrics in OTFTs.

## 2. Experimental section

The 2,2-bis(hydroxymethyl)butyl-3-phenylacrylate was synthesized according to a described method [8], and p-6P sample was synthesized according a described method [15] and the purified twice by thermal gradient sublimation prior to the experiments. The 4-biphenylboronic acid was purchased from Beijing OuHe Co., Ltd., 2-(4-bromophenyl)ethanol was purchased from Alfa Aesar Chemical Co., Propionic acid was purchased from Aladdin Chemistry and Bromotris(triphenylphosphine)Copper(I) was purchased from Aldrich Chemical Co. THF and  $\text{Et}_3\text{N}$  were purified by refluxing with and distilling from sodium to remove water and other impurities.

NMR spectra were measured on a Bruker AVANCE NMR spectrometer at a resonance frequency of 500 MHz for  $^1\text{H}$ . The chemical shifts of the  $^1\text{H}$  spectra were referenced to TMS at 0 ppm. Atomic force microscopy (AFM) observations of the film surfaces were conducted using an SPI3800 N (Seiko Instrument Inc.) with tapping mode under ambient conditions at room temperature. The capacitance of the photosensitive polymer was measured with an Agilent E4980A LCR meter. Current–voltage measurements were performed under ambient conditions at room temperature with two Keithley236 source measurement units.

The preparation of I is illustrated in Scheme 1. 4-Biphenylboronic acid (8.9136 g, 45 mmol), toluene (180 ml), 2 M  $\text{K}_2\text{CO}_3$  (45 ml) solution and ethanol (90 ml) were mixed in a 500 ml three-neck boiling flask. After the 4-Biphenylboronic was completely dissolved, 2-(4-bromophenyl)ethanol (7.302 g, 36.32 mmol) and

$\text{Pd}[\text{P}(\text{Ph})_3]_4$  were added in the mixture under  $\text{N}_2$  protection. The mixture was then stirred at  $90\text{ }^\circ\text{C}$  for 12 h. After cooling to room temperature (RT), the above solution was washed with water and ethanol and the white solid was obtained. To purify the crude product, it can be solved in THF and filtered, the solvent was removed under reduced pressure and the resulting solid product weighed 7.466 g (75%). Fig. 1 shows the  $^1\text{H}$  NMR (500 MHz, DMSO, TMS) of terphenyl ethanol.

Terphenyl ethanol (1.37 g, 5 mmol) and anhydrous THF (500 ml) were mixed in a 1 L three-neck boiling flask. After terphenyl ethanol was completely dissolved, DMAP (0.1222 g, 1 mmol) in 40 ml anhydrous THF and propionic acid (930  $\mu\text{l}$ , 15 mmol) were added, afterward, DCC (3.0932 g, 15 mmol) in anhydrous THF (100 ml) was added dropwise at RT under  $\text{N}_2$  protection. The mixture was stirred at the same temperature overnight. The resulting precipitate was removed and washed with THF and  $\text{CH}_2\text{Cl}_2$ . The filtrates were combined and the solvent was removed under reduced pressure, the resulting solid was purified by column chromatography over silica using ethyl

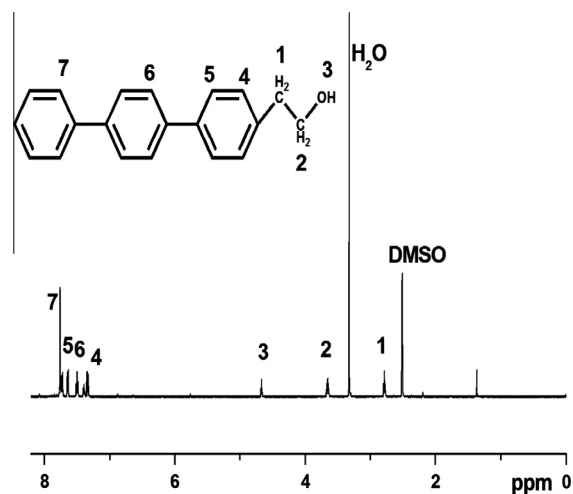
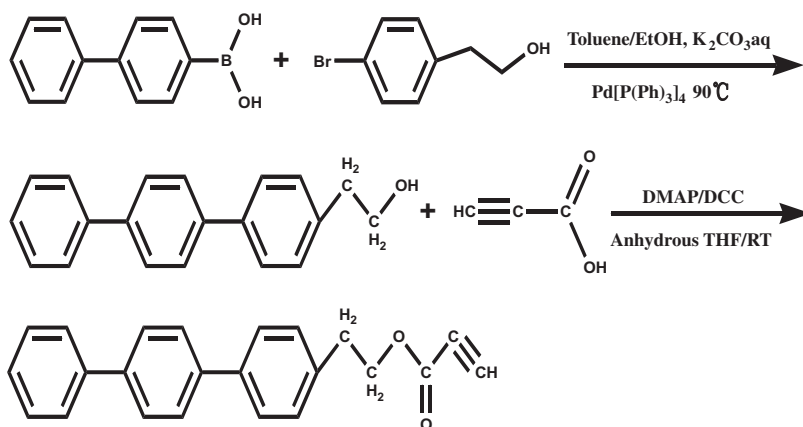


Fig. 1.  $^1\text{H}$  NMR spectrum of terphenyl ethanol.



Scheme 1. Synthesis of propionic acid terphenyl ethyl ether (I).

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