

## Logarithmic organic photodetectors



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### ABSTRACT

A novel photodetector of Al/GO:C8-BTBT/n-Si/Au with various graphene oxide (GO) contents has been investigated. The electrical properties of the diodes were characterized by current–voltage ( $I$ – $V$ ) and capacitance–voltage ( $C$ – $V$ ) measurements. The values of barrier height, ideality factor, and series resistance of the diodes were determined from  $I$ – $V$  characteristic curves by using Norde's equations. The photocurrent properties of the diode were studied under various illumination intensities. The photoconducting mechanism of the diodes is controlled by the traps. The photoresponse properties of the diodes are increased with GO contents. The obtained results indicate that graphene oxide doped 2,7-dioctyl[1]benzothieno[3,2-*b*][1]benzothiophene/n-Si heterojunctions can be used as a photodetector for optoelectronic applications.

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## 1. Introduction

Organic semiconductor-based optoelectronic devices have attracted a great deal of attention in the field of semiconductors and optoelectronics. These devices have several advantages such as low-cost, flexibility, and facile structure/device modification, and they can be fabricated on various plastic and inorganic substrates at room temperature by using solution processing techniques [1]. In addition, unique optical and electrical properties can be obtained with organic materials compared to inorganic-based semiconductors [2–13]. Recently, a series of  $\pi$ -extended heteroarene semiconductors including [1]benzothieno[3,2-*b*][1]benzothiophene (BTBT), dinaphtho[2,3-*b*:20,30-*f*]thieno[3,2-*b*]thiophene (DNNT), and dianthra-[2,3-*b*:20,30-*f*]thieno[3,2-*b*]thiophene (DATT) have been developed based on fused aromatic units, and charge carrier mobilities exceeding those of amorphous silicon (0.5–1 cm<sup>2</sup>/V s) have been achieved. [14]. Specifically, solution-processable p-type semiconductor, 2,7-dioctyl[1]benzothieno[3,2-*b*][1]benzothiophene (C8-BTBT) (Fig. 1), has shown impressive hole mobilities up to 25 cm<sup>2</sup>/V · s, which is one of the

highest results achieved in organic semiconductor materials [14]. Organic/inorganic multi-layered hybrid devices have recently attracted great attention, which has been facilitated by the availability of various organic semiconducting materials [11–14]. In these devices, organic/inorganic heterojunctions play key roles in controlling the device operation and performance, and the rational integration of high performance organic materials in these devices may significantly enhance their optoelectronic characteristics. Over the past decade, graphene oxide (GO) has attracted widespread attention because of its remarkable properties such as superior mechanical strength [15], excellent charge carrier mobility [16], high thermal conductivity [17], and large specific surface area [18,19].

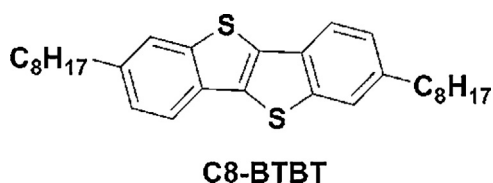
The motivation in the present work is to study photodiodes of Al/C8-BTBT/n-Si/Au based on a p-type small molecule semiconductor, [1] benzothieno[3,2-*b*][1]benzo-thiophene (C8-BTBT), and to investigate the effect of graphene oxide doping in Al/C8-BTBT:GO/n-Si/Au diode structure for low-cost, high-performance optical sensing applications.

## 2. Experimental details

All reagents were purchased from commercial sources and used without further purification unless otherwise noted. Conventional

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**Fig. 1.** Chemical structure of 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8-BTBT).

Schlenk techniques were used, and reactions were carried out under  $N_2$  unless otherwise noted. NMR spectra were recorded on a Bruker 400 spectrometer ( $^1H$ , 400 MHz and  $^{13}C$ , 100 MHz). Elemental analyses were performed by ERU-Technology Research and Application Center on LECO/TruSpec Micro-Elemental Analysis Instrument.

#### Synthesis and characterization

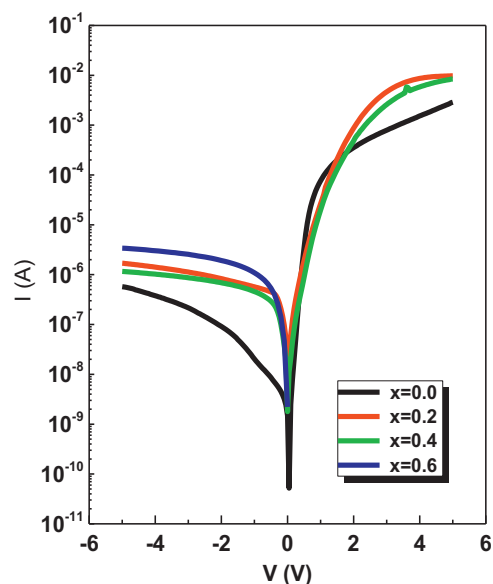
The synthetic route to C8-BTBT is shown in Scheme 1. The synthesis of Benzothieno[3,2-b][1]benzothiophene (BTBT, **1**) was performed according to the reported procedure [20].

#### Synthesis of ([1]benzothienopheno[3,2-b]benzothienophene-2,7-diyl) bis(octan-1-one) (**2**)

To a solution of [**1**] benzothieno[3,2-b][1]benzothiophene (1.0 g, 4.16 mmol) in 50 mL of anhydrous dichloromethane,  $AlCl_3$  (3.07 g, 23 mmol) was added at  $-10^\circ C$  under nitrogen. The resulting mixture was stirred at  $-10^\circ C$  for 30 min, and then octanoylchloride (3.41 g, 21 mmol) was added dropwise. The reaction mixture was stirred at  $-10^\circ C$  for 1 h, and then at room temperature for 2 days. The reaction mixture was poured into water to yield a white precipitate. The precipitate was collected by vacuum filtration, and washed with water and methanol, respectively, to give **2** (1.3 g, 64% yield) as a white crystalline solid. This compound was directly used in the next step without any further purification.  $^1H$  NMR  $\delta$  8.56 (d,  $J=1.3$  Hz, 2H), 8.06 (dd,  $J=8.0, 1.3$  Hz, 2H), 7.95 (d,  $J=8.0$  Hz, 2H), 3.07 (t,  $J=7.0$  Hz, 4H), 1.75–1.83 (m, 4H), 1.20–1.52 (m, 16H), 0.90 (t,  $J=7.2$  Hz, 6H).

#### Synthesis of 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8-BTBT)

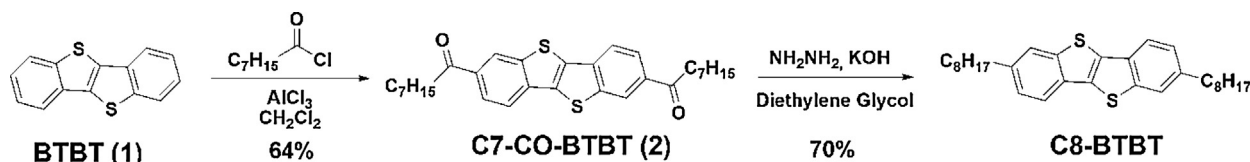
To a solution of ([1]benzothienopheno[3,2-b]benzothienophene-2,7-diyl) bis(octan-1-one) (**2**, 1.0 g, 2.03 mmol) and potassium hydroxide (626 mg, 11.2 mmol) in diethyleneglycol (50 mL) was added hydrazine hydrate (3.2 mL, 51.75 mmol) under nitrogen, and the resulting solution was heated to  $110^\circ C$  for 1 h and then further heated at  $210^\circ C$  for 5 h. After cooling to room temperature, a white precipitate formed, which was filtered and washed with water and methanol to yield a white crude solid. The



**Fig. 2.** Dark  $I$ - $V$  characteristics of Al/GO:C8-BTBT/n-Si/Au diode with for various  $x$  contents ( $x$ =GO:C8-BTBT)

crude compound was next chromatographed on silica, eluting with hexanes to yield C8-BTBT as a white crystalline solid (0.66 g, 70% yield).  $^1H$  NMR  $\delta$  7.76 (d,  $J=8.0$  Hz, 2H), 7.71 (d,  $J=1.0$  Hz, 2H), 7.27 (dd,  $J=8.0$  Hz, 1.0 Hz, 2H), 2.76 (t,  $J=8.0$  Hz, 4H), 1.71 (m, 4H), 1.26–1.36 (m, 20H), 0.89 (t,  $J=6.8$  Hz, 6H);  $^{13}C$  NMR  $\delta$  143.2, 140.5, 132.7, 131.1, 124.9, 122.9, 121.2, 36.2, 31.9, 32.0, 29.2, 29.1, 28.8, 22.5, 13.9; Anal. Calc. for  $C_{30}H_{40}S_2$ : C, 77.53; H, 8.67. Found: C, 77.20; H, 8.50.

Firstly, graphene oxide was prepared by Hummer's method. The C8-BTBT was dissolved in dichlorobenzene and GO was ultrasonically dispersed in chloroform for 2 h. The composites were prepared using C8-BTBT and GO solutions for various weight ratios of ( $x$ =GO:C8-BTBT) ( $x=0.0, 0.2, 0.4$  and  $0.6$ ). The C8-BTBT and GO solutions were coated onto surface of n-Si substrate using drop casting method. Before coating procedure, the silicon substrates were etched by HF and then rinsed in deionized water using an ultrasonic bath for 10–15 min. The silicon wafers were chemically cleaned with methanol and acetone baths, respectively. Top contact of the diodes was prepared by Au metal. For this, Au metal was evaporated by sputtering system in the form of circles giving a diode contact area of  $3.14 \times 10^{-2} \text{ cm}^2$ . Surface morphology of the films was investigated using a scanning electron microscopy (SEM). The current–voltage ( $I$ - $V$ ) characteristics of the diodes were performed using a KEITHLEY 4200 semiconductor characterization system (SCS). Photoresponse measurements were performed using a solar simulator and KEITHLEY 4200 SCS. The intensity of the illumination was measured using a solar power meter (TM-206).



**Scheme 1.** Synthetic route to C8-BTBT.

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