

# Ambipolar small molecular semiconductor-based heterojunction diode



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## ABSTRACT

A heterojunction diode based on an ambipolar organic semiconductor 2,8-bis(5-(2-octyldodecyl)thien-2-yl)indeno[1,2-*b*]fluorene-6,12-dione (2OD-TIFDKT) was fabricated on p-Si using a drop-casting technique. The current–voltage and capacitance–voltage characteristics of Al/2OD-TIFDKT/p-Si/Al devices with aluminized contacts were investigated under dark and 100 mW/cm<sup>2</sup> illumination intensity. The result is a novel interface-state controlled diode device that is shown to be rectifying. In the forward bias it has a current that depends on the illumination intensity at constant bias, showing potential application in low-power solar cell application. In the reverse bias, it has a response that depends on the illumination intensity regardless of the applied reverse bias. This suggests a potential use as a sensor in photoconductive applications. Between 0 and 0.7 V forward bias, the ideality factor, series resistance and barrier height average at 2.35, 67.6 k $\Omega$  and 0.842 eV, respectively, regardless of illumination.

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## 1. Introduction

Organic semiconductors consisting of  $\pi$ -conjugated small molecules ( $\pi$ -SM) have emerged over the past few decades as promising candidates for application in low-cost, flexible, and large area organic devices. Such devices include, but are not limited to organic light-emitting diodes (OLED), organic thin-film transistors (OTFT), organic photovoltaics (OPV), light-emitting transistors (OLET), sensors and large-area integrated circuits. These devices, because of their inherently low electron and hole mobilities, are not intended to compete with silicon-based technologies, but are desired to be lower-cost alternatives. At the same time, significant developments continue to boost organic device performance due to improved synthetic methods, optimization techniques and device engineering [1]. In contrast to larger  $\pi$ -conjugated macromolecular systems,  $\pi$ -SMs have the advantages of easier synthesis, better purity and yield, good solubility in common organic solvents and better thin-film crystallinity. These traits lead to good reliability and device fabrication reproducibility

[2]. One current design approach for semiconducting organic molecules is to attempt the realization of a good balance between solution processability, highest occupied molecular orbital (HOMO)/lowest unoccupied molecular orbital (LUMO) energies, optical band gaps and actual device packaging and metallization [1,2]. With the continuing rapid development of new semiconducting organic materials, the need to better understand and rationalize electronic structure, charge transport and organic molecule/metal interfaces become increasingly important [3]. With the emergence of organic semiconductors in optoelectronic devices, metal/organic interfaces were initially studied in the context of the established metal/semiconductor interface theory developed by Schottky-Mott for ideal metal/inorganic semiconductor interfaces. Recent studies have revealed that such interfaces are dominated by large dipole interactions that lead to significant Fermi-level pinning and manipulation barrier height [4]. In order to realize optimized high-performance devices, lowered charge-injection barrier height with minimized localized trap states at the organo-metal interface is needed. Ideally, the organic-metal interface should offer low charge injection barriers for Ohmic contact, which depends on the difference in energy between the metal work function and the HOMO level for hole injection or LUMO level for electron injection. This condition can be closely approached for organic-metal interfaces if there is direct contact

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between the metal and the  $\Pi$ -electrons in the conjugated small molecule. The metal atoms are evaporated and condensed onto the organic film, where diffusion of metal atoms up to several nanometers of the p organic surface can occur during deposition [5]. However, the exact nature of the interface relies on the deposition method, which controls the surface morphology. The work of Liu et al. [1] also suggests that engineering the contact surface with an ultra-thin, insulating layer before the deposition of  $\pi$ -SMs, the performance of the device could be enhanced substantially. Specifically, they studied the role of 0–6 nm thicknesses of  $\text{Si}_3\text{N}_4$  for two different organic compounds. Their findings suggest that the onset of various device behaviors—from rectifying to quasi-Ohmic to tunneling—could be controlled by tuning the insulator thickness, which is in agreement with the theory of Fermi-level pinning or de-pinning [6]. The distribution of the localized states close to the organic-metal interface was found to change due to the improved protection of the organic-film from physical interference during metallization. In addition, the improved modulation of the charge injection led to reduced barrier height. The use of  $\text{Si}_3\text{N}_4$  was motivated by the need to avoid oxygen during a thermally activated deposition process, which could lead to oxidation artifacts arising from the organic compound. Also, the wider band gap of the insulator is chosen to be far from the HOMO/LUMO energies thereby eliminating charge injection into these levels [1]. On the account of the band gap of  $\text{SiO}_2$ , ( $\sim 9$  eV), which is much higher than that of metal oxides and  $\text{Si}_3\text{N}_4$  ( $\sim 5.3$  eV) would be ideal [7]. However, the aforementioned need to avoid oxygen precludes it. Another currently held general principle of organic molecule design for practical applications is to attempt to achieve lower band gap and to enhance  $\pi$ -conjugation by engineering donor-acceptor molecular architectures while minimizing the torsional forces between the rings coupling the  $\pi$ -electron deficient acceptors and the  $\pi$ -electron rich donor moieties [8–11]. The role of substituents such as chlorine has also been investigated by Ryu et al. [12]. Their findings suggest that such substitutions may extend the application of organic semiconductors in optoelectronic devices by enhancing physicochemical and optoelectronic properties through the band gap and relative positions of HOMO/LUMO energies. This also has direct implications to the possible metallization contacts. Some effects of annealing on the thermal and electrical properties were

investigated by Wang et al. [13], who found that annealing small organic molecules at temperatures between  $24^\circ\text{C}$  and  $140^\circ\text{C}$  led to a gradual increase in the field effect mobility while shifting the threshold voltage from positive to negative. Some efforts have been spent by others to functionalize the organic-metal interface through work-function modification by employing higher ordering of self-assembled molecular mono-layers [14]. In this paper, we report on the synthesis and application of an organic small molecule based on an indeno[1,2-*b*]fluorene-6,12-dione-thiophene ladder-type ambipolar building block, which demonstrates good charge transport ability and device stability. The molecule is designed based on the repositioning the  $\beta$ -substituents to a molecular terminal referred to as  $\alpha$ ,  $\omega$ -disubstitution, which has improved the stereochemistry by enhancing the  $\pi$ -core planarity of the semiconductor backbone through reduced steric interactions. The resulting ambipolar organic compound is 2,8-bis(5-(2-octyldodecyl)thien-2-yl)indeno[1,2-*b*]fluorene-6,12-dione (2OD-TIFDKT), which has been recently reported as an ambipolar semiconductor in OTFTs with good charge transport characteristics ( $\mu_e = 0.12 \text{ cm}^2/\text{Vs}$ ,  $\mu_h = 0.02 \text{ cm}^2/\text{Vs}$ ). We present the results of the fabrication and measurements of a novel metalized rectifying 2OD-TIFDKT/p-Si junction diode, which has few precedents and has a potential use as in sensor applications.

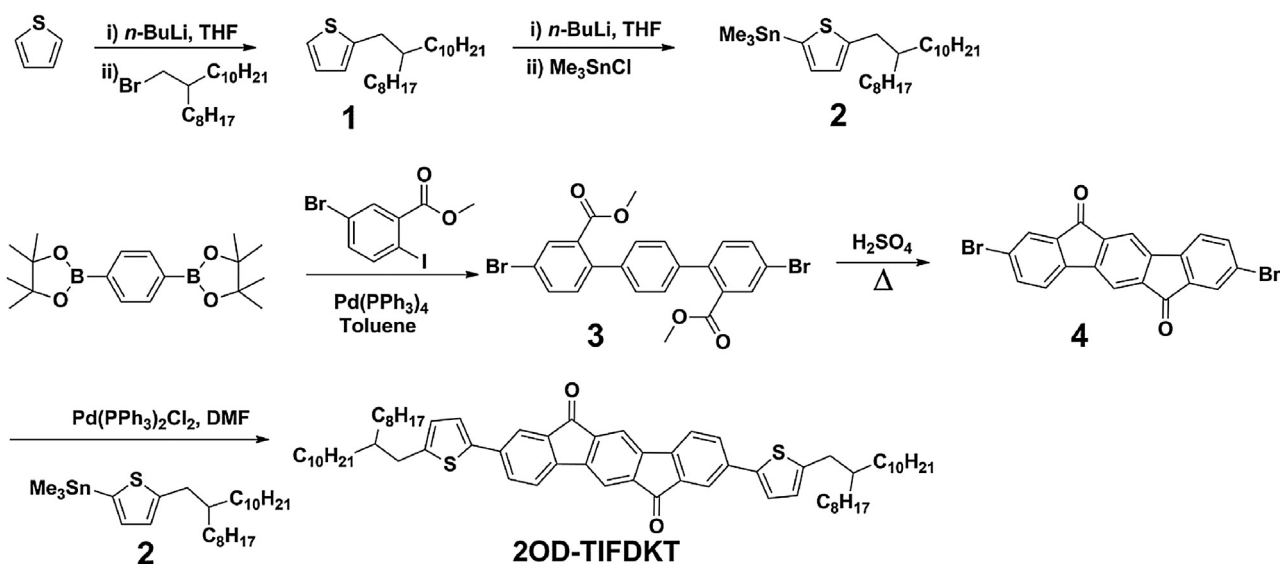
## 2. Experimental

### 2.1. Synthesis of ambipolar 2OD-TIFDKT

#### Scheme 1

### 2.2. Materials and methods

The reagents used in this synthetic work were acquired from commercial sources and used as received. Vacuum/nitrogen manifold was used for the reactions that will be carried out under  $\text{N}_2$  and for drying processes.  $^1\text{H}$  NMR (400 MHz) and  $^{13}\text{C}$  NMR (100 MHz) spectra were recorded on a Bruker 400 spectrometer. LecoTruspec Micro model instrument and Bruker Microflex LT MALDI-TOF-MS Instrument were used for elemental and MALDI-TOF analyses.



Scheme 1. Synthesis of 2,8-bis(5-(2-octyldodecyl)thien-2-yl)indeno[1,2-*b*]fluorene-6,12-dione (2OD-TIFDKT).

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