Performance improvement of organic phototransistors by using polystyrene microspheres

Bilei Zhou, Jiachen Zhou, Yantao Chen, Yingli Chu, Jia Huang and Xiaohan Wu*

ABSTRACT Organic phototransistors (OPTs) have been intensively studied in recent years due to the combined advantages of phototransistors and organic semiconductors (OSCs). However, the electrical performance of OPTs is largely limited by OSCs themselves, posing a challenge to further improve the performance of the devices. Preparing nano/micro-structures of OSCs is considered as an effective way to improve the performance of OPTs. Polystyrene (PS) microsphere, as a kind of insulating and low-cost material, is extensively used in fabricating nano/microporous structures, and the resulting devices exhibit high response to external stimuli. Therefore, we combined PS microspheres with OSCs to fabricate PS/OSC OPTs, and the $I_{\text{light}}/I_{\text{dark}}$ ratio was enhanced by two orders of magnitude compared with the pristine counterparts, which can be modulated from 46 to 1800 by controlling the diameters of PS microspheres. This strategy paves a way for developing high-performance OPTs with nano/microporous structures with potential applications in organic optoelectronics.

Keywords: phototransistor, organic semiconductor, polystyrene microsphere, nanostructure

INTRODUCTION

Organic phototransistors (OPTs) have been intensively studied in recent years owing to the combined advantages of phototransistors and organic semiconductors [1–7]. Phototransistors, as one type of the photodetectors, possess a gate and a dielectric layer for amplifying the electrical signals and improving the corresponding photosensitive parameters like responsivity and detectivity. They usually adopt field-effect transistor (FET) configuration, in which charge carrier density (or electrical conductivity) can be effectively modulated by the incident light intensity [8–14]. Organic semiconductors (OSCs), especially π -conjugated organic molecules have potential

in the next-generation flexible and printable electronics due to the appealing merits including low-temperature processability, simple fabrication, light-weight, mechanical flexibility, large-area coverage rate and low cost [15–17]. More importantly, some OSCs with suitable optical band-gaps can be fabricated into phototransistors for light detection ranging from ultraviolet (UV) to near infrared (NIR). It is therefore essential to study OSC-based phototransistors [2,18–23]. Developing the OSCs films with nano/micro-structures has attracted interest over the recent years, which is effective to improve the performances of the OPTs [24,25].

Polystyrene (PS) microspheres have been widely used in fabricating nano/microporous structures due to their low cost, high stability and insulation [26,27]. Qi and colleague [28] have prepared an inverse-opal two-dimensional organic-inorganic hybrid perovskite photonic crystal. And the unique structure has endowed the perovskite thin films with vivid colors and promising optical properties.

In our study, the performance of OPTs was significantly enhanced by nano/micro-structured OSC films using PS microspheres. Dinaphtho[2,3-b:2',3'f]thieno[3,2-b]thiophene (DNTT) and 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8BTBT) were selected because of their relatively high mobility and stability [29,30]. By incorporating PS microspheres, PS microspheres/DNTT-based phototransistors (PS/DNTT OPTs) enhanced photosensitivity (ratio of photo current to dark current, simplified as $I_{\text{light}}/I_{\text{dark}}$ ratio) by two orders of magnitude compared with DNTT OPTs. When changing the diameter of PS microspheres ($d = 0.5, 1$ and $10 \mu\text{m}$, respectively), we found that the PS/DNTT OPTs with the smallest diameter (0.5-PS/DNTT OPTs) exhibited the best optoelectronic performance with $I_{\text{light}}/I_{\text{dark}}$ ratio (1,800). The PS/DNTT OPTs also showed highly tunable

School of Materials Science and Engineering, Tongji University, Shanghai 201804, China

* Corresponding author (email: xiaohanwu@tongji.edu.cn)

photoresponsivity from 1 to 2700 A W⁻¹ by varying gate voltage (V_G) and incident light intensity. Meanwhile, PS film/DNTT-based OPTs (PS film/DNTT OPTs) were fabricated to verify our suggested mechanism. We have also fabricated PS microspheres/C8BTBT-based UV-OPTs (PS/C8BTBT OPTs), with a great enhancement of $I_{\text{light}}/I_{\text{dark}}$ ratio as compared with the C8BTBT-based phototransistors (C8BTBT OPTs). Our findings have provided an effective and convenient way to fabricate high performance organic phototransistors with nano/microporous structures, expanding the applications of OSCs in photosensors.

EXPERIMENTAL SECTION

Materials preparation

PS microspheres (2.5 wt% dispersed in water) with three different sizes (0.5 μm , 1 μm and 10 μm) were purchased from Alfa Aesar. PS powders with average molar mass (M_w) of 35,000 g mol⁻¹ was purchased from Aldrich. DNTT was synthesized in our lab according to the previous report, followed by vacuum sublimation for purification [29]. C8BTBT was purchased from Suna Tech. Inc. with further purification by vacuum sublimation.

Devices fabrication

The PS/OSC OPTs were fabricated on heavily doped n-type silicon wafers with a 300 nm SiO₂ layer. The Si wafers were treated to be hydrophilic by a plasma cleaner (Harrick Plasma, PDC-32G) for 5 min. The 0.5 μm and 1 μm PS microspheres dispersions were diluted to concentrations of 0.25 wt% and 0.5 wt% respectively, and the 10 μm PS microspheres dispersion kept its original concentration. Such concentrations were adjusted to form monolayer PS microspheres with the surface coverage of ca. 20%. The diluted PS microspheres dispersion was drop-coated on Si wafers. The excess water was removed by a vacuum freeze-dryer (Ningbo Scientz, 10N) for 2.5 h to obtain a uniform coating of PS microspheres. The PS film was deposited *via* a solution method. 50 mg mL⁻¹ chloroform (Acros, 99.8%) solution of PS powders was spin coated onto Si wafers at 3000 rpm for 60 s followed by thermal annealing at 60°C for 1 h. Subsequently, 50 nm DNTT layer was deposited on PS microspheres or PS film by a vacuum thermal evaporator at the rate of 0.2 Å s⁻¹. The temperature of substrate was controlled at 60°C during the whole evaporation process. Finally, a 50 nm gold (Au) layer was thermally evaporated as the source and drain electrodes with the channel length and width of 200 (or 5) μm and 6 (or 1) mm, respectively. The

PS/C8BTBT OPTs and C8BTBT OPTs have been fabricated by the same procedures.

Device characterization

The distribution of PS microspheres and the microstructure of DNTT films were measured by scanning electron microscope (SEM, Quanta 200, FEI) and atomic force microscope (AFM, SEIKO SPA-300 HV). The UV-vis absorption spectrum of DNTT was obtained by using Agilent Technologies, Cary 60. The coverage of PS microspheres was calculated by Photoshop. The *I*-*V* characteristics of the devices were measured by a Keithley 4200 semiconductor characterization system with a vacuum probe station. The photo-responsivity of the devices under different light intensities was estimated by using a xenon lamp light source with a double grating monochromator (Ommo330150, Beijing NBeT).

RESULTS AND DISCUSSION

Fig. 1a shows the molecular structure of DNTT and structural formula of PS microspheres. The PS/DNTT OPTs (Fig. 1b) adopted bottom gate top contact device FET architecture. SEM image of PS microspheres (10 μm) (Fig. 1d, left) demonstrated a random coating of PS microspheres on SiO₂/Si substrates. The SEM images of different PS microspheres distributions and the sectional image of monolayer PS microspheres (10 μm) are showed in Fig. S1, proving that PS microspheres were uniformly dispersed. DNTT was then thermally evaporated on the monolayer PS-coated SiO₂/Si substrates to form a discontinuous thin film. AFM was further employed to observe the specific microstructure of the DNTT film. As depicted in Fig. 1d (right), DNTT thin film presented island-like microstructure with obvious grain boundaries and average grain size of ca. 200 nm. It is worth noting that the DNTT surface coverage rate of all devices was controlled to ca. 80% with different diameters of PS microspheres (0.5, 1 and 10 μm , respectively). Fig. 1c shows the UV-vis absorption spectrum of DNTT, which demonstrated a characteristic absorption peak at 448 nm corresponding to the optical band gap of 2.55 eV. Thus, we applied white light to PS/DNTT OPTs and DNTT OPTs in the following test, ensuring that DNTT could be excited.

We evaluate the optoelectronic performance of PS/DNTT OPTs in dark and under white light illumination of 4.8 mW cm⁻². As shown in Fig. 2, the output and transfer curves of both 0.5-PS/DNTT OPTs and DNTT OPTs demonstrated a typical p-type behavior of OFETs.

Fig. 2a and b indicate that the output current of 0.5-PS/

Figure 1 (a) Molecular structure of DNTT and structural formula of PS microspheres. (b) Schematic structure of PS/OCSs OPTs (the scale on the diagram does not represent the actual size). (c) UV-vis absorption spectrum of DNTT. (d) SEM images of PS/DNTT thin film (left) and amplified AFM image of DNTT thin film (right).

Figure 2 I_D - V_D output curves of 0.5-PS/DNTT OPTs of (a) in dark and (b) under 4.8 mW cm⁻² light illumination. (c) I_D - V_G transfer curve of 0.5-PS/DNTT OPTs. I_D - V_D output curves of DNTT OPTs of (d) in dark and (e) under 4.8 mW cm⁻² light illumination. (f) I_D - V_G transfer curve of DNTT OPTs.

DNTT OPTs was increased significantly by light. By contrast, the DNTT OPTs presented a slight increase of output current at the same testing condition (see Fig. 2d, e). Fig. 2c, f depict transfer curves of two types of the OPTs, showing the high photosensitivity of 0.5-PS/DNTT OPTs compared with the DNTT OPTs. Although the output current of 0.5-PS/DNTT OPTs was lower than that of pristine counterparts, 0.5-PS/DNTT OPTs presented excellent optoelectronic performance.

The photosensing properties of PS/DNTT OPTs with different diameters of PS were further investigated. Fig. 3 exhibits the transfer curves (I_D - V_G) and their corresponding $I_{\text{light}}/I_{\text{dark}}$ ratios of PS/DNTT OPTs with different

diameters of PS at various light intensity. As depicted in Fig. 3a, 0.5-PS/DNTT OPTs exhibited outstanding photosensitivity under various light intensity. The $I_{\text{light}}/I_{\text{dark}}$ ratios (Fig. 3e) increased gradually with enhancing light illumination, and the maximum value reached 1,800 at 50 mW cm⁻². Similarly, for 1-PS/DNTT OPTs and 10-PS/DNTT OPTs, both the transfer curves show the photosensitivity and the maximum $I_{\text{light}}/I_{\text{dark}}$ ratios were up to be 1,000 and 350 (Fig. 3f, g) at 50 mW cm⁻², respectively. However, the DNTT OPTs (Fig. 3d, h) exhibited low photosensitivity with the maximum $I_{\text{light}}/I_{\text{dark}}$ ratio of about 46 under the same testing conditions. After integrating with PS microspheres, the maximum $I_{\text{light}}/I_{\text{dark}}$ ratio was increased by two orders of magnitude for 0.5-PS/DNTT OPTs and 1-PS/DNTT OPTs, as well as one order of magnitude for 10-PS/DNTT OPTs. Fig. 3i plots the maximum $I_{\text{light}}/I_{\text{dark}}$ ratios versus different diameters of PS spheres under different light intensity, where PS/DNTT OPTs presented much higher photosensitivity than DNTT OPTs, especially for the PS diameter of 0.5 μm. More importantly, the photosensitivity could be modulated from 46 to 1,800 by controlling the diameters of PS microspheres. The reduction of PS diameters and the enhancement of light intensity improved the output performance of PS/DNTT OPTs. To further trace the origin of the photosensing enhancement for PS/DNTT OPTs, we have removed the PS simply by adhesive tapes. The $I_{\text{light}}/I_{\text{dark}}$ ratio was almost comparable to that without removing the PS microspheres for 10-PS/DNTT OPTs (Fig. S2). The result indicates that the photosensing enhancement of PS/DNTT OPTs comes from the nano/

Figure 3 I_D - V_G transfer curves of PS/DNTT OPTs with different diameters of (a) 0.5 μm , (b) 1 μm , and (c) 10 μm . (d) I_D - V_G transfer curves of DNTT OPTs. $I_{\text{light}}/I_{\text{dark}}$ ratios as functions of V_G and light intensities of PS/DNTT OPTs with different diameters of (e) 0.5 μm , (f) 1 μm , and (g) 10 μm . (h) $I_{\text{light}}/I_{\text{dark}}$ ratios as functions of V_G and light intensities of DNTT OPTs. (i) The comparison of the maximum $I_{\text{light}}/I_{\text{dark}}$ ratios of different PS/DNTT OPTs and the DNTT-OPT under various light intensity.

microstructures of DNTT. The PS microspheres do not affect the discontinuity of the OSCs films because: 1) they are insulators and will cause discontinuous conducting channels and 2) the polar groups on the PS microspheres will introduce defects and traps in the OSC conducting channels. Therefore, the PS microspheres were retained in the DNTT film to simplify the device fabrication process.

Besides the photosensitivity, photoresponsivity (R) has also been investigated for our devices, which can be calculated by [31,32]:

$$R = \frac{|I_{\text{light}} - I_{\text{dark}}|}{P_{\text{light}} \times A}, \quad (1)$$

where P_{light} is the light intensity, A is the effective area of the devices. Fig. S3 is the dependence of R and light intensities with different diameters of PS microspheres. The PS/DNTT OPTs with the smallest diameters showed the highest R values, in agreement with the $I_{\text{light}}/I_{\text{dark}}$. Because R is inversely proportional to the channel length (L), a higher R value can be obtained with reducing the channel

length. Therefore, we have narrowed the channel length to 5 μm to further improve the photoresponsivity. Fig. 4a shows the transfer curves of the devices, demonstrating the high photosensitivity which was consistent with Fig. 3a. Fig. 4b shows the R is dependent on different light intensity and V_G . When illuminated at 0.17 mW cm^{-2} , the device showed R of 2,700 A W^{-1} . At the same time, the devices showed tunable photoresponsivity from 1 to 2,700 A W^{-1} by varying V_G and incident light intensities.

Detectivity (D^*) is another important parameter for phototransistors in unit of Jones ($\text{cm Hz}^{1/2} \text{W}^{-1}$), which can be expressed as [33–35]:

$$D^* = R \left(\frac{A}{2qI_{\text{dark}}} \right)^{1/2}, \quad (2)$$

where q is the elementary charge. Fig. 4c presents the detectivity of 0.5-PS/DNTT OPTs with V_G fixed at -60 V . The devices showed a D^* of 3×10^{12} Jones at 0.17 mW cm^{-2} , manifesting the high capability to detect weak light.

The photosensitivity enhancement of PS/DNTT OPTs

Figure 4 (a) I_D - V_D output curves of 0.5-PS/DNTT OPTs. (b) Responsivity of 0.5-PS/DNTT OPTs as functions of light intensity with the V_G fixed at $-60 V$ and $0 V$. (c) Detectivity of 0.5-PS/DNTT OPTs with the V_G fixed at $-60 V$. Channel length and width are $5 \mu m$ and $1 mm$, respectively.

Figure 5 I_D - V_G transfer curves in dark and at UV (365 nm) light (a) 0.5-PS/C8BTBT OPTs and (b) C8BTBT OPTs. (c) I_{light}/I_{dark} ratios as functions of V_G of 0.5-PS/C8BTBT OPTs and C8BTBT OPTs under 365 nm UV light illumination of $0.001 mW cm^{-2}$.

compared to DNTT OPTs is suggested as following reasons, which is different from the mechanism reported by Qi [28]. In their work, the improved photocurrent and photosensitivity mainly come from the increased light harvesting. In our work, the PS microsphere causes the discontinuous nano/microstructures and introduces defects and traps in the DNTT films, leading to decreased current for the PS/DNTT OPTs in dark. Upon light illumination, the photo-generated carriers in DNTT will compensate the trapped carriers and refill the DNTT conducting channel, contributing to the significantly increased photocurrent. Given the decreased dark current and the increased photocurrent at the same time, the PS/DNTT OPTs exhibit remarkably improved photosensitivity. The higher the light intensity is, the larger the number of the photo-generated carriers is, and the higher the drain current is. Furthermore, ca. 20% surface coverage of all the PS microspheres with different diameters were fabricated by controlling the concentrations of the PS microspheres dispersions. Thus, the PS microsphere dispersion with smaller diameters formed more nano/micropores, leading to discontinuous DNTT films and more defects and traps. The resulting PS/DNTT OPTs presented lower dark currents and enhanced photosensitivities. We can expect that in a certain range of PS

microsphere coverage, a discontinuous DNTT will be favor of enhancing the photosensitivity of the devices. And the relationship of coverage and photosensitivity requires further exploration. To verify the proposed mechanism, we fabricated bilayer-structured PS film/DNTT OPTs in which a uniform PS film was spin-coated on SiO_2/Si substrate, while the optoelectronic performance was not superior to that of nano/microporous PS/DNTT OPTs, as shown in Fig. S4. The devices presented lower photocurrent and dark current than our PS microspheres/DNTT OPTs, and the maximum I_{light}/I_{dark} ratio was only 160 at light intensity of $50 mW cm^{-2}$. The underlying mechanism can be explained by the less defects and traps in continuous DNTT films induced by uniform PS films.

In order to further confirm the role of PS microspheres in PS/DNTT OPTs, we also fabricated 0.5-PS/C8BTBT OPTs and C8BTBT OPTs to investigate the photo-response. C8BTBT presented a high sensitivity to ultraviolet (UV) light (400 nm) and nearly no photo-response to visible light (Fig. S5), therefore, 365 nm UV light source was employed. Fig. 5a and b show the transfer curves of 0.5-PS/C8BTBT OPTs and C8BTBT OPTs under 365 nm UV light with the intensity of $1 \mu W cm^{-2}$ at a fixed V_D of $-30 V$. As we can see, 0.5-PS/C8BTBT OPTs presented lower photocurrent and dark current but higher photo-

response than C8BTBT OPTs, which is consistent with the result of PS/DNTT OPTs and the above-mentioned mechanism. The $I_{\text{light}}/I_{\text{dark}}$ ratio as functions of V_G at the light intensity of $1 \mu\text{W cm}^{-2}$ is shown in Fig. 5c. The maximum $I_{\text{light}}/I_{\text{dark}}$ ratio of 0.5-PS/C8BTBT OPTs (1,900) is much larger than that of C8BTBT OPTs (80). The photo-response can be improved by integrating PS microspheres with C8BTBT.

CONCLUSIONS

In conclusion, we have successfully fabricated polystyrene microspheres/DNTT-organic phototransistors based on the interfacial trapping effect. By incorporating PS microsphere in DNTT, the PS/DNTT OPTs exhibited high $I_{\text{light}}/I_{\text{dark}}$ ratio of 1,800, which increased by two orders of magnitude compared with DNTT OPTs. Moreover, the effect of the PS microsphere's diameters on the output performance of PS/DNTT OPTs has been systematically studied, indicating that the smaller the PS microspheres diameter is, the higher the R and the $I_{\text{light}}/I_{\text{dark}}$ ratio are. In our study, the highest R and D^* of the devices were $2,700 \text{ A W}^{-1}$ and 3×10^{12} Jones, respectively, by modulating the gate voltages and incident light intensity. In addition, the photoresponse of PS/C8BTBT OPTs have also been investigated, demonstrating the validity of this strategy. Our study provides a new strategy to develop high-performance organic phototransistors.

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Author contributions Zhou B, Huang J and Wu X designed the devices and experiments; Chen Y synthesized the organic semiconductors; Chu Y drew the schematic structure of phototransistors; Zhou B performed the experiments; Zhou B analyzed the data; Zhou B, Zhou J and Wu X wrote the paper.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

Bilei Zhou received her bachelor degree from the School of Materials Science and Engineering, Wuhan University of Technology, Hubei, China. She is currently pursuing Master degree in the School of Materials Science and Engineering, Tongji University, Shanghai, China. Her main research includes the preparation and optoelectronic performance of photodetectors based on composite semiconductors.

Xiaohan Wu is now an assistant professor in the School of Materials Science and Engineering at Tongji University in Shanghai. He received the PhD degree in chemistry and biology for health at the University of Montpellier I, France. His research interests refer to organic semiconductors and biomaterials, biocompatible and flexible electronics; organic field-effect transistors; highly sensitive temperature and optical sensors; self-assembly of amphiphilic bio-copolymers; nano particles for drug controlled release.

使用聚苯乙烯微球提高有机光敏晶体管性能

周碧蕾, 周佳晨, 陈彦涛, 褚莹莉, 黄佳, 吴小晗*

摘要 有机光敏晶体管(OPTs)因结合了有机半导体和光敏晶体管的优势,引起了科研工作者的广泛兴趣.然而,OPTs性能很大程度上受限于有机半导体本身的性质,这对进一步提高OPTs性能提出了挑战.制备具有微纳结构的有机半导体被认为是提高器件性能的一种有效途径.聚苯乙烯(PS)微球作为一种绝缘和低成本的材料,可应用于制备微纳结构的器件,使器件实现对外部刺激的超高响应.本文中,我们利用PS微球杂化有机半导体来制备具有微纳结构的OPTs,使其光电流/暗电流(光灵敏度)提高了两个数量级,并且通过调控PS微球的直径,使光灵敏度从46增大到1800.此研究结果为制备具有微纳孔结构的OPTs提供了新方法,扩大了其在有机光电子中的应用.