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materials [30-32].

Synthesis of GO

Experimental section

yield of the reaction was 90%.

Synthesis of F-GQD

of F-GQD to 5.26 eV [29] from 5.01 eV of GO. The PSC device

with PTB7 and [6,6]-phenyl-C71-butyric acid methyl ester

(PC71BM) as the active layer and F-GQD as the HEL is

demonstrated to exhibit a PCE of 7.91%, outperforming its

counterparts based on the GO (6.33%) and PEDOT:PSS (7.46%)

HEL. To our best knowledge, the PCE thus achieved among

the highest values reported for PSCs containing graphene

A mixture of graphite powder (1.00 g, 325 mesh), concen-

trated H₂SO₄ (25 mL), sodium nitrate (0.50 g) was stirred for

30 min in an ice-water bath and was slowly added KMnO₄

(4.50 g) under vigorously stirring, followed by stirring at 35 °C

for 30 min and at 90 °C for another two hours. After cooled

down, the mixture was slowly added into de-ionized water

(100 mL) and H_2O_2 (30 mL). Centrifugation was carried out to

precipitate the solid, which was washed with 1 M hydrochlo-

ric acid for three times and de-ionized water for another

three times. The resultant solid was then dispersed in water

by ultrasonication for 30 min, followed by centrifugation at

6000 rpm for 30 min. The supernatant was collected and

purified through dialysis to afford aqueous GO solution (ca.

200 mL). To determine the concentration, 20 mL of the GO

solution was filtered with a 0.22- μ m membrane and then the

dried in vacuum to give a solid content of 0.09 g. Hence, the

concentration of the GO solution was 4.5 mg/mL and the

Vulcan VXC-72 carbon black (5.00 g, purchased from Cabot

Corporation) was added into a mixture of concentrated H₂SO₄

(200 mL) and concentrated HNO₃ (100 mL). The mixture was

stirred and refluxed for 24 h. After cooled down to room

temperature, the mixture was at first neutralized with

Na₂CO₃ powder and then acidified with hydrochloric acid to

give pH=1. The resulting mixture was added into acetone

(300 mL), followed by filtration to remove the precipitate.

The filtrate was collected and the solvent was removed by

rotary evaporation. The resulting deep yellow solid was dried

in vacuum to afford F-GQD. Yield: 1.90 g, 38%.

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recombination at the interfaces between the active laver and the cathode/anode electrodes. Therefore, a holeextraction laver (HEL) between the anode and active laver and/or an electron-extraction layer (EEL) between the cathode and active layer are often required for highperformance PSCs [6-8]. However, the state-of-the-art HEL, poly(3,4-ethylenedioxythiophene) doped with poly (styrene sulfonate) (PEDOT:PSS), is still suffering from strong acidity and hygroscopicity to cause the device instability [9]. In order to maximize the PCE without the detrimental effect on device stability, several inorganic semiconductors, such as MoO₃, V₂O₅, NiO, WO₃ and RuO₂, have been used as the HEL in high-performance PSCs [10-14]. However, the use of these inorganic semiconductors increases the manufacturing cost as they need to be vacuum deposited. Recently, graphene oxide (GO) and its derivatives have emerged as a promising class of HEL materials with advantages of solution processability and low cost [15-24].

GO, a derivative of the one-atom-thick graphene with 19 hydroxyl (OH) and epoxy groups on its basal plane and 21 carboxyl groups (COOH) at the edge (see Scheme 1), often has a lateral size larger than several hundred nanometers and the work function of 4.7-5.0 eV [25,26]. Since the work 23 function of GO matches well with the highest occup-25 ied molecule orbital (HOMO) level of the prototype donor polymer poly(3-hexylthiophene) (P3HT, HOMO: -4.9 eV) [20] for hole extraction, GO has been used as HEL in PSCs 27 by several groups including our own one [15-19]. Due to the 29 poor film-forming property intrinsically associated with the difficulties for large flakes of GO to uniformly cover the rough surface of indium-tin oxide (ITO) anode [16], the 31 device performance of GO HEL cannot surpass that of 33 PEDOT: PSS. Moreover, GO is not suitable for PSCs based on highly efficient donor polymers with deep HOMO levels 35 (< -5.1 eV) [15,19]. The relatively low work function of GO induces an energy barrier for hole extraction and greatly decreases the PSC device efficiency. Therefore, solution-37 processable graphene materials with high work function and 39 excellent film-forming property should be developed for high-efficiency PSC devices [21-24]. 41

In this manuscript, we report the use of few-layered graphene quantum dots (F-GQD) with a small size of about 4 nm and high content of periphery COOH groups (see, Scheme 1) as excellent HEL in PSCs based on a highly efficient donor polymer, poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl) carbonyl]thieno[3,4-b]thiophenediyl]] (PTB7). The small size of F-GQD ensures an excellent film-forming capability [27,28] while the abundant COOH groups increase the work function Download English Version:

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