



Synthesis of carbon quantum dots by chemical vapor deposition approach for use in polymer solar cell as the electrode buffer layer



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ABSTRACT

Fluorescent carbon quantum dots (C-CQDs) with graphitic structure were synthesized by chemical vapor deposition (CVD) for the first time. With this approach, the synthesized C-CQDs exhibit excellent crystalline graphitic nature with an average diameter of 3.5 nm and rich of hydrophobic $-\text{CH}_3$ terminal groups. Optical band gap of the C-CQDs was determined to be 3.16 eV, whereas low-lying HOMO/LUMO energy levels of $-7.00/-3.84$ eV were measured for these C-CQDs by cyclic voltammetry. Polymer solar cells using solution-processed C-CQDs as the electron transporting layer (ETL) were fabricated and tested. Results indicate the C-CQDs based devices exhibit similar or slightly improved device performance as the LiF based devices. Lowered series resistance (R_s) was found for these C-CQDs based devices, suggesting a better interfacial connection between the Al electrode and polymer with this ETL. More importantly, improved thermal stability was confirmed for the CQD-based devices, which was ascribed to the less diffusion possibility of C-CQDs in solid film. The current work confirmed that the CVD synthesized C-CQDs could serve as a solution processed ETL for use in organic solar cells with improved device stability.

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1. Introduction

Owing to the advantages of low cost, light weight, mechanical flexibility and solution processability, polymer solar cells (PSCs) have the potential to become new alternative sources of energy using spin-coating, inkjet printing, or roll-to-roll printing techniques to fabricate large area photovoltaic devices on flexible and light weight substrates [1–3]. To date, the main challenges for PSCs are efficiency and stability [4]. Significant improvements in efficiency have been achieved in PSCs through materials design [5–7], device optimization processing [8–10] and device interface engineering [11–13], and power conversion efficiency over 10% has

been reported in the literature [14,15]. Long-term stability of PSCs is becoming the next key challenge for the successful commercialization of PSCs.

Typically, bulk heterojunction PSCs are fabricated by sandwiching a photoactive layer between a transparent conductive anode and a low work function metal cathode. Besides, electrode interfacial buffer layers are also essential for PSCs, because they play a critical role in improving device performance of PSCs [11–13]. Ideal interfacial buffer layer can facilitate charge extraction and prevent charge recombination at the interfaces between the photoactive layer and the cathode/anode electrodes, originating from a better energy alignment between these functional layers [16,17]. What's more, ideal interfacial buffer layer can also act as a barrier layer for limiting the diffusion of oxygen, moisture and Al atoms into the organic layers. Therefore, interfacial buffer layer can also improve device stability of PSCs. So far, LiF and Ca are the two most commonly used electron transport layers (ETLs) for PSCs and have been proved to be able to increase both the open circuit

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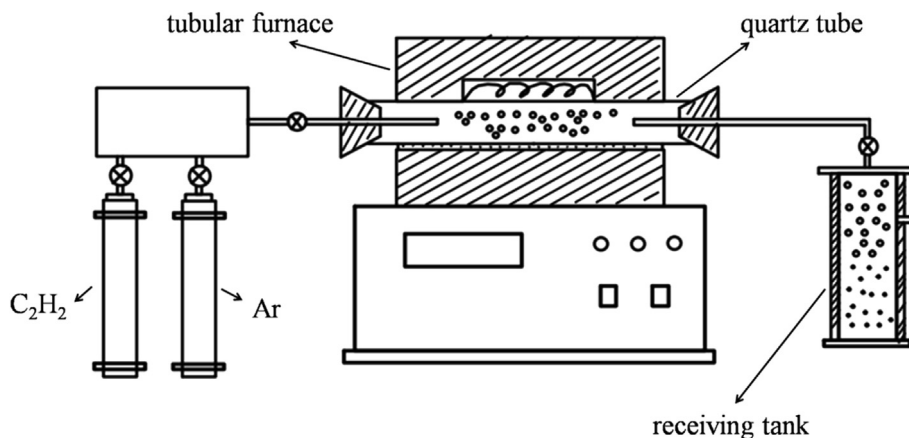


Fig. 1. Schematic of CVD experimental apparatus.

voltage (V_{OC}) and fill factor (FF) of PSCs [16,18,19]. However, because of its insulating nature, the optimized LiF layer thickness is usually limited to few nanometers (1–2 nm), which is on one hand too thin to function as an effective protection barrier layer against oxygen and moisture [13,20], on the other hand, is more difficult to be precisely controlled during thermal evaporation. In addition, the fast diffusion rate of Li^+ and F^- would also cause the inferior device stability and operation lifetime [21,22]. Insertion of a thin Ca layer between Al electrode and the photoactive layer has been believed to form an ohmic contact and therefore result in an increase of device FF [23,24]. However, Ca is very sensitive to oxygen and moisture, which is a potential drawback for the stability of PSCs. In order to maximize the efficiency without posing detrimental effect on stability of PSCs, various kinds of inorganic semiconductors, such as ZnO [25], TiO_x [26], graphene quantum dots (GQDs) [27,28], and polymers [29,30] have been used as the ETLs in PSCs, and high device performance have been achieved for these ETL based devices.

Carbon quantum dots (CQDs), as novel carbon nanomaterials, have been reported for use in solution processed photovoltaic devices as electron acceptor recently [31,32], due to their excellent photoelectric properties, solution processability, simple preparation and low cost [33,34]. Till now, CQDs reported in the literature were mostly synthesized via hydrothermal approach, which possess advantages of excellent chemical and thermal stability, low toxicity, intensive photoluminescence performance, low photobleaching and good biocompatibility. However, CQDs synthesized by hydrothermal method (H-CQDs) usually contain multiple hydrophilic terminal groups or with long alkyl side chain, which might hinder the charge transport in the blended film. Chemical vapor deposition (CVD) is an alternative method to synthesize carbon nanomaterials with precise CQD property control, however, there is no report on the synthesis of CQDs by CVD approach till now. In this paper, we report the synthesis of carbon quantum dots (C-CQDs) via a simple and fast CVD method for the first time. The synthesized C-CQDs have crystalline graphite structure with an average diameter of 3.5 nm. The synthesized C-CQDs can be used as the ETL in polymer solar cells. Results showed that the C-CQDs-based devices have similar device performance as the LiF based devices, whereas improved thermal stability was found for the C-CQD-based cells when compared to LiF based devices, which was attributed to lower molecular diffusion rates of C-CQDs. The current work proved that C-CQDs prepared by CVD approach could be an alternative ETL materials for polymer solar cells.

2. Experimental

2.1. Materials

Highly pure C_2H_2 and Ar (>99.99%) were purchased from Leap Gas Co. Ltd. *N,N*-Dimethylformamide (DMF) was purchased from Tianjin Fuchen Chemical Reagents. *n*-Hexane and cyclohexane were supplied by Tianjin Guangfu Technology Development Co. Ltd. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS; Clevis PVP Al 4083) was purchased from Heraeus Precious Metals GmbH & Co. KG. Regioregular poly(3-hexylthiophene) (P3HT; $M_n = 5.0 \times 10^4$ g/mol, PDI = 1.7, regioregularity $R_r = 95\%$) was purchased from Solarmer Energy, Inc. (Beijing). Poly[[4,8-bis[(2-ethylhexyl)oxy]-benzo[1,2-*b*:4,5-*b'*]dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)-carbonyl]thieno[3,4-*b*]thiophenediyl]] (PTB7) and poly[[2,6'-4,8-di(5-ethylhexylthienyl)benzo[1,2-*b*:3,3-*b'*]dithiophene][3-fluoro-2[(2-ethylhexyl)carbonyl]thieno[3,4-*b*]thiophenediyl]] (PTB7-Th) were purchased from 1-Material and Solarmer Materials Inc., respectively. Phenyl- C_{61} -butyric acid methyl ester (PC₆₁BM) and [6,6]-phenyl- C_{71} -butyric acid methyl ester (PC₇₁BM) were purchased from Solenne B.V. H-CQDs were prepared following the literature [35].

2.2. Preparation of C-CQDs

C-CQDs were synthesized by CVD in Ar atmosphere using C_2H_2 as carbon source (see Fig. 1 for the experiment set-up). The preparation was conducted in a quartz tube (32 mm i.d. and 1 m long), which was mounted in a horizontal tubular furnace. At the beginning of synthesis, the quartz tube was heated to 1000 °C in Ar atmosphere. Then a mixture of C_2H_2 /Ar was introduced into the reactor at a rate of 70/700 mL min^{-1} for 2 h. After the reaction, the quartz tube was cooled to room temperature under the protection of flowing Ar. The crude product was collected in a receiving tank, and dissolved in 200 mL of DMF by sonication for 30 min. The DMF solution was filtered through a filter paper (in middle speed) and syringe filter (Polytetrafluoroethylene (PTFE), 0.45 μm). The organic solvents were removed under vacuum to give the C-CQDs.

2.3. Preparation of solar cell

Photovoltaic devices were fabricated with the configuration of ITO/PEDOT:PSS/active layer/ETL/Al. The patterned substrates were cleaned in an ultrasonic bath using detergent, deionized water,

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