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RAPID COMMUNICATION

A fluorescent quenching performance enhancing principle for carbon nanodot-sensitized aqueous solar cells

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Abstract
We report a fluorescent quenching principle capable of markedly enhancing the conversion efficiency of carbon nanodots (CNDs)-sensitized aqueous solar cells (CNDs-ASCs). A conversion efficiency of 0.529%, over 4-times of the best conversion efficiency reported for CNDs-sensitized solar cells, is achieved with a cell constructed using CNDs as the sensitizer and aqueous I^-/I_3^- electrolyte serving a dual-function as the recombination blocker (fluorescent quencher) and redox mediator. The results confirm that the significantly enhanced utilization efficiency of the photo-excited electrons resulting from the efficiently quenched fluorescent emission of CNDs sensitizer by I^- is responsible for the markedly improved conversion efficiency of CNDs-ASCs. The findings of this work validate a principle that could be widely applicable for enhancing the performance of solar cells employing other fluorescent quantum dots/nanodots sensitizers.
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Introduction

A variety of quantum dots (QDs) and nanodots (NDs) has been utilized as sensitizers for solar cells [1-6]. Although QDs/NDs-sensitized solar cells (QDs/NDs-SSCs) have many attractions, in general they possess lower conversion efficiency (<7%) when compare to dye-sensitized solar cells and newly reported perovskite-based solar cells [7-10]. It is well known that the majority of QDs/NDs are fluorophores [11-13]. When they are used as sensitizers in solar cells, the fluorescent emission is, in effect, a charge recombination pathway, leading to a dramatically reduced utilization efficiency of the photo-excited electrons for electricity generation, which is a major factor responsible for low conversion efficiencies of QDs/NDs-SSCs. On this basis, we hypothesize that the utilization efficiency of the photo-excited electrons generated by the fluorescent sensitizers could be significantly enhanced by a suitable fluorescent quenching mechanism, which can be a widely applicable performance enhancing principle for QDs/NDs-SSCs. The validation of such a principle would be highly valuable for design and development of high performance QDs/NDs-SSCs.

Carbon quantum dots (CQDs) and nanodots (CNDs) are a new class of metal-free fluorophores [14-17]. Recent studies revealed that the fluorescent emission of CQDs and CNDs can be efficiently quenched by the electron accepters such as Cu^{2+}, Hg^{2+} and Fe^{3+} [18-22]. Such quenching phenomena have been successfully utilized to sensitively detect the electron accepter type of quenchers [18-22]. However, the vast majority of the reported CQDs and CNDs based fluorophores are fabricated using carbon sources that are originated from fossil fuels. It would be highly attractive to fabricate such a new class carbon materials from cheap, plentiful and renewable natural biomasses [18,23,24]. We recently confirmed that the fluorescent emission of graphitic CNDs obtained from grass can also be efficiently quenched by electron donors such as I^- for sensitive I^- determination in aqueous solution [24]. With graphene QDs sensitizers, an overall conversion efficiency of 0.056% was reported in an earlier study [25], and the conversion efficiency was further improved to 0.13% with CNDs sensitizers in subsequent studies (Table S1) [26-28].

In this work, N-doped CNDs synthesized from grass are chosen as the testing case to validate the proposed fluorescent quenching performance enhancement principle. The aqueous solar cells (ASCs) were assembled by a CNDs-sensitized TiO_2 photoanode and a commercial Pt counter electrode with aqueous I^-/I_3^- being used as electrolyte to serve a dual-function as the redox mediator and recombination blocker (fluorescent quencher) (denoted as CNDs-ASCs) in this work. An overall conversion efficiency of 0.529% can be achieved from a CNDs-ASC, significantly higher than the best conversion efficiency (0.13%) obtained from the CNDs-sensitized solar cells with organic solvent I^-/I_3^- electrolyte (Table S1) [25-28].

Experimental section

Synthesis of N-doped carbon nanodots (CNDs)

N-doped CNDs were synthesized by a facile hydrothermal method [18]. In a typical synthesis, 30 g fresh Monkey Grass (*Ophiopogon japonicus*) was firstly cut into pieces and added into 60 mL of deionized water (Millipore Corp., 18 MΩ), and then the mixture was transferred into a 100 mL of Teflon lined autoclave. The hydrothermal reaction was kept at 180 °C for 6 h. After hydrothermal reaction, the obtained product was collected, respectively, by filtration (0.2 μm cellulose membrane) and centrifugation at 4500 rpm and 14,000 rpm for 15 min to remove large-sized carbon products. The obtained nanodots suspension solution was preserved for further characterization and use. In this work, the fabricated N-doped CNDs solution has a concentration of *ca.* 35 mg/mL with a production yield of around 7.0%.

Sensitization

For more meaningful comparison, the commercially available nanocrystalline TiO_2 films with a thickness of *ca.* 10 μm (DYESOL, Australia) were used as photoanode material and firstly treated at 500 °C for 30 min prior to sensitizing with N-doped carbon nanodot solution at room temperature for 24 h. The carbon nanodot-sensitized TiO_2 films were then dried in a nitrogen stream for further use in solar cell measurements. The TiO_2 film sensitized with N-doped carbon nanodot solution was denoted as CNDs-TiO_2. The fabricated film was further treated in 100 mM I^- aqueous solution (NaI, Sigma-Aldrich) for 30 min to form I^- modified CNDs-TiO_2 film (denoted as CNDs-TiO_2-I). After that, the CNDs-TiO_2-I film was adequately rinsed using deionized water and then dried at room temperature in a nitrogen stream for 12 h. For comparison, the CNDs-TiO_2 film was also immersed in acetonitrile containing 100 mM I^- for 30 min for further measurements. All photoanodes were preserved for further characterization and measurement in solar cells.

Characterization

A scanning electron microscope (SEM, JSM-7001F), transmission electron microscopy (TEM, Philips F20), and X-ray diffraction (XRD, Shimadzu, XRD-6000, diffractometer, equipped with a graphite monochromator) were employed for characterizing the sample structures. The UV-vis absorption of nanodot solution was measured using a Varian Cary 4500. Diffuse reflectance spectra of the photoanode films were recorded on a Varian Cary 5E ultra violet-visible-near infrared (UV-vis-NIR) spectrophotometer. FT-IR spectra of carbon nanodot sample were measured to investigate structural information and specific molecule-groups information (Perkin-Elmer 2000). Chemical compositions of the samples were analyzed by X-ray photoelectron spectroscopy (XPS, Kratos Axis ULTRA incorporating a 165 mm hemispherical electron energy analyzer). The photoluminescent (PL) spectra of carbon nanodot samples were measured on F-7000 Fluorescence Spectrophotometer (Hitachi). Carbon nanodot loading amount on nanocrystalline TiO_2 film was measured by carefully calculating the weight of TiO_2 film samples before and after sensitization.

Measurements

All investigated solar cells were fabricated with traditional sandwich type configuration by using a carbon nanodot-sensitized TiO_2 film and a platinum counter electrode deposited on FTO conducting glass (DYESOL, Australia). The organic

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