Layered Graphene/Quantum Dots: Nanoassemblies for Highly Efficient Solar Cells

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Because of the alarming rate at which global energy consumption has grown, due to rapid worldwide economic expansion, population increase, and the ever-increasing reliance on energy-based appliances, there is an urgent need for the development of renewable energy sources. This urgency for developing new types of energy is further emphasized by the limited supply of today's main energy sources (i.e., oil, coal, and uranium) and their detrimental long-term effects on the environment. Fortunately, the sun provides us with renewable energy sources that neither run out nor have any significant harmful environmental effects. Ever since the French scientist Alexandre-Edmond Becquerel discovered the photovoltaic effect in 1839,[1] scientists and engineers have devoted considerable efforts to realizing mankind’s dream of being able to exploit this effect and meet our daily energy needs by converting the energy of sunlight directly into electricity.[2] However, after more than 170 years this dream still has to be realized. Although inorganic silicon-based photovoltaic devices have demonstrated sufficiently high power conversion efficiencies (>10%) for the production of electricity from sunlight, the use of conventional silicon-based photovoltaic devices is still not widespread because of difficulties in modifying the bandgap of Si crystals and the high costs associated with the elaborate fabrication processes, which involve elevated temperatures and high-vacuum conditions.[3] Carbon nanomaterials of suitable band energies, such as fullerenes and single-walled carbon nanotubes (SWNTs), have been used as efficient electron acceptors in QD solar cells.[5, 6] However, even the highest incident-photon-to-charge-carrier generation efficiency (IPCE = 5%, under 100 mW cm⁻² illumination) reported so far for these carbon-based QD solar cells[6] is still too low to meet the requirements for commercialization. Recent developments in nanomaterials and nanotechnology have opened up new frontiers in materials science and device engineering, to create novel carbon nanomaterials with tailored bandgaps and new devices with desirable structures for enhancing the solar cell performance.[6, 7]

A significant advance in the development of layered graphene/quantum dots for highly efficient solar cells was recently reported by Li and co-workers.[8] Instead of using one-dimensional, cylindrically shaped carbon nanotubes to construct CdS QD solar cells[6c, d] with an inhomogeneous CdS distribution, the group of Li used two-dimensional, single-atom-thick graphene sheets to support the CdS QDs. Graphene sheets offer many competitive advantages compared to carbon nanotubes, including versatility for solution-based large-scale fabrication, good transparency, high charge mobility, large specific surface area, and low costs.[9] By creating a novel layered nanofilm of graphene/CdS QDs on an ITO glass electrode (Figure 1a) from all-aqueous solutions through sequential electrophoretic and chemical...
deposition, Li et al.\textsuperscript{[3]} successfully increased the IPCE of the QD solar cell by a factor of more than 3, up to 16\% (Figure 1b).

As demonstrated by Li et al.,\textsuperscript{[3]} graphene shows a relatively good band energy match with CdS QD compared to SWNTs (Figure 1c) for an effective charge separation, and allowed for a uniform distribution of the QD nanoparticles, thereby maximizing the solar absorption and charge collection.\textsuperscript{[3]} Indeed, Figure 2a clearly shows a conformal graphene layer on ITO glass, as produced by the electrophoretic deposition. Subsequent chemical reduction of Cd\textsuperscript{2+} from CdCl\textsubscript{2} in an aqueous solution of Na\textsubscript{2}S led to the formation of crystalline CdS QDs of ca. 5 nm in diameter, which were uniformly distributed over the predeposited graphene film (Figure 2b and 2c). The resulting ITO/graphene/CdS QD bilayer photovoltaic device exhibited an IPCE of 5\%; a value that is much higher than the highest IPCE = 0.45\% reported for its ITO/SWNT/CdS QD counterpart.\textsuperscript{[3, 4]}

To further improve the device performance, Li and co-workers prepared a multilayer (graphene/CdS QD)\textsubscript{n} nanoassembly while largely retaining the intimate contact and large interface between each of the adjacent graphene and CdS QD layers (Figure 3a), by sequentially depositing graphene and CdS QD layers onto each other through electrophoretic and chemical deposition, respectively.

\begin{figure}[h]
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\includegraphics[width=\textwidth]{fig2.png}
\caption{a) Scanning electron microscopy (SEM) image of the graphene layer electrophoretically deposited on an ITO-coated glass. b, c) Transmission electron microscopy (TEM) images of CdS QDs on the graphene layer at different magnifications. Reproduced with permission from Ref. [8].}
\end{figure}

The aforementioned innovative approach not only significantly increased the solar absorption efficiency by increasing the total film thickness in a controllable manner (Figure 3b),\textsuperscript{[3a, 7a]} but also offers the potential for minimizing recombination losses of charge carriers by independently tuning the thickness and morphology for each of the constituent layers. As a consequence, IPCEs of up to 16\% (Figure 3c), a value 3-fold higher than that of the ITO/graphene/CdS QD bilayer (5\%), were obtained in the study of Li et al.,\textsuperscript{[8]} and even higher efficiencies may be achievable. The IPCEs of the multilayer graphene/CdS QD nanoassemblies are far superior to those of other carbon-based QD solar cells reported so far, for example, 0.45\% for SWNT/CdS QD solar cells,\textsuperscript{[6c, d]} 4.5\% for fullerene/CdSe QD composites,\textsuperscript{[5]} 5\% for stacked carbon nanocup/CdSe QD solar cells,\textsuperscript{[6a]} and 1.2\% for SWNT/pyrene/CdTe.\textsuperscript{[6i]}

The study highlighted herein clearly indicates that nanotechnology can indeed serve as an enabling technology, creating novel carbon nanomaterials with tailored bandgaps and new photovoltaic devices with desirable structures for significantly improved performance. Continued research in this important area could overcome some of the major hurdles that solar cells are facing in the race to the technological marketplace.

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\begin{thebibliography}{99}
\bibitem{5} S. Sun, N. S. Sariciftci, Organic Photovoltaics, CRC Press, Boca Raton, 2005.
\end{thebibliography}
HIGHLIGHTS

Nanoassemblies for Highly Efficient Solar Cells


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