Fabrication and investigation of the optoelectrical properties of MoS$_2$/CdS heterojunction solar cells

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Abstract

Molybdenum disulfide (MoS$_2$)/cadmium sulfide (CdS) heterojunction solar cells were successfully synthesized via chemical bath deposition (CBD) and chemical vapor deposition (CVD). The as-grown CdS film on a fluorine tin oxide (FTO) substrate deposited by CBD is continuous and compact. The MoS$_2$ film deposited by CVD is homogeneous and continuous, with a uniform color and a thickness of approximately 10 nm. The optical absorption range of the MoS$_2$/CdS heterojunction covers the visible and near-infrared spectral regions of 350 to 800 nm, which is beneficial for the improvement of solar cell efficiency. Moreover, the MoS$_2$/CdS solar cell exhibits good current-voltage ($I$-$V$) characteristics and pronounced photovoltaic behavior, with an open-circuit voltage of 0.66 V and a short-circuit current density of 0.227 $\times$ 10$^{-6}$ A/cm$^2$, comparable to the results obtained from other MoS$_2$-based solar cells. This research is critical to investigate more efficient and stable solar cells based on graphene-like materials in the future.

Keywords: Molybdenum disulfide; CdS; Solar cells; CVD; CBD; $I$-$V$ behaviors

Background

Single-layer (SL) and few-layer (FL) molybdenum disulfide (MoS$_2$) recently became attractive alternative semiconductor materials for next-generation nanoelectronic applications due to their large electron mobility, large bandgap [1-5], excellent stability, and the absence of dangling bonds [6]. MoS$_2$ has been widely studied and applied in many areas, such as field-effect transistors [6-13], energy harvesting [14,15], optoelectronics [16-18], photocatalysts [19-21], and counter electrodes [22,23]. Moreover, single and multilayer MoS$_2$ phototransistors have been demonstrated with an on/off ratio of approximately 10$^3$ and a carrier mobility of 80 cm$^2$/Vs [17,18], which indicates that MoS$_2$ is a promising candidate for photovoltaic solar cells. Gourmelon et al. previously reported on the use of MoS$_2$ in solar cells [14], but the report did not draw much interest until recently. Yu et al. reported a TiO$_2$/MoS$_2$/P$_3$HT bulk heterojunction solar cell with a short-circuit current density of 4.7 mA/cm$^2$, an open-circuit voltage of 560 mV, and a power conversion efficiency of 1.3%, as well as MoS$_2$ nanomembrane-based Schottky-barrier solar cells with a power conversion efficiency of 0.7% for approximately 110-nm MoS$_2$ and 1.8% for approximately 220-nm MoS$_2$ [24,25]. Clearly, the optical current, voltage, and energy transfer efficiency of these cells are low, and further investigations of MoS$_2$-based solar cells are significant and necessary.

It is well known that cadmium sulfide (CdS), with a large direct bandgap of 2.4 eV [26-28], is a viable material and widely used in solar cells as a window layer. Zhang et al. have demonstrated MoS$_2$/CdS heterojunction by photoelectrochemical methods and studied the photocatalytic and contact interface properties [15,19,29,30]. However, the photoelectric characteristics and conversion efficiency of MoS$_2$/CdS heterojunction solar cells have not been demonstrated. And the complexity of these methods or the poor morphologies and structures of samples limited its use. Here, we present the fabrication of MoS$_2$-based solar cells composed of p-MoS$_2$ and n-CdS by simply using chemical bath deposition (CBD) and chemical vapor deposition (CVD). CBD is considered to be a low-cost and simple approach, which can produce reproducible, uniform, and adherent CdS films [31-33]. Additionally, CVD has been recognized as one of the best techniques for the fabrication of large-area homogeneous MoS$_2$ films [12,13,34-36]. Moreover, we systematically analyzed the individual films' surface morphologies, structures and electrical and optical properties, as well as the photovoltaic...
properties of the MoS2/CdS films and heterojunction solar cells.

**Methods**

MoS2/CdS heterojunction solar cells were synthesized, as shown in Figure 1, in a three-step process: (i) CBD of CdS on a fluorine tin oxide (FTO)-coated glass substrate using the reaction between CdAc2 and H2NCSNH2, (ii) CVD of MoS2 on CdS, and (iii) sputtering of Ni electrodes on MoS2. CdS thin films were firstly deposited via CBD on FTO substrates that had been ultrasonically cleaned with deionized water, and then dried at 80°C in a drying oven. The FTO substrates were immersed in a solution composed of 0.007 M cadmium acetate (Cd(CH3COO)2·2H2O) and 0.05 M thiourea (H2NCSNH2) and maintained at 80°C for 60 min with stirring to obtain uniform deposition. After deposition, the CdS films were ultrasonically washed to remove the loosely adhered CdS particles on the surface and subsequently dried and annealed at 400°C for 60 min in N2 to improve the crystalline quality. Some CdS films were set aside as representative samples for characterization of surface morphologies and structures, and the others were used to synthesize MoS2/CdS heterojunction solar cells.

MoS2/CdS heterojunctions were formed by further CVD of a MoS2 thin film on the pre-existing CdS film. The CVD experimental setup consisted of a horizontal quartz tube furnace, an intake system, a vacuum system, and a water bath. The substrates were placed in the center of the furnace, and subsequently, the furnace was pumped down to 10⁻² Pa and heated up to 550°C for 30 min. A mixed solution comprising 1 g analytical grade MoS2 micro powder, 1 g analytical grade silver nitrate (AgNO3) powder, and 200 mL of diluted sulfuric acid (H2SO4) was formed by stirring for 5 min and maintained at 70°C via the water bath. Ar gas was then flowed through the mixed solution with a flow rate of 20 standard cm³/min, carrying silver-doped MoS2 molecules into the furnace. The adsorption and deposition of MoS2 molecules onto the CdS films yielded MoS2/CdS thin films. After the completion of the deposition, the samples were annealed at 600°C for 30 min in an Ar atmosphere. Furthermore, to investigate the material properties of MoS2 films, MoS2 samples were deposited on quartz crystalline slides by the same method.

To construct a MoS2/CdS heterojunction solar cell, Ni electrodes were sputtered onto the corner of the MoS2/CdS thin films using magnetron sputtering. The surface morphologies and crystalline structures of MoS2 and CdS films were characterized using atomic force microscopy (AFM) and X-ray diffraction (XRD), respectively. The electrical properties of the samples were analyzed by a Hall Effect Measurement System (HMS-3000, Ecopia, Anyang, South Korea) at room temperature. The UV-visible absorption spectra of the samples were investigated by a UV-visible spectrophotometer (Shimadzu UV-3600, Kyoto, Japan). Photovoltaic measurements of the MoS2/CdS heterojunction solar cells were taken using a Keithley 4200 semiconductor characterization system (Keithley Instruments, Inc., Cleveland, OH, USA), both in the dark and under standard AM 1.5 illumination (100 mW/cm²).

**Results and discussion**

Figure 2 shows the AFM images of the CdS film and the MoS2 film on a quartz crystalline slide. The surface of the CdS shown in Figure 2a is continuous and compact, and some nanoparticles are present on the top layer, which
can effectively promote the absorption of light. Additionally, many MoS$_2$ quantum dots around 100 nm in diameter, shown in Figure 2b, are uniformly deposited on the surface of the MoS$_2$ film. Under the quantum dots, the MoS$_2$ film is homogeneous and continuous, with a uniform color and a thickness of about 10 nm, which is equal to a few layers of MoS$_2$. This growth mode, called the layer-quantum dot mode, corresponds to the hexagonal crystalline structure of MoS$_2$.

The crystal structures of the samples were characterized by XRD. The XRD pattern of the CdS film is illustrated in Figure 3a. Only the (111) diffraction peak, appearing at $26.2^\circ$, belongs to cubic CdS; the others, located at $24.8^\circ$, $28.2^\circ$, $43.7^\circ$, and $50.8^\circ$, correspond to the (100), (101), (110), and (112) diffraction planes of a hexagonal CdS, respectively, which is more suitable to be an n-type window layer for solar cells, due to its high transmission and electrical conductivity [37]. Moreover, these observed diffraction peaks are rather sharp, especially the (111) and (101) peaks, which indicate good crystallinity. Figure 3b shows the XRD pattern of the MoS$_2$ film. Six sharp diffraction peaks are located at $14.7^\circ$, $29.3^\circ$, $33.1^\circ$, $47.8^\circ$, $54.6^\circ$, and $56.4^\circ$, corresponding to the (002), (004), (100), (105), (106), and (110) crystal planes of MoS$_2$, respectively, which show that the MoS$_2$ film exhibits a variety of crystal structures. In addition, it has to be noted that no silver diffraction peaks are observed, indicating that the silver doping does not change the crystal structure of the MoS$_2$ film.

Figure 4 shows the ultraviolet-visible (UV-vis) absorption spectra of the CdS, MoS$_2$, and MoS$_2$/CdS samples in the wavelength region of 350 to 800 nm. The CdS film has a strong optical absorption peak at 490 nm, and the optical absorption covers the wavelength region of 350 to 510 nm, consistent with the previously reported findings [29,38]. Over the region 510 to 800 nm, the absorptivity of the CdS film decreases abruptly, and no other absorption peaks are observed, indicating that the CdS film is transparent to light in this range. However, there is an absorption peak observed for the MoS$_2$ film located at 735 nm, which corresponds to the MoS$_2$ bandgap of about 1.69 eV. The optical absorption range of the MoS$_2$ film almost covers the range that the CdS film does not absorb light, demonstrating that MoS$_2$/CdS solar cells enhance the absorption of light, compared with silicon-based solar cells. Moreover, the optical absorption range of the MoS$_2$/CdS sample covers the visible and near-infrared spectral regions of 350 to 800 nm, which is beneficial for the improvement of solar cell efficiency.

We measured surface current-voltage (I-V) properties, carrier mobilities, and Hall coefficients of the MoS$_2$ and CdS samples using a Hall Effect measurement system. Figure 5 shows the surface I-V behaviors of the two measured points on the samples. The extracted voltages between the two points show a linear dependency on the
applied current, indicating that the MoS$_2$ and CdS films have good conductivity, with few surface defects or impurities. The electron mobilities in the MoS$_2$ and CdS films are $1.579 \times 10^3$ cm$^2$/Vs and $7.68 \times 10^2$ cm$^2$/Vs, respectively. Note that the mobility value for the MoS$_2$ film is higher than previously reported [39,40], which may be due to lower phonon and lattice scattering. Furthermore, the Hall coefficients of the MoS$_2$ and CdS films are $6.379 \times 10^6$ cm$^3$/C and $-3.257 \times 10^2$ cm$^3$/C, respectively, showing that MoS$_2$ is a p-type semiconductor, and it can form a p-n junction with n-type CdS, as demonstrated in previous studies [15,19,29,41-43].

Figure 6 displays the energy band diagram of the fabricated MoS$_2$/CdS heterojunction solar cell. $E_{C1}$, $E_{C2}$, $E_{V1}$, and $E_{V2}$ denote the conduction bands and valence bands of CdS and MoS$_2$, respectively. $E_F$ is the Fermi level energy. $\chi_1$ and $\chi_2$ are the electron affinities of CdS (3.8 eV) [38] and MoS$_2$ (4.0 eV), respectively. $V_0$ is the built-in potential, and $E$, with the direction from n-CdS to p-MoS$_2$, is the built-in electric field. Because of the Fermi level difference between n-CdS and p-MoS$_2$, electrons diffuse from n-CdS to p-MoS$_2$, and simultaneously, holes in p-MoS$_2$ move to n-CdS, leading to the formation of a space-charge region and built-in electric field with the direction from n-CdS to p-MoS$_2$ at the contact interface. The built-in electric field, $E$, prevents carriers from diffusing and makes them drift in the opposite direction, and finally, the heterojunction comes to thermal equilibrium with a unified Fermi level. Under light illumination, the photo-generated electrons and holes are quickly separated and driven into n-CdS and p-MoS$_2$, respectively, under the acceleration of $E$, which gives rise to the generation of the photocurrent.

Figure 7a shows the dark current density-voltage ($J$-$V$) characteristics of the fabricated MoS$_2$/CdS heterojunction solar cell. Remarkably, the current curve of the device shows an exponential dependence on the applied positive voltage, and tends to be almost zero under the reverse voltage, indicating that the MoS$_2$/CdS solar cell exhibits good rectification characteristics, and forms a well-defined p-n junction, as demonstrated by the previous reports [15,19,29].

Figure 7b displays the light-illuminated $J$-$V$ characteristics of the fabricated MoS$_2$/CdS heterojunction solar cell. The solar cell exhibits pronounced photovoltaic behavior, with an open-circuit voltage ($V_{oc}$) of 0.66 V and a short-circuit current density ($J_{sc}$) of $0.227 \times 10^{-6}$ A/cm$^2$. We can see that $V_{oc}$ is much larger than the results obtained from other MoS$_2$-based solar cells [24,25], but $J_{sc}$ is much lower than that of common solar cells [24,25], which is likely attributed to the large resistances for the device. The fill factor (FF) can be obtained based on the relationship of
FF = \frac{I_m V_m}{I_{sc} V_{oc}} \text{ where } I_m \text{ and } V_m \text{ are the current density and voltage at the maximum power output, respectively. In this instance, FF is approximately 0.22, comparable to previously reported values [25]. These results show that to improve the light energy efficiency of the MoS$_2$/CdS heterojunction solar cells it is necessary to lower the contact resistance of the cell, which is also critical to solar cells based on graphene-like materials.}

Conclusions

We have fabricated heterojunction solar cells composed of p-MoS$_2$ and n-CdS films using CBD and CVD methods and studied the surface morphologies, structures, and electrical and optical properties, as well as the photovoltaic properties. The MoS$_2$ film is homogeneous and continuous, with a thickness of around 10 nm, which is equal to a few layers of MoS$_2$. The as-grown CdS film is continuous and compact. The optical absorption range of the MoS$_2$/CdS film covers the visible and near-infrared spectral regions of 350 to 800 nm, which is beneficial for improving solar cell efficiency. Moreover, the MoS$_2$/CdS solar cell exhibits good rectification characteristics and pronounced photovoltaic behavior, with a short-circuit current density of $0.227 \times 10^{-6}$ A/cm$^2$ and an open-circuit voltage of 0.66 V, comparable to the results obtained from other MoS$_2$-based solar cells.

Competing interests

The authors declare that they have no competing interests.

Authors’ contributions

WG participated in the fabrication of MoS$_2$/CdS heterojunction solar cells, measured the electrical properties of the solar cells, and wrote the manuscript. FY, CW, and YZ studied the surface morphologies, structures, and electrical and optical properties of the samples and participated in the analysis of the results of the solar cells. XM designed the structure of the solar cell and analyzed the results. All authors read and approved the final manuscript.

Authors’ information

WG is a graduate student major in fabrication of new semiconductor nanometer materials. FY, CW, and YZ are undergraduate. MS is a professor and PhD-degree holder specializing in semiconductor materials and devices, especially expert in nanoscaled optical-electronic materials and optoelectronic devices.

Acknowledgements

This work was supported in part by the Innovation Program for Postgraduate of Suzhou University of Science and Technology (No. SKX135_053), the Priority Academic Program Development of Jiangsu Higher Education Institutions, the USTS Cooperative Innovation Center for Functional Oxide Films and Optical Information, and the Education Reform of Jiangsu University (No. JGLX13_091).

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Received: 2 September 2014 Accepted: 25 November 2014

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doi:10.1186/1556-276X-9-662
Cite this article as: Gu et al.: Fabrication and investigation of the optoelectrical properties of MoS2/CdS heterojunction solar cells. Nanoscale Research Letters 2014 9:662.