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Effective CdS/ZnO Nanorod Arrays as Antireflection Coatings for Light Trapping in c-Si Solar Cells

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Effective CdS/ZnO nanorod arrays (NRAs) as antireflection coatings for light trapping in c-Si solar cells were prepared by chemical bath deposition method which is a mature technology suitable for mass production. Field emission scanning electron microscopic (FESEM) reveals that CdS nanoparticles with a diameter of ~65 nm are deposited on the top of ZnO NRAs with an average length of 380 nm and a
10 diameter of ~40 nm. The cell with CdS/ZnO NRAs coatings exhibits lower reflectance and better light trapping ability than the cell with ZnO NRAs. PL spectra reveal the formation of CdS nanoparticles can suppress the outward broad defect emission of ZnO NRAs peaking around 575 nm so that green-yellow light emission of ZnO NRAs can be transmitted into c-Si solar cell for photovoltaic conversion. The J-V curves obtained under the AM1.5G simulator illumination condition further indicate the improved
15 photovoltaic properties of c-Si solar cells due to CdS/ZnO NRAs antireflection coatings.

1. Introduction

In the past few years, nanostructure materials have been widely used to make photovoltaic devices. Among them, one-dimensional (1-D) materials have attracted peoples' interests.
20 One-dimensional metal-oxide-based nanostructures open up a new area in developing advanced functional materials and photovoltaic devices. Typically, ZnO is considered as one of the promising candidates among the one-dimensional materials. It is also a typical n-type transparent semiconducting material with a
25 band gap of 3.37eV (300K) which mainly absorbs the UV light and hardly absorbs the visible light. 1-D ZnO nanocrystals have been widely used in dye-sensitized solar cells (DSSCs)[1, 2], ultraviolet laser[3], light-emitting diodes (LED)[4], and gas sensors[5]. They were synthesized by various chemical routes, including
30 chemical vapor deposition (CVD)[6], metal-organic chemical vapor deposition (MOCVD)[7], vapor-liquid-solid (VLS) process[8], and wet chemical methods[9-11]. Solution grown ZnO nanorod arrays (ZnO NRAs) can work as an efficient antireflection layers in the solar cell[12].

35 Cadium sulfide (CdS) is among the first discovered semiconductors and it is generally an n-type semiconductor with a band gap of 2.42eV (300K) in the visible light region. The remarkable fundamental properties of high transmissivity, stability and low cost make it to be used as traditional yellow
40 light optical flitters and buffer layer materials for fabricating solar cells[13-15]. Some fundamental researches have been performed on the combination of CdS nano materials and ZnO NRAs[16-18] , but there have been no reports on the CdS/ZnO NRAs composited structure applied to monocrystalline silicon (c-Si) solar cells.

45 It is commonly acknowledged that silicon is the second most abundant element in the earth's crust with a band gap of 1.12eV (300K). The polished silicon surface can reflect appropriate 30-35% of the sunlight. Generally, the front surface of crystalline silicon solar cell is textured with inverted pyramids textures to
50 reduce the reflection and make practical silicon cells [19] and then is covered with different inorganic materials such as TiO_2[20], In_2O_3[21], SiN_x[22], and ZnO[23] through CVD method to work as an antireflection film. Although crystalline silicon solar cells have been the mainstream productions for the photovoltaic market,
55 taking over 80% of total cell production in the recent years[24], their low efficiency and corresponding high costs still limit their comprehensive success for mass production until now[25].

In our research, effective CdS/ZnO NRAs as antireflection coatings for light trapping were successfully prepared by
60 chemical bath deposition. The as-prepared coating with a novel structure fabricated on the front surface of the c-Si solar cells is consisted of mass oriented vertically aligned ZnO NRAs and CdS nanoparticle layers above the ZnO layers. It shows that CdS/ZnO NRAs coatings improve the photovoltaic properties of c-Si cells,
65 compared to ZnO NRAs coatings. Furthermore, the formaiton of CdS nanoparticles can suppress the outward defect emission of ZnO NRAs so that green yellow light emission of ZnO NRAs is transmitted to c-Si solar cell.

2. Experimental

70 **2.1 Preparation of CdS/ZnO NRAs antireflection coatings on c-Si**

The CdS/ZnO NRAs antireflection coatings were deposited

uniformly on the surface of the c-Si solar cells by chemical bath solution method[26]. The surface of the Si wafer was first cleaned by rinsing with double distilled water and ethanol before deposition to eliminate any impurities. Then, the wafer was dried, and the ZnO seed layers were deposited on the surface of the Si wafers through spin-assisted deposition methods. In this step, firstly, 0.05 mL of 5 mM zinc acetate dihydrate ($Zn(CH_3COO)_2 \cdot 2H_2O$) ethanol solution were dropped onto the Si surface under the speed of 1700 rpm for 40 s, and 10 times of this operation were needed to ensure the Zn-precursor got the substrate full covered. Then the wafer was put into an oven at 200 °C for 30 min to decompose $Zn(CH_3COO)_2$ particles into ZnO seeds. The above progress was repeated to ensure the ZnO seeds were fully distributed on the silicon's surface. Secondly, the wafer was immersed into solution A which contained 0.1M $Zn(CH_3COO)_2$ and 0.1 M methenamine (HMTA). Ammonia was used to adjust the pH value of solution A to ~10. The Si wafer was immersed in solution A in a drying oven at 90°C for 20 min to form ZnO NRAs. Thirdly, the wafer was taken out and immersed in solution B which contained 0.02 M Cadmium acetate dihydrate [$Cd(CH_3COO)_2 \cdot 2H_2O$] and 0.04 M thiourea (TU) in a drying oven at 80 °C for 35 min. The color change of solution B from transparent to faint yellow indicated the formation of CdS particles. Finally, the sample was dried at 80 °C to form the c-Si solar cell with CdS/ZnO NRAs antireflection coatings.

The c-Si solar cells employed in this work were fabricated on boron-doped (100) c-Si substrates with the thickness of 200 μm by a standard commercial producing procedure including RCA clearing, surface texturization, thermal diffusion of phosphorus, phosphorus silicon glass(PSG) removal, screen printed back surface reflection, screen printed front sliver electrodes, and a high temperature firing step. All chemicals (Shanghai Chemicals Co. Ltd.) were of analytical reagent grade and used as received. All the aqueous solutions were prepared using double distilled water.

2.2 Characterization

Field emission scanning electron microscopic (FESEM, Hitachi S-4800) was used to observe the morphology of the coatings. The coatings were scraped from the surface of cell and prepared on copper grids to observe the shape of nanoparticles and nanorods using a high resolution transmission electron microscope (HRTEM, Hitachi H-80) equipped with energy dispersive X-ray (EDX) spectroscope. X-ray diffraction (XRD) analysis for ZnO NRAs and CdS /ZnO NRAs was performed on a Rigaku D/max 2000 diffractometer equipped with Cu Kα radiation (λ = 1.5405 Å) (40 kV, 40 mA). The reflection and absorption spectra of the sample were measured by ultraviolet-visible- near infrared (UV-Vis-NIR) spectrophotometer (Varian, Carry 500). Photoluminescence (PL) spectra were measured at room temperature using fluorescence spectrophotometer (Cary Eclipse Spectrophotometer) with xenon lamp as excitation source (λ = 347nm).

2.3 Cell measurement

The current destiny–voltage (J-V) characteristics of the solar cell were measured with a cell active area of 2.5×2.5 cm^2 using an electrochemical workstation (Zahner, Zennium) with two-electrode configuration under 100 mW/cm^2 calibration which is performed using a Class A AM 1.5G spectral distributed Abet Technologies Sun 2000 Solar Simulator.

3. Results and discussion

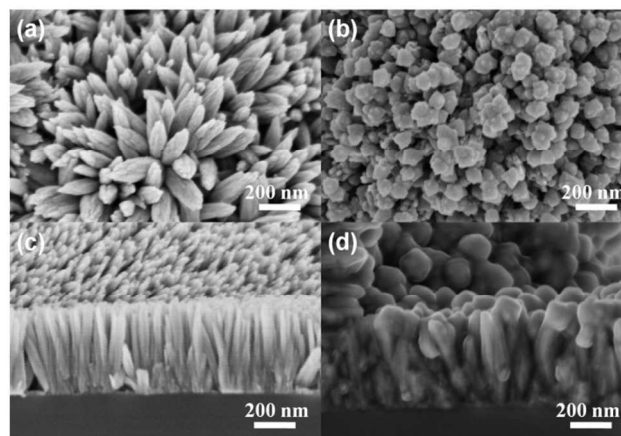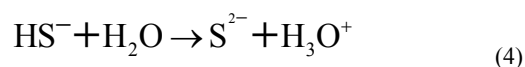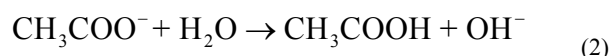### 3.1 The morphology of ZnO NRAs and CdS/ZnO NRAs

Fig. 1 FESEM images of: (a) the plan view of ZnO NRAs coatings; (b) the plan view of CdS/ ZnO NRAs coatings; (c) the cross section of ZnO NRAs coatings; (d) the cross section of CdS/ZnO NRAs coatings.

As shown in Fig. 1a, ZnO nanorods with tapered shape top were fabricated by employing CBD method. Tip shape can be observed on the top of each nanorod. After the substrate being immersed in the reaction solution containing cadmium and sulfide, the top surface of the ZnO NRAs layers was covered with CdS nanoparticles with a diameter of ~65 nm. It may be accounted that during the decomposition process of thiourea, the concentration of sulfide ions in the solution gradually increased, and when the multiplication of sulfide ions and cadmium ions was larger than the Ksp(CdS), CdS nanopricles began to depose on the top surface of the ZnO NRAs layers. The growing mechanism of CdS nanoparticles can be explained by the following reaction equation[27, 28]:

$$H_2O \rightarrow H^+ + OH^- \quad (1)$$

$$CH_3COO^- + H_2O \rightarrow CH_3COOH + OH^- \quad (2)$$

$$(NH_2)_2 CS + OH^- \rightarrow CH_2N_2 + H_2O + HS^- \quad (3)$$

$$HS^- + H_2O \rightarrow S^{2-} + H_3O^+ \quad (4)$$

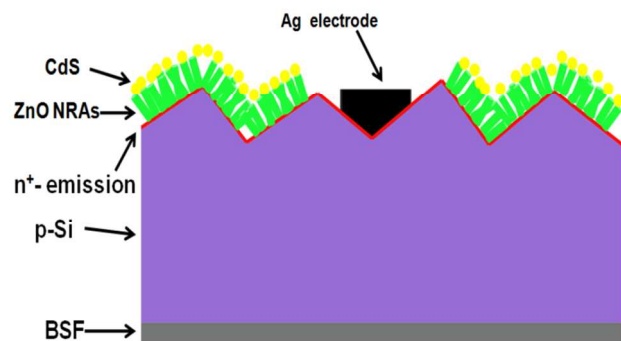$$Cd^{2+} + S^{2-} \rightarrow CdS \quad (5)$$

Fig. 2 Schematic of the c-Si solar cell configured with CdS/ZnO NRAs antireflection coatings.

As shown in the plan view of CdS/ZnO NRAs (Fig. 1b), only CdS nanoparticles can be observed. Fig. 1c shows the section cross of ZnO NRAs layer. ZnO nanorods with an average length of 380 nm and a diameter of ~40 nm grew vertically on the substrate to form ordered ZnO NRAs layer. Fig. 1d illustrates that the CdS nanoparticles on the top of the ZnO NRAs contact closely with the ZnO nanorods to form the CdS/ZnO NRAs layer. The total thickness of the CdS/ZnO NARs layer is about 450 nm. Fig. 2 displays the schematic illustration of the c-Si solar cell configured with CdS/ZnO NRAs antireflection coatings.
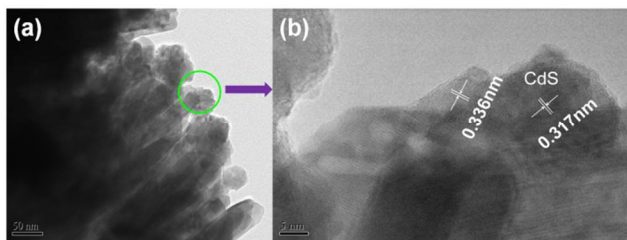

Fig.3 (a) TEM image of the CdS /ZnO nanorods scraped from the surface of the silicon solar cell and (b) HRTEM image of CdS /ZnO nanorods.

TEM and HRTEM were used to further observe the morphology of the CdS/ZnO nanorods. Fig. 3a shows the TEM image of CdS/ZnO nanorods. The structure of the nanorods was clearly recorded. It was found that ZnO nanorods were close to each other with CdS nanoparticles deposited on the top of nanorods. Fig. 3b shows the HRTEM image of CdS/ZnO nanorods. The CdS nanoparticle at the top portion of the ZnO nanorod has a polycrystalline structure with a few nanometers of finger like domain. The plane spacings of 0.317 and 0.336 nm for hexagonal CdS (101) lattice plane and CdS (002) lattice plane were clearly observed, respectively.

3.2 EDX spectrum and XRD pattern

As shown in EDX spectrum of CdS/ZnO NRAs (Fig. 4a), the sample contains the elements of Oxygen (O), Zinc (Zn), Sulfur (S), Cadmium (Cd), Carbon (C), and Copper (Cu). The Cu and C peaks can be attributed to the copper grids which are used to support the CdS /ZnO NRAs coatings. Fig. 4b shows the X-ray diffraction of the ZnO NRAs coated cell and the CdS/ZnO NRAs coated cell. Compared to the standard JCPDs card(#36-1451), the formed ZnO NRAs have hexagonal wurtzite crystal structure with a series of diffraction peaks (100), (002), (101), and (102) planes of hexagonal ZnO. The X-ray diffraction pattern of CdS/ZnO NRAs proves that CdS nanoparticles have been formed around the ZnO nanorods. A series of new peaks at around 24.8°, 26.6°, 28.1° and 51.8° correspond to (100), (002), (101) and (112) plane of the JCPDs card(#41-1049), indicating the crystalline structure of hexagonal phase CdS. Compared with the pattern of ZnO NRAs, the intensity of the XRD peaks is weak due to the short time for depositing the CdS nanoparticles on ZnO NRAs and the rough surface of the sample. Both samples have two obvious diffraction peaks which correspond to the JCPDs card (#65-2871) for the existence of silver electrodes on the front surface of the c-Si solar cell.

3.3 Reflectance and absorption spectrum

The reflection and absorption properties for the cell with ZnO NRAs and cell with CdS/ZnO NRAs in the range of 300 to 1200 nm were investigated by using a UV-Vis-NIR spectrophotometer. Fig. 5a shows the reflectance spectra for c-Si solar cell, cell with ZnO NRAs, and cell with CdS/ZnO NRAs, respectively.
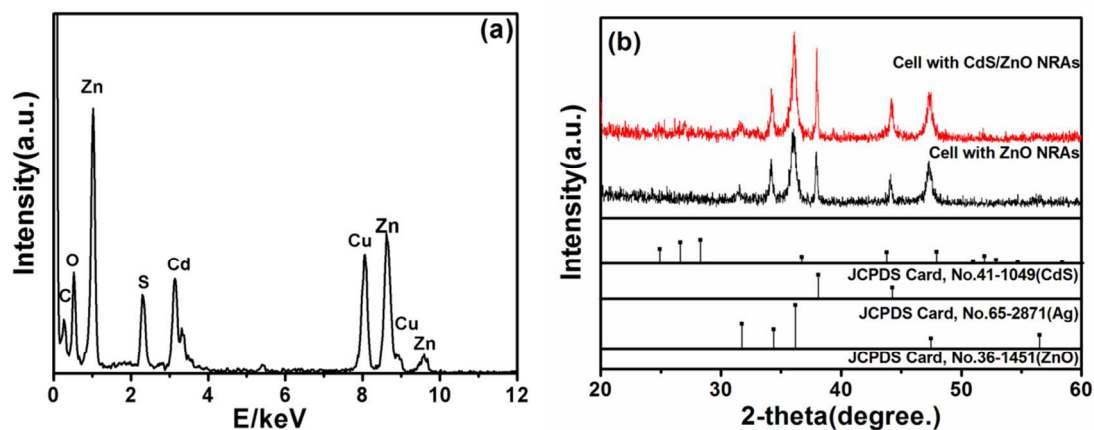

Fig. 4 (a) EDX spectrum of CdS /ZnO nanorods and (b) XRD pattern of ZnO NRAs and CdS/ZnO NRAs on silicon solar cell
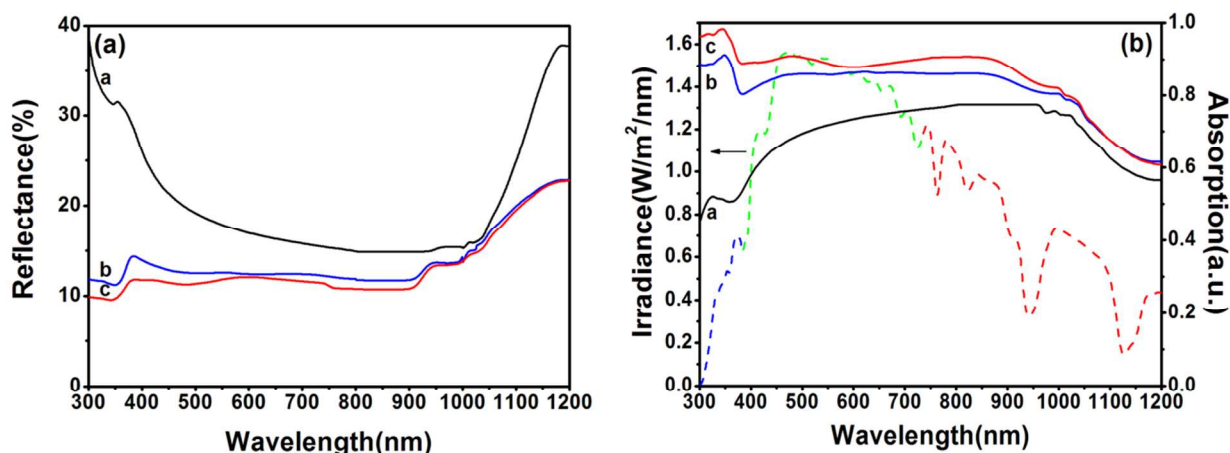
Fig. 5 (a) UV-Vis-NIR reflectance spectra and (b) UV-Vis-NIR absorption spectra with solar energy distribution spectrum in the range from 300 to 1200nm for curve (a) c-Si solar cell; curve (b) cell with ZnO NRAs; curve (c) cell with CdS/ZnO NRAs.

Silicon has an energy band gap of 1.1eV (300K), so all the curves of samples have an abruptly raise shift at the wavelength of 1100 nm. For c-Si solar cell, it has the lowest reflectance to 15% in the range of 800-900 nm. Compared to the reflectance of nearly 35% for polished silicon surface, the reduction of reflectance can be attributed to the surface texturization procedure which makes light reflection repeat between the pyramid structures on the surface of the c-Si, and thus reduces the light reflection out into the air. It was reported that ZnO NRAs can suppress broadband reflection[12]. After the deposition of ZnO NRAs, the reflectance has been significantly reduced in UV region for the reason that ZnO can absorb short wave UV light. The sample has the lowest reflectance to 11% at the wavelength of 347 nm and this is corresponding to the absorption spectrum in Fig. 5b. In the region of Vis-NIR region, the lowest reflectance of 11.7% was obtained in the range of 800-900 nm. The ZnO nanorod has a high rate of length to diameter, and this ensures more light enter the ZnO NRAs layers. Under the circumstance similar to the surface texturization procedure, light is much harder to escape from the NRAs layers. Compared to Cell with ZnO NRAs, Cell with CdS/ZnO NRAs has lower reflectance to 9.6% in the UV region and suppresses the reflectance to 10.7% in the range of 800-900nm. The enhanced antireflection properties are achieved by reason that the reflected light from the void spaces of the ZnO NRAs can re-reflect at the interface between CdS and ZnO NRAs and then pass through the inner of coatings. Thus, light is perfectly captured by the CdS/ZnO NRAs coatings though they seem to have a less roughness surface consisting of mass particles.

For both cell with ZnO NRAs and cell with CdS/ZnO NRAs, the spectra have an abrupt raise shift which is located at the wavelength of 900 nm, and this can be contributed to the silver nanoparticles deposited on the surface of the sensitized solar cell due to the solution of silver from the silver electrodes in the reaction solution[29, 30].

Fig. 5b shows the absorption spectra of the samples with the solar energy distribution spectrum in the range of 300 to 1200 nm. The solar distribution spectrum was given out according to the date of standard AM1.5 solar radiation distribution value. The maximum power peak of the sun light energy distribution spectrum is located at 475 nm. The solar light energy level in the range of 400-800 nm exceeds the half value of the maximum power, so the absorption improvement of the solar energy in this wavelength range is essential for the improvement in photovoltaic proprieties of the solar cells[31, 32].

As shown in Fig. 5b, except for c-Si solar cell, the spectral absorptions of other two samples have abrupt down shift at the wavelengths between 900 and 1100 nm. The down shift in the absorption spectrum corresponding to the raise shift in the reflection spectrum is attributed to the absorption of sliver particles and the instruct energy band gap absorption of silicon. Compared to the spectral absorption of the c-Si solar cell, the spectral absorption of the cell with ZnO NRAs is significantly improved in all the light absorption range for silicon, which is a result of the antireflection effect of the ZnO NRAs and is in accordance with the reflection spectrum in Fig. 5a. Because ZnO is a typical n-type semiconducting material with a band-gap of 3.37eV, the maximum peak of the absorption spectrum for ZnO NRAs appears at 347 nm in the UV region and an inflexion appears at 384 nm, which is in accord with the instinct absorption and emission of the free excitons in ZnO. It is expected that the optical absorption properties will be changed after the deposition of CdS particles on the ZnO NRAs since CdS has a band gap of 2.42 eV which is narrower than that of ZnO. From the light absorbance data of Fig. 5b, the absorption of the CdS/ZnO NRAs is further improved in range of 300 to 1100 nm, compared to that of ZnO NRAs. Also, except for absorption peak at 347 nm and an inflexion at 384 nm which is the same as those of ZnO NRAs, an absorption peak and an inflexion appear at 483 and 600 nm, respectively. The outcome illustrates that more light is captured by coatings, but the optical properties are affected by the energy band absorption of CdS, leading to the decrease of light absorption from 483 to 600 nm in the spectrum. In summary, the absorption proprieties of CdS/ZnO NRAs are the results of the enhanced light trapping capacity of coatings and the absorption of CdS at the center of 483 nm.
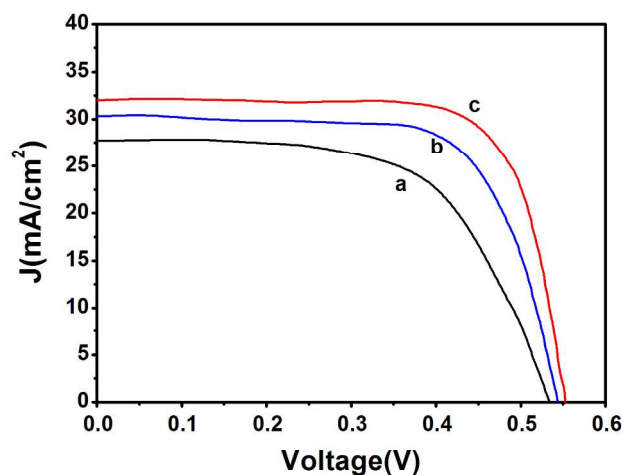
3.4 J-V character

Fig. 6 J-V character for (a) c-Si solar cell; (b) cell with ZnO NRAs; (c) cell with CdS/ZnO NRAs.

The measured J-V curves for c-Si solar cell, cell with ZnO
5 NRAs, and cell with CdS/ZnO NRAs under the air mass 1.5 global (AM 1.5G) simulator illumination condition are shown in Fig. 6. Besides, the detailed values of the photo voltage proprieties are listed in Table 1.

Table 1 Photovoltaic parameters of the c-Si solar cell with and without
10 antireflection coatings.

Sample	Jsc(mA/cm^2)	Voc(mV)	FF(%)	η(%)
c-Si solar cell	27.7	0.534	61.7	9.13
Cell with ZnO NRAs	30.3	0.544	69.8	11.5
Cell with CdS/ZnO NRAs	32.0	0.553	74.6	13.2

By compared with c-Si solar cell, a larger short circuit current density (J_{sc}) and an improved open circuit photo voltage (V_{oc}) were obtained for the cell with anti-reflectance coatings. The improvements for both photocurrent and open circuit photo
15 voltage indicate the enhanced photovoltaic proprieties. The fill-factor (FF) increasing from 61.7% to 74.6% means the decrease of series resistance. The photovoltaic conversion efficiency(η) of the c-Si solar cell increased by 26% with the effect of ZnO NRAs coatings is mainly due to the suppression of reflection of incident
20 light for optical optimization[33, 34]. Meanwhile, cell with CdS/ZnO NRAs reached maximum power conversion efficiency value of 13.2%, exhibiting a 45% enhancement compared to that of the c-Si solar cell. Since absorption is proportional to the short-circuit current density[35], this result can be mainly attributed to the
25 improved absorption properties of the CdS/ZnO NRAs coatings in the wavelength range of high density solar photon flux of 450-600nm for the reason that an absorption peak and an inflexion appeared at 483 and 600 nm in the absorption sprectum while no obvious peak was recorded in this range for cell with ZnO NRAs.

30 **3.5 PL spectrum**

Fig. 7 shows the photoluminescence (PL) spectra for c-Si solar cell, cell with ZnO NRAs, and cell with CdS/ZnO NRAs at the room temperature (300K). The c-Si solar cell showed no obvious PL emission from 350 to 600 nm.
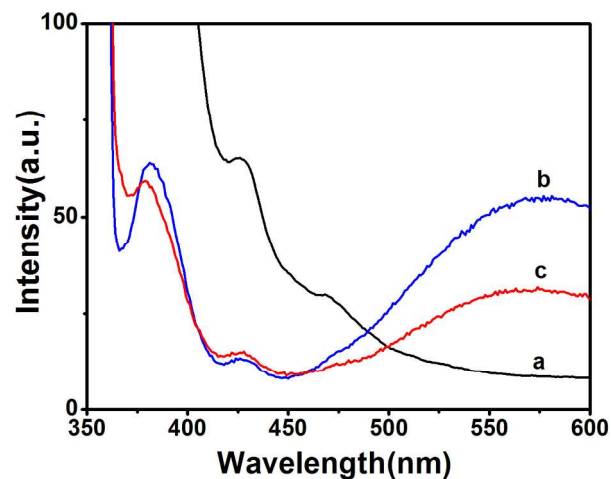
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Fig. 7 Photoluminescence (PL) spectra for (a) c-Si solar cell; (b) cell with ZnO NRAs; (c) cell with CdS/ZnO NRAs.

Two PL emission peaks for cell with ZnO NRAs were observed. The marked UV emission at 381nm corresponds to the
40 decay of free excitons in ZnO, while a broad visible luminescence emission was observed from 450 to 600 nm[36, 37]. For a typical solar cell, photoluminescence would be beneficial if short wavelength UV absorption could be shifted into the emission over 500 nm[38]. The centre of the broad visible
45 luminescence emission was at about 575 nm, which is contributed to the oxygen vacancies in the ZnO NRAs. The defect luminescence in the visible light is beneficial to the photo voltage effect of the c-Si solar cell, because c-Si has a higher light absorption in the visible region than in the UV region. The small
50 peak at 425nm is attributed to the light scatter on the silicon wafer, compared to the spectrum of the c-Si solar cell. The PL emission spectrum for CdS/ZnO NRAs is similar to that for the ZnO NRAs except the differences in the changes for location and the intensity of the luminescence emission peak[16]. A 2 nm blue
55 shift was observed in the UV region, which can be contributed to the part formation of the CdS/ZnO heterojunction. The defect luminescence of ZnO NRAs in the green-yellow region has been significantly suppressed, which is much more obvious than the decrease of the intensity of the emission in the UV region, due to
60 the visible light scatter on the CdS nanoparticles. It indicates that the emitting green–yellow light was absorbed by the c-Si for photovoltaic conversion.

4. Conclusions

In summary, oriented, densely packed CdS/ZnO NRAs
65 coatings were fabricated on the c-Si solar cell by chemical bath deposition method. The cell with CdS/ZnO NRAs coatings shows lower reflectance than the cell with only ZnO NRAs. The light absorption is enhanced with the contribution of enhanced light trapping capacity of coatings and the absorption of CdS
70 nanoparticles. CdS nanopartials can suppress the outward broad defect emission at central wavelength of approximately 575 nm of ZnO NRAs so that the emitting green-yellow light of ZnO NRAs can be absorbed by c-Si solar cell. The J-V curves obtained under the AM1.5G indicate the improved photovoltaic

properties of c-Si solar cells. It is believed that the synthesized method and the novel layer structure of the ZnO/CdS NRAs can open a new approach to exploit effective antireflection coatings for solar cells.

Acknowledgements

Notes and references

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† Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/b000000x/

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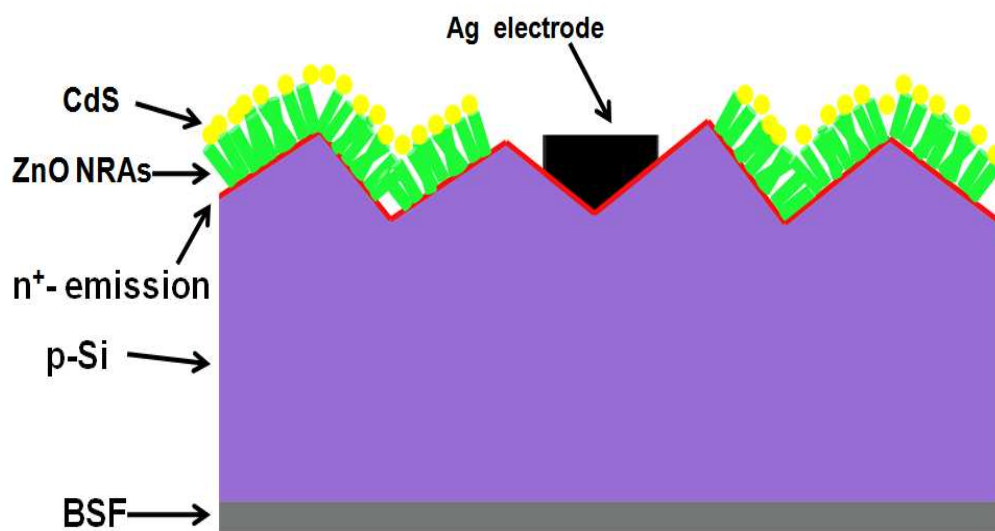
The cell with CdS/ZnO nanorod arrays (NRAs) antireflection coatings exhibits

lower reflectance and better light trapping ability than the c-Si solar cell.