



## Regular Article

# Synthesis of uniform cadmium sulphide thin film by the homogeneous precipitation method on cadmium telluride nanorods and its application in three-dimensional heterojunction flexible solar cells



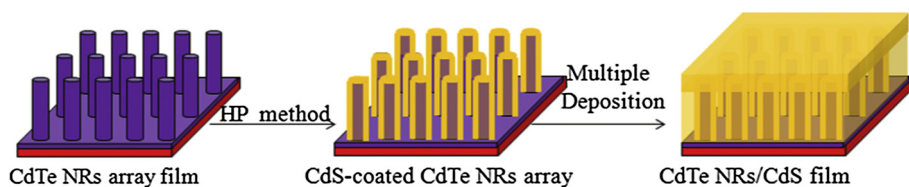
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## GRAPHICAL ABSTRACT

Homogeneous precipitation process can effectively ensure a uniform and sufficient coverage of CdS on CdTe NRs array film.



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## ABSTRACT

High-density CdTe nanorod arrays are successfully embedded in a uniform and compact CdS layer, forming a novel three-dimensional (3D) CdTe NRs/CdS heterojunction structure. The CdS layer is prepared by homogeneous precipitation (HP) method using decomposition of urea. The effects of temperature and concentration of reactants on the growth and composition of CdS film are investigated in detail. The results demonstrate that the temperature affects the thermal decomposition of urea significantly, and the concentration of  $\text{CdCl}_2$  and  $\text{CS}(\text{NH}_2)_2$  plays an essential role in the compositional ratio of CdS film. Further investigations reveal that, in comparison with the traditional precipitation method, a better coverage of CdS on the surface of CdTe NRs can be obtained by HP method due to the slow and even hydrolysis of urea. Moreover, photovoltaic performance of the novel CdTe NRs/CdS 3D photovoltaic device is also investigated. This study demonstrates that the 3D heterostructure has potential application in thin film solar cells, and the successful deposition of CdS layer on the surface of CdTe NRs by HP method suggests a promising technique for large-scale fabrication of these solar cells.

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## 1. Introduction

CdTe-based solar cells have long been of interest for terrestrial usage because of their high potential conversion efficiency (in the

range of 18–24%) with low-cost manufacturability. Due to the effective optical transmittance, wide band gap ( $\sim 2.42$  eV) and electrical properties, CdS remains the best heterojunction partner in CdTe solar cells [1–4]. The conventional CdTe thin film solar cells are usually constructed from planar junctions of p-type CdTe and n-type CdS semiconductors. These CdS–CdTe thin film solar cells have shown photoconversion efficiencies in the range of 6–22% [5].

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Various methods have been reported for the synthesis of CdS thin film such as thermal evaporation method, close spaced sublimation method, metal organic chemical-vapor deposition, electrodeposition, chemical pyrolysis deposition, successive ionic layer adsorption and reaction, and chemical bath deposition (CBD) method [6–12]. Among all the synthetic methods, CBD is considered as the best method to obtain low-cost CdS thin films that have optimal performance for photovoltaic device applications. An excellent efficiency reported for CdTe-based solar cells is obtained when CBD method is used to grow the CdS window layer [13]. The traditional CBD method is based on the slow release of  $\text{Cd}^{2+}$  ions and  $\text{S}^{2-}$  ions in an ammoniacal alkaline bath and the subsequent condensation of these ions on substrates [14–16]. This method gives little control over the precipitate particle shape and size because of the rapid change of solution concentration and the localized, discontinuous nature of the introduction of ammonia [17,18]. It is generally known that controlled synthesis of nanocrystals with specific structures and research into their structure-based properties are important subjects in nanoscience [19]. An alternative to further improve performance will highly rely on creation of different nanostructures for solar cell devices [20]. Therefore, the synthesis of CdS film with controlled particle size and uniformity is necessary to improve the performance of the solar cells even further. Recently this concept is applied to CdS–CdTe nanostructured solar cells by Javey and his co-workers. They demonstrate a photovoltaic structure that incorporates three-dimensional, single-crystalline n-CdS nanopillars, embedded in polycrystalline thin films of p-CdTe, to enable high absorption of light and efficient collection of the carriers [21,22]. However, this CdS–CdTe nanopillar solar cells are made by employing anodic aluminium oxide (AAO) membranes to produce uniform arrays of the nanopillars. Due to the rigidity of the AAO, this method is still not the most lucrative high-throughput. More importantly, the template removal usually requires harsh chemical treatment in strong base which might be detrimental to the film composition. Thus, an AAO-free method for producing the different nanostructures might be very useful for advancing the solar cells. In addition, another challenge for the 3D heterojunction solar cells is that the absorber layers are hard to deposit into the spaces among the high-density nanostructured arrays with small separations and only in contact with the top of the arrays, even though the nanostructured arrays possess desirable larger surface area.

In fact, a better control of synthesis for CdS thin film with controlled particle size and uniformity can be achieved by a template-free method only if the ligand, and precipitates are generated simultaneously and uniformly throughout the solution. Generally, this process is termed as the homogeneous-precipitation process [23]. This method has attracted much attention due to its highly uniform increase in the pH of the solution. The uniformity is achieved by gradual and even thermal decomposition of urea. When the reaction solution, in which urea has been dissolved evenly, is gently heated, a highly uniform thermal decomposition of urea occurs, and the release of ammonia into the solution becomes slow. The gradual and uniform rise in pH can result in the nucleation and growth of uniformly nanosized particles [24–27]. Many powders have been synthesized by HP method, such as ZnO [28], NiO [29],  $\text{TiO}_2$  [30], and  $\text{Y}_2\text{O}_3$  [31]. These nanostructures exhibit novel chemical and physical properties, and functionality due to their nanoscale size and uniformity. Nevertheless, it should be pointed out that there are very few reports on the synthesis of CdS film by HP method for photovoltaic application.

In this paper HP method is firstly employed to prepare CdS film on the surface of CdTe NRs in urea aqueous solution, forming a novel 3D CdTe NRs/CdS heterojunction structure. The effects of growth conditions, including temperature and concentration of reactants, on the growth of CdS film are investigated carefully. In

addition, traditional precipitation method is also applied to prepare CdS film on CdTe NRs. Through these comparisons it can be found that HP method can ensure a better coverage and growth of CdS on the surface of CdTe NRs, indicating the possibility for large scale production. What's more, the photoelectric performance of this novel CdTe NRs/CdS 3D flexible solar cell is also measured. The 3D CdTe NRs/CdS solar cell shows a short-circuit current density ( $J_{sc}$ ) of  $7 \text{ mA cm}^{-2}$  and an open-circuit voltage ( $V_{oc}$ ) of 0.37 V, yielding an overall energy-conversion efficiency of 0.88% with a fill factor (FF) of 0.34. Though the efficiency is rather low for industrial production, successful preparation of 3D heterojunction solar cells on flexible substrates demonstrates a potential application in nanostructured solar cells, and the HP method also represents a promising technique for fabricating these novel solar cells.

## 2. Experimental section

### 2.1. Preparation of 3D CdTe NRs/CdS thin film

Firstly, high density vertically CdTe NRs are synthesized on the Ni substrates by electrodeposition method. The electrodeposition process is carried out in a three electrode electrochemical cell, using the Ni substrate as the cathode, a graphite plate as the counter electrode and a saturated silver/silver chloride as the reference electrode. The fabrication of CdTe NRs has been reported in our previous study [32]. After deposition, the as-prepared CdTe films are annealed at  $300^\circ\text{C}$  for 0.5 h under high-purity nitrogen ( $\text{N}_2$ ) atmosphere, and used for deposition of CdS thin film. After heat treatment, the CdTe films are mounted vertically and immersed into the chemical bath solution. Chemical baths used for the deposition of CdS films consist of cadmium chloride ( $\text{CdCl}_2$ ), urea, thiourea ( $\text{CS}(\text{NH}_2)_2$ ) and deionized water. To study the effect of deposition temperature on the growth of CdS film, the deposition is performed for 40 min at 70, 80, 90, and  $95^\circ\text{C}$ , respectively. Similarly, in order to investigate the influence of the concentration of reactants on the synthesis of CdS film, the concentrations of  $\text{CdCl}_2$  and  $\text{CS}(\text{NH}_2)_2$  are varied while the concentration of urea ( $\sim 1.5 \text{ M}$ ) is kept constant in each experiment. The concentration of  $\text{CdCl}_2$  and  $\text{CS}(\text{NH}_2)_2$  is varied from 0.01 M and 0.1 M to 0.02 M and 0.2 M, then to 0.04 M and 0.4 M, and finally to 0.06 M and 0.6 M. After each deposition, samples are taken out of the reaction bath, washed ultrasonically to remove the loosely adhered CdS particles on CdTe NRs and finally dried in  $\text{N}_2$ . The as-synthesized CdS films are found to be well adherent to the CdTe NRs.

### 2.2. Photovoltaic device fabrication

The as-synthesized 3D CdTe NRs/CdS composite thin film on Ni substrate is employed for device fabrication. Device architecture is substrate configuration Ni/CdTe NRs/CdS/ITO/Au. Magnetron sputtered ITO layer is grown after the deposition of CdS. The ITO layer is deposited for 15 min with a power of 50 W. Au electrode are also deposited by sputtering. No antireflecting coating is used and the active area of our devices is  $3 \times 4 \text{ cm}^2$ .

### 2.3. Characterization and measurements

The morphology and elemental composition of thin films are characterized by field emission scanning electron microscope (FESEM, JEOL JSM-6700F) and energy dispersive X-ray spectroscopy (EDS). The structural properties of thin films are evaluated using Rigaku D/Max-2500V/PC (Japan) X-ray diffractometer with Cu K $\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ) in the span of angle between  $20^\circ$  and  $80^\circ$  at a scan rate of  $4^\circ \text{ min}^{-1}$ . The photocurrent density–voltage (J–V) characteristics of the solar cells are recorded with a

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