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# Interfacial engineering for high efficiency solution processed $Sb_2Se_3$ solar cells

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ABSTRACT

As a binary semiconductor, $Sb_2Se_3$ possesses appropriate band gap, high absorption coefficient and remarkable air and moisture stability, promising for light absorption material in a practical solar cell. This study demonstrates that interfacial enginnering of $TiO_2/Sb_2Se_3$ by CdS is able to considerably enhance the photovoltaic performance in planar heterojunction solar cells. Mechanistic investigations show that the optimized energy alignment upon the introduction interfacial layer is responsible for the improved device performance. With this approach, $Sb_2Se_3$ based solar cell delivers a power conversion efficiency of 3.9% with a high current density of 27.2 mA $cm^{-2}$, which is the highest efficiency in solution processed $Sb_2Se_3$ solar cells. This research provides basic understanding regarding the interfacial engineering of $Sb_2Se_3$ solar cell and practical strategy for improving the device efficiency.

## 1. Introduction

To alleviate the reliance on conventional fossil fuels, intensive efforts have been carried out to the development of new generation solar cells. The major target is to find photovoltaic materials that are cost effective with high power conversion efficiency (PCE) and long term stability. To this end, many potential materials have been examined for device application, such as $AgBiS_2$ [1], $Sb_2S_3$ [2–4], $Sb_2Se_3$ [5,6], $Sb_2S_{3-x}Se_x$ [7–9], $CuSbS_2$ [10], and SbSI [11]. As a binary semiconductor, $Sb_2Se_3$ possesses appropriate band gap of 1.1–1.3 eV, with high absorption coefficient, excellent carrier mobility and remarkable air and moisture stability [12]. According to Shockley–Queisser limit, PCE of the device based on $Sb_2Se_3$ light absorption material can be boosted to 32% [13]. The investigation on the $Sb_2Se_3$ solar cells focused primarily on the development of various synthetic methods for $Sb_2Se_3$ films, for instance, electrochemical deposition [14], thermal evaporation [5,15] and solution methods [16–18], aiming at improving the PCE. Notably, the vacuum deposition method has been demonstrated very successful for fabrication of planar heterojunction $Sb_2Se_3$ solar cells and PCE as high as 7.6% has been obtained [5,12,19,20]

. In the solution processing, the Seok group fabricated $Sb_2Se_3$ sensitized solar cells, where a Sb-Se complex was synthesized [17]. The thermal decomposition leads to $Sb_2Se_3$ sensitizers. In this research, a PCE of 3.2% was obtained, which is also the state-of-the-art efficiency in solution processed $Sb_2Se_3$ solar cells. The Tang group firstly applied hydrazine based solution method for the fabrication of $Sb_2Se_3$ thin films, a PCE of 2.26% was achieved in a planar heterojunction solar cell [16]. In addition, a few attempts have been made for the synthesis of $Sb_2Se_3$ film by thiol-amine complex method [21–23], while these studies do not report power conversion efficiency (PCE) in complete device. In this perspective, it is a challenge to fabricate $Sb_2Se_3$ absorber material together with suitable device construction for achieving high efficiency energy conversion.

Here we employ a chemical method to prepare molecular precursor solution for the synthesis of $Sb_2Se_3$. The dissolution of element antimony and selenium is conducted in a mixture of ethylenediamine (EN) and ethanethiol (EtSH). $Sb_2Se_3$ thin film is then obtained by spin coating the antimony–selenium (Sb-Se) molecular solution and subsequent thermal annealing. A planar heterojunction solar cell is then constructed. After the optimization of $Sb_2Se_3$ absorber film and embellishment of CdS as interfacial layer between compact $TiO_2$ and $Sb_2Se_3$, we identify that the hole blocking layer CdS is one of the main factors for enhancement of the device performance.

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## 2. Experimental

### 2.1. Materials

Antimony (Sb, 99.999%, Sinapharm Chemical Reagent Co., Ltd.), selenium (Se, ≥ 99.0%, Sinapharm Chemical Reagent Co., Ltd.), anhydrous ethylenediamine (En, Analytical Reagent (AR), Sinapharm Chemical Reagent Co., Ltd.), cadmium nitrate tetrahydrate , ($Cd(NO_3)_2{\cdot}4H_2O$, AR, Sinapharm Chemical Reagent Co., Ltd.) and ammonium hydroxideare ($NH_3{\cdot}H_2O$, 25–28%, Sinapharm Chemical Reagent Co., Ltd.), 2,2,7,7-tetrakis(N,N-di-p-methoxyphenylamine)-9,9-spirobifluorene (Spiro-OMeTAD, 99.8%, Yingkou Youxuan Trade Co. Ltd.), titanium (IV) isopropoxide ($C_{12}H_{28}O_4Ti$, 97%, J&K), thiourea (Tu, 98%, J&K), ethanethiol (EtSH, 98%, J&K), chlorobenzene ($C_6H_5Cl$, 99.8%, J&K), acetonitrile ($C_2H_3N$, 99.9%, J&K), lithium bis(trifluoromethylsulfonyl) imide (Li-TFSI, 98%, J&K) and 4-tert-butylpyridine (tBP, 96%, J&K) were all directly used without any further purification.

### 2.2. Preparation of precursor solutions

$Sb_2Se_3$ precursor solutions was synthesized by referring to the reported method with further modifications [21]. In specific, antimony and selenium powders with a stoichiometric ratio are dissolved in a mixed solvent of ethanediamine and ethanethiol with the volume ratio of 4:1.25. The mixture was then stirred and heated to 90 °C at least 30 min. It turns into a clear dark yellow colored solution after cooling down.

### 2.3. Solar cells fabrication

The compact $TiO_2$ layer was deposited on patterned FTO-coated glass using our reported method [24]. A thin hole blocking layer CdS thin film was prepared by traditional CBD method at 65 °C for 6 min [25]. In brief, 10 mL $Cd(NO_3)_2$ (15 mM) and 13 mL ammonium hydroxide are mixed and stirred for 2 min. Then, 6.4 mL Tu (1.2 M) and 70 mL deionized water are added into above solution in sequence under stirring.

$Sb_2Se_3$ thin film was fabricated by spin-coating $Sb_2Se_3$ precursor solution at a speed of 6000 rpm. The film was immediately pre-heated on a hot plate in $N_2$-purged glove box at 200 °C for 1 min. $Sb_2Se_3$ precursor film was then annealed at a preheated hotplate 400 °C for 2 min in glove box. Afterwards, the Spiro-OMeTAD solution prepared by dissolving 36.6 mg Spiro-OMeTAD powder, 14.5 μL tBP and 9.5 μL Li-TFSI solution (520 mg $mL^{-1}$ in acetonitrile) in 1 mL chlorobenzene was spread out on $Sb_2Se_3$ thin film and spin-coated at a speed of 3000 rpm for 30 s. Then, the Spiro-OMeTAD layer was baked at 100 °C for 10 min. Finally, the Au counter electrode was deposited by a thermal evaporator under a pressure of $5.0 \times 10^{-4}$ Pa. The active area of the device was defined as 0.12 $cm^2$.

### 2.4. Characterization and measurements

Thermogravimetric analysis (TGA) was used to study the weight loss of $Sb_2Se_3$ by a TGA Q5000 instrument under nitrogen atmosphere at a heating rate of 10 °C/min with temperature increasing from 25 °C to 800 °C. The crystallinity of samples were investigated by X-ray diffraction (XRD) using a Bruker Advance D8 diffractometer equipped with graphite-monochromatized Cu K (radiation (λ = 1.5406 Å). The surface and cross-section morphologies of the $Sb_2Se_3$ thin films were characterized by Field emission SEM (FE-SEM siron 200). UV–vis spectroscopy was characterized by UV–vis–NIR 3600 spectrometer. UPS (Synchrotron radiation photoemission spectroscopy, He I excitation, 30 eV, referenced to the Femi edge of argon etched gold) was implemented to detect the Femi level and valence band of $Sb_2Se_3$ film. The valance band (VB) spectra were measured with photon energy of 30 eV. A sample bias of − 5 V was applied to observe the secondary electron cutoff (SEC). The work function (ϕ) can be determined by the difference between the photon energy and the binding energy of the secondary cutoff edge. Capacitance–voltage (C-V) measurements were conducted by electrochemical workstation (Zahner Mess Systeme PP211) with a frequency of 10 kHz at room temperature in darkness and the AC amplitude was 5 mV. DC bias voltage was changed from −0.5 to 1.0 V. Electrochemical impedance spectroscopy (EIS) was performed using the same electrochemical workstation at a bias potential of − 0.3 V in darkness with the frequency ranging from 1 Hz to 4 MHz. Finally, current–voltage measurements of $Sb_2Se_3$ solar cell were performed in a Newport Sol3A Class AAA Solar Simulator (450 W, Oriel, model 9119). Test was under an AM1.5 illumination to produce a 100 mW $cm^{-2}$ solar irradiation at room temperature. The external quantum efficiency (EQE, Model SPIEQ. 200) was measured using a single source illumination system (halogen lamp) combined with a monochromator.

## 3. Results and discussion

In the preparation of molecular precursor solution, the volume ratio of EN and EtSH is set to 4:1.25. To seek optimal Sb/Se ratio for device fabrication based on absorption layer of $Sb_2Se_3$, Sb/Se ratios of 1:1.25, 1:1.5, and 1:1.75 in the precursor solution are investigated. It turns out that elemental antimony cannot be completely dissolved when Sb/Se ratio is lower than 1:1.5. On the other hand, when Sb/Se ratios exceeds 1:1.5, $Sb_2Se_3$ thin films show poor surface morphology and in turn leading to poor surface coverage and a relative low efficiency (Fig. S1, Fig. S5a). Therefore, we conduct the film fabrication using precursor solution with Sb/Se ratio of 1:1.5. In this case, Sb and Se powders are quickly dissolved in the mixed solvent and form optically transparent solution (Fig. 1a, inset), which is stable for months. For film deposition, the solution is spin-coated onto the $TiO_2$ compact layer coated FTO

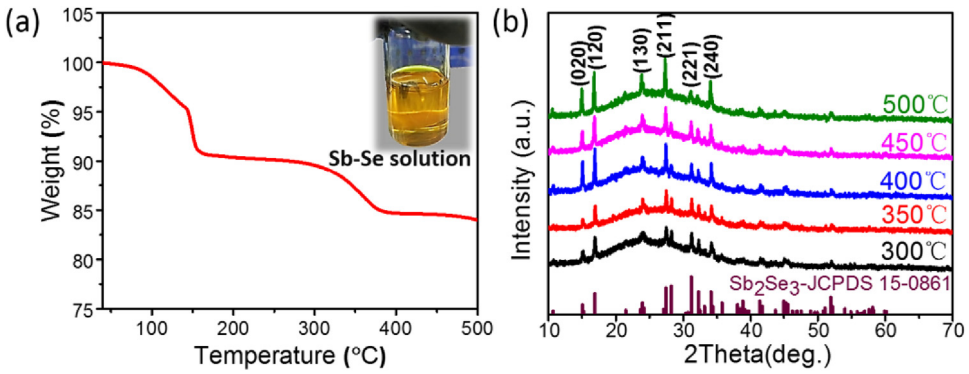


**Fig. 1.** (a) Thermogravimetric analysis of solidified Sb-Se precursor solution (inset showing Sb-Se solution) (b) X-ray diffraction patterns of the $Sb_2Se_3$ precursor thin films annealing at different temperatures from 300 °C to 500 °C in $N_2$ filled glove box.

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