Facile growth of a Sb$_2$Se$_3$ nanorods array induced by a MoSe$_2$ interlayer and its application in 3D p–n junction solar cells

Co-evaporation process was used to deposit uniformly grown Sb$_2$Se$_3$ nanorods array using a MoSe$_2$ interlayer. The MoSe$_2$ interlayer acted as a crucial role as a seed layer for the preferential growth of the Sb$_2$Se$_3$ nanorods and also improved the contact quality between the Sb$_2$Se$_3$ nanorods array and a Mo substrate. The application of the Sb$_2$Se$_3$ nanorods array to 3D p–n junction high-efficiency Sb$_2$Se$_3$ solar cells was also presented.
Facile growth of a Sb$_2$Se$_3$ nanorod array induced by a MoSe$_2$ interlayer and its application in 3D p–n junction solar cells

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A uniformly grown Sb$_2$Se$_3$ nanorod array, with the introduction of a MoSe$_2$ interlayer, obtained by a co-evaporation process and its application in three-dimensional (3D) p–n junction high-efficiency Sb$_2$Se$_3$ solar cells were investigated in this study. The MoSe$_2$ interlayer played a crucial role as a seed layer for the preferential growth of Sb$_2$Se$_3$ crystals, which facilitated the formation of a Sb$_2$Se$_3$ nanorod array regardless of the process conditions. 3D p–n junction between the Sb$_2$Se$_3$ nanorod array and the CdS buffer layer improved the short-circuit current of Sb$_2$Se$_3$ solar cells due to improved carrier transportation from the Sb$_2$Se$_3$ absorber to the CdS buffer. The MoSe$_2$ interlayer also improved the contact quality between the Sb$_2$Se$_3$ nanorod array and the Mo substrate by forming a quasi-ohmic contact, which resulted in a higher open-circuit voltage due to a reduced contact barrier and series resistance in Sb$_2$Se$_3$ solar cells. The crystal growth rate of Sb$_2$Se$_3$ was controlled by the source evaporation rate and substrate temperature to tune the final nanostructure and crystalline orientation of the co-evaporated Sb$_2$Se$_3$ nanorods array. 3D p–n junction solar cells based on an ordered and (hkl) preferentially oriented Sb$_2$Se$_3$ nanorod array showed a power conversion efficiency of 5.637%. Therefore, by including a MoSe$_2$ interlayer, it is possible to achieve high-efficiency 3D p–n junction Sb$_2$Se$_3$ solar cells.

Introduction

Antimony selenide (Sb$_2$Se$_3$) is one of the most promising semiconductor materials for thin-film solar cells because of its outstanding optoelectronic properties. Additionally, it is a non-toxic, relatively low-cost material with high absorption coefficient and low processing temperature.\(^1\)\(^-\)\(^6\) Therefore, significant research has been conducted in the past decade to replace the conventional multinary compound semiconductors in thin-film solar cells for high efficiency solar cells.\(^7\)\(^-\)\(^11\) However, the best power conversion efficiency (PCE) reported so far for Sb$_2$Se$_3$ solar cells is approximately 10%,\(^12\) which is similar to those of thin-film solar cells made of conventional materials. In order to overcome the efficiency limitation of the Sb$_2$Se$_3$ solar cells, the application of three-dimensional (3D) nanostructured Sb$_2$Se$_3$ thin films for the formation of the 3D p–n junction solar cells could be one of the promising solutions.\(^13\)\(^-\)\(^15\)

Carrier transportation and p–n junction formation in Sb$_2$Se$_3$ solar cells are closely related to the Sb$_2$Se$_3$ nanostructure. Therefore, the latter is one of the critical factors determining the photovoltaic performance of Sb$_2$Se$_3$ solar cells. However, there is little study on the nanostructure control of the Sb$_2$Se$_3$ thin films for high efficiency solar cells. Li et al. improved the short-circuit current (J$_{SC}$) of a solar cell using a 1000 nm-high Sb$_2$Se$_3$–nanorod array with superior light-harvesting capacity and reduced optical reflection.\(^16\) In our previous work, we investigated the evolution of Sb$_2$Se$_3$ nanorods by controlling the process temperature of co-evaporation and proposed the photovoltaic performance enhancement of Sb$_2$Se$_3$ solar cells based on 3D p–n junction formation.\(^17\) However, the temperature window for the formation of Sb$_2$Se$_3$ nanorods was narrow and the orientation of the Sb$_2$Se$_3$ nanorods was random. From these previously reported studies, it was found that the application of 3D nanostructured Sb$_2$Se$_3$ absorber layers would be very useful to overcome of the performance limitation of Sb$_2$Se$_3$ solar cells based on the 3D p–n junction solar cell concept. However, to the best of the authors’ knowledge, there is no detailed investigation of the nanostructure evolution of Sb$_2$Se$_3$ thin films reported in the literature.
The objective of this work was to study the evolution of a nanostructured \( \text{Sb}_2\text{Se}_3 \) thin film fabricated using a \( \text{MoSe}_2 \) interlayer and the co-evaporation process. Additionally, a detailed investigation of the controlling factor to achieve fine-tuned \( \text{Sb}_2\text{Se}_3 \) nanorod arrays is presented in this work. In our work, we proposed the evolution of nanostructured \( \text{Sb}_2\text{Se}_3 \) thin films using a \( \text{MoSe}_2 \) interlayer and detailed investigation on the control factor of the co-evaporation process for the fine-tuning of the \( \text{Sb}_2\text{Se}_3 \) nanorod array. By introducing a \( \text{MoSe}_2 \) interlayer between the \( \text{Mo} \) substrate and \( \text{Sb}_2\text{Se}_3 \), it was possible to form uniformly ordered packed \( \text{Sb}_2\text{Se}_3 \) nanorod formation using co-evaporation of \( \text{Sb}_2\text{Se}_3 \) and \( \text{Se} \) sources. The \( \text{MoSe}_2 \) interlayer has locally distributed dangling bonds, which play the role of nucleation seed for \( \text{Sb}_2\text{Se}_3 \) crystalline growth perpendicular to \( (\text{CBD}) \) of \( \text{n-type CdS} \) buffer layers on the \( \text{Sb}_2\text{Se}_3 \) nanorod array. In addition, the formation of a quasi-ohmic contact between \( \text{Sb}_2\text{Se}_3 \) and \( \text{MoSe}_2 \) was another improving point of open-circuit voltage \( (V_{\text{OC}}) \) of \( \text{Sb}_2\text{Se}_3 \) solar cells compared with \( \text{Sb}_2\text{Se}_3 \) solar cells without the \( \text{MoSe}_2 \) interlayer.

The nanostructure of the \( \text{Sb}_2\text{Se}_3 \) nanorod arrays on the \( \text{MoSe}_2 \) interlayer was also sensitively affected by the conditions of the co-evaporation processes, such as the evaporation rate of sources and substrate temperature. The growth behavior of \( \text{Sb}_2\text{Se}_3 \) crystals during the co-evaporation process was closely related with the final nanostructure of \( \text{Sb}_2\text{Se}_3 \) nanorod array, which was the main factor that determines the short-circuit current \( (J_{\text{sc}}) \) of \( \text{Sb}_2\text{Se}_3 \) solar cells. The crystalline orientation of co-evaporated \( \text{Sb}_2\text{Se}_3 \) thin films, which is a crucial factor for high efficiency \( \text{Sb}_2\text{Se}_3 \) solar cells, was also affected by the substrate temperature.\(^{18-25} \) After chemical bath deposition (CBD) of n-type CdS buffer layers on the \( \text{Sb}_2\text{Se}_3 \) nanorod array, the 3D p–n junction between \( \text{Sb}_2\text{Se}_3 \) and \( \text{CdS} \) was formed and the 3D p–n junction was effective for the enhancement of \( J_{\text{sc}} \). By introducing the \( \text{MoSe}_2 \) interlayer, the growth of a uniformly ordered \( \text{Sb}_2\text{Se}_3 \) nanorod array and the quasi-ohmic contact between \( \text{Sb}_2\text{Se}_3 \) and \( \text{MoSe}_2 \) were possible and 3D p–n junction \( \text{Sb}_2\text{Se}_3 \) solar cells with 5.637% efficiency was achieved.

### Experimental

#### \( \text{MoSe}_2 \) interlayer and \( \text{Sb}_2\text{Se}_3 \) thin-film preparation

The structure of the studied \( \text{Sb}_2\text{Se}_3 \) thin films consisted of a soda-lime glass (SLG) substrate and a 600 nm-thick Mo layer as the back-contact layer. Before deposition, the substrate was sequentially cleaned in an ultrasonic bath using acetone, methanol, and deionized water. The Mo layer was deposited on the SLG substrate via direct current (DC) magnetron sputtering using a Mo target with a purity of 99.99%.

To form the \( \text{MoSe}_2 \) interlayer on Mo substrates, the latter were treated in a tube furnace in a Se atmosphere. Vacuum \( (10^{-3} \text{ Torr}) \) was applied to the furnace to remove moisture and impurities. Then, Ar gas was introduced into the furnace, allowing the pressure inside the furnace to reach the atmospheric pressure. The furnace was heated to 430 °C with Se pellets inside for 10 min and then annealed for 10 min. The \( \text{MoSe}_2 \) layer obtained was approximately 100 nm-thick.

The \( \text{Sb}_2\text{Se}_3 \) thin films were prepared on bare Mo substrates or \( \text{MoSe}_2/\text{Mo} \) substrates using a co-evaporator system using \( \text{Sb}_2\text{Se}_3 \) and \( \text{Se} \) sources. The substrate temperature was controlled in the range of 250–290 °C. The substrate was heated at 0.53 °C s\(^{-1}\) from room temperature (\( \sim 25 °C \)) to the desired temperature. The evaporation rate of \( \text{Sb}_2\text{Se}_3 \) and \( \text{Se} \) sources varied from 50–200 Å s\(^{-1}\). \( \text{Sb}_2\text{Se}_3 \) was deposited to obtain thin films (thickness: approximately 500 nm). After deposition, the samples were allowed to cool to room temperature naturally.

#### Solar cell fabrication

Chemical bath deposition (CBD) was used to cover the \( \text{Sb}_2\text{Se}_3 \) thin films obtained with a 50 nm-thick n-type cadmium sulfide (CdS) buffer layer, and radiofrequency (RF) sputtering was used to sequentially deposit a 50 nm-thick intrinsic ZnO window layer and a 300 nm-thick Al-doped ZnO (AZO) A transparent conducting oxide (TCO) layer was deposited on top of the buffer layer to fabricate the solar cells. Thermal evaporation was then used to deposit a 1 μm-thick Al collection grid on top of the device.

#### Characterization of \( \text{Sb}_2\text{Se}_3 \) solar cells

The \( \text{Sb}_2\text{Se}_3 \) thin films were analyzed using various techniques to determine the influence of substrate temperature on the chemical composition and morphology of the films, and consequently, on the photovoltaic characteristics of the \( \text{Sb}_2\text{Se}_3 \) solar cells. The surface morphology and chemical composition of the \( \text{Sb}_2\text{Se}_3 \) thin films were observed using field emission scanning electron microscopy (FE-SEM, Hitachi, SU8020) and transmission electron microscopy (FE-TEM, Hitachi, HF-3300) with a focused ion beam (FIB) system (Hitachi, NB 5000). The crystalline structure of the \( \text{Sb}_2\text{Se}_3 \) thin films was determined using X-ray diffraction (XRD) analysis (Panalytical, Empyrean). The photovoltaic characteristics of the solar cells were characterized using a source meter (Keithley, 2400) unit and a solar simulator (Newport, 94022A) to simulate 1.5AM solar irradiation.

### Results and discussion

To obtain a controlled \( \text{Sb}_2\text{Se}_3 \) crystalline structure in \( \text{Sb}_2\text{Se}_3 \) solar cells, an \( \text{MoSe}_2 \) interlayer between the \( \text{Sb}_2\text{Se}_3 \) absorber and the Mo substrate was adopted. Additionally, the \( \text{MoSe}_2 \) interlayer between \( \text{Sb}_2\text{Se}_3 \) and the Mo substrate played an important role in the growth control of the \( \text{Sb}_2\text{Se}_3 \) crystalline structure. To investigate the role and effect of the \( \text{MoSe}_2 \) interlayer on \( \text{Sb}_2\text{Se}_3 \) crystal growth, \( \text{Sb}_2\text{Se}_3 \) thin films were deposited on bare Mo substrates and \( \text{MoSe}_2/\text{Mo} \) substrates using the co-evaporation process at different source evaporation rates (50–200 Å s\(^{-1}\)). The source evaporation rate is closely related to the \( \text{Sb}_2\text{Se}_3 \) crystal growth mechanism. Fig. 1 shows...
the cross-sectional SEM images of Sb$_2$Se$_3$ thin films with different substrates and at different evaporation rates.

In the case of co-evaporated Sb$_2$Se$_3$ thin films on bare Mo substrates, thin films deposited at a slow source evaporation rate (50 Å s$^{-1}$) showed a small and randomly oriented Sb$_2$Se$_3$ nanorod structure (Fig. 1a). However, as the source evaporation rate increased to 100 Å s$^{-1}$, Sb$_2$Se$_3$ thin films showed a densely packed columnar structure with some long Sb$_2$Se$_3$ nanorods (Fig. 1b). At a source evaporation rate of 200 Å s$^{-1}$, a flat Sb$_2$Se$_3$ thin-film structure comprising densely packed grains was observed (Fig. 1c). Generally, the nucleation rate is closely related to the evaporation rate. Thus, a easily formed continuous film under a high evaporation rate was expected. For film materials having an anisotropic crystal structure, such as Sb$_2$Se$_3$, rod-shaped film formation is highly probable when the evaporation rate is low. Under these conditions, the impingement rate of adatoms on the substrate decreases and their diffusion length may increase, causing the formation of a non-continuous film. Based on these results, authors believe that controlling the evaporation rate is critical to developing the Sb$_2$Se$_3$ rod array for 3D p–n junction solar cells.

Contrary to the co-evaporated Sb$_2$Se$_3$ thin films on Mo substrates, Sb$_2$Se$_3$ thin films prepared on MoSe$_2$/Mo substrates showed a different nanostructure. All Sb$_2$Se$_3$ thin films on MoSe$_2$/Mo substrates showed non-flat thin-film structures, regardless of the source evaporation rate. At an evaporation rate of 50 Å s$^{-1}$, thin films showed a vertically oriented narrow Sb$_2$Se$_3$ nanorod array of length 500 nm (Fig. 1d). The diameter and length of the Sb$_2$Se$_3$ nanorods increased with increasing evaporation rate from 50 to 100 Å s$^{-1}$ (Fig. 1e). At an evaporation rate of 200 Å s$^{-1}$, the thin-film showed ordered arrays of thick Sb$_2$Se$_3$ nanorods with a diameter of 100 nm (Fig. 1f). Sb$_2$Se$_3$ thin films deposited on MoSe$_2$ substrates showed a Sb$_2$Se$_3$ nanorod array structure regardless of the source evaporation rate. Additionally, the Sb$_2$Se$_3$ nanorod array nanostructure was tuned by varying the source evaporation rate. The formation of Sb$_2$Se$_3$ nanorod array on the MoSe$_2$ interlayer might be attributed to different Sb$_2$Se$_3$ crystal growth mechanisms (Fig. 2). Moreover, the MoSe$_2$ interlayer has unique anisotropic structural properties. For example, the (00l) plane of MoSe$_2$ has no surface dangling bond, making the nucleation process difficult while facilitating the diffusion and desorption of adatoms on the (00l) plane of MoSe$_2$. However, the nucleation process of Sb$_2$Se$_3$ could be easily formed on a plane having a high-dangling bond density, such as the one perpendicular to the (00l) plane. Therefore, the MoSe$_2$ interlayer was expected to provide a selective nucleation site, facilitating the formation of the Sb$_2$Se$_3$ rod array on the MoSe$_2$ interlayer. To investigate in
detail the interface between Sb$_2$Se$_3$ and MoSe$_2$, a high-angle annular detector dark-field scanning transmission electron microscopy (HADDF-STEM) image of the Sb$_2$Se$_3$/MoSe$_2$/Mo sample was observed (Fig. S3, ESI†). The MoSe$_2$ showed a two-dimensional crystal structure with weak van der Waals bonding forces and without dangling bonds in the c-plane of MoSe$_2$. Overall, the layer structure was oriented perpendicular to the Mo substrate. Interestingly, in our case, the density of the MoSe$_2$ interlayer bunch seemed relatively low. Thus, the orientation of the flexible bunch of the MoSe$_2$ layer was not perfectly aligned. The exposed-MoSe$_2$ layer on the surface consisted of a c-plane without dangling bonds and another plane with dangling bonds.

To confirm the effect of the substrate type on the morphology of the co-evaporated Sb$_2$Se$_3$ thin films, we also deposited Sb$_2$Se$_3$ thin films on indium tin oxide (ITO) substrates using the same co-evaporation process. Unlike Mo substrates, all the co-evaporated Sb$_2$Se$_3$ thin films on ITO substrates showed a similar dense thin-film morphology, regardless of the source evaporation rate (Fig. S1, ESI†) and substrate temperature (Fig. S2, ESI†). These results show that the type of substrate had a significant effect on the formation of Sb$_2$Se$_3$ nanorods by the co-evaporation process.

To evaluate the photovoltaic characteristics of the co-evaporated Sb$_2$Se$_3$ thin films on different Mo and MoSe$_2$/Mo substrates, three different Sb$_2$Se$_3$ solar cells with a planar device structure of SLG/Mo/MoSe$_2$/Sb$_2$Se$_3$/CdS/i-ZnO/AZO/Al were fabricated. Fig. 3 shows the cross-sectional TEM and energy dispersive X-ray spectroscopy (EDS) mapping images of the Sb$_2$Se$_3$ solar cells fabricated at different source evaporation rates and with different substrates. The Sb$_2$Se$_3$ solar cells without the MoSe$_2$ interlayer showed a planar structure (Fig. 3a and b), due to the flat Sb$_2$Se$_3$ thin-film structure shown in Fig. 1c. The CdS buffer, i-ZnO window, and AZO TCO layers were uniformly deposited on the flat Sb$_2$Se$_3$ absorber layer. However, the Sb$_2$Se$_3$ solar cells prepared on MoSe$_2$/Mo substrates showed completely different nanostructures than those using the Mo substrate. The Sb$_2$Se$_3$ absorber layer showed a complex nanorod array, and the CdS buffer layer was deposited on the Sb$_2$Se$_3$ nanorods conformally. The ZnO and AZO layers deposited on the rough CdS/Sb$_2$Se$_3$ interface showed irregular nanostructures. At the bottom of the Sb$_2$Se$_3$ absorber layer, a uniformly formed MoSe$_2$ interlayer was confirmed. Moreover, the source evaporation rate showed a considerable effect on the nanostructure of the Sb$_2$Se$_3$ absorber layer. The Sb$_2$Se$_3$ solar cells prepared using a 200 Å s$^{-1}$ source evaporation rate on MoSe$_2$/Mo substrates showed an irregular and agglomerated Sb$_2$Se$_3$ grain structure (Fig. 3c and d). However, when the evaporation rate decreased to 100 Å s$^{-1}$, the Sb$_2$Se$_3$ absorber layer showed a more ordered and regular nanorod array (Fig. 3e and f). These different Sb$_2$Se$_3$ nanorod array structures observed according to the source evaporation rate were consistent with the SEM images (Fig. 1e and f).

Table 1 lists the photovoltaic properties of the Sb$_2$Se$_3$ solar cells with and without the MoSe$_2$ interlayer. Despite the planar and uniform device structure of the Sb$_2$Se$_3$ solar cells without the MoSe$_2$ interlayer, the solar cells with the MoSe$_2$ interlayer showed a superior photovoltaic performance. The higher open-circuit voltage ($V_{OC}$) showed by the Sb$_2$Se$_3$ solar cells with the MoSe$_2$ interlayer might be attributed to the quasi-ohmic contact formation between Sb$_2$Se$_3$ and MoSe$_2$. The quasi-ohmic contact formation of the MoSe$_2$ interlayer is well known for copper indium gallium diselenide (CIGS) solar cells.27–31 This quasi-ohmic contact could reduce the contact barrier and series resistance of Sb$_2$Se$_3$ solar cell devices. The higher $J_{SC}$ could be related to the 3D p–n junction formation between the Sb$_2$Se$_3$ absorber and CdS buffer layers. Due to the 3D p–n junction structure, photo-generated carriers can easily escape into the CdS buffer layer. Thus, nanostructured Sb$_2$Se$_3$ solar cells show a higher $J_{SC}$ than flat thin-film Sb$_2$Se$_3$ solar cells. This superior photovoltaic performance was consistent with the results reported by Park et al. and was also attributed to the 3D p–n junction formation between the Sb$_2$Se$_3$ and CdS buffer layers.17

Between the two Sb$_2$Se$_3$ solar cells with a MoSe$_2$ interlayer, the one fabricated at an evaporation rate of 100 Å s$^{-1}$ showed higher $J_{SC}$ and fill factor (FF) than that at 200 Å s$^{-1}$, which might be attributed to the more regular and ordered 3D p–n junction formation between Sb$_2$Se$_3$ and CdS.

A previous report showed that high substrate temperatures induce the evolution of Sb$_2$Se$_3$ nanorod arrays, demonstrating the close relationship between the nanostructure of
co-evaporated Sb$_2$Se$_3$ thin films and substrate temperature. Therefore, we have tried to tune the nanostructure of co-evaporated Sb$_2$Se$_3$ thin films by controlling the substrate temperature at a source evaporation rate of 100 Å s$^{-1}$ on MoSe$_2$/Mo substrates. We also prepared co-evaporated Sb$_2$Se$_3$ thin films on bare Mo at 315°C, which is the requirement for Sb$_2$Se$_3$ nanorod array formation on bare Mo substrates. Fig. 4 shows the top and cross-sectional SEM images of co-evaporated Sb$_2$Se$_3$ thin films at different substrate temperatures on bare Mo or MoSe$_2$/Mo.

The co-evaporated Sb$_2$Se$_3$ thin films on the MoSe$_2$/Mo substrate did not show the abrupt morphology change, from the flat thin-film structure to a nanorod array, observed in our previous report on Mo substrates. Because the MoSe$_2$ interlayer already easily induces the formation of the Sb$_2$Se$_3$ nanorod array, the substrate temperature showed little effect on the formation of the Sb$_2$Se$_3$ nanorod array. However, the detailed nanostructure of the Sb$_2$Se$_3$ nanorods was affected by the substrate temperature. As the substrate temperature decreased from 290 to 250°C, the shape regularity and the degree of ordering of the Sb$_2$Se$_3$ nanorods improved. As the substrate temperature decreases, the Sb$_2$Se$_3$ nuclei density increases, suppressing the rapid growth of Sb$_2$Se$_3$ nanorods. Therefore, the growth of Sb$_2$Se$_3$ nanorods could be controlled effectively to form an ordered Sb$_2$Se$_3$ nanorod array by adjusting the substrate temperature on MoSe$_2$/Mo. Moreover, using the MoSe$_2$ interlayer, it was possible to control the nanostructure of co-evaporated Sb$_2$Se$_3$ thin films by varying both the evaporation rate and substrate temperature. Co-evaporated Sb$_2$Se$_3$ thin films on a bare Mo substrate at 315°C also showed a similar Sb$_2$Se$_3$ nanorod structure to that on MoSe$_2$/Mo substrates, which is consistent with the results from our previous work. The bare Mo substrate required a higher substrate temperature to form a similar Sb$_2$Se$_3$ nanorod array to the one obtained using MoSe$_2$/Mo substrates and the co-evaporation process.

The crystalline orientation of Sb$_2$Se$_3$ thin films is well known to be crucial for determining the photovoltaic properties of Sb$_2$Se$_3$ solar cells due to the intrinsic one-dimensional ribbon structure of Sb$_2$Se$_3$. Therefore, the crystalline structure of co-evaporated Sb$_2$Se$_3$ thin films on MoSe$_2$/Mo substrate at different substrate temperatures was analyzed by XRD analysis. All the co-evaporated Sb$_2$Se$_3$ thin films showed characteristic XRD peaks of Sb$_2$Se$_3$. However, the relative intensity of specific peaks corresponding to different crystalline orientations was different for each substrate temperature. Particularly, at 260°C where the (020) and (120) peaks were not observed, and the intensity of the (211) and (221) peaks increased (Fig. 5a). To further investigate the crystalline orientation of the co-evaporated Sb$_2$Se$_3$ thin films, their texture coefficient was calculated. At 260°C the texture coefficient of the (211) peak was remarkably higher than that at other substrate temperatures (Fig. 5b). Additionally, all other Sb$_2$Se$_3$ thin films showed relatively higher texture coefficients of (hk0) than those at 260°C. The (hk1) preferential orientation of the Sb$_2$Se$_3$ thin
film at a substrate temperature of 260 °C could help in improving the photovoltaic performance of Sb$_2$Se$_3$ solar cells.

Sb$_2$Se$_3$ solar cells were fabricated at different substrate temperatures to investigate the effect of substrate temperature of co-evaporated Sb$_2$Se$_3$ thin films on their photovoltaic performance. Fig. 6 shows the $I$–$V$ curves of the Sb$_2$Se$_3$ solar cells at a source evaporation rate of 100 Å s$^{-1}$ and different substrate temperatures, and Table 2 lists the photovoltaic properties obtained for the Sb$_2$Se$_3$ solar cells. All the Sb$_2$Se$_3$ solar cells using the MoSe$_2$ interlayer showed a $V_{OC}$ > 0.4 V, regardless of the substrate temperature. Because the MoSe$_2$ interlayer already improved the $V_{OC}$, the structural change in the Sb$_2$Se$_3$ nanorods had little effect on the $V_{OC}$. The Sb$_2$Se$_3$ solar cell using a bare Mo substrate showed a slightly lower $V_{OC}$ (0.37 V) than that with MoSe$_2$/Mo substrates. These results show that the introduction of the MoSe$_2$ interlayer significantly improves the $V_{OC}$ of Sb$_2$Se$_3$ solar cells. The Sb$_2$Se$_3$ solar cell prepared at a substrate temperature of 260 °C showed the highest $J_{SC}$. This might be attributed to the (hkl) preferential orientation of the Sb$_2$Se$_3$ absorber layer. Moreover, the ordered and regular Sb$_2$Se$_3$ nanorod array could help increase the area of the 3D p–n junction between Sb$_2$Se$_3$ and CdS, improving the $J_{SC}$ of the Sb$_2$Se$_3$ solar cells. By the way, it showed comparable $J_{SC}$ (25.138 mA cm$^{-2}$) to those of the Sb$_2$Se$_3$ solar cell with MoSe$_2$/Mo substrates because of the densely packed Sb$_2$Se$_3$ nanorod array. Interestingly, Sb$_2$Se$_3$ solar cells using a

![Fig. 6](image_url) $I$–$V$ curves of Sb$_2$Se$_3$ solar cells at a source evaporation rate of 100 Å s$^{-1}$ and different substrate temperatures: (a), 250; (b), 260; (c), 270; and (d), 290 °C using MoSe$_2$/Mo and bare Mo substrates.

Move the source evaporation rate of 100 Å s$^{-1}$, and Table 2 lists the photovoltaic properties of the co-evaporated Sb$_2$Se$_3$ nanorod array by temperature is remarkable (Fig. 4), which might be closely related to the $J_{SC}$ and $FF$ of Sb$_2$Se$_3$ solar cells. At 250 °C, the width of Sb$_2$Se$_3$ nanorods is smaller than that at 260 °C, which is disadvantageous for sufficient light absorption. The length uniformity of Sb$_2$Se$_3$ nanorods at 270 and 290 °C is inferior compared to that at 260 °C. The irregular nanostructure of the Sb$_2$Se$_3$ nanorods induces insufficient light absorption and photo-generated carrier transportation. The thinner rods (250 °C) and increased empty space (270 °C) could deteriorate the FF by increasing the shunt paths too.

To investigate the correlation between the 3D p–n junction structure and the photovoltaic properties of Sb$_2$Se$_3$ solar cells, TEM analysis of the best Sb$_2$Se$_3$ solar cell, with the evaporation rate of 100 Å s$^{-1}$ and the substrate temperature of 260 °C, was performed. Fig. 7 shows the TEM-EDS mapping images of the Sb$_2$Se$_3$ solar cell device with a nanorod array with a PCE of 5.637% (Table 2). The Sb$_2$Se$_3$ solar cell using a MoSe$_2$/Mo substrate showed a 1.225% higher PCE than the one using a bare Mo substrate, which might be attributed to the improved $V_{OC}$. A densely packed, uniformly distributed, Sb$_2$Se$_3$ nanorod array and the 3D p–n junction between CdS and Sb$_2$Se$_3$ nanorods were observed. These properties are advantageous for the enhancement of $J_{SC}$. Additionally, the uniform MoSe$_2$ interlayer between the Mo substrate and the Sb$_2$Se$_3$ nanorod array is beneficial for the formation of a quasi-ohmic contact between the layers.

![Fig. 7](image_url) TEM-EDS mapping images of Sb$_2$Se$_3$ nanorod array solar cell with PCE of 5.637%.

### Table 2: Photovoltaic characteristics of Sb$_2$Se$_3$ solar cells using MoSe$_2$/Mo and bare Mo substrates at different temperatures (T). Open-circuit voltage ($V_{OC}$), short-circuit current ($J_{SC}$), fill factor (FF), power conversion efficiency (PCE), and series and shunt resistances ($R_s$, $R_{sh}$)

| Substrate | $V_{OC}$ [V] | $J_{SC}$ [mA cm$^{-2}$] | FF | $R_s$ [$\Omega$ cm$^2$] | $R_{sh}$ [$\Omega$ cm$^2$] |
|-----------|--------------|--------------------------|----|----------------|--------------------------|
| MoSe$_2$  | 250          | 0.40 22.167              | 43.99 3.886 5.48 | 67.59                    |
| MoSe$_2$  | 260          | 0.43 27.430              | 47.35 5.637 5.89 | 118.26                   |
| MoSe$_2$  | 270          | 0.42 23.834              | 44.47 4.464 6.04 | 80.89                    |
| MoSe$_2$  | 290          | 0.45 23.661              | 40.27 4.288 8.49 | 70.32                    |
| Mo         | 315          | 0.37 25.138              | 47.11 4.412 5.69 | 112.25                   |
The presented results in this work reveal that the MoSe2 interlayer between a Mo substrate and a Sb2Se3 absorber layer improves the performance of Sb2Se3 solar cells in two relevant ways. First, the contact quality between Sb2Se3 and Mo layers improved due to quasi-ohmic contact formation, which was confirmed by the higher \( V_{OC} \) values of Sb2Se3 solar cells using MoSe2/Mo substrates compared to those using a bare Mo substrate; second, a facile formation of the Sb2Se3 nanorod array and a lower substrate temperature were achieved using the co-evaporation process, which was followed by the 3D p–n junction Sb2Se3 solar cell fabrication. Moreover, the MoSe2 interlayer played a key role in improving the photovoltaic performance of the Sb2Se3 nanorod array solar cell by increasing both \( V_{OC} \) and \( J_{SC} \). The quasi-ohmic contact caused by the MoSe2 interlayer effectively improved the contact quality between Sb2Se3 layers and Mo substrates. Moreover, the MoSe2 interlayer also played a critical role as a seed layer for the uniform growth of the Sb2Se3 nanorod array.

The Sb2Se3 solar cells prepared by co-evaporation at a source evaporation rate of 100 Å s\(^{-1}\) and substrate temperature of 260 °C on a MoSe2/Mo substrate were analyzed by an external certified laboratory to verify our PCE laboratory measurements. We calculated integrated \( J_{SC} \) from EQE data and compared with the \( J_{SC} \) of the \( J-V \) curve (Fig. 8a) and external quantum efficiency (EQE) data (Fig. 8b) showed similar results (PCE of 5.484%) to our lab-scale photovoltaic measurements. We calculated integrated \( J_{SC} \) from integrated EQE (25.89 mA cm\(^{-2}\)) and \( J_{SC} \) of the \( J-V \) curve (26.05 mA cm\(^{-2}\)).

**Conclusions**

This work investigated the role of the MoSe2 interlayer on the growth of the Sb2Se3 nanorod array and the contact quality between Sb2Se3 thin films and Mo substrates. By introducing a MoSe2 interlayer between Sb2Se3 layers and Mo substrates, uniformly ordered densely packed Sb2Se3 nanorods were successfully prepared. The MoSe2 interlayer, with locally distributed high surface energy planes, played a crucial role as a seed layer for the preferential growth of Sb2Se3 crystals perpendicular to the substrate, which facilitated the formation of a Sb2Se3 nanorod array on the MoSe2 interlayer. The latter also significantly improved the photovoltaic performance of Sb2Se3 solar cell devices by forming a quasi-ohmic contact between Sb2Se3 absorber layers and Mo substrates, resulting in higher \( V_{OC} \) and \( J_{SC} \). The nanostructure and crystalline orientation of the Sb2Se3 nanorods array were affected by the substrate temperature and the source evaporation rate during the co-evaporation process. 3D p–n junction solar cells based on an ordered and (hk1) preferentially oriented Sb2Se3 nanorods array showed a PCE of 5.637%.

**Conflicts of interest**

There are no conflicts to declare.

**Acknowledgements**

This work was supported by the DGIST R & D Programs of the Ministry of Science and ICT, Republic of Korea (21-CoE-ET-01, 21-ET-08). We thank Mr Eun and Mr Cheon at the Center for Core Research Facilities (CCRF) of DGIST for FIB sampling and STEM measurements.

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