Supporting Information

(Bi$_x$Sb$_{1-x}$)$_2$Se$_3$ thin films for short wavelength infrared region solar cells

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Supporting information S1

Table S1. Composition of (Bi\textsubscript{x}Sb\textsubscript{1-x})\textsubscript{2}Se\textsubscript{3} thin film obtained by CSS method.

<table>
<thead>
<tr>
<th></th>
<th>X\textsubscript{target} = Bi/(Bi + Sb)</th>
<th>0.00</th>
<th>0.10</th>
<th>0.30</th>
<th>0.50</th>
<th>0.65</th>
</tr>
</thead>
<tbody>
<tr>
<td>X\textsubscript{exp} = Bi/(Bi + Sb)</td>
<td>0.00</td>
<td>0.01 ± 0.003</td>
<td>0.02 ± 0.01</td>
<td>0.06 ± 0.01</td>
<td>0.07 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>Se/(Bi + Sb)</td>
<td>1.53 ± 0.06</td>
<td>1.55 ± 0.03</td>
<td>1.60 ± 0.03</td>
<td>1.50 ± 0.02</td>
<td>1.50 ± 0.06</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>X\textsubscript{target} = Bi/(Bi + Sb)</th>
<th>0.75</th>
<th>0.85</th>
<th>0.95</th>
<th>1.00</th>
</tr>
</thead>
<tbody>
<tr>
<td>X\textsubscript{exp} = Bi/(Bi + Sb)</td>
<td>0.11 ± 0.01</td>
<td>0.20 ± 0.01</td>
<td>0.30 ± 0.01</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Se/(Bi + Sb)</td>
<td>1.33 ± 0.06</td>
<td>1.45 ± 0.03</td>
<td>1.46 ± 0.05</td>
<td>1.66 ± 0.09</td>
<td></td>
</tr>
</tbody>
</table>

Supporting information S2

To quantitatively understand the behaviour of the preferred orientation, the texture coefficient (TC) of the Sb\textsubscript{2}Se\textsubscript{3} thin film has been calculated using equation (S1)

\[
TC_{hkl} = \frac{I_{hkl}}{I_{0,hkl}} \sum_{i=1}^{N} \frac{I_{h_i k_i l_i}}{I_{0,h_i k_i l_i}}
\]

(S1)

Where \(I_{hkl}\) is the intensity of the diffraction peaks corresponding to hkl reflection \(I_{0,hkl}\) is the intensity of hkl reflection of the standard Sb\textsubscript{2}Se\textsubscript{3} (JCPDS no. 051-0861) \(N\) is the number of reflections considered for the calculation.

Figure S2. Texture coefficient (TC) of the Sb2Se3 thin-film estimated for (hk0) and (hk1) family of planes
Supporting information S3

Williamson-Hall Method

Crystallite domain size and microstrain in the thin films were estimated from the XRD peak broadening.

\[ \beta_l = \frac{k\lambda}{L \cos\theta} \]  

(1)

\[ \beta_e = 4\varepsilon \tan\theta \]  

(2)

Here, wavelength of X-ray \( \lambda = 1.5406 \, \text{Å} \), \( L \) = crystallite domain size, \( K = 0.94 \), \( \beta_l \) = peak broadening due to crystallite dome size, \( \varepsilon \) = microstrain, \( \beta_e \) = peak broadening due to microstrain. \( \theta \) = Bragg diffraction angle

Total boarding in the XRD peaks is the sum of (1) and (2)

\[ \beta_{total} = \beta_l + \beta_e = \frac{k\lambda}{L \cos\theta} + 4\varepsilon \tan\theta \]

(3)

If was make a plot between \( \beta_{total} \cos\theta \) and \( 4\sin\theta \) than the intercept on the y-axis will give the crystallite domain size, and the slope will give us microstrain present in the film.

Here, \( \beta_{total} \) is the full width at half maximum after subtracting the instrument broadening \( \beta_i \).

The instrument broadening \( \beta_i \) was estimated after subtracting the FWHM of the standard silicon reference sample as per the (4). Here \( \beta \) is the FWHM of the XRD peaks of (Bi\(_x\)Sb\(_{1-x}\))\(_2\)Se\(_3\) thin film
\[ \beta_{\text{total}} = \sqrt{(\beta - \beta_i)(\beta^2 - \beta_i^2)} \quad (4) \]

Figure S3. The plots of \( \beta_{\text{total}} \cos \theta \) versus \( 4 \sin \theta \) for \((\text{Bi}, \text{Sb})_{1-x} \text{Se}_3\) thin films. Strain is extracted from the slope, and crystallite size is extracted from the y-intercept.

**Supporting information S4**

Figure S4. Raman spectrum of the Bi\(_2\)Se\(_3\) thin film. Inset is the vibrational modes of Bi\(_2\)Se\(_3\) [1-2].


Supporting information S5

The optical bandgap of the (Bi$_x$Sb$_{1-x}$)$_2$Se$_3$ thin films was estimated by measuring the diffuse reflectance on UV-Vis-NIR spectrophotometer.

Using Kubelka-Munk (KM) function the reflectance spectrum can be transformed to the corresponding absorption spectrum using eq. (1) [1]

\[ F(R) = \frac{\alpha}{S} = \frac{(1 - R)^2}{2R} \]

Using Tauc method the energy dependent absorption coefficient (\(\alpha\)) can be expressed by following eq. (2) [2-3]

\[ (\alpha hv)\frac{1}{n} = B(hv - E_g) \]

Combining eq1 and eq2

\[ \left[F(R)hv\right]^{\frac{1}{n}} = B(hv - E_g) \]

The value of n is equal to 1/2 or 2 for the direct and indirect transition bandgaps, respectively.

The bandgaps were estimated by a linear fit of the linear part of the KM plot. Errors for the bandgaps are estimated by shifting the fitting range by ± 10 meV.
Figure S5. (a) Measured diffused reflectance and (b) and (c) shows the corresponding KM function for the estimation of direct and indirect bandgap, respectively.


Supporting information S6

Table S6. (BiₓSb₁₋ₓ)₂Se₃ thin film solar cell performance parameters. PCE is power conversion efficiency, Jₛₑ is short circuit current, FF is fill factor, Rₛ is shunt resistance, Rₛ is series resistance.

<table>
<thead>
<tr>
<th>xₑₜ</th>
<th>Jₛₑ (mA/cm²)</th>
<th>Vₛₑ (mV)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
<th>Rₛ (Ω·cm²)</th>
<th>Rₛ (Ω·cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>0.68 ± 0.28</td>
<td>187.42 ± 46.21</td>
<td>31.04 ± 2.64</td>
<td>0.68 ± 0.28</td>
<td>25.75 ± 8.59</td>
<td>10.68 ± 2.52</td>
</tr>
<tr>
<td>Best cell</td>
<td>14.19</td>
<td>244.95</td>
<td>35.23</td>
<td>1.23</td>
<td>31.33</td>
<td>8.40</td>
</tr>
<tr>
<td>0.01</td>
<td>3.14 ± 0.64</td>
<td>39.33 ± 5.23</td>
<td>23.74 ± 0.22</td>
<td>0.03 ± 0.009</td>
<td>12.46 ± 1.39</td>
<td>13.31 ± 1.42</td>
</tr>
<tr>
<td>Best cell</td>
<td>3.67</td>
<td>45.4</td>
<td>23.57</td>
<td>0.039</td>
<td>11.99</td>
<td>12.78</td>
</tr>
<tr>
<td>0.02</td>
<td>21.72 ± 1.93</td>
<td>139.96 ± 5.9</td>
<td>28.79 ± 0.69</td>
<td>0.88 ± 0.10</td>
<td>8.32 ± 0.65</td>
<td>4.81 ± 0.59</td>
</tr>
<tr>
<td>Best cell</td>
<td>22.73</td>
<td>146.04</td>
<td>29.15</td>
<td>0.97</td>
<td>8.41</td>
<td>4.56</td>
</tr>
<tr>
<td>0.06</td>
<td>16.16 ± 1.03</td>
<td>136.45 ± 0.0354</td>
<td>27.90 ± 1.43</td>
<td>0.633 ± 0.216</td>
<td>10.19 ± 2.88</td>
<td>6.33 ± 1.09</td>
</tr>
<tr>
<td>Best cell</td>
<td>17.14</td>
<td>169.52</td>
<td>29.63</td>
<td>0.861</td>
<td>13.11</td>
<td>6.53</td>
</tr>
<tr>
<td>0.07</td>
<td>18.68 ± 8.71</td>
<td>59.87 ± 0.01</td>
<td>25.23 ± 0.33</td>
<td>0.27 ± 0.11</td>
<td>4.39 ± 2.59</td>
<td>4.03 ± 2.29</td>
</tr>
<tr>
<td>Best cell</td>
<td>27.78</td>
<td>54.34</td>
<td>24.94</td>
<td>0.376</td>
<td>2.00</td>
<td>1.88</td>
</tr>
</tbody>
</table>