

## Electronic Supplementary Information

### Self-Oriented Sb<sub>2</sub>Se<sub>3</sub> Nanoneedle Photocathodes for Water Splitting Obtained by Simple Spin-Coating Method

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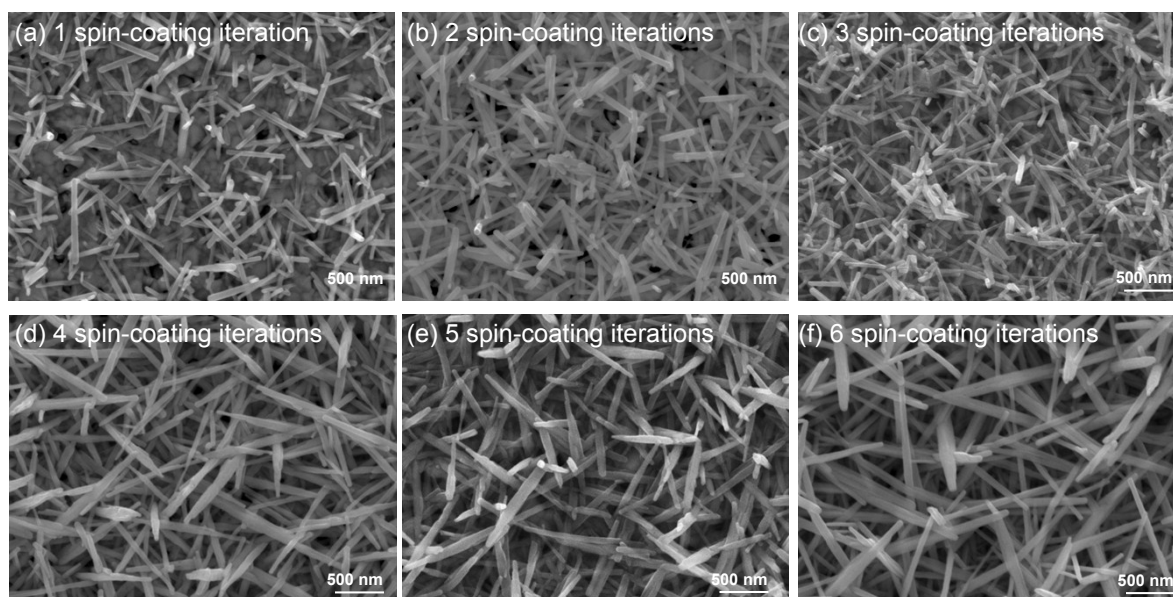
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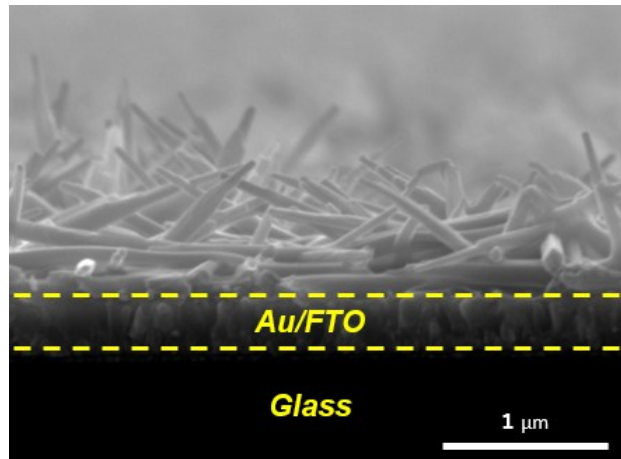
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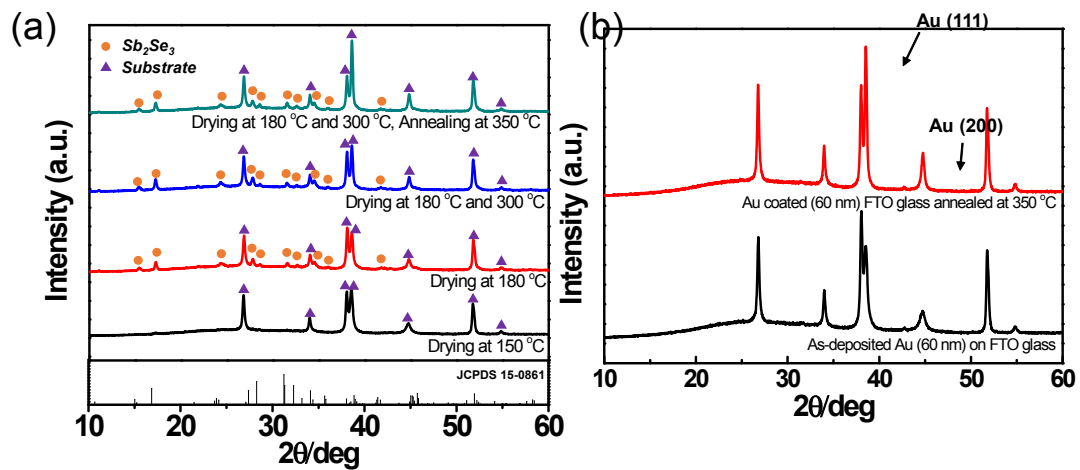
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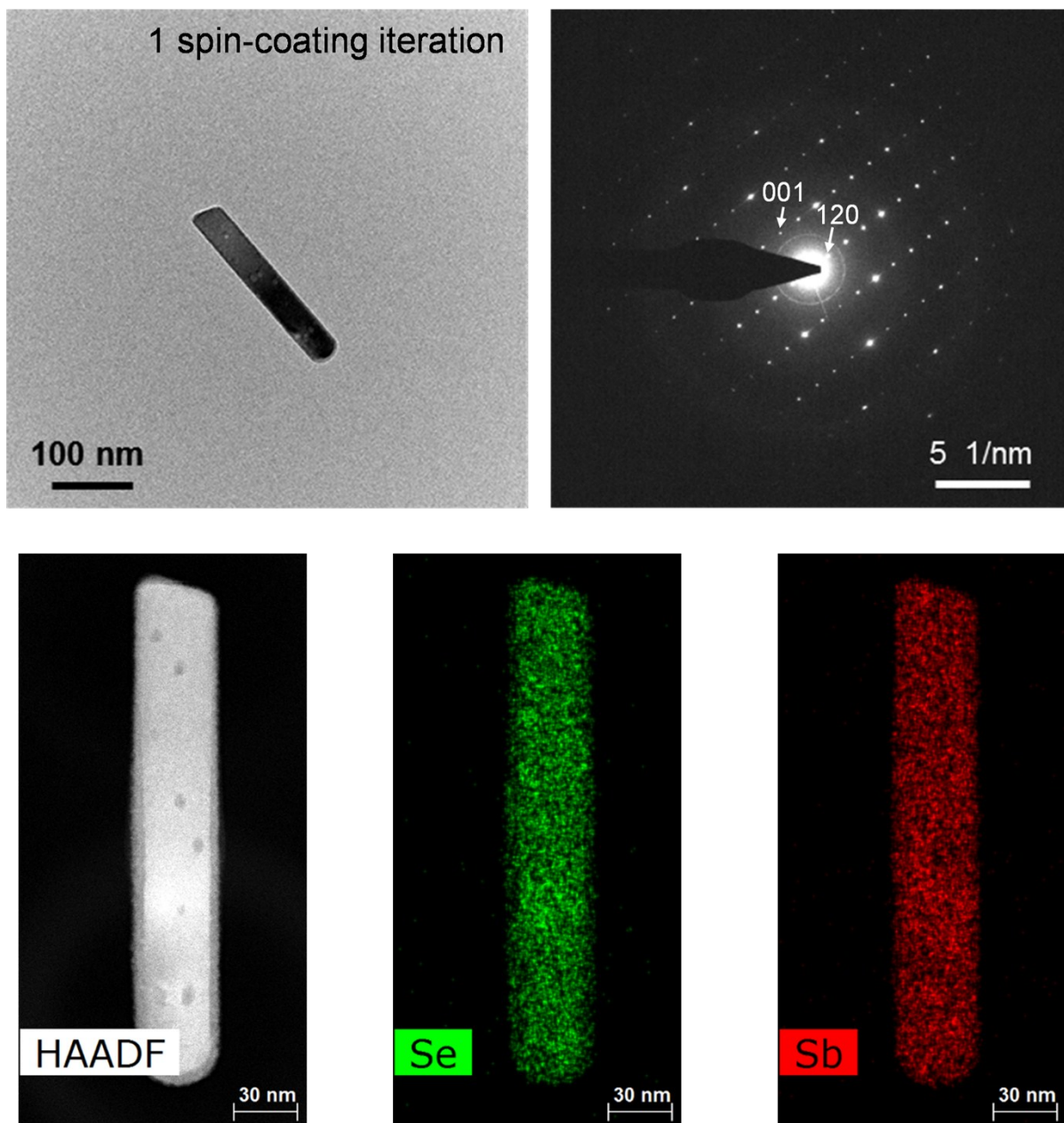
**Fig. S1** Top-view SEM images of  $\text{Sb}_2\text{Se}_3$  nanoneedles after different numbers of spin-coating iterations.



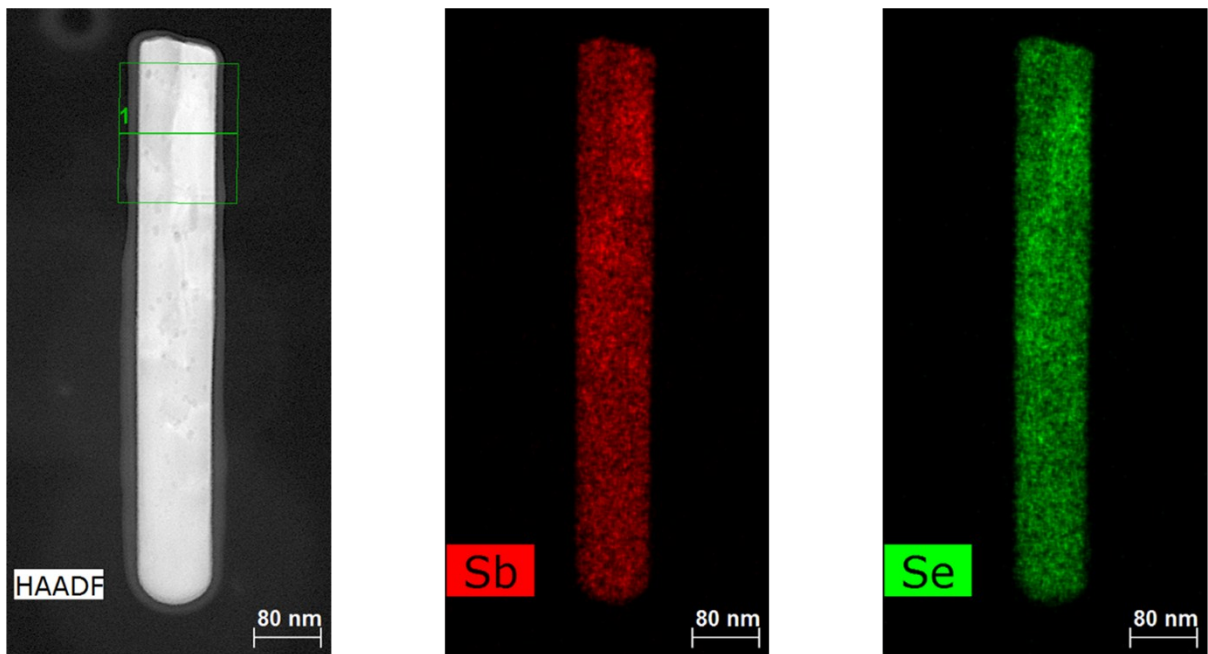
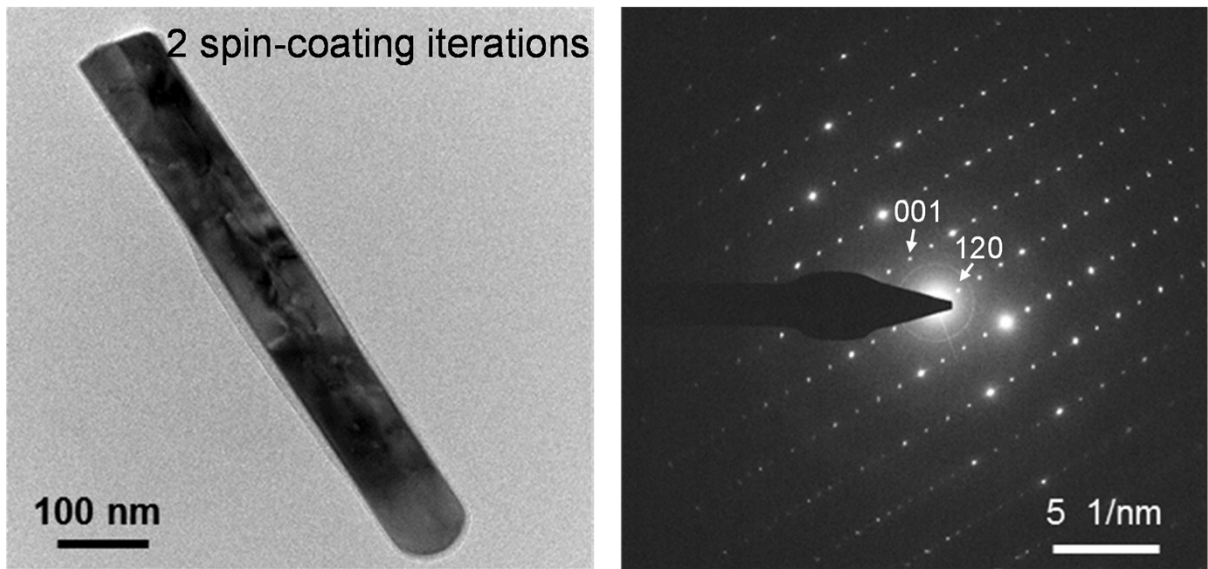
**Fig. S2.** Cross-sectional SEM image of grass-like  $\text{Sb}_2\text{Se}_3$  nanoneedle arrays directly grown from the as-spin-coated precursor film.



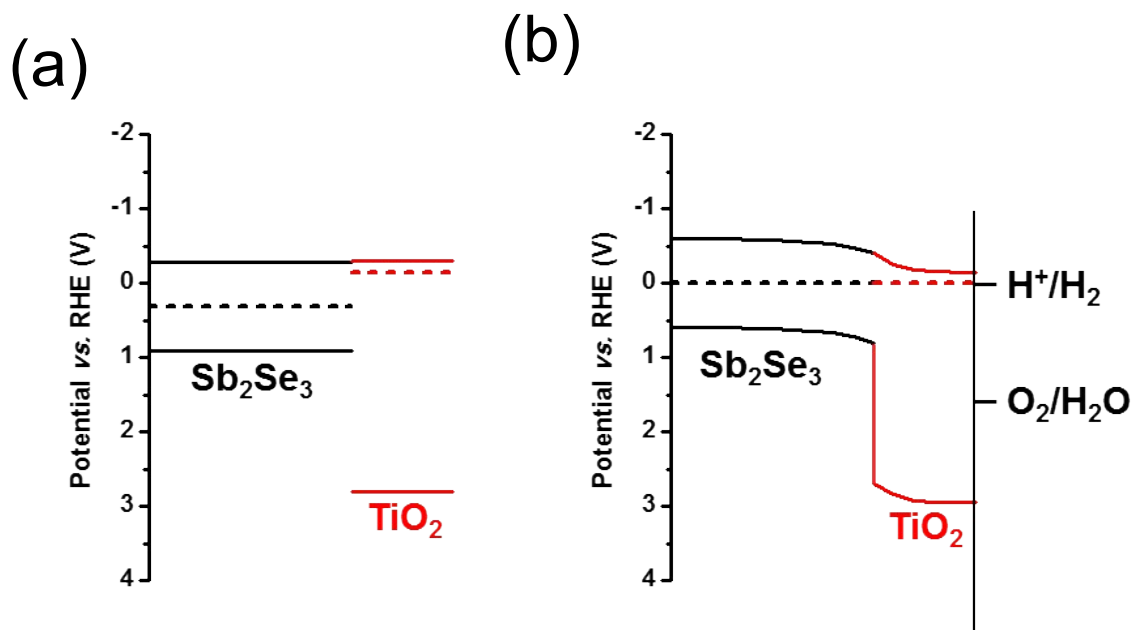
**Fig. S3** XRD analysis of (a)  $Sb_2Se_3$ /Au (60 nm)/fluorine-doped tin oxide (FTO) as a function of annealing temperature and (b) as-deposited Au (60 nm) on FTO glass and Au-coated (60 nm) FTO glass after annealing at 350 °C.



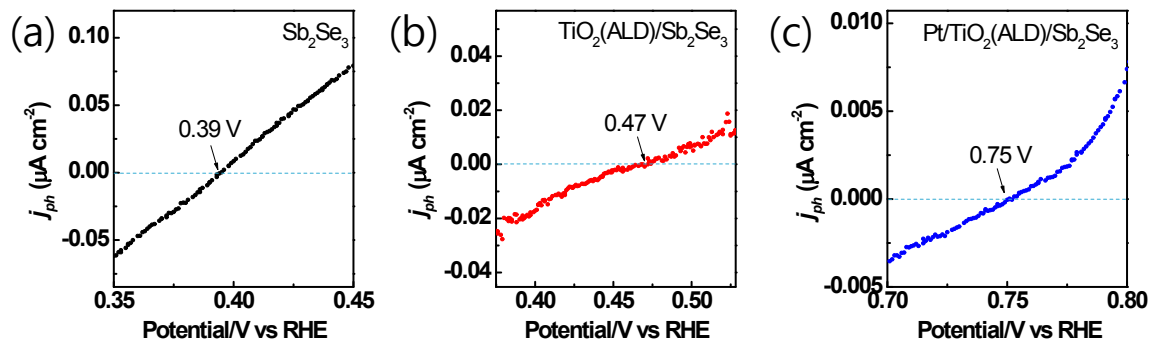
**Fig. S4** Low-resolution TEM image and the corresponding selected area electron diffraction (SAED) pattern as well as high-angle annular dark-field (HAADF) and energy-dispersive spectroscopy (EDS) elemental mapping for single  $\text{Sb}_2\text{Se}_3$  nanorod after one spin-coating iteration.



**Fig. S5** Low-resolution TEM image and corresponding SAED pattern as well as HAADF and EDS elemental mapping for single  $\text{Sb}_2\text{Se}_3$  nanorod after two spin-coatings.

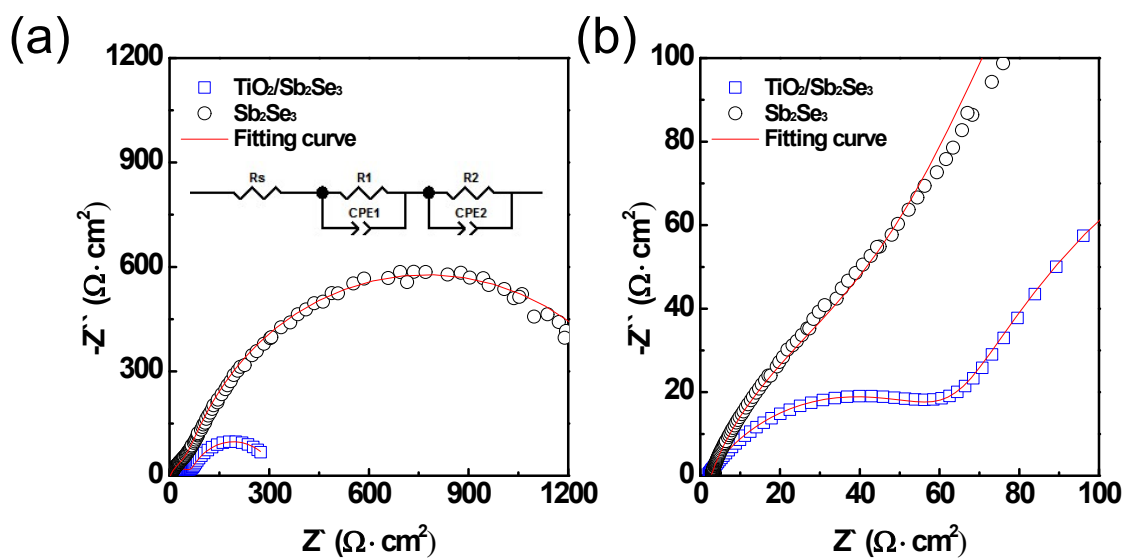


**Fig. S6.** Schematics of band diagram depicting the energetics of the  $\text{TiO}_2/\text{Sb}_2\text{Se}_3$  photocathode before (a) and after (b) equilibrium in the electrolyte.



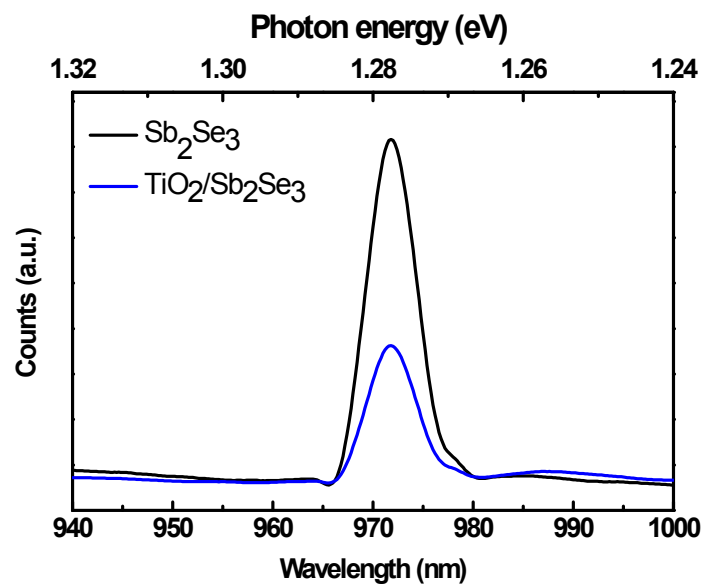
**Fig. S7** Onset potential measurement for (a)  $\text{Sb}_2\text{Se}_3$ , (b)  $\text{TiO}_2/\text{Sb}_2\text{Se}_3$ , and (c)  $\text{Pt}/\text{TiO}_2/\text{Sb}_2\text{Se}_3$  determined by obtaining the intercept between the  $j_{ph}$  versus  $E$  curve and the  $x$  axis.



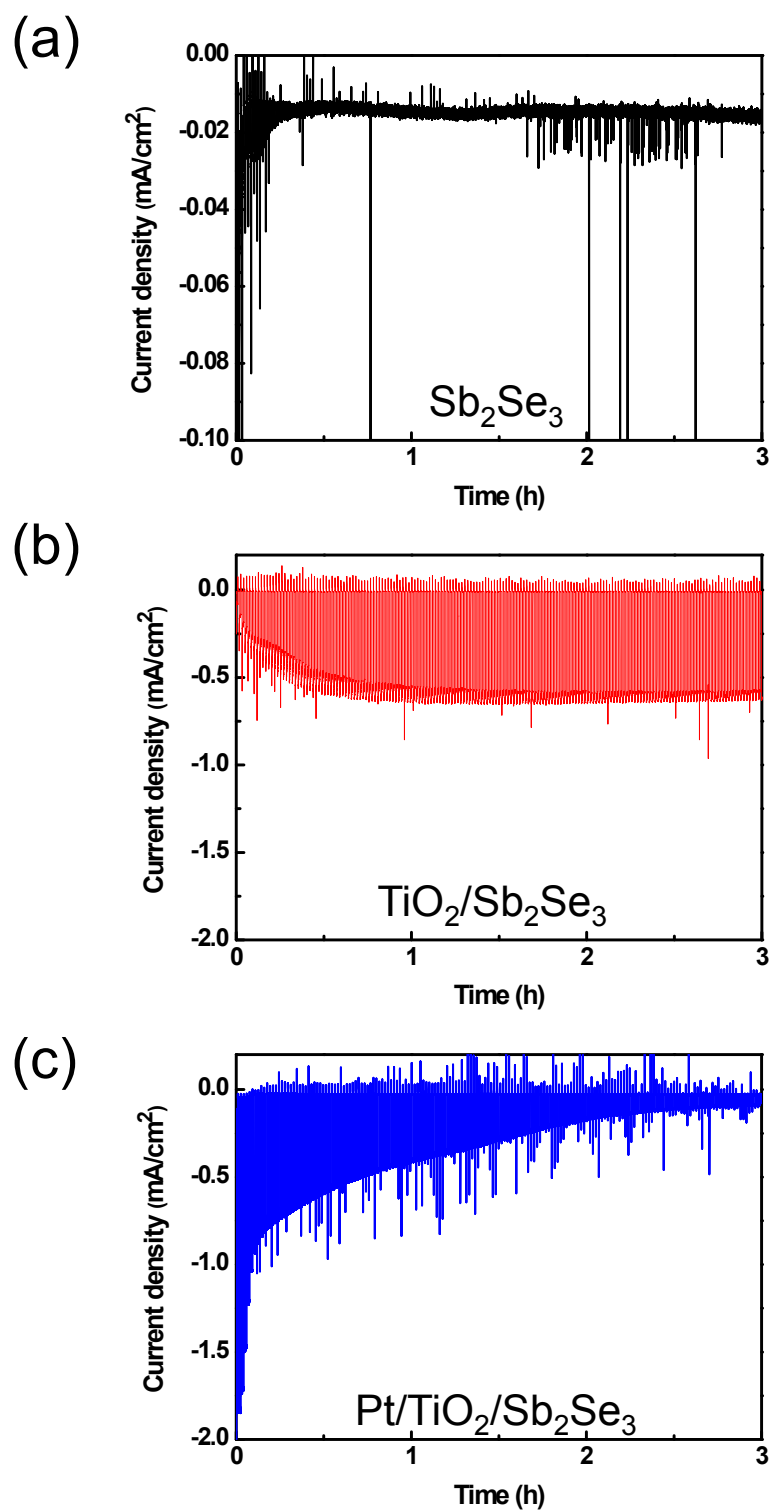


	$R_S$ ( $\Omega \text{ cm}^{-2}$ )	$R_1$ ( $\Omega \text{ cm}^{-2}$ )	$CPE_1$ ( $\text{F cm}^{-2}$ )	$R_2$ ( $\Omega \text{ cm}^{-2}$ )	$CPE_2$ ( $\text{F cm}^{-2}$ )
$\text{Sb}_2\text{Se}_3$	2.75	48.4	$1.39 \times 10^{-4}$	1558	$1.77 \times 10^{-4}$
$\text{TiO}_2/\text{Sb}_2\text{Se}_3$	2.1	65.1	$2.91 \times 10^{-4}$	253	$3.92 \times 10^{-3}$

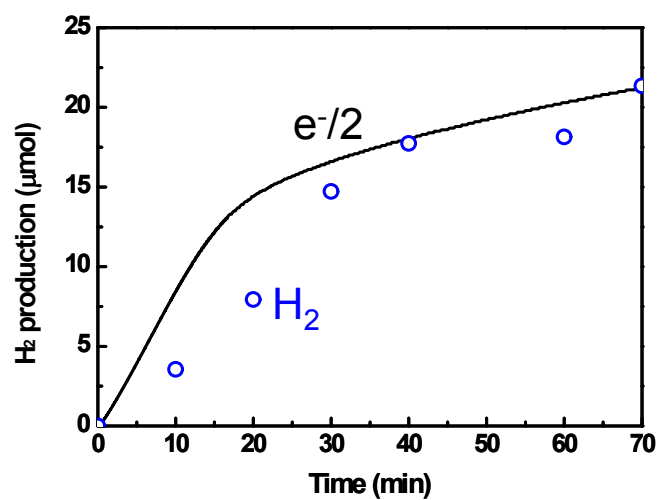
**Fig. S8** Nyquist plots of  $\text{Sb}_2\text{Se}_3$  and  $\text{TiO}_2/\text{Sb}_2\text{Se}_3$  under simulated solar light illumination: (a) full scale range and (b) enlarged spectra in high-frequency range.



**Fig. S9** Photoluminescence emission spectra of  $\text{Sb}_2\text{Se}_3$  and  $\text{TiO}_2/\text{Sb}_2\text{Se}_3$ .



**Fig. S10** Current–time curves for (a)  $\text{Sb}_2\text{Se}_3$ , (b)  $\text{TiO}_2/\text{Sb}_2\text{Se}_3$ , and (c)  $\text{Pt}/\text{TiO}_2/\text{Sb}_2\text{Se}_3$  photoelectrodes submerged in 0.5 M  $\text{H}_2\text{SO}_4$  solution at 0  $V_{\text{RHE}}$  under illumination.



**Fig. S11.** Time course curve of H<sub>2</sub> evolution over the Pt/TiO<sub>2</sub>/Sb<sub>2</sub>Se<sub>3</sub> photocathode under simulated sunlight (AM 1.5G) at 0 V vs RHE. The solid line denotes the time course curve for one-half of the electrons passing through the outer circuit ( $e^-/2$ ).