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**Supporting Information** 

For

## Interfacial Engineering Enabling Solution - Processed Cu: NiO<sub>x</sub>/Sb<sub>2</sub>Se<sub>3</sub>/TiO<sub>2</sub>/Pt Photocathode for

## Highly Efficient Photoelectrochemical Water – Splitting

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Fig. S1 XRD of Si/NiO<sub>x</sub> (black) and Si/Cu: NiO<sub>x</sub> (red).



Fig. S2 UV-vis absorbance spectra for the three different spin coating times of Sb<sub>2</sub>Se<sub>3</sub>: Sb<sub>2</sub>Se<sub>3</sub>-2

(black), Sb<sub>2</sub>Se<sub>3</sub>-3 (red), and Sb<sub>2</sub>Se<sub>3</sub>-5 (blue).



Fig. S3 LSV curves: (a) Spin-coating times of bare-Sb<sub>2</sub>Se<sub>3</sub> NPs (The insertion depicts an enlarged



curve) and (b) different TiO<sub>2</sub> spin-coating times.

Fig. S4 (a) LSV curves and (b) the photocurrent under 0 V<sub>RHE</sub> of Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-x/Pt-6.



Fig. S5 LSV curves of Sb<sub>2</sub>Se<sub>3</sub>-based photocathodes with different TiO<sub>2</sub> NPs impregnation times: (a) 2

times and (b) 5 times.



Fig. S6 (a) Different Pt impregnation times and (b) the comparison of photocurrent for Sb<sub>2</sub>Se<sub>3</sub>-5,



Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3 and Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3/Pt-6.

Fig. S7 The Cross-view SEM image of Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3/Pt-6.



Fig. S8 Chronoamperometry stability measurement on the Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3/Pt-6 with a 0  $V_{RHE}$  in an

electrolyte with a pH 1 under AM 1.5G simulated sunlight.



Fig. S9 XPS spectra of (a) Sb 3d, (b) Pt 4f, and (c) Ti 2p, and (d) XRD patterns of Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3/Pt-



6 after chronoamperometry stability test.

Fig. S10 (a-b) Top-view SEM image and (c-d) cross-view SEM images of Sb<sub>2</sub>Se<sub>3</sub>-5/TiO<sub>2</sub>-3/Pt-6 before



(first row) and after (second row) chronoamperometry stability measurement.

Fig. S11 The UPS spectra for FTO/NiO<sub>x</sub> and FTO/Cu: NiO<sub>x</sub>, obtained using He I radiation at 21.21 eV

for determination: (a)  $E_{\text{cutoff}}$  and (b)  $E_{\text{edge}}$ .