Supporting Information

A Pseudo-Metal-Free Strategy for Constructing High Performance Photoelectrodes

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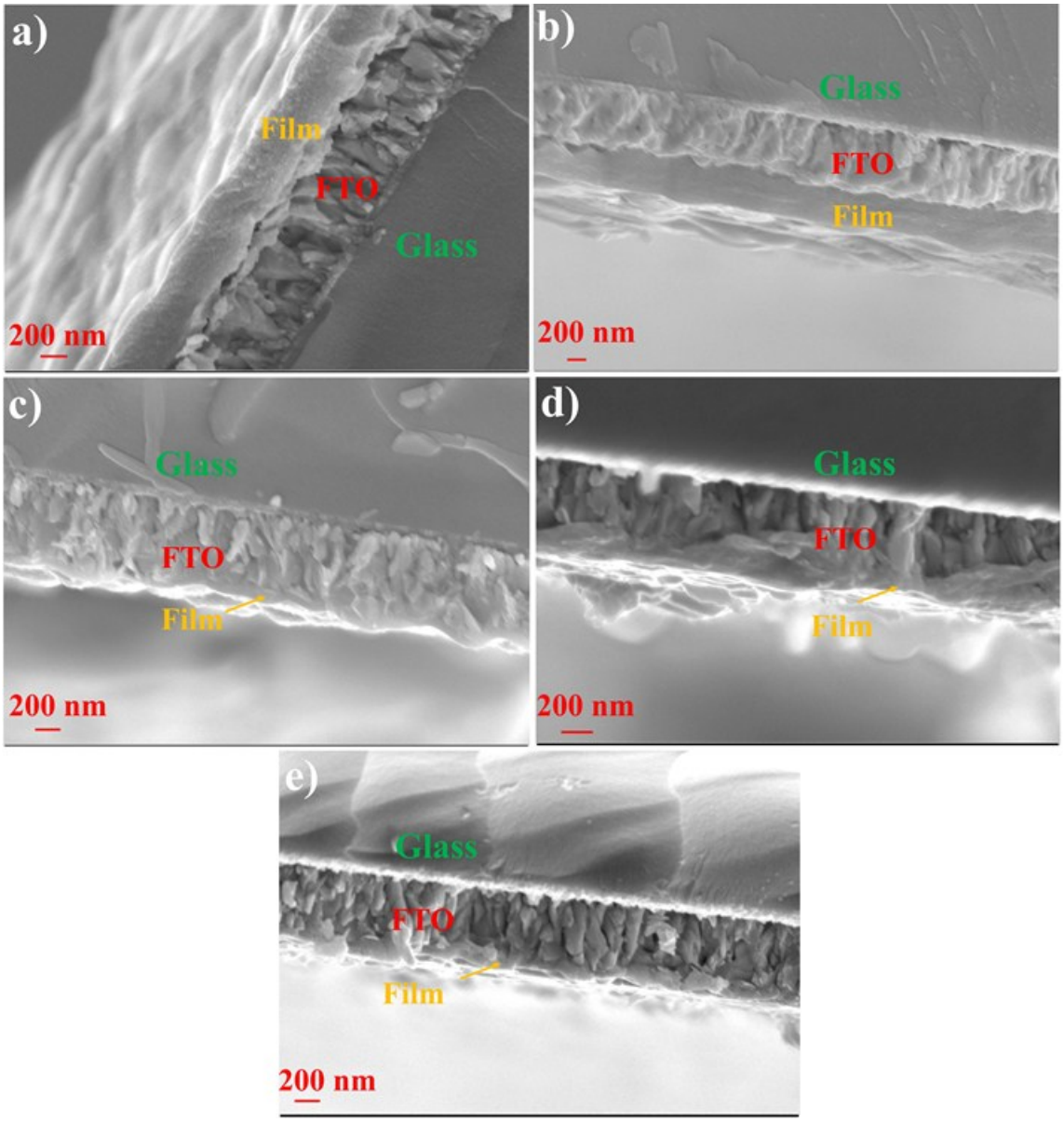

Figure S1. Cross-sectional SEM images of a) Thio, b) Thio-0.8F, c) Thio-1.2F, d) Thio-1.6F and e) Thio-2F films.

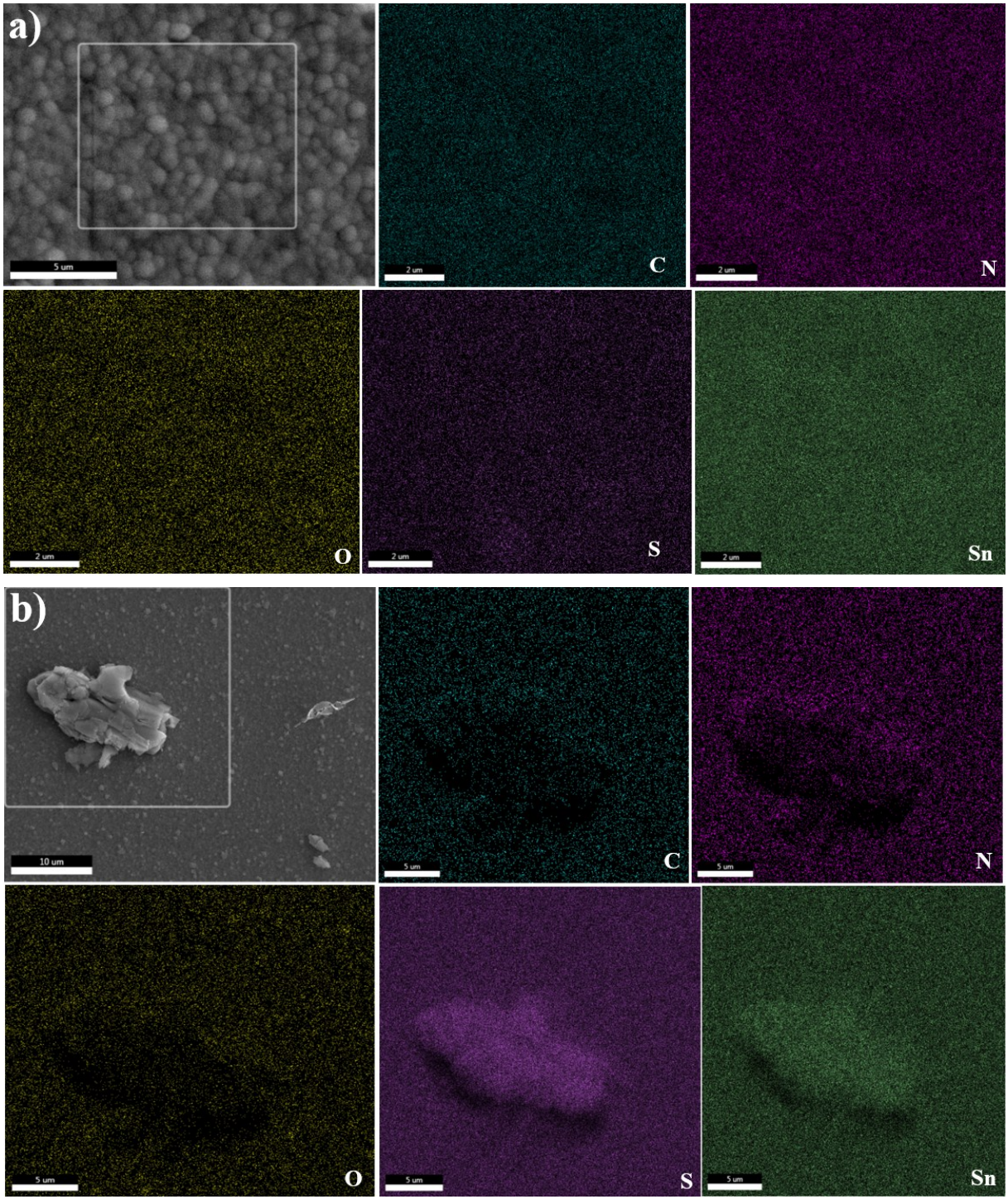

Figure S2. EDS mappings of a) Thio and b) Thio-1.6F films.

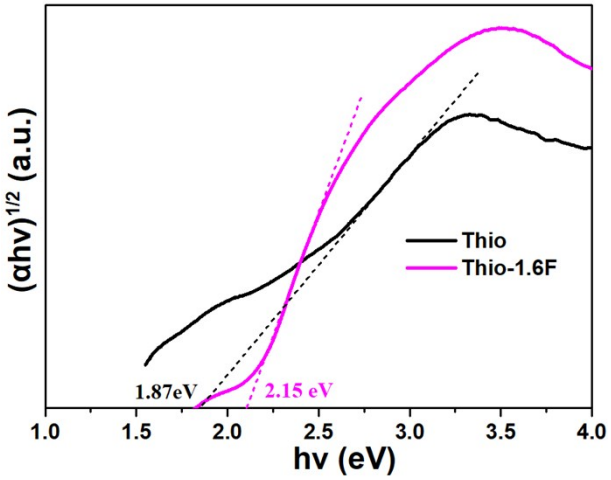

Figure S3. Tauc plots analysis of Thio and Thio-1.6F films.

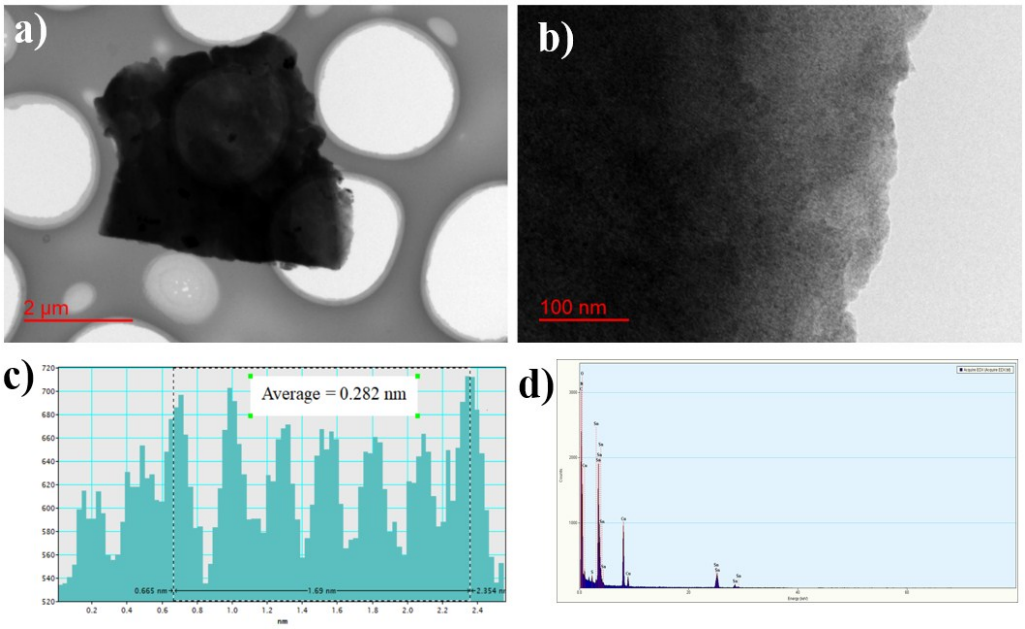

Figure S4. a, b) Top view of TEM images (scraped sample flake), c) average distance of lattice fringes, d) EDS spectrum of the Thio film.

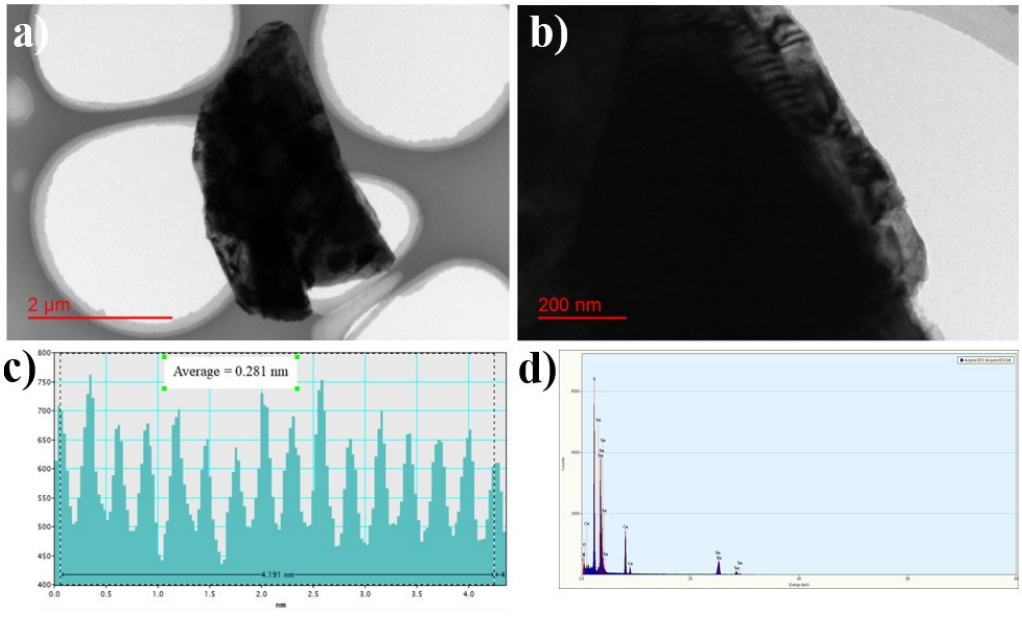

Figure S5. Top view of TEM images (scraped sample flake), c) average distance of lattice fringes, d) EDS spectrum of the Thio-1.6F film.

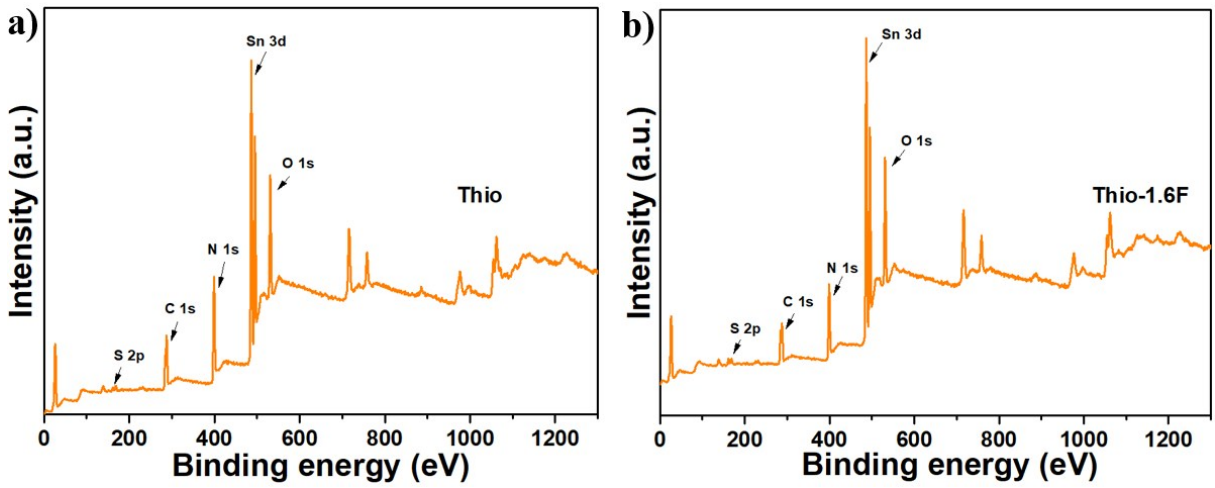

Figure S6. XPS full spectra of a) Thio, b) Thio-1.6F films.

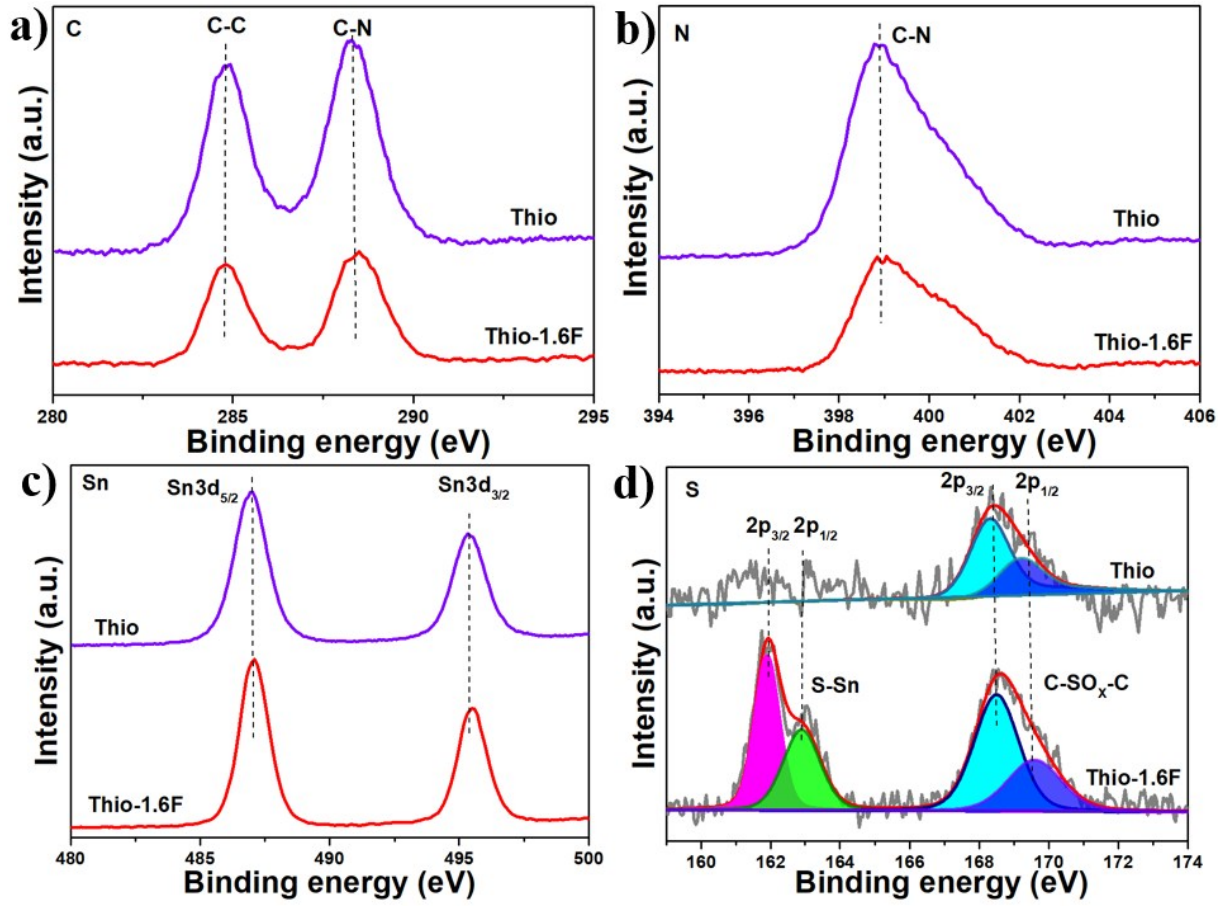

Figure S7. High resolution of XPS spectra of a) C 1s, b) O 1s, c) Sn 3d and d) S 2p with fitted peaks of Thio and Thio-1.6F films.

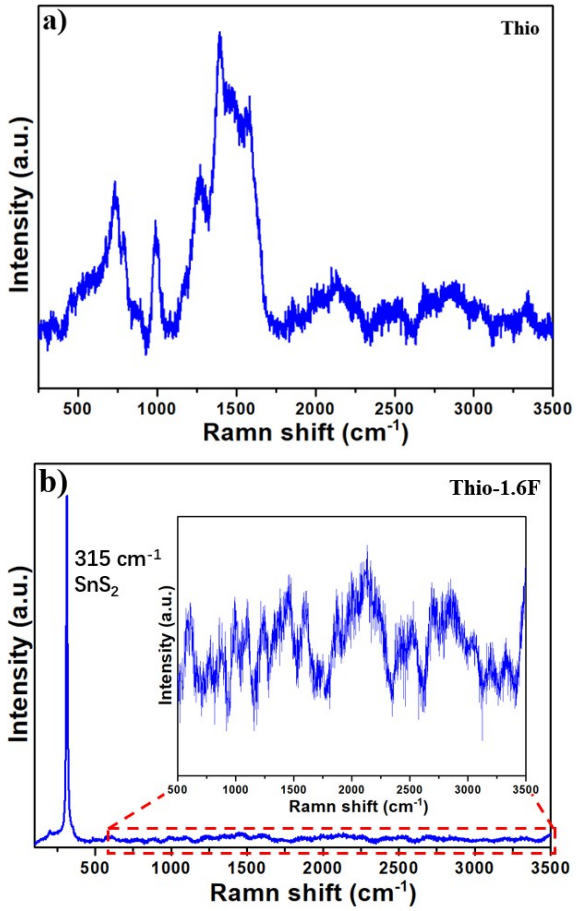

Figure S8. Raman spectra of a) Thio and b) Thio-1.6F films.

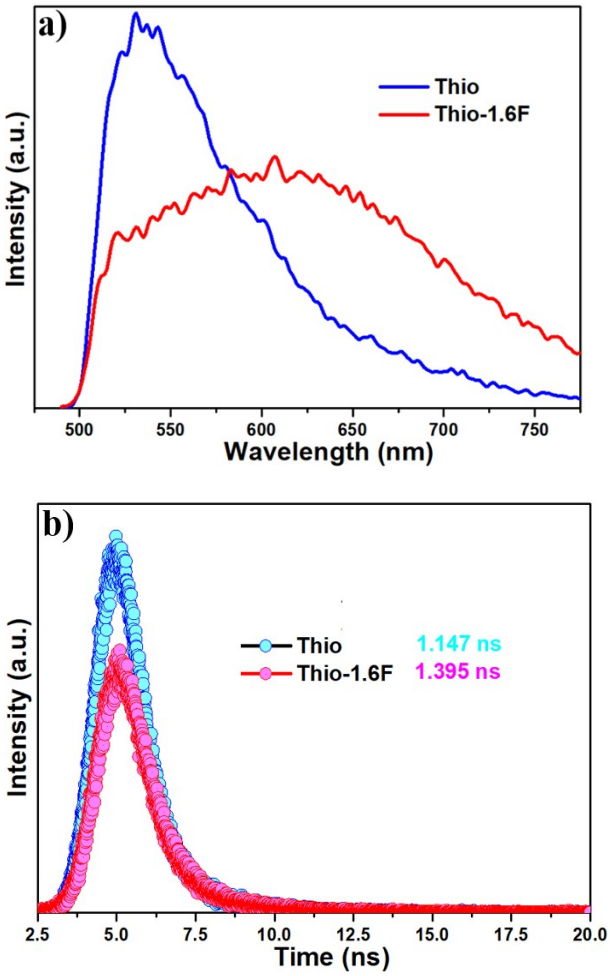

Figure S9. Steady-state PL spectra and time-resolved PL decay curves of Thio and Thio-1.6F films.

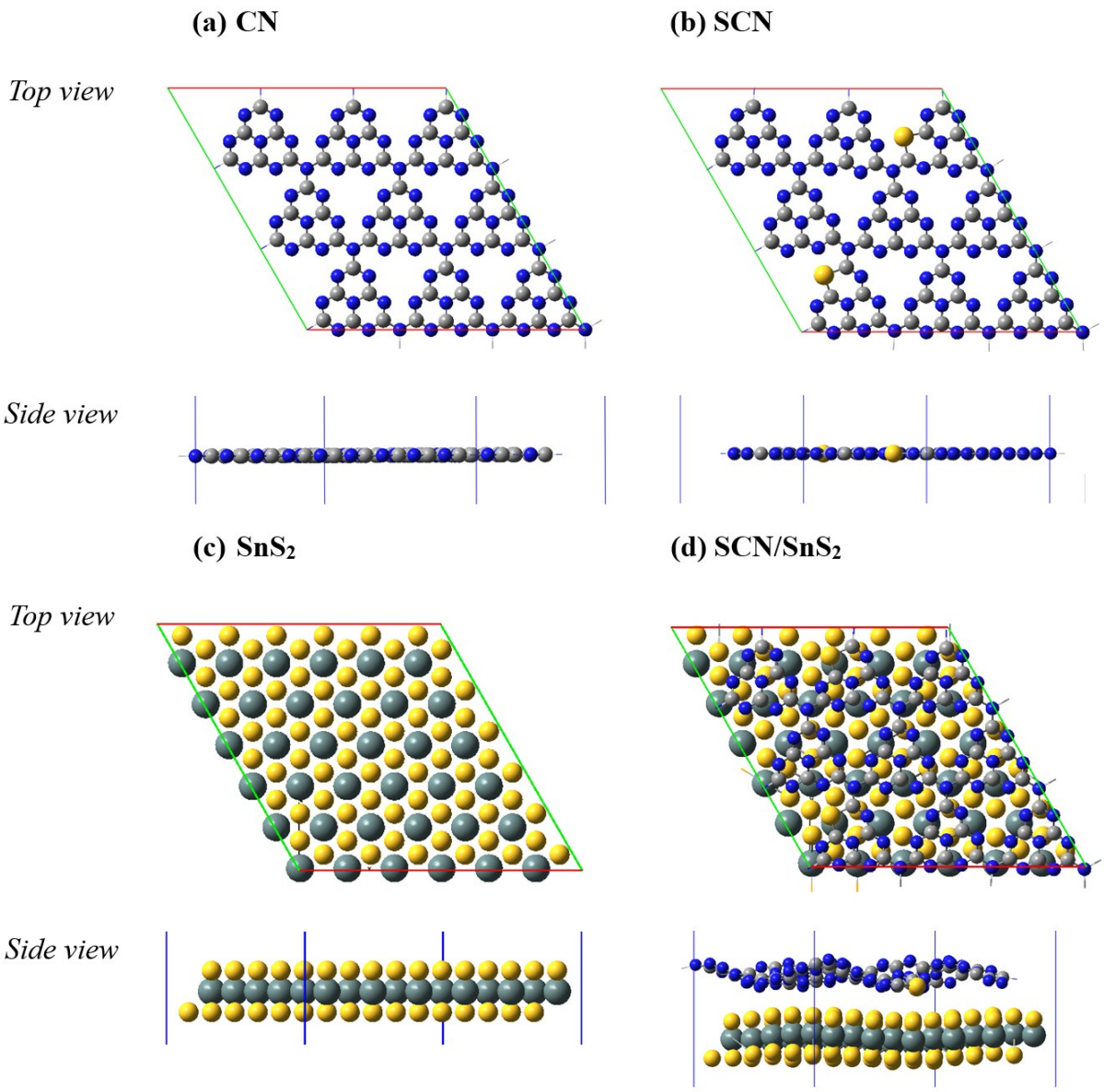

Figure S10. Top and side views of S-doped g-CN (SCN) film with two N substituted by S atoms, SnS2 film, and SCN/SnS2 interface with the interlayer distance ~3.5 Å. The binding energy between SCN and SnS2 films is about -27.6 eV. The supercell geometries are optimized in a lattice constant of a=b=21.04 Å, c=20.0 Å, and α=β=90°, γ=120° by first-principles DFT calculations. The grey, blue, yellow, and green colors respectively stand for C, N, S, and Sn atoms.

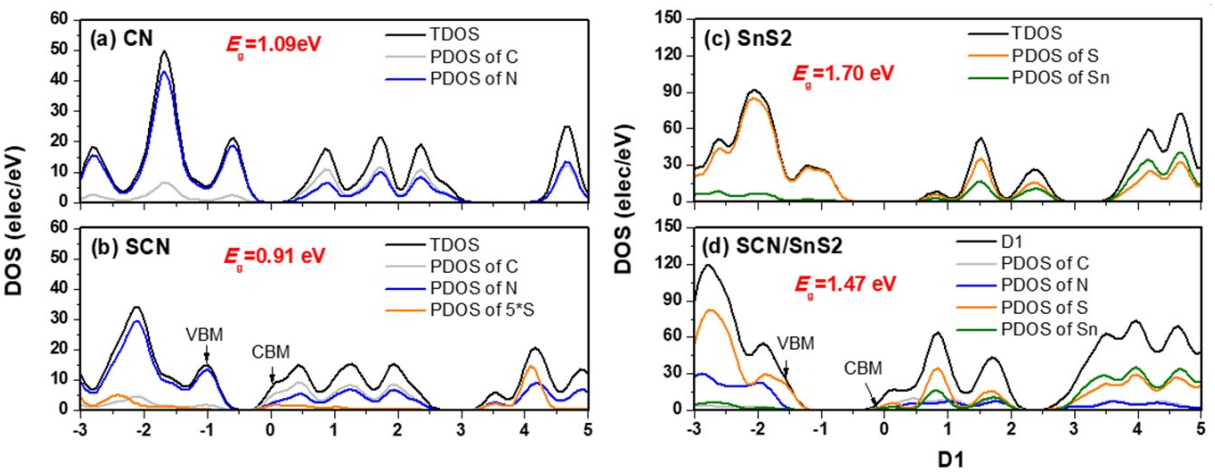

Figure S11. DFT calculated total and atomic density of states (DOS) of (a) CN (b) SCN (c) SnS_2 and (d) SCN/SnS_2 films, the structures of which are shown in Figure 1 above. The partial DOS of S atoms in SCN film is enlarged to 5 times.

Table S1 Comparison of PEC performance of our work and the state-of-the-art reported g-CN based photoelectrodes.

g-CN based photoelectrode	Photocurrent density (μA/cm2)	Light source	Potential	Sacrificial agent	Ref. source
g-CN/SnS_2	844.6	AM 1.5G	1.23 V vs RHE	No	This work
g-CN	30.2	AM 1.5G	1.23 V vs RHE	No	Adv. Mater.[1]
g-CN	17	AM 1.5G	-0.2 V vs Ag/AgCl	No	Angew. Chem.-Int Edit.[2]
Sulfur doped g-CN	60	AM 1.5G	1.23 V vs RHE	No	Adv. Funct. Mater.[3]
Sulfur doped g-CN	89	AM 1.5G	1.1 V vs RHE	No	Appl. Catal. B-Environ.[4]
g-CN	120	AM 1.5G	1.55 V vs RHE	Yes	Nano Energy[5]
Carbon doped g-CN	100	AM 1.5G	1.23 V vs RHE	Yes	Adv. Energy Mater.[6]
g-CN	12	AM 1.5G	1.23 V vs RHE	No	Angew. Chem.-Int Edit.[7]
Boron doped g-CN	103.2	AM 1.5G	1.23 V vs RHE	No	Angew. Chem.-Int Edit.[8]
Sulfur doped g-CN	110	AM 1.5G	1.23 V vs RHE	No	Appl. Catal. B-Environ.[9]
Sulfur doped g-CN	100	AM 1.5G	1.23 V vs RHE	No	ACS Catal.[10]
g-CN	116	AM 1.5G	1.23 V vs RHE	No	Angew. Chem.-Int Edit.[11]
g-CN	110	AM 1.5G	1.23 V vs RHE	No	Angew. Chem.-Int Edit.[12]
Boron doped	55	AM 1.5G	1.23 V vs	No	Appl.

g-CN			RHE		Catal. B-Environ.[13]
g-CN/ graphene oxide	660	AM 1.5G	1.23 V vs RHE	Yes	Adv. Energy Mater.[14]
g-CN/ZnO	400	AM 1.5G	1.23 V vs RHE	No	Angew. Chem.-Int Edit.[15]
g-CN/$BiVO_4$	420	AM 1.5G	1.23 V vs RHE	No	Appl. Catal. B-Environ.[16]
g-CN/SnS_2/RGO	1450	AM 1.5G	1.23 V vs RHE	No	Int. J. Hydrog. Energy[17]

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