

Supplementary Materials: Electrodeposition of Copper Oxides as Cost-Effective Heterojunction Photoelectrode Materials for Solar Water Splitting

Tai-Hsin Yin ^{1,†}, Bu-Jine Liu ^{1,†}, Yu-Wei Lin ¹, Yi-Syuan Li ¹, Chih-Wei Lai ¹, Yu-Pin Lan ¹, Changsik Choi ^{2,*}, Han-Chen Chang ^{3,*} and YongMan Choi ^{1,*}

¹ College of Photonics, National Yang Ming Chiao Tung University, Tainan 71150, Taiwan

² Clean Energy Conversion Research Center, Institute for Advanced Engineering, Yongin 17180, Republic of Korea

³ Green Energy and Environment Research Laboratories, Industrial Technology Research Institute, Tainan 71150, Taiwan

* Correspondence: cschoi@iae.re.kr (C.C.); yessey@itri.org.tw (H.-C.C.); ymchoi@nycu.edu.tw (Y.C.)

† These authors contributed equally to this work.

Table S1. Compilation of copper oxide-based photocathodes used for PEC water splitting.

Photocathode	Efficiency (%)	Max. Current Density (mA/cm ²)	Condition	Publication Year, Reference
NiO/CuO/MoS ₂ /ITO	0.10	-2.14 (-0.55 V vs. Ag/AgCl)	1.0 M KOH	2020, [26]
Cu ₂ O/ZnO/NiO _x /ITO	0.11	0.84 (0 V vs. RHE)	0.1M NaPi	2020, [27]
Cu ₂ O/FTO	0.12	-0.21 (-0.21 V vs. RHE)	0.5 M Na ₂ SO ₄	2016, [28]
g-C ₃ N ₄ /CuO/FTO	0.12	-0.85 (0 V vs. RHE)	0.1 M Na ₂ SO ₄	2021, [29]
Cu-CuBi ₂ O ₄ /TiO ₂ /FTO	0.16	0.33 (0.5 V vs. RHE)	0.1 M phosphate buffer solution	2020, [30]
Cu ₂ O/CuO/ITO	0.28	2.80 (0 V vs. RHE)	1.0 M Na ₂ SO ₄	2018, [31]
CuO/FTO	0.43	-	0.5 M Na ₂ SO ₄	2016, [28]
CuO/Cu ₂ O/FTO	0.55	3.15 (0.40 V vs. RHE)	0.5 M Na ₂ SO ₄	2016, [28]
BiVO ₄ /Ag/Cu ₂ O/FTO	0.56	3.36 (1.23 V vs. RHE)	0.2 M Na ₂ SO ₄	2022, [32]
NiO-doped CuO/FTO	1.00	-1.9 (0.6 V vs. RHE)	2 M KOH	2022, [33]
Cu ₂ O/ITO	1.10	2.00 (0.65 V vs. RHE)	0.5 M Na ₂ SO ₄	2021, [34]
A-GO/Cu ₂ O/ITO	1.42	2.74 (-1.1V vs Ag/AgCl)	0.1 M NaHCO ₃	2022, [35]
CuO/ITO	1.48	1.20 (-0.55 V vs. Ag/AgCl)	1 M KOH	2011, [36]
NiO _x /Cu ₂ O/AZO/MoO _x /FTO	1.75	6.10 (0 V vs. RHE)	1.0 M Na ₂ SO ₄	2022, [37]

Table S2. Compilation of heterostructured copper oxide-based photoelectrodes.

Structure	Photocurrent Density (mA/cm ²)	Electrolyte	Publication Year, References
Cu ₂ O/CuO	0.45 at 0 V _{RHE}	0.5M Na ₂ SO ₄ (pH = 6)	2017, [38]
Cu ₂ O/CuO	1.4 at 0 V _{RHE}	1 M Na ₂ SO ₄	2017, [39]
Cu ₂ O/CuO	1.2 at 0 V _{RHE}	0.1 M Na ₂ SO ₄ (pH = 6.25)	2022, [40]
Cu ₂ O/CuO	1.03 at 0 V _{RHE}	0.2 M Na ₂ SO ₄ (pH = 6.2)	This work

Table S3. EDS analysis results of Cu₂O, CuO/Cu₂O, and CuO thin films.

Thin Film	Cu	O	Ratio
Cu ₂ O	69.78	30.22	2.30
CuO/Cu ₂ O	60.42	39.58	1.52
CuO	51.96	48.04	1.08

Table S4. Band gaps of Cu₂O, CuO/Cu₂O, and CuO thin films.

Sample	Band Gap (eV)
Cu ₂ O	2.35
CuO/Cu ₂ O	1.81
CuO	1.45

Table S5. EIS parameters obtained by fitting the Nyquist plots with the equivalent circuit.

Sample	R _s (Ω)	R _{sc} (Ω)	R _{ct} (Ω)
Cu ₂ O	75.2	47.9	45613
CuO/Cu ₂ O	30.7	1281.0	11446
CuO	31.9	862.2	27602

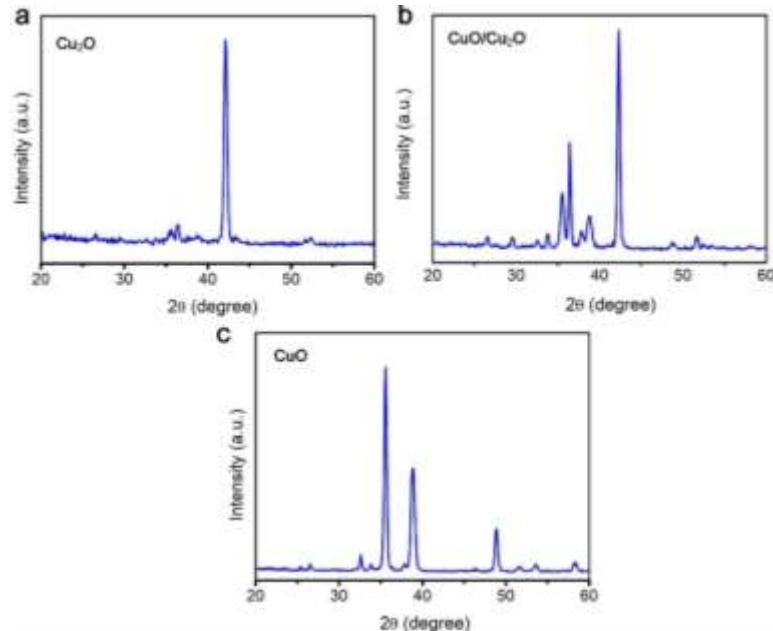


Figure S1. XRD patterns of (a) Cu₂O, (b) CuO/Cu₂O, and (c) CuO thin films.

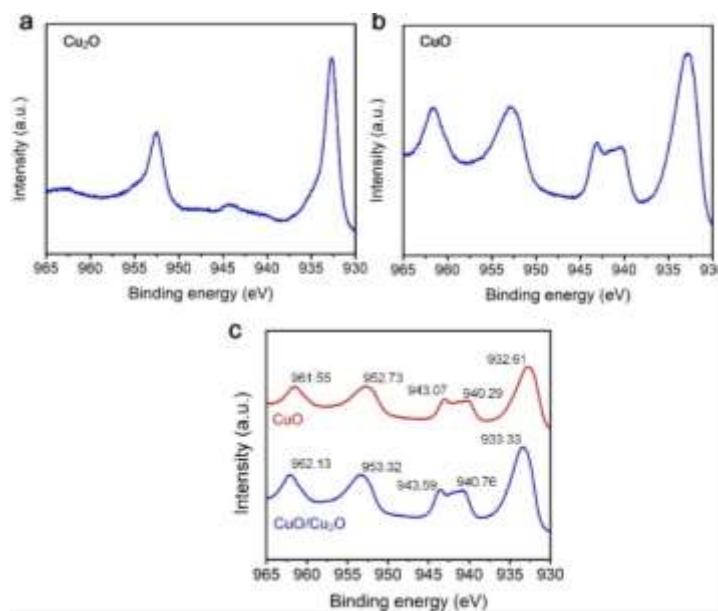


Figure S2. XPS spectra for Cu 2p of (a) Cu₂O and (b) CuO thin films and (c) comparison of CuO and CuO/Cu₂O thin films.