Supporting Information

Design of an Amorphous TaO_x Multifunctional Interfacial Layer onto Photocathodes for Photoelectrochemical H_2 Evolution

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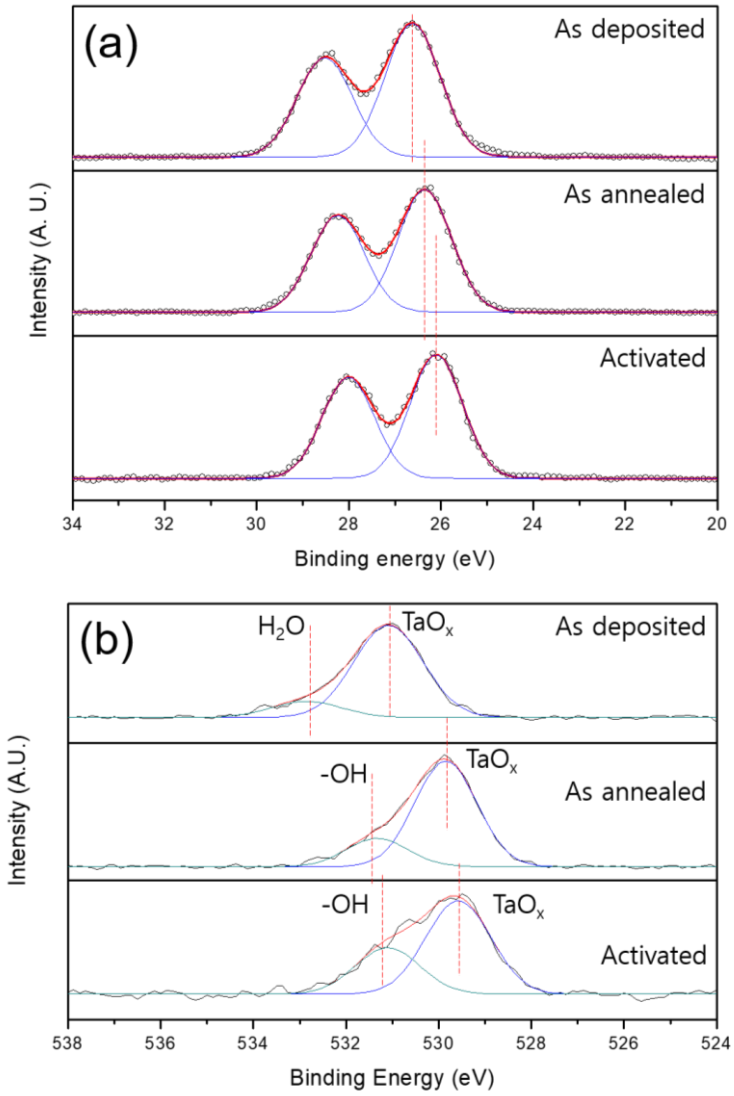

Fig. S1. XPS spectra of $CuInS_2/TaO_x$ with different conditions (a) Ta 4f and (b) O 1 s

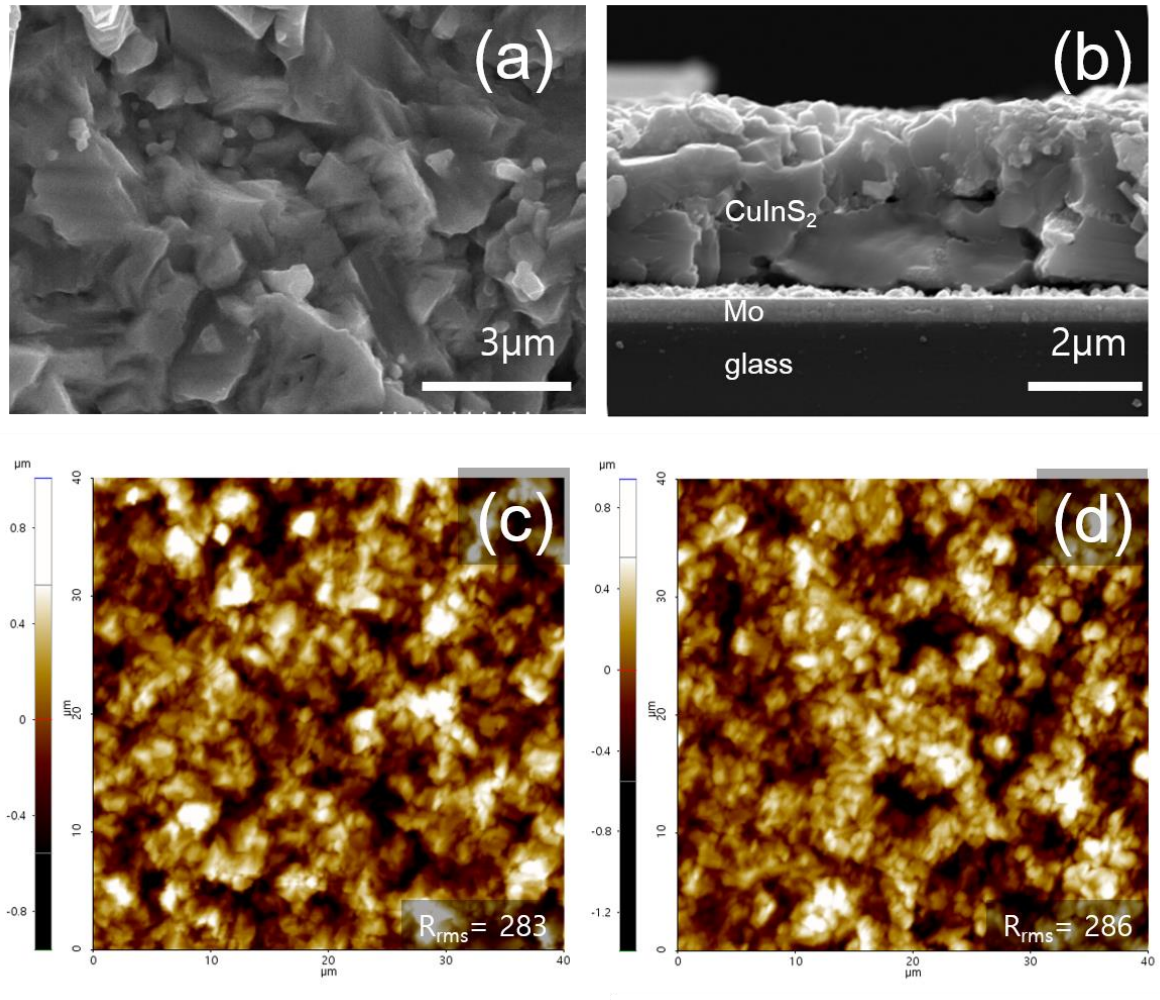

Fig. S2. SEM images of (a) top view and (b) cross-section of $CuInS_2$. AFM topography images of $CuInS_2/TaO_x$ (c) as deposited and (d) activated samples.

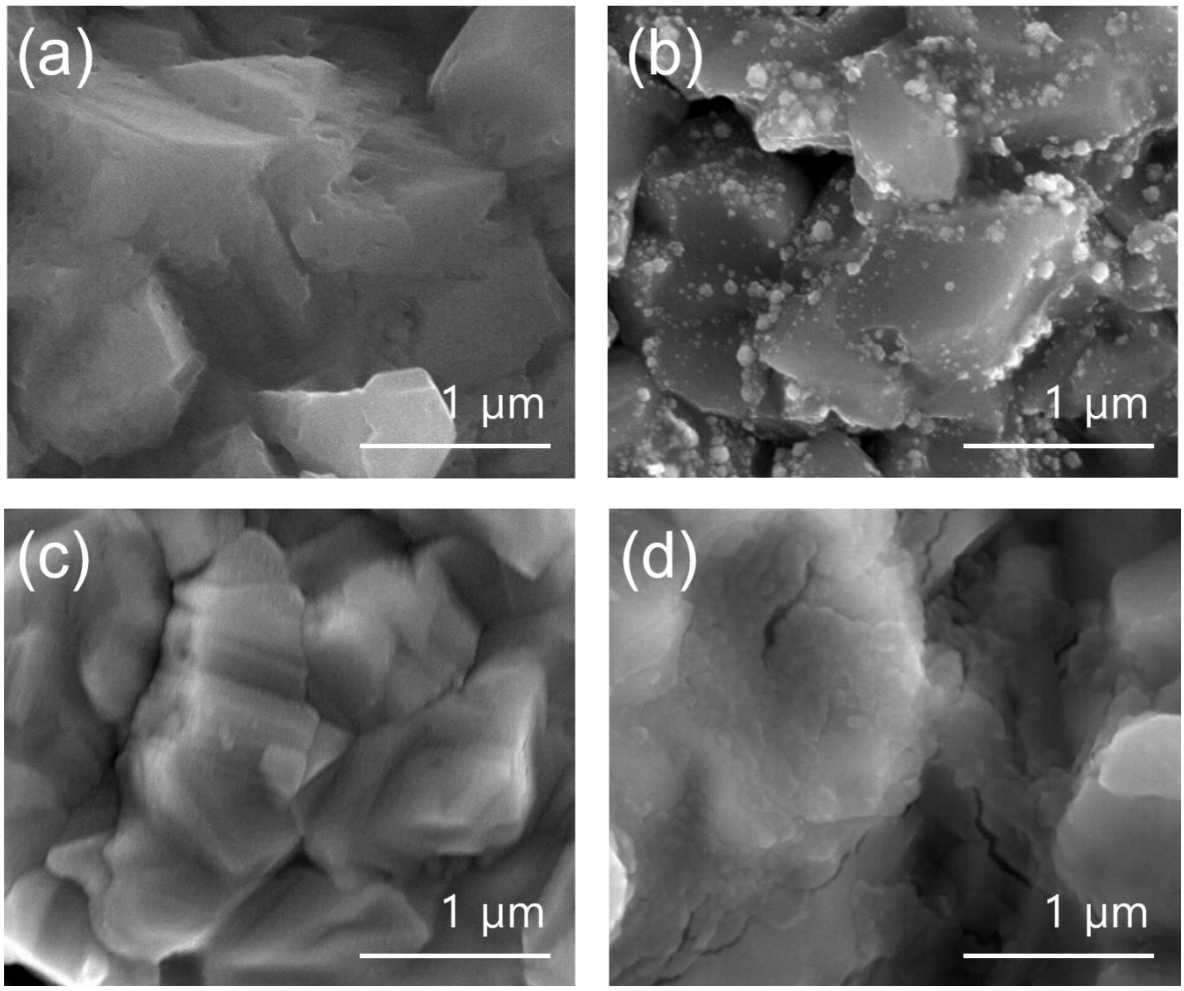

Fig. S3. SEM images of (a) $CuInS_2$. (b) $CuInS_2$/Pt (c) $CuInS_2/TaO_x$ (as prepared) (d) $CuInS_2/TaO_x$ (activated)

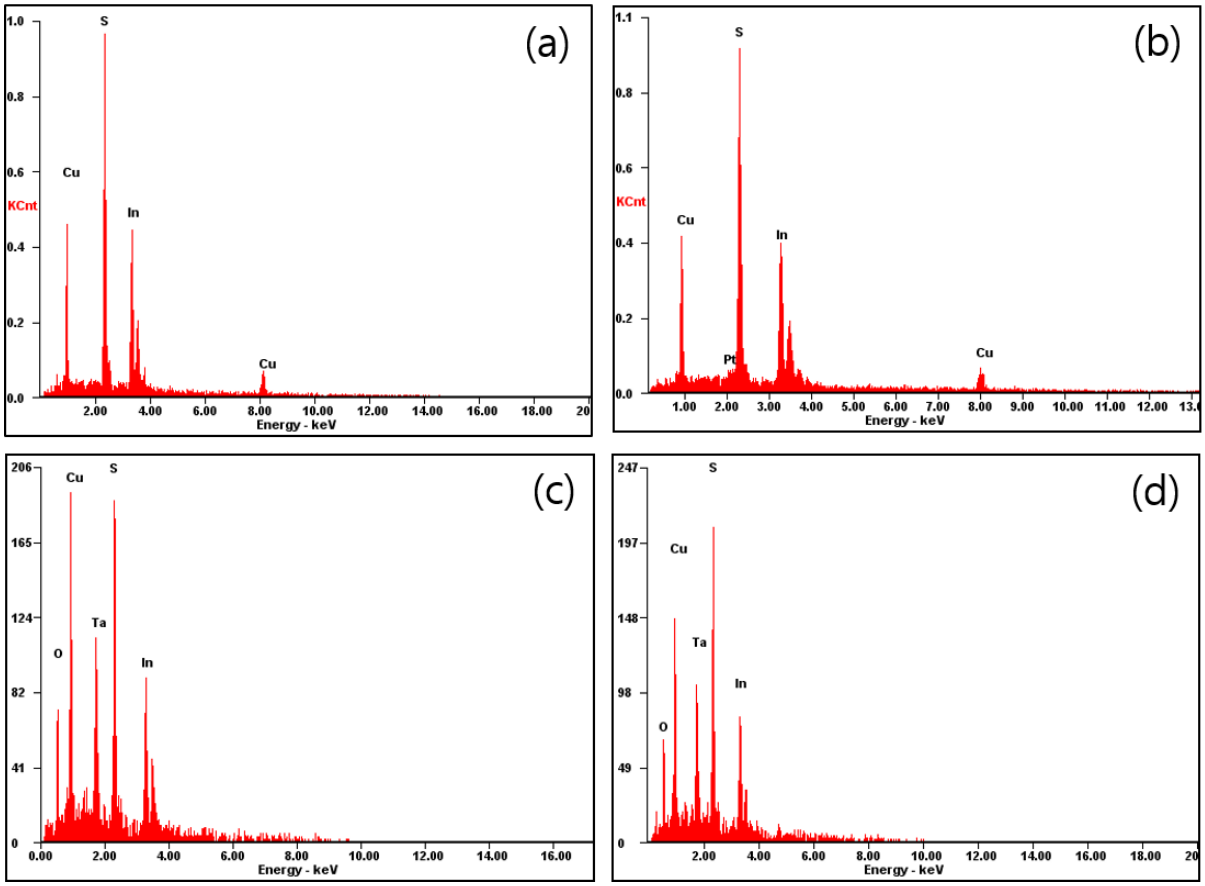

Fig. S4. EDS spectra of (a) $CuInS_2$. (b) $CuInS_2$/Pt (c) $CuInS_2/TaO_x$ (as prepared) (d) $CuInS_2/TaO_x$ (activated)

Table S1. Atomic percent of photocathodes from EDS results

	Cu (%)	In (%)	S (%)	Pt (%)	Ta (%)	O (%)
$CuInS_2$	26.10	24.63	49.27	-	-	-
$CuInS_2$/Pt	24.15	24.73	50.03	1.09	-	-
$CuInS_2/TaO_x$ (as prepared)	20.23	11.70	26.89	-	8.28	32.90
$CuInS_2/TaO_x$ (activated)	20.30	11.29	28.43	-	8.35	31.63

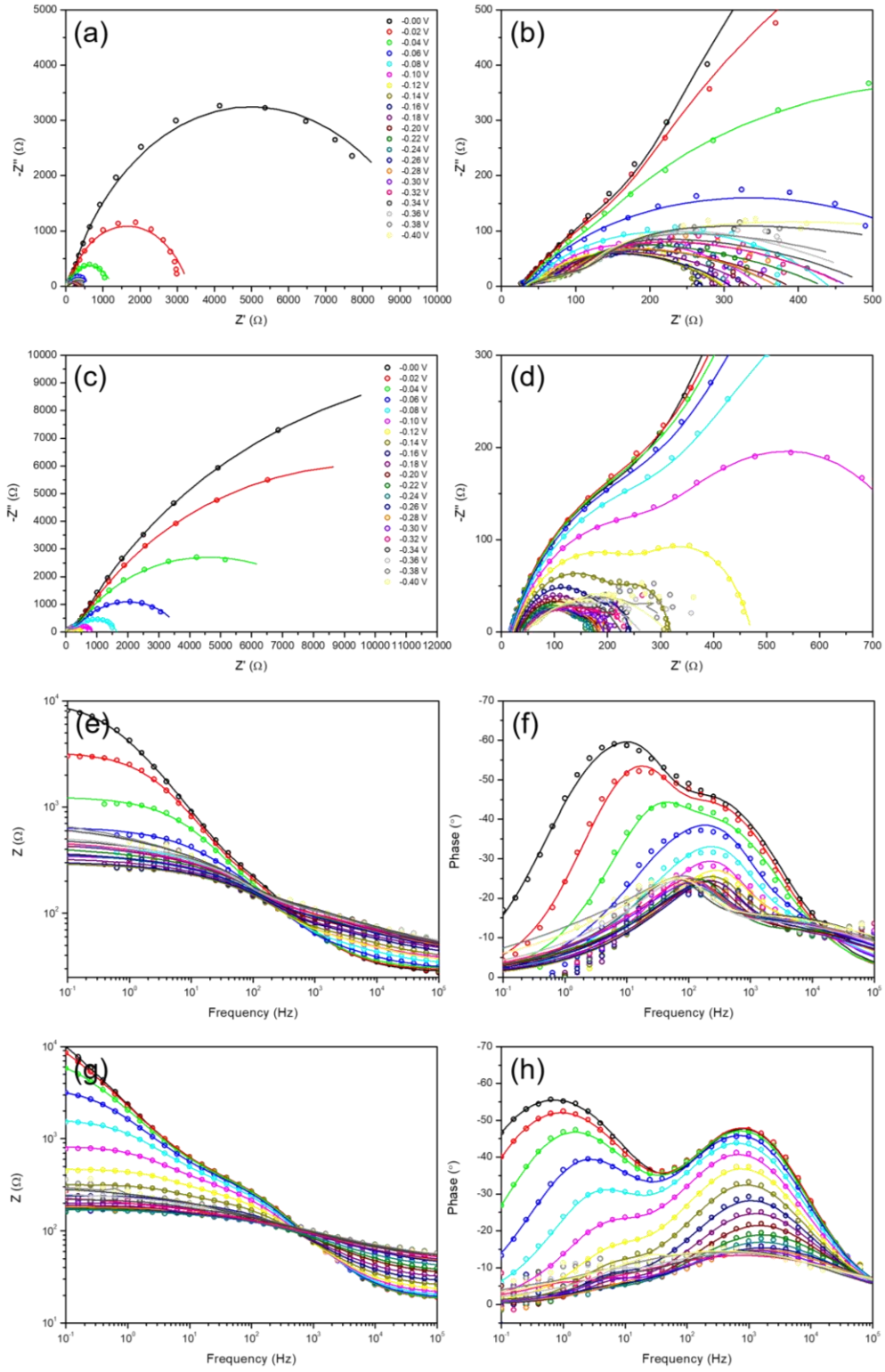

Fig. S5. Nyquist plots of electrochemical impedance spectroscopy results for $CuInS_2$/Pt ((a), (b)) and $CuInS_2/TaO_x$ ((c), (d)) on a different scale. Bode plots of electrochemical impedance spectroscopy for $CuInS_2$/Pt ((e), (f)) and $CuInS_2/TaO_x$ ((g), (h)) on a different scale. The dotted data is obtained from experiments and the solid lines indicate the fitted results.

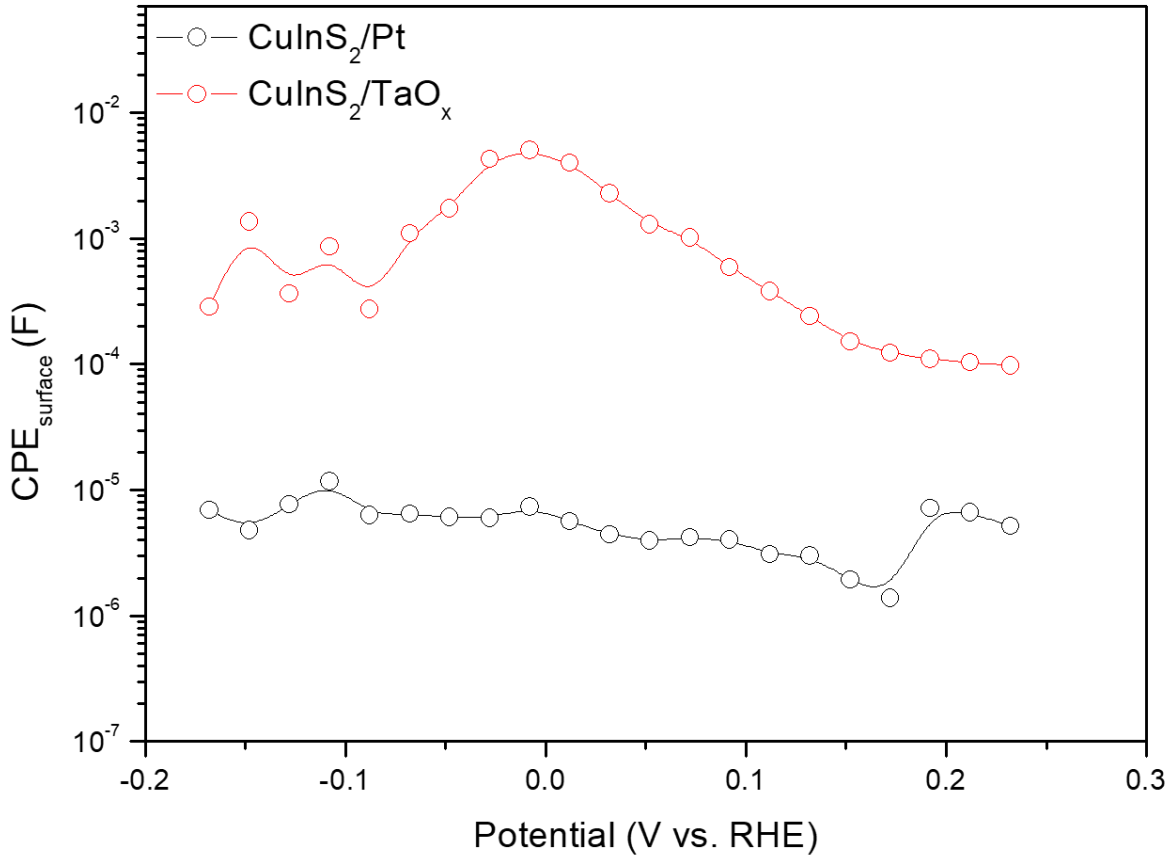

Fig. S6. Capacitance of surfaces for $CuInS_2$/Pt and $CuInS_2/TaO_x$

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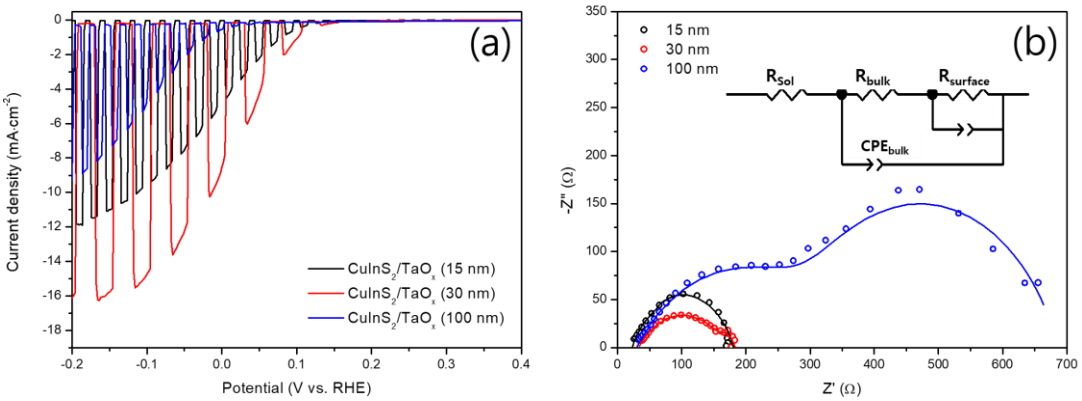

Fig. S7. LSV (a) and Nyquist plots (b) of $CuInS_2/TaO_x$ films depending on the thickness at -0.2 V vs. Ag/AgCl.

Table S2. Fitted values from data of electrochemical impedance spectroscopy for $CuInS_2/TaO_x$

TaO_x thickness (nm)	R_{sol} (Ω)	R_{bulk} (Ω)	$R_{surface}$ (Ω)	CPE_{bulk} (F)	$CPE_{surface}$ (F)
15	22.98	130.5	27.38	1.19×10^{-5}	1.53×10^{-7}
30	32.7	137.5	13.28	8.58×10^{-5}	2.29×10^{-3}
100	29.72	323.4	336.4	1.01×10^{-4}	7.25×10^{-4}

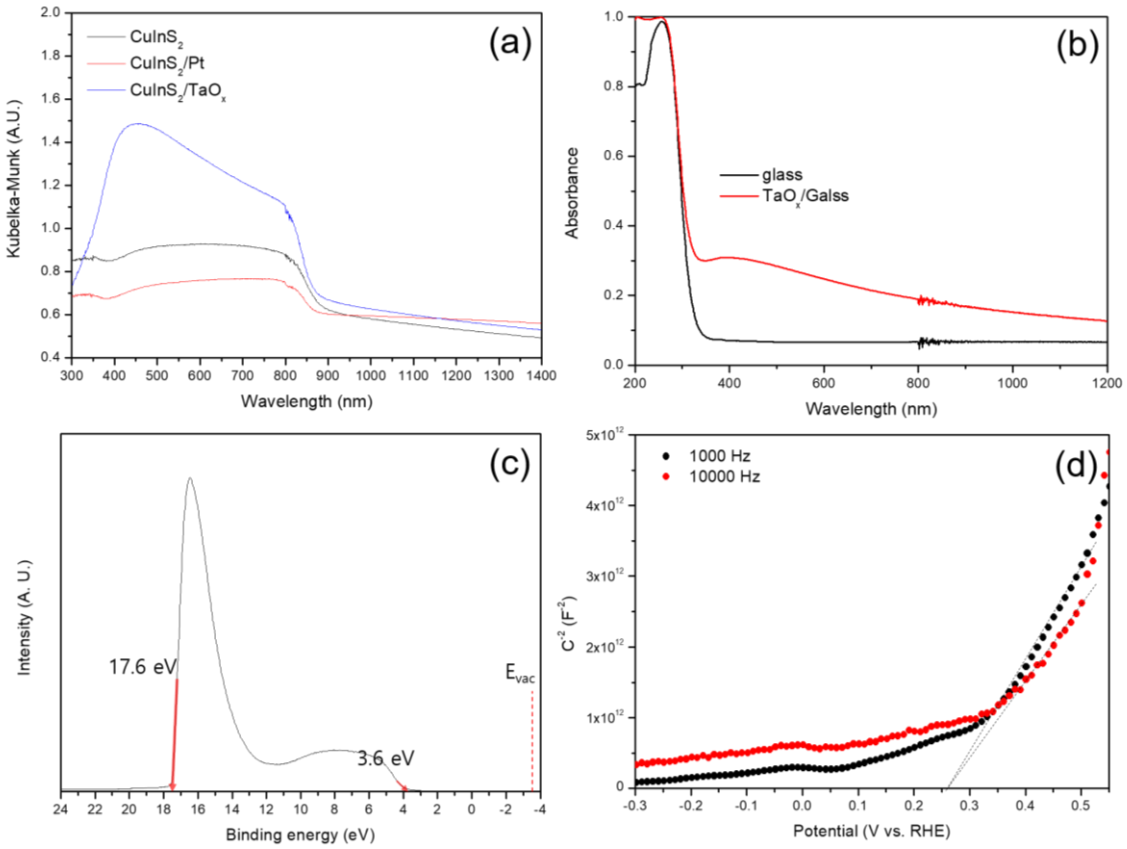

Fig. S8. (a) Kubelka-Munk absorption spectra of photocathodes (b) absorbance of TaO_x deposited on glass (c) UPS result of TaO_x deposited on $CuInS_2$ (d) Mott-Schottky plot of TaO_x

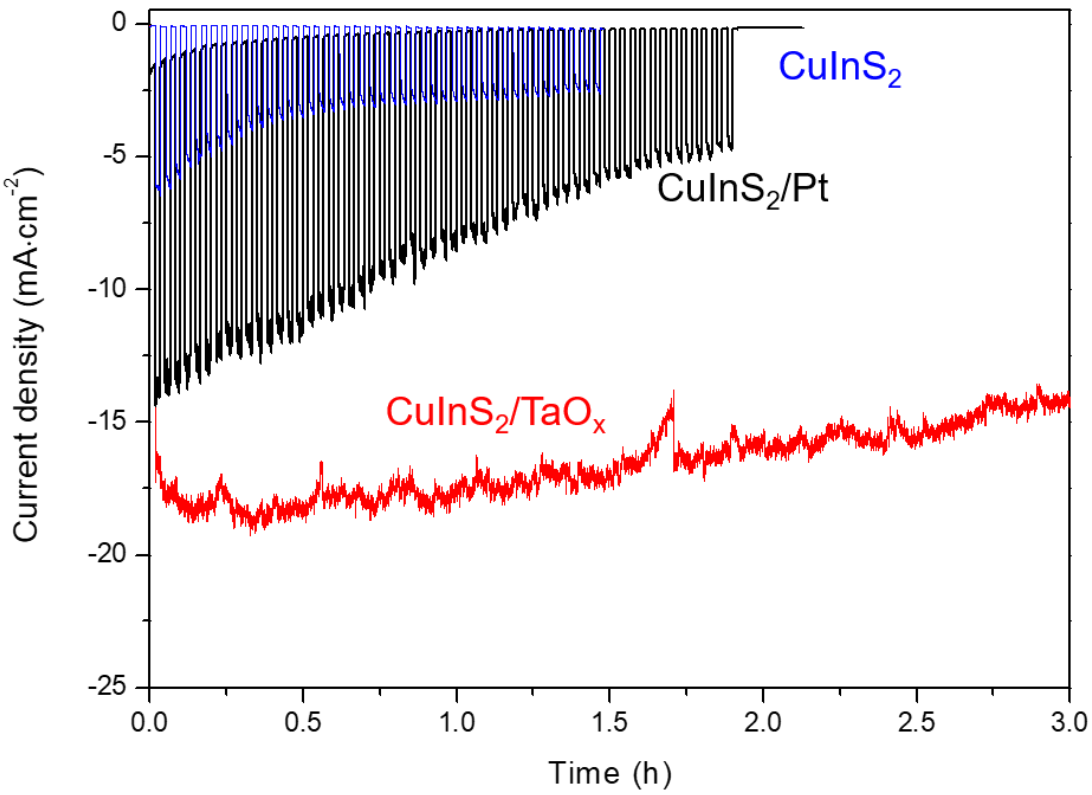

Fig. S9. Chronoamperometry results of $CuInS_2$, $CuInS_2$/Pt, and $CuInS_2/TaO_x$ photocathodes under light illumination.

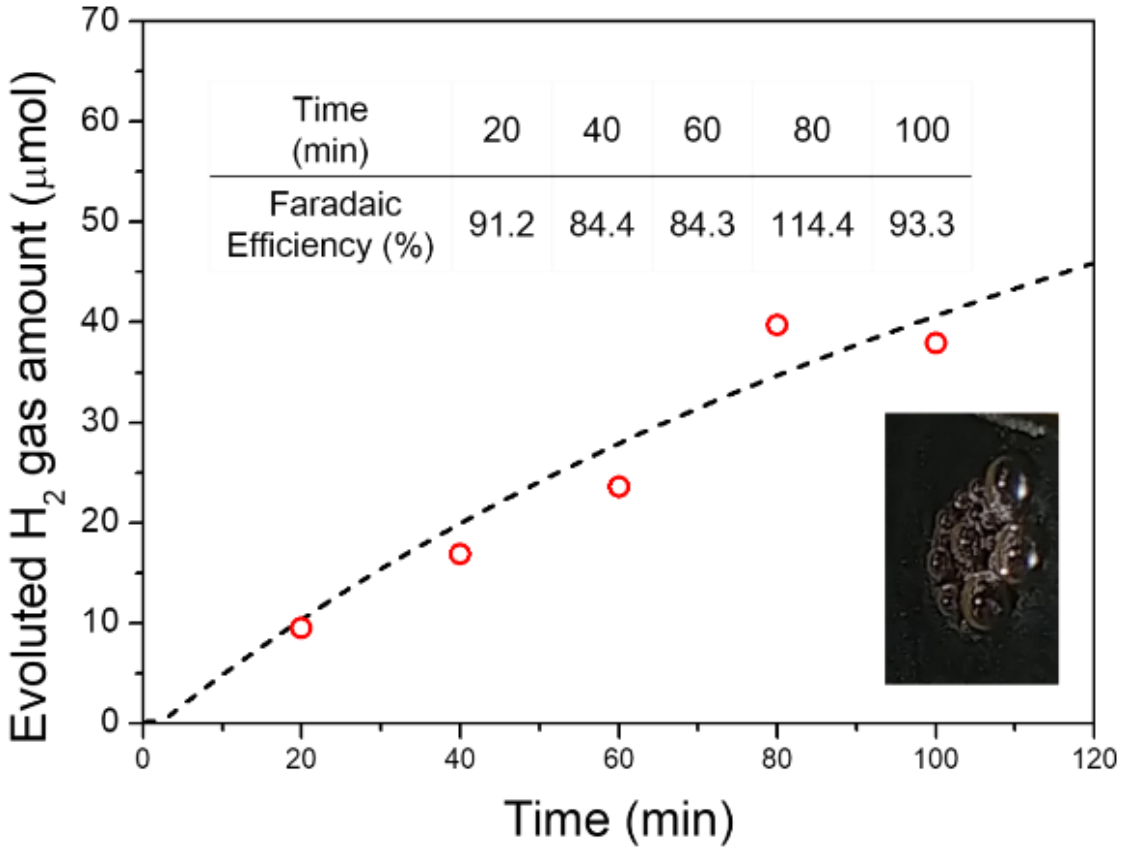

Fig. S10. Evolved H_2 gas measurement for a $CuInS_2/TaO_x$ photocathode during chronoamperometry. Inset photograph shows the bubble coalescence of H_2 gas during the photoelectrochemical reaction.