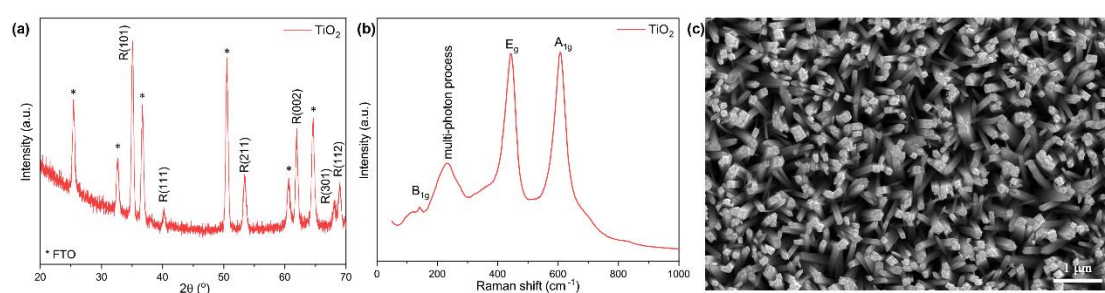


## Supporting information for

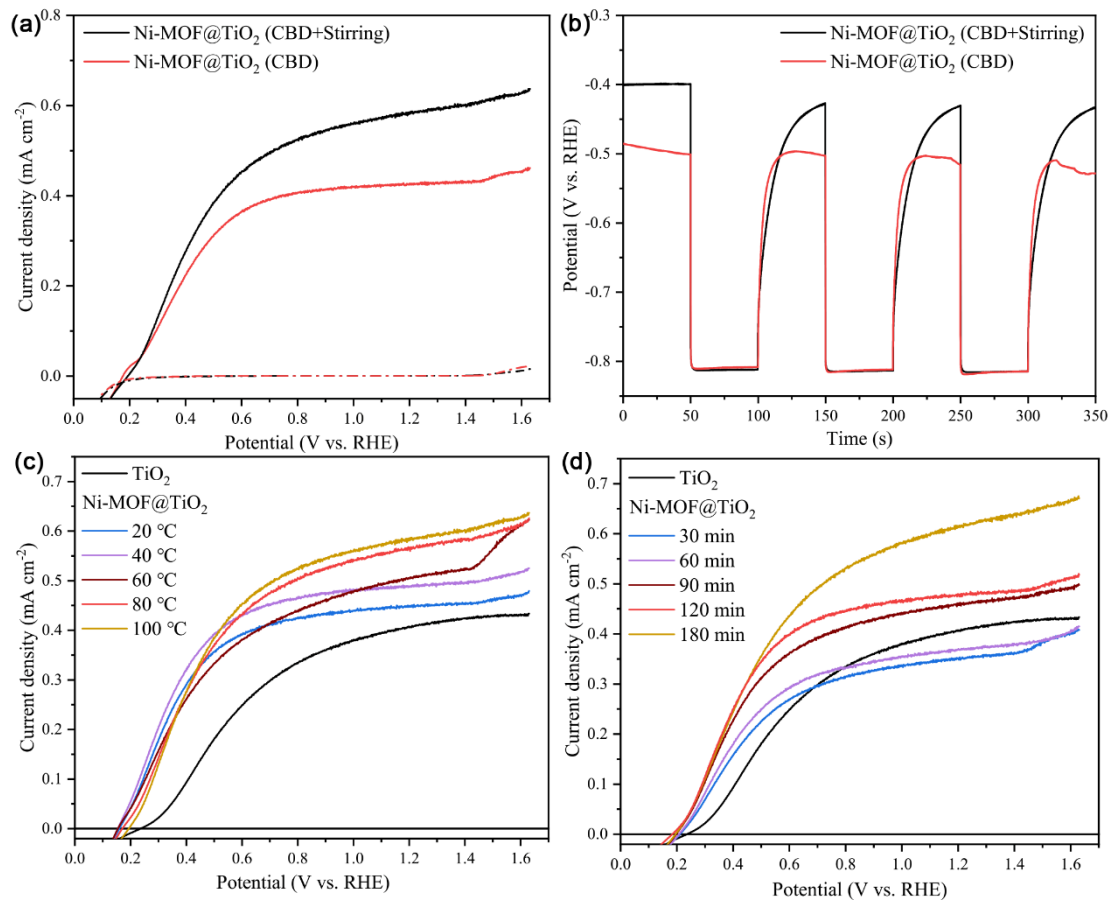
Content (10 Pages)

Supplementary Figures ..... S1–S9

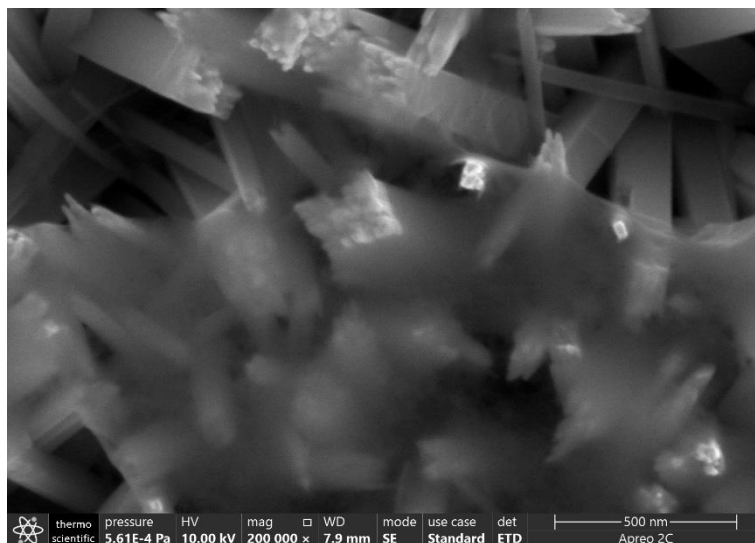
Supplementary Tables ..... S10



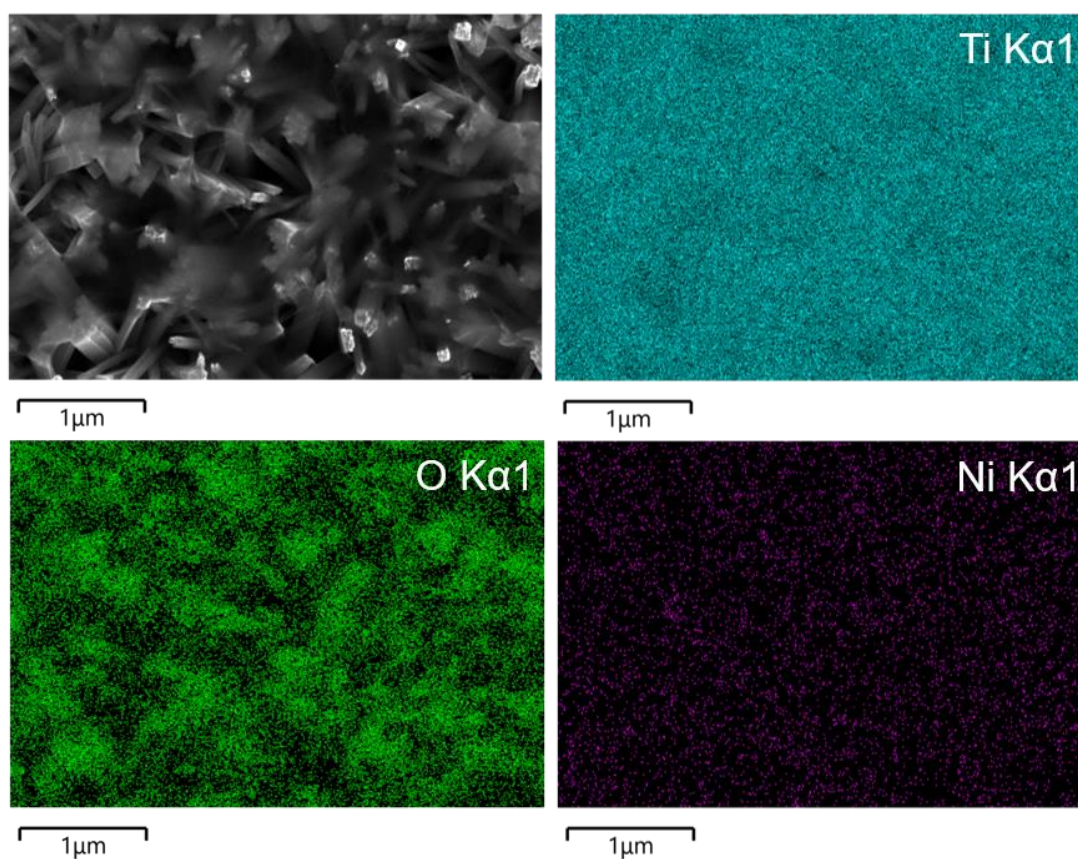
**Figure S1.** (a) XRD pattern, (b) Raman spectrum, and (c) SEM image of TiO<sub>2</sub> nanorod arrays film on FTO glass.



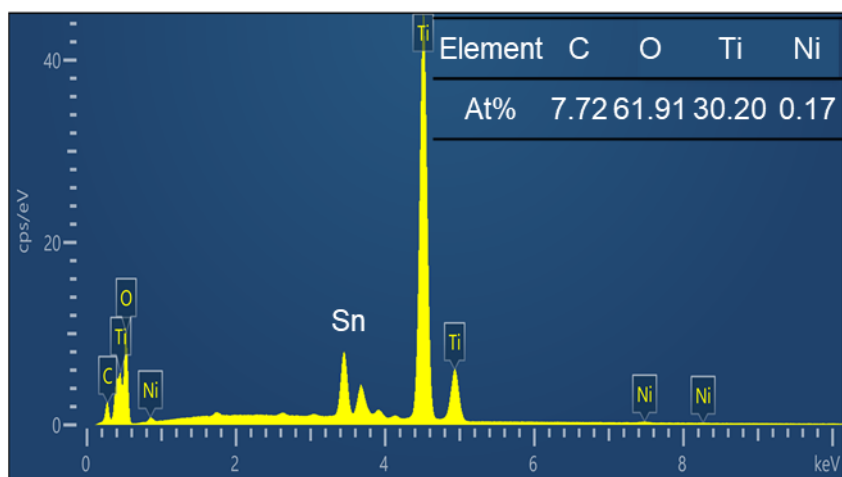
**Figure S2.** (a) LSV curves and (b) chopped OCP curves of Ni-MOF@TiO<sub>2</sub> prepared through CBD method with/without stirring. (c) LSV curves of Ni-MOF@TiO<sub>2</sub> prepared at different temperatures. (d) LSV curves of Ni-MOF@TiO<sub>2</sub> prepared with various times. The electrolyte is 1 M KOH/0.33 M urea.



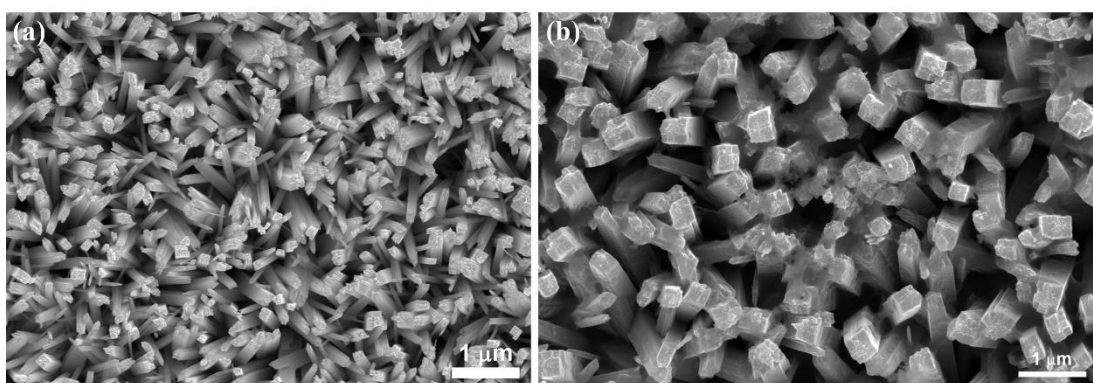
**Figure S3.** SEM image of Ni-MOF@TiO<sub>2</sub> film.



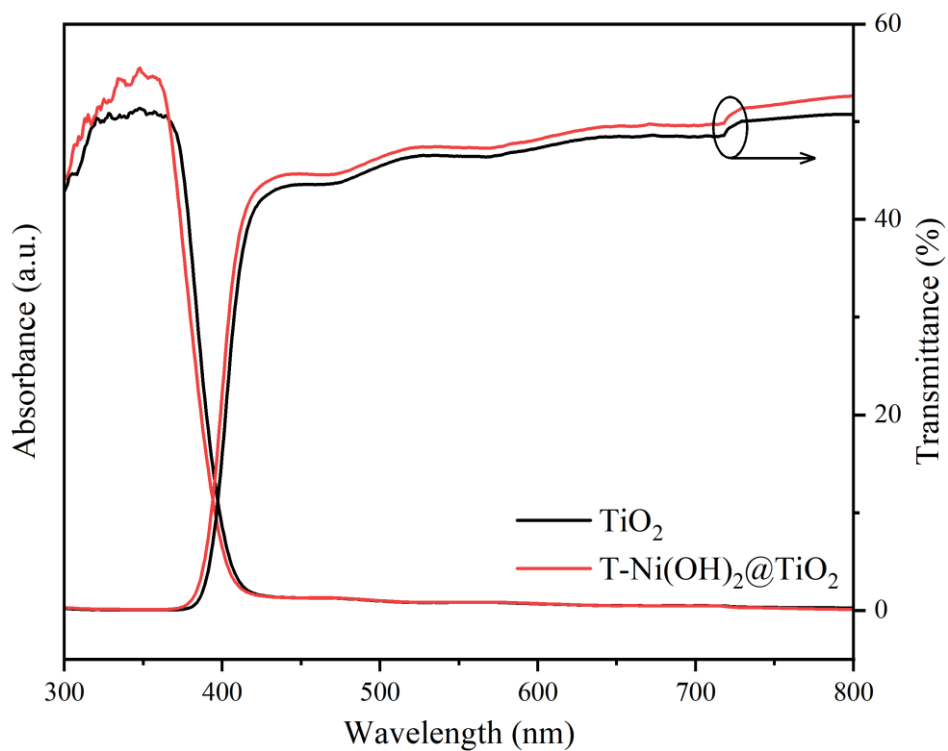
**Figure S4.** EDS mapping of T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> film.



**Figure S5.** EDS pattern of T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> film on FTO glass.

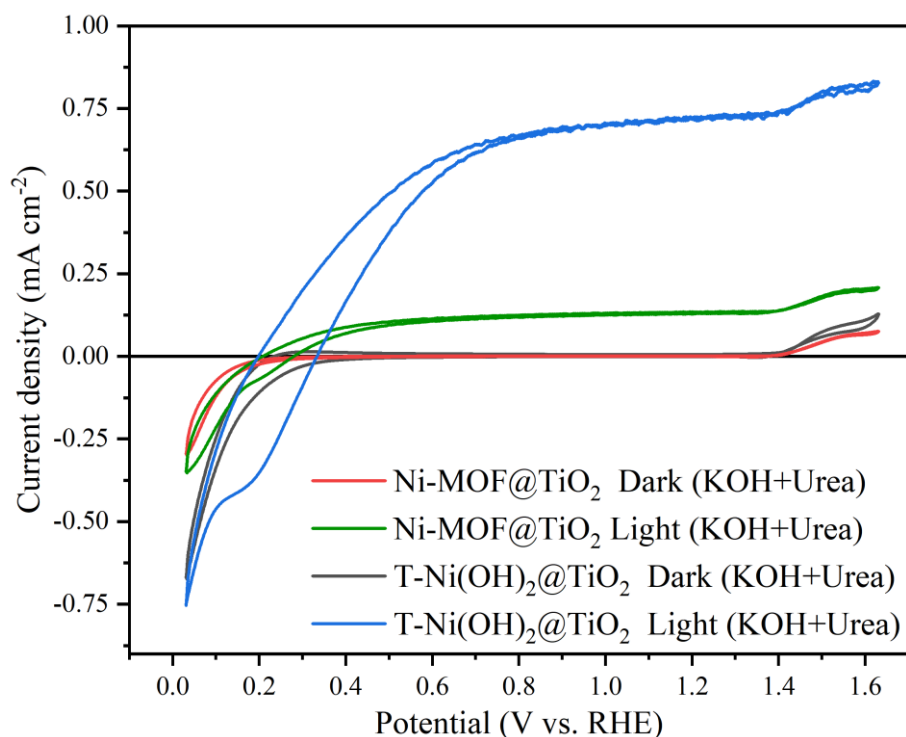


**Figure S6.** Top-view SEM images of (a) pristine TiO<sub>2</sub> and (b) Ni(OH)<sub>2</sub>@TiO<sub>2</sub> films.

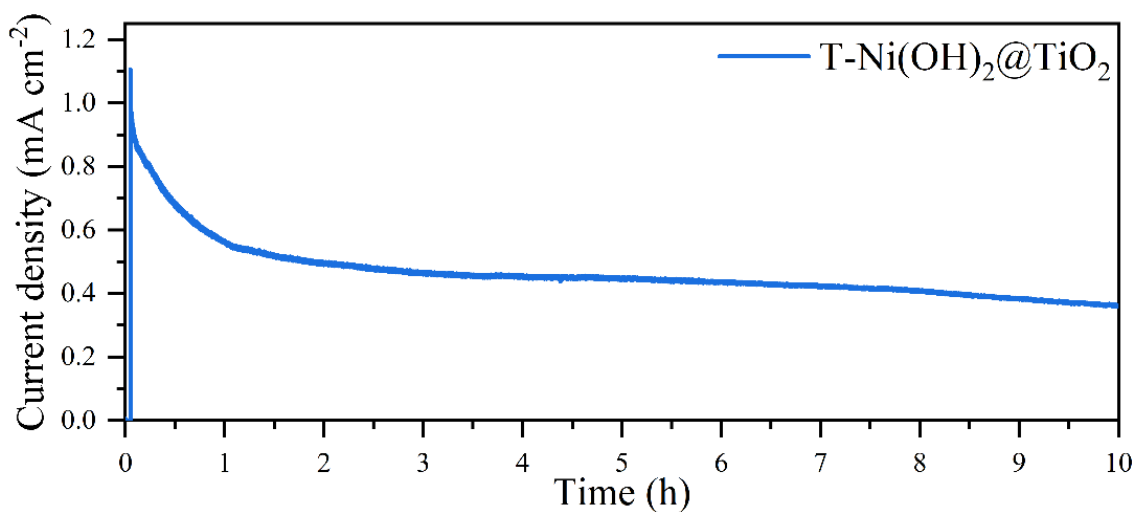


**Figure S7.** UV-vis absorption and transmittance spectra of TiO<sub>2</sub> and T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> with 6 h EL

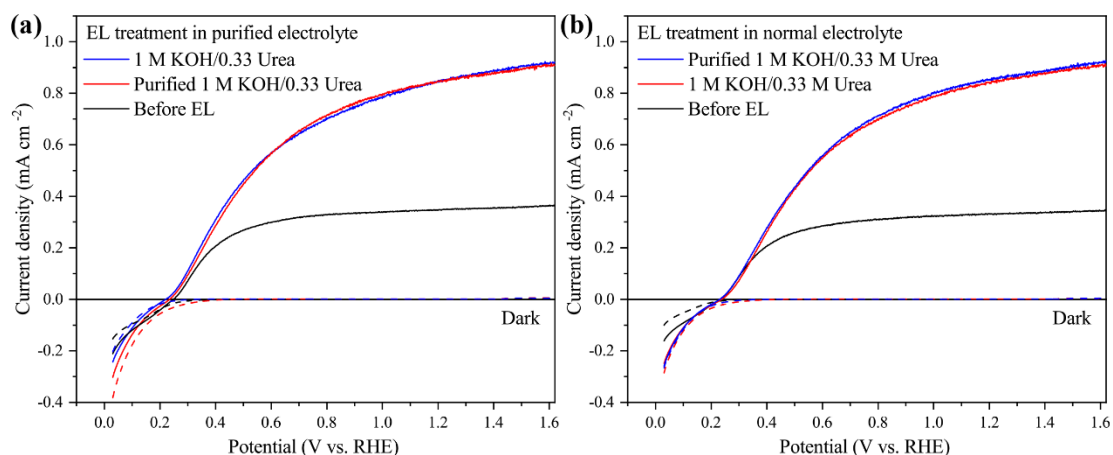
treatment.



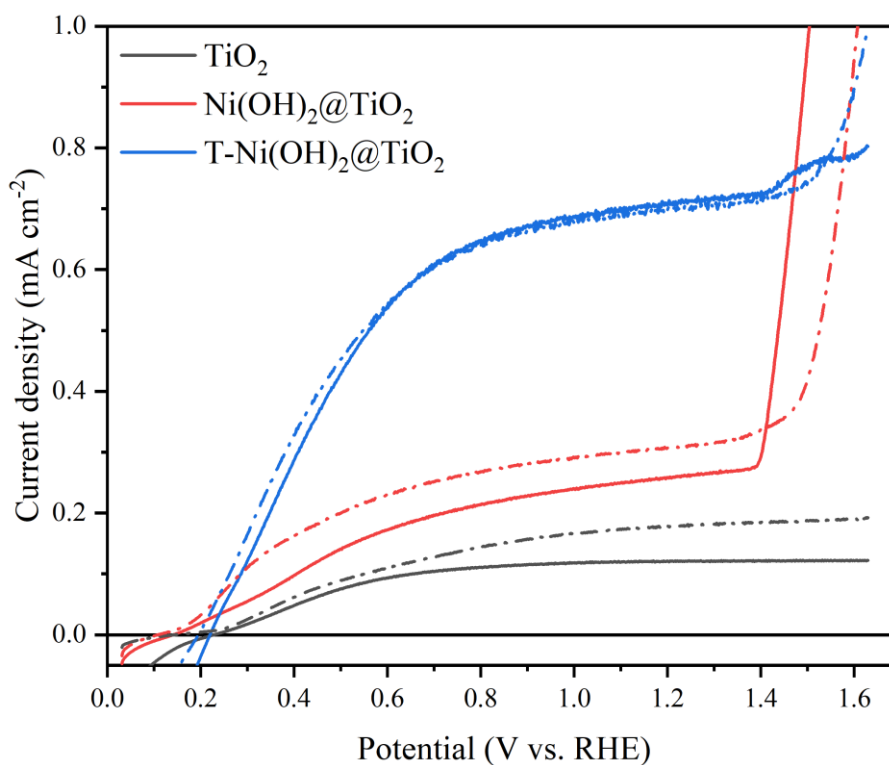
**Figure S8.** CV curves of Ni-MOF@TiO<sub>2</sub> and T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanodes with 1 h EL treatments under AM1.5G illumination and in the dark. Electrolyte: 1.0 M KOH and 0.33 M urea. Scan rate: 20 mV s<sup>-1</sup>.



**Figure S9.** Long-term stability of T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanode at 1.23 V<sub>RHE</sub> under AM1.5G illumination.

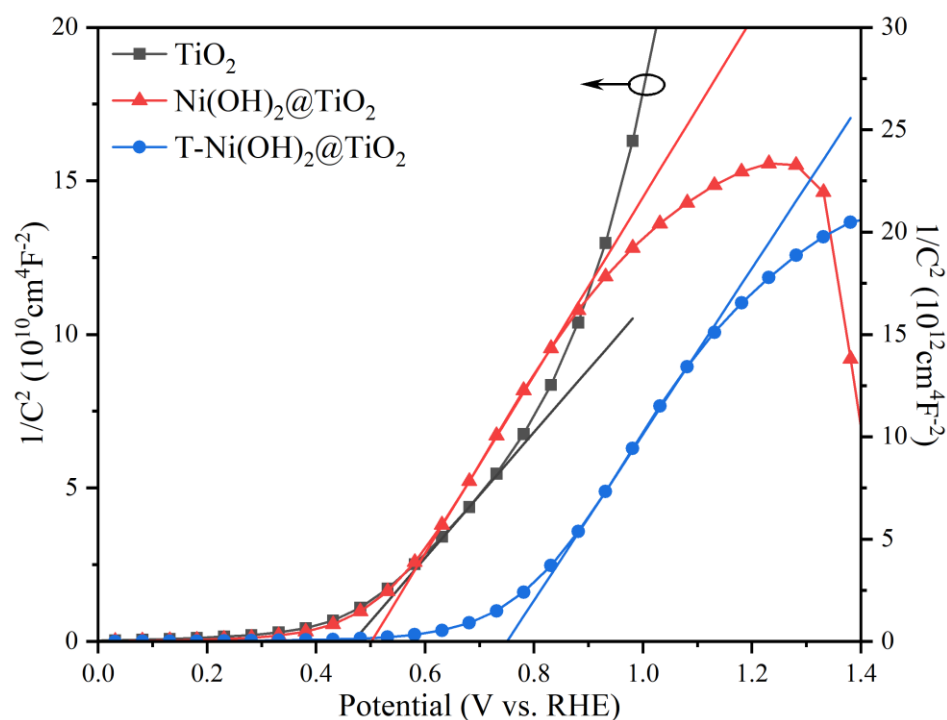


**Figure S10.** (a) LSV curves of T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanode with 1 h EL treatment in purified electrolyte. (b) LSV curves of T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanode with 1 h EL treatment in normal electrolyte. Scan rate: 20 mV s<sup>-1</sup>. Illumination: AM1.5G illumination.

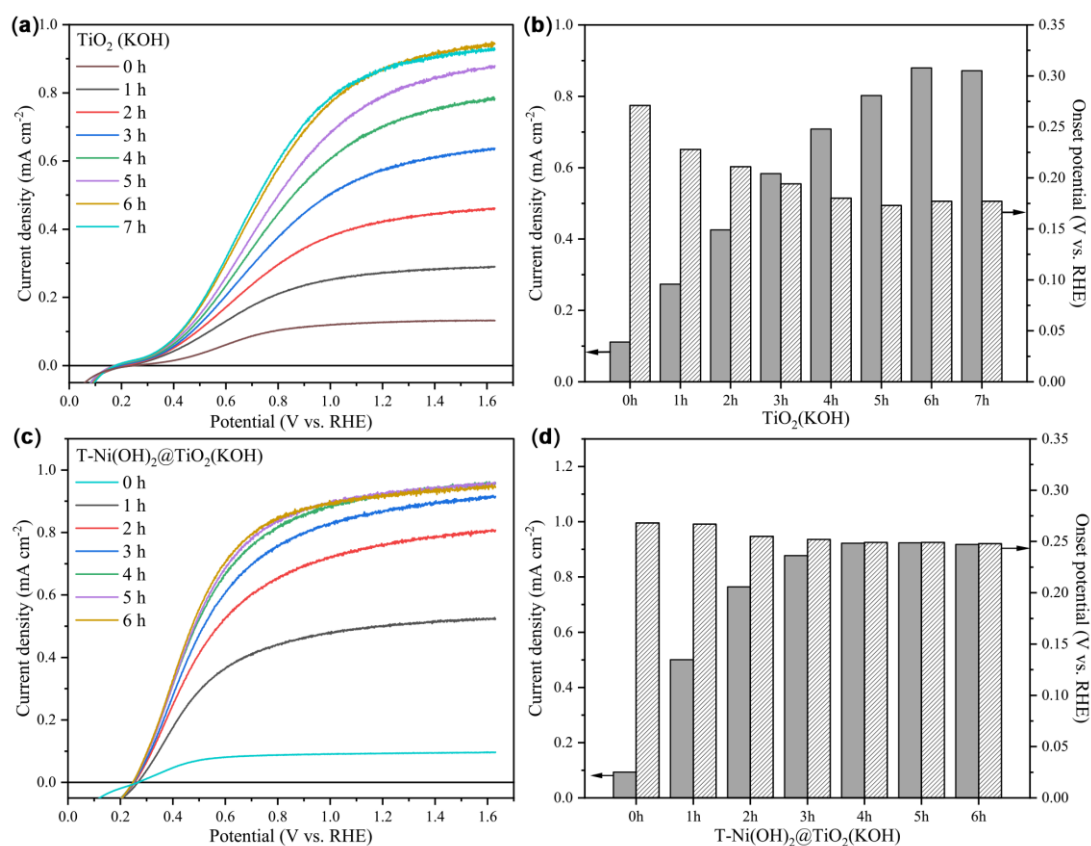


**Figure S11.** LSV curves of TiO<sub>2</sub>, Ni(OH)<sub>2</sub>@TiO<sub>2</sub>, and T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanodes measured in 1 M KOH/0.33 M urea (Solid line) and 1 M KOH/0.33 M urea containing 0.5 M Na<sub>2</sub>SO<sub>3</sub> (Dash line).



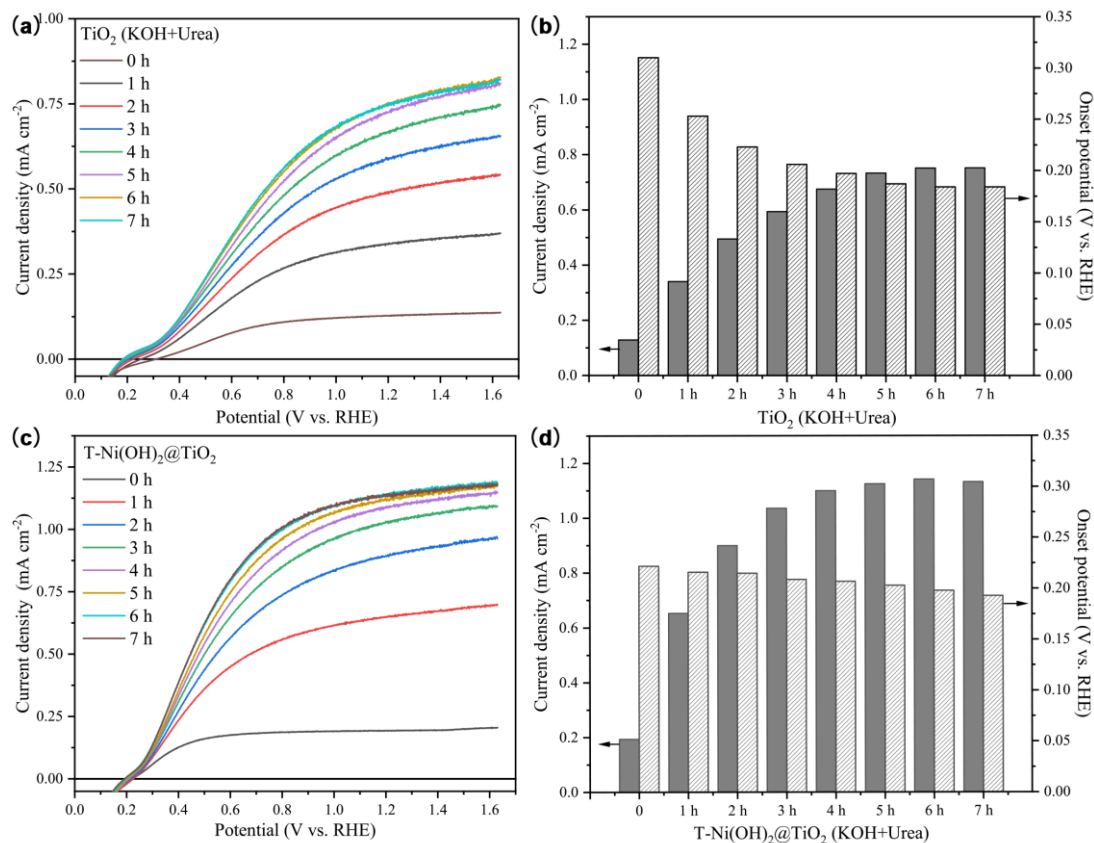


**Figure S12.** MS plots of  $\text{TiO}_2$ ,  $\text{Ni}(\text{OH})_2@\text{TiO}_2$ , and  $\text{T-Ni}(\text{OH})_2@\text{TiO}_2$  photoanodes measured at a fixed frequency of 1000 Hz in the dark.

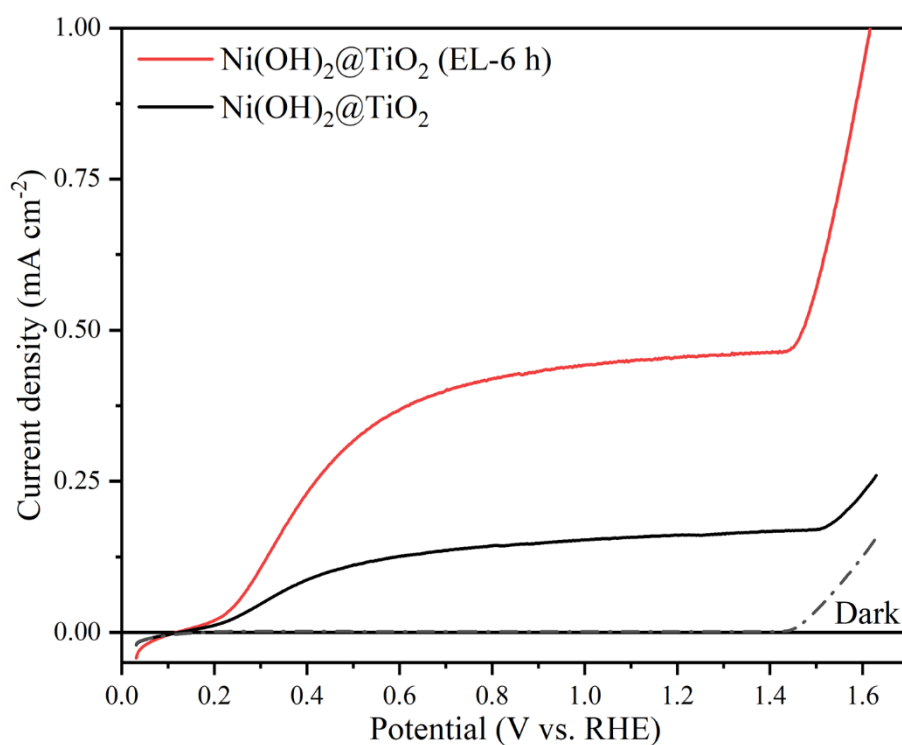


**Figure S13.** (a) LSV curves of  $\text{TiO}_2$  after EL treatment with various times. (b) Histogram of

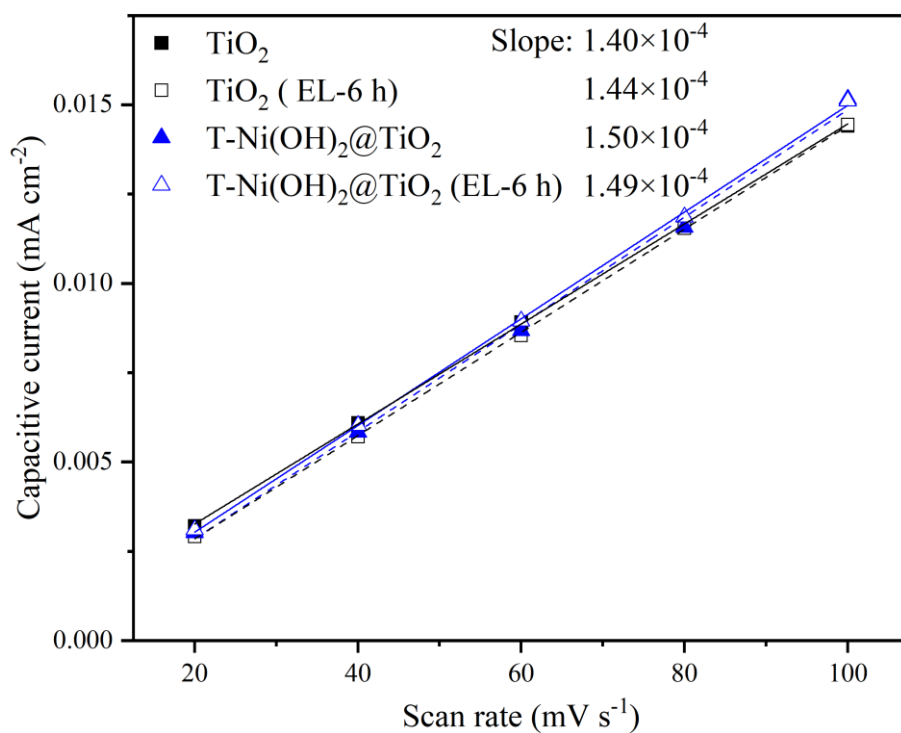
photocurrent density and onset potential of the above  $\text{TiO}_2$  photoanodes. (c) LSV curves of  $\text{T-Ni(OH)}_2@\text{TiO}_2$  after EL treatment with different times. (d) Histogram of photocurrent density and onset potential of the above  $\text{T-Ni(OH)}_2@\text{TiO}_2$  photoanodes. Electrolyte: 1 M KOH. Light illumination: AM1.5G illumination. Scan rate:  $20 \text{ mV s}^{-1}$ .



**Figure S14.** (a) LSV curves of  $\text{TiO}_2$  after EL treatment with various times. (b) Histogram of photocurrent density and onset potential of the above  $\text{TiO}_2$  photoanodes. (c) LSV curves of  $\text{T-Ni(OH)}_2@\text{TiO}_2$  after EL treatment with different times. (d) Histogram of photocurrent density and onset potential of the above  $\text{T-Ni(OH)}_2@\text{TiO}_2$  photoanodes. Electrolyte: 1 M KOH/0.33 M urea. Light illumination: AM1.5G illumination. Scan rate:  $20 \text{ mV s}^{-1}$ .

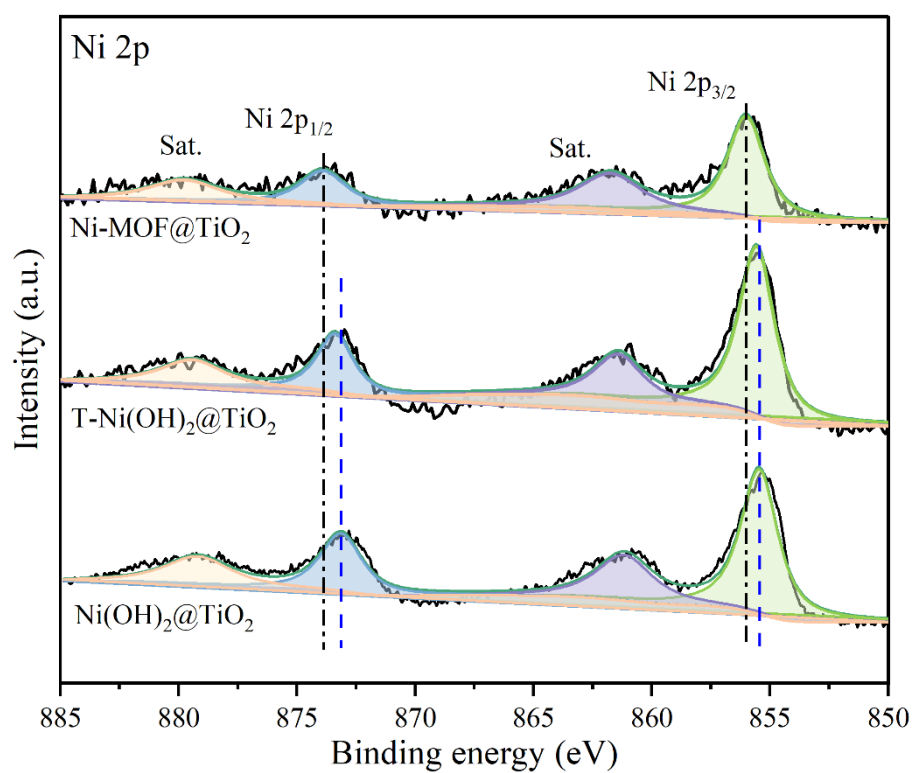


**Figure S15.** LSV curves of  $\text{Ni(OH)}_2@\text{TiO}_2$  photoanode with/without EL treatment of 6 h. Electrolyte: 1 M KOH/0.33 M urea. Light illumination: AM1.5G illumination. Scan rate:  $20 \text{ mV s}^{-1}$ .



**Figure S16.** The relationship curves between the capacitance current and the scan rate of  $\text{TiO}_2$  based photoanodes.





**Figure S17.** Ni 2p XPS spectra of Ni-MOF@TiO<sub>2</sub>, T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> and Ni(OH)<sub>2</sub>@TiO<sub>2</sub>.

## Supplementary Tables

**Table S1.** PEIS fitting parameters of TiO<sub>2</sub>, Ni(OH)<sub>2</sub>@TiO<sub>2</sub> and T-Ni(OH)<sub>2</sub>@TiO<sub>2</sub> photoanodes.

Photoanode	R <sub>s</sub> (Ω)	R <sub>bulk</sub> (Ω)	CPE <sub>bulk</sub> (F)	R <sub>ct</sub> (kΩ)	CPE <sub>ct</sub> (F)
TiO <sub>2</sub>	23.35	/	/	45.37	2.29E-5
Ni(OH) <sub>2</sub> @TiO <sub>2</sub>	9.47	1727.00	3.33E-6	28.54	4.05E-6
T-Ni(OH) <sub>2</sub> @TiO <sub>2</sub>	10.72	569.90	1.64E-6	15.58	2.75E-6