## Supporting information

## Preparation of Nanostructured Ta<sub>3</sub>N<sub>5</sub> Electrodes by Alkaline Hydrothermal Treatment Followed by NH<sub>3</sub>-annealing and Their Improved Water Oxidation Performance

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Figure S1. Schematic diagram of the steps of samples preparation.

Ta <sub>3</sub> N <sub>5</sub> morphology	Modifying layer	Onset potential (V <sub>RHE</sub> )	Initial current @1.23V <sub>RHE</sub> (mA cm <sup>-2</sup> )@AM1.5G	Stability test: I <sub>final</sub> / I <sub>intial</sub> ; Measuring time	Ref.
nanoparticles	Ni:CoFeO <sub>x</sub>	0.76	5.3 @ 100 mW/cm <sup>2</sup> (Xenon lamp)	90%, 7 h	This work
nanorods	Ni-Fe Layered Double Hydroxides (LHD)	0.9	1.7	90 %; 2h	1
nanorods	Co-Pi + Co(OH)x	0.8	5;	20 %; 2h	1
nanorods	IrO <sub>3</sub>	0.8	3	20 %; 20 min	2
nanorods	Co-Pi	0.7	4 @ 0.9V	62 %; 20 min	2,3
nanorods	Co(OH) <sub>x</sub>	0.85	3	93 %; 20 min	4
nanorods	Co-Pi	0.8	3.6	No stability test	5
nanorods	Co-Pi+Co(OH)x/NiFe LDH	0.7	7	71 %, 2 h	1
nanotubes	Co(OH) <sub>x</sub>	0.7	6	96 %; 200 sec	6
nanotubes	Co <sub>3</sub> O <sub>4</sub>	0.8	5	20 %; 6 min	7
nanotubes	Co <sub>3</sub> O <sub>4</sub>	0.95	5	20 %; 1 h	8

Table S1. Water splitting PEC performance of catalyst-modified  $Ta_3N_5$  nanostructures electrodes that recently reported.



Figure S2. SEM images (top-view) of the  $Ta_3N_5$  derived from  $(Na,Ta)O_x$  grown in 23 wt% NaOH solution at (a) 200 °C for 8 h and (b) 250 °C for 5 h.



Figure S3. Current-voltage curves of Ni:CoFeO<sub>x</sub>-loaded Ta<sub>3</sub>N<sub>5</sub> whose precursor electrodes grown in 23 wt% NaOH solution at 250  $^{\circ}$ C for 5 h (black), 200  $^{\circ}$ C for 8 h (red) under dark (broken line) and light (solid line) conditions (100 mW•cm<sup>-2</sup>) in 1 M NaOH (pH 13.6) electrolyte.



Figure S4. SEM image (top-view) of the  $Ta_3N_5$  derived from (Na,Ta)O<sub>x</sub> grown in 16 wt% NaOH solution at 250 °C for 5 h.



Figure S5. SEM image (top-view) of the  $Ta_3N_5$  derived from  $NaTaO_3$  grown in 23 wt% NaOH solution at 250 °C for 3 h.



Figure S6. Current-voltage curves of Ni:CoFeO<sub>x</sub>-loaded Ta<sub>3</sub>N<sub>5</sub> whose precursor electrodes grown in 16 wt% KOH solution at 150  $^{\circ}$ C for 1 h (black), 150  $^{\circ}$ C for 3 h (red), 120  $^{\circ}$ C for 2 h (blue) under dark (broken line) and light (solid line) conditions (100 mW•cm<sup>-2</sup>) in 1 M NaOH (pH 13.6) electrolyte.



Figure S7. SEM image (cross-sectional view) of the  $Ta_3N_5$  derived from (K<sub>1</sub>Ta)O<sub>x</sub> grown in 16 wt% KOH solution at 150 °C for 3 h.



Figure S8. XRD pattern of the NaTaO<sub>3</sub> precursor electrode grown in 23 wt% NaOH solution at 250  $^{\circ}$ C for 5 h.



Figure S9. XRD pattern of the  $K_2Ta_2O_6$  precursor electrode grown in 16 wt% KOH solution at 150 °C for 3 h.



Figure S10. SEM images (top-view) of the  $Ta_3N_5$  derived from  $NaTaO_3$  grown in 23 wt% NaOH solution at 250  $^\circ\!C$  for 5 h.



Figure S11. SEM image (top-view) of the  $Ta_3N_5$  derived from  $K_2Ta_2O_6$  grown in 16 wt% KOH solution at 150 °C for 1h.



Figure S12. (a) XPS spectra of Co2p, (b) Ni2p and (c) peak-fitting Fe2p core levels measured at the surface of Ni:CoFeO<sub>x</sub>-modified Ta<sub>3</sub>N<sub>5</sub> derived from NaTaO<sub>3</sub> prepared in 23 wt% NaOH solution at 250 °C for 5 h.





Figure S13. Current-voltage curves of Ni:CoFeO<sub>x</sub>-loaded Ta<sub>3</sub>N<sub>5</sub> (black) and bare Ta<sub>3</sub>N<sub>5</sub> (red), whose precursor electrodes grown in 23 wt% NaOH solution at 250  $^{\circ}$ C for 5 h, under dark (broken) and light (solid) conditions (100 mW•cm<sup>-2</sup>) in 1 M NaOH (pH 13.6) electrolyte.



Figure S14. Mott-Schottky plots of (a) bare S-NaOH and (b) bare S-KOH. The experiments were conducted under dark condition in 1 M NaOH solution (pH 13.6) as the electrolyte, a Pt wire as the counter electrode, and Ag/AgCl as a reference electrode.



Figure S15. IPCE values (black) of Ni:CoFeO<sub>x</sub>/Ta<sub>3</sub>N<sub>5</sub> photoanode at 1.23 V<sub>RHE</sub> potential under irradiation of Xenon light (100 mW.cm<sup>-2</sup>) and the optical absorbance (red) of the bare Ta<sub>3</sub>N<sub>5</sub>. The precursor NaTaO<sub>3</sub> was grown in 23 wt% NaOH solution at 250 °C for 5 h.

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