

## Supporting Information

# Activation of 2D Titanate Nanosheets Photocatalyst by Nitrogen Doping and Solution Plasma Processing

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## 1. Experimental Section

### 1.1. Characterization

The properties of the materials were investigated by the scanning electron microscope (JEOL JSM 4800F SEM) and transmission electron microscopy (TEM) experiment (JEM-2100F microscope at an acceleration voltage of 200 kV). Elemental analysis (Ti, O, N) was performed on a Vario EL microanalyzer. X-ray diffraction (XRD) patterns were collected using a powder X-ray diffractometer (Rigaku D/MAX-2500 with filtered Cu-K $\alpha$  radiation). Raman spectra were conducted using a JobinYvon HR800 micro-Raman spectrometer with 488 nm laser radiation. X-ray photoelectron spectroscopy (XPS) experiments were performed to verify the electronic states of elements with an Al-K $\alpha$  achromatic X-ray source, and the binding energy was calibrated by the C 1s peak (284.8 eV) of the exotic contaminated carbon. The UV-vis diffuse reflectance spectra were recorded using a JASCO V-770, with Light source D2/WI. Electron spin resonance (ESR) spectra were obtained at room temperature using a Bruker EMXnano spectrometer. Ultraviolet photoelectron spectroscopy (UPS) measurement was carried out with He (I) (21.22 eV) discharge lamp in an ultra-high vacuum chamber by X-ray photoelectron spectroscopy/ESCA. Photoluminescence (PL) spectra were measured on a Jobin-Yvon HR800 spectrophotometer with the excitation wavelength of 325 nm.

### 1.2. Measurement of catalytic activity

The experiment was performed in 500 mL Pyrex glass vessel under solar light. In the photocatalytic test, 20 mg of HTiO catalysts were spread over a square of 2.5 cm $\times$ 2.5 cm glass sheet with 0.2 mL deionized water. The coated glass was put into glass vessel and then the glass vessel was filled with high purity air passing through a water bottle to keep the water vapor concentration at about 70% relative humidity. 5  $\mu$ mol (200 ppm) of acetaldehyde gas was injected into the reactor and dark adsorption for 30 min. The concentrations of CH<sub>3</sub>CHO and CO<sub>2</sub> were measured using a gas chromatograph (GC) (SP-2100A) equipped with a flame ionization detector (FID). Gas samples were collected using a syringe and directly injected into the GC.

### 1.3. Synthesis of photocatalyst

#### 1.3.1. Synthesis of titanate nanosheets (HTiO)

The layered protonic titanate, H<sub>x</sub>Ti<sub>2-x/4</sub>○<sub>x/4</sub>O<sub>4</sub>·H<sub>2</sub>O (x $\sim$ 0.7), was prepared by repeating twice the heat treatment (800 °C, 20 h) of a stoichiometric mixture of Cs<sub>2</sub>CO<sub>3</sub> (3.26 g) and TiO<sub>2</sub> (4.24 g). The interlayer Cs ions were extracted by stirring  $\sim$ 5 g of Cs<sub>x</sub>Ti<sub>2-x/4</sub>○<sub>x/4</sub>O<sub>4</sub> in a 1 mol L<sup>-1</sup> HCl solution (100 mL). After three cycles of acid exchange, the solid was washed with copious water to remove excess acid, filtered, and then collected by centrifugation and lyophilization.

#### 1.3.2. Synthesis of N-doped titanate nanosheets

The N-doped titanate nanosheets were prepared by heat treatment (400 °C, 2 h) of a stoichiometric mixture of urea and HTiO, yellow nitrogen-doped titanate nanosheet layers were obtained. The stoichiometric ratios of HTiO and urea were 4:1, 2:1 and 1:1, respectively, and the obtained samples were recorded as N-HTiO-L, N-HTiO and N-HTiO-H.

### 1.3.3. SPP modified N-doped titanate nanosheets

Plasma was generated by a bipolar-DC pulsed power supply (Kurita, Japan). The voltage, pulse width, and frequency were  $\pm 2.5$  kV, 2.0  $\mu$ s and 20 kHz, respectively. Besides, the solution temperature was kept at 10 °C with cooling circulating water. N-HTiO-SPP was prepared by adding 0.1 g pre-synthesized N-HTiO to 100 mL deionized water. The solution was subjected to ultrasonic treatment for 30 min. N<sub>2</sub> was introduced into the reactor through the hollow tungsten electrode. The flow rate of nitrogen is 2 L min<sup>-1</sup>. The solution plasma process lasted for 1 h. Solution plasma-treated samples were collected by centrifugation and lyophilization.

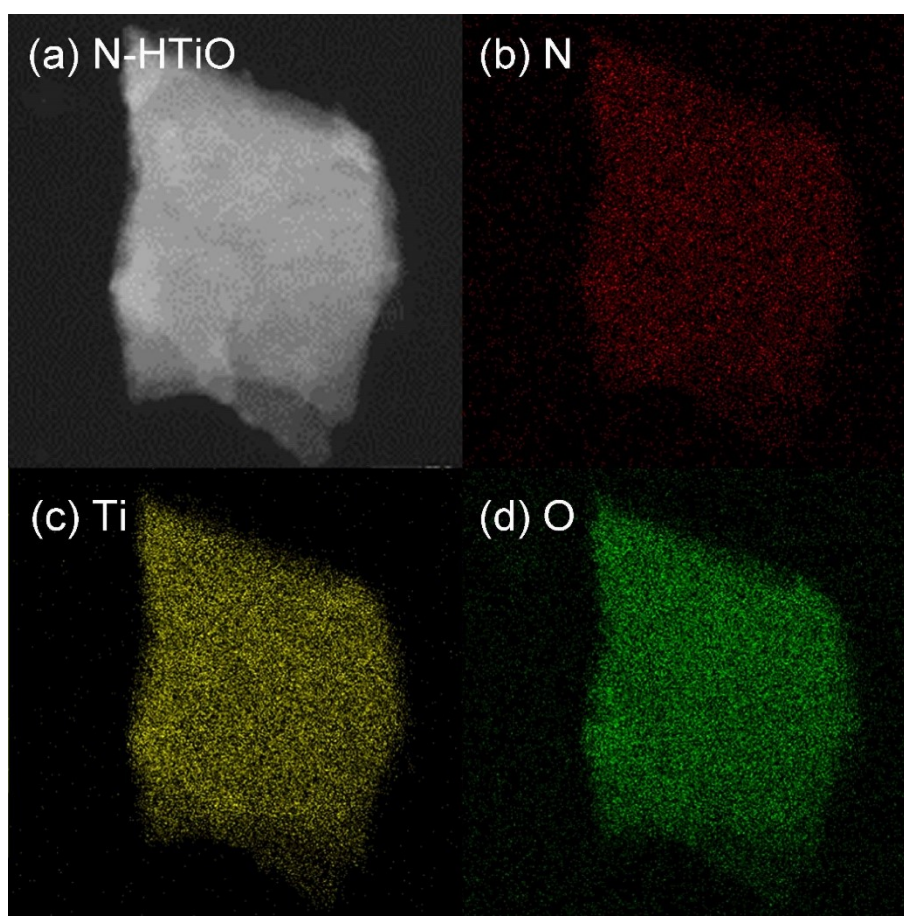


Figure S1. EDS analysis of N-HTiO shows the locations of Ti, O, N.

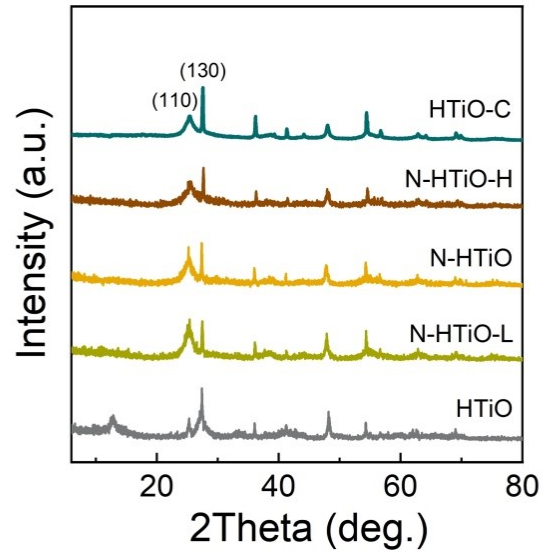


Figure S2. XRD patterns of HTiO, N-HTiO-L, N-HTiO and N-HTiO-H.

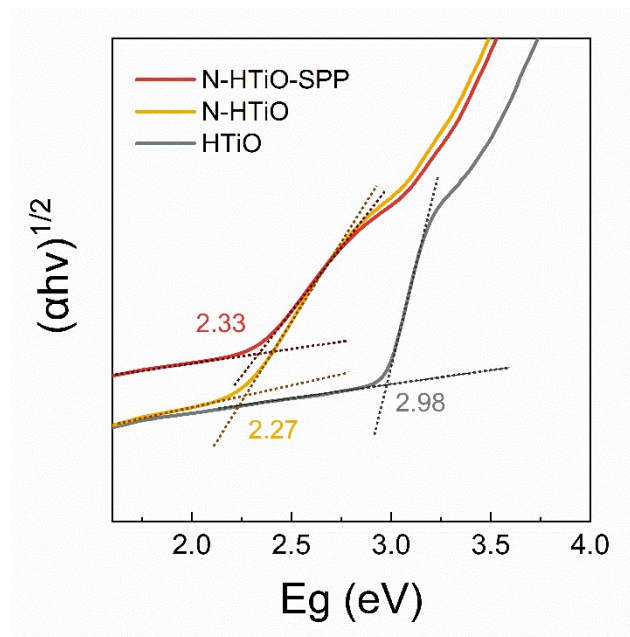


Figure S3. The band gap energies of HTiO, N-HTiO and N-HTiO-SPP, respectively.

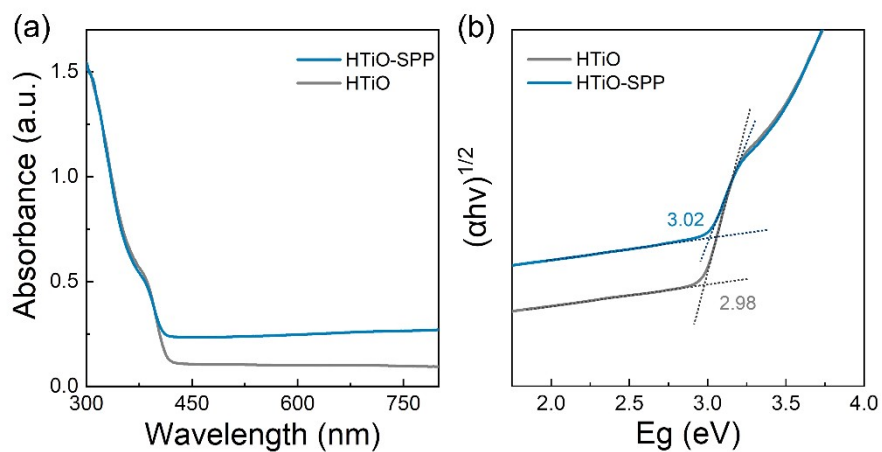


Figure S4. (a) UV-vis absorption spectra and (b) the corresponding band gap energies of HTiO and HTiO-SPP.

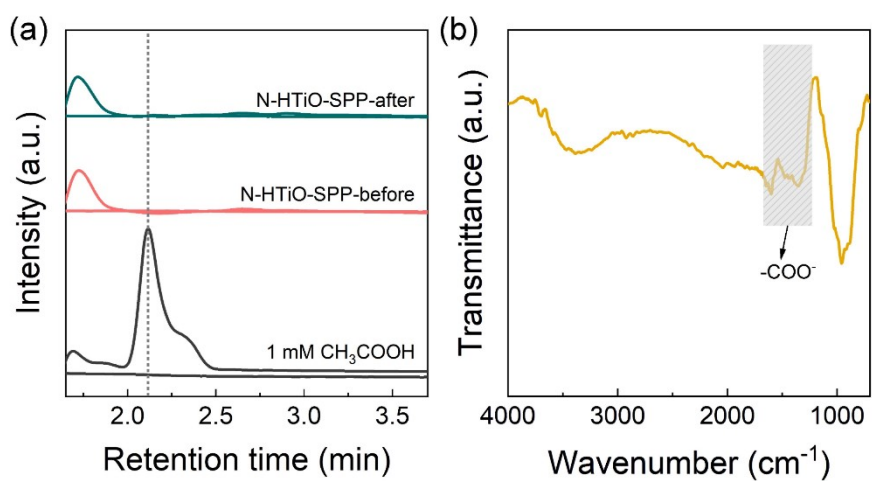


Figure S5. (a) Ion chromatogram of N-HTiO-SPP surface acetic acid before and after acetaldehyde degradation and 1 mM acetic acid standard; (b) DRIFTS spectra collected at room temperature while performing the acetaldehyde photodegradation on N-HTiO-SPP.

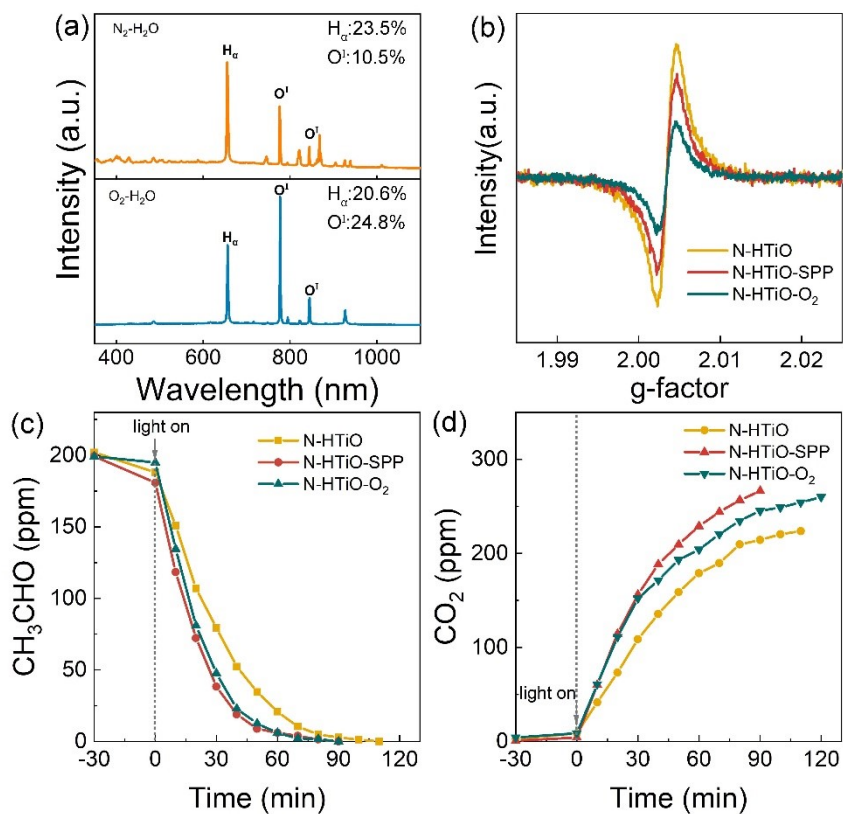


Figure S6. (a) The atomic emission spectrum of the synthesis process of N-HTiO-SPP and N-HTiO-O<sub>2</sub>; (b) The ESR spectra of N-HTiO, N-HTiO-SPP and N-HTiO-O<sub>2</sub>; Time-dependent profiles for the photocatalytic oxidation of (c) CH<sub>3</sub>CHO and concurrent production of (d) CO<sub>2</sub> on N-HTiO, N-HTiO-SPP and N-HTiO-O<sub>2</sub>.

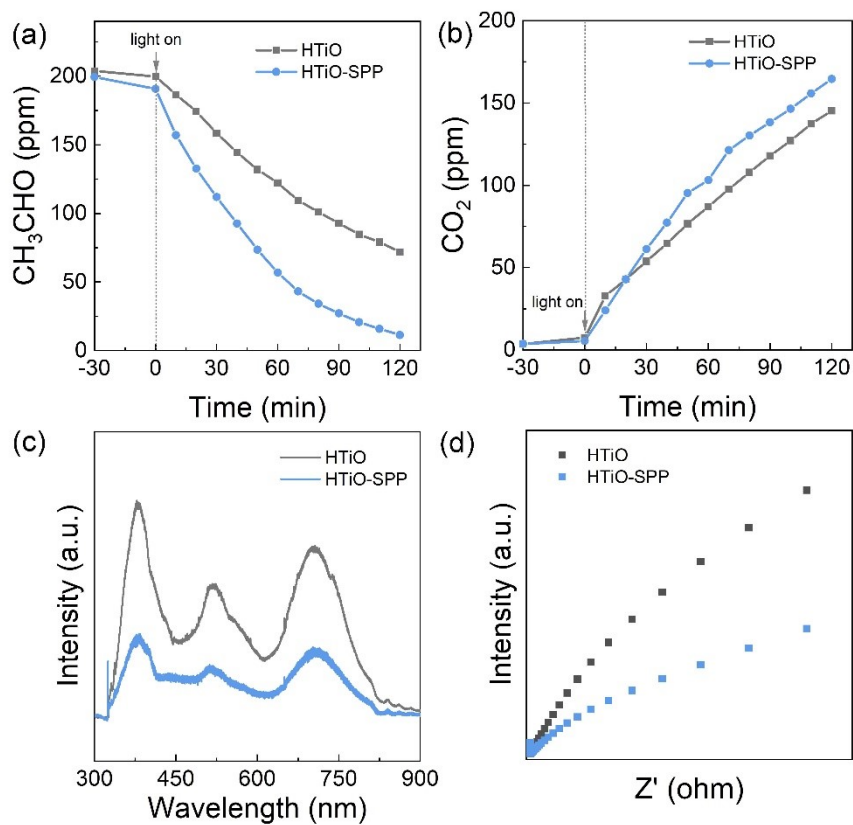


Figure S7. Time-dependent profiles for the photocatalytic oxidation of (a) CH<sub>3</sub>CHO and concurrent production of (b) CO<sub>2</sub> on HTiO and HTiO-SPP; (c) Photoluminescence spectra of HTiO and HTiO-SPP; (d) EIS Nyquist plots of HTiO and HTiO-SPP.

**Table S1** Elemental analysis data of HTiO, N-HTiO and N-HTiO-SPP.

Sample	Ti/wt%	O/wt%	N/wt%
HTiO	46.20	53.80	-
N-HTiO	52.47	46.08	1.45
N-HTiO-SPP	49.20	48.67	2.13