## **Supporting Information**

## **Experimental Section**

Highly ordered  $Ta_2O_5$  NTs were prepared by anodizing Ta foil (99.9%, 0.1 mm, Advent) in a two electrode electrochemical cell by using a Pt sheet as counter electrode. Prior to anodization, the Ta foils were cleaned by sonication in acetone and ethanol for 20 min followed by rinsing in deionized (DI) water and drying in a N<sub>2</sub> stream. The anodization experiments were performed at 60 V at room temperature in an electrolyte consisting of 0.8 wt.% NH<sub>4</sub>F, 13.6 vol% DI water, and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, 98%). The maximum current density is chosen at 0.1 mA cm<sup>-2</sup>. In order to remove the initial layer, the first anodized Ta<sub>2</sub>O<sub>5</sub> NTs were removed ultrasonically in deionized water, and the underlayering Ta was exposed. The second anodization was performed in fresh electrolyte at 60 V for the same time. Then, the prepared NTs were immersed in ethanol for 5 min and dried in N<sub>2</sub>.

For producing Ta<sub>2</sub>O<sub>5</sub> nanotube membranes, the as-prepared Ta<sub>2</sub>O<sub>5</sub> nanotube layers were annealed in air at 450 °C for 1 h. Then the Ta<sub>2</sub>O<sub>5</sub> NT samples were anodized at 80 V for a certain time in the same fresh electrolyte. Afterwards, the nanotube layers were detached from the Ta substrate by immersing the tube layers in an aqueous 5% HF solution for 30-60 min at room temperature. The nanotube layers formed by the third anodization step were still amorphous and therefore underwent preferential chemical dissolution in the HF solution, so that the nanotube layers formed by the second anodization step and pre-annealed could be lifted off as free-standing membranes with open bottom. Then, the Ta<sub>2</sub>O<sub>5</sub> NT membranes were sandwiched between two quartz glasses, followed by annealing in a gaseous atmosphere of NH<sub>3</sub> at 900 °C to obtain Ta<sub>3</sub>N<sub>5</sub> NTs. The temperature was ramped up with a heating rate of 10 °C min<sup>-1</sup>, kept at the desired temperature for 1 h, and finally the furnace was cooled down to the room temperature. Flow-through photocatalytic activities of the photocatalysts were evaluated by degradation of Methylene blue (MB) dye in an aqueous solution (10 mg/L) under visible light irradiation. The nitride membrane was mounted on a PVC holder where the round membrane of 1 cm diameter was glued along its perimeter on a 0.5 cm diameter opening in the holder using epoxy resin. The holder was placed as a separator wall in a two compartment cell with PVC. A 300 W Xeon lamp with a cut-off filter ( $\lambda$ >420 nm, 100 mW cm<sup>-2</sup>), and 532 nm (150 mW) laser point were used as light sources. When the remaining MB and water needed to be measured, at given irradiation time intervals (60 min), 0.5 mL aliquots were collected. Then, for the photocatalytic test of MB and DI water, the absorption UV-vis spectra of the solution were recorded on a spectrophotometer (Lambda Bio XLS, Perkin Elmer). The gold nanoparticles (Sigma-Aldrich, 10 nm diameter, stabilized suspension in buffer) with DI water were prepared for the membranes of flow-through tests.

X-ray diffraction (X'pert Philips MPD with a Panalytical X'celerator detector, Germany) was carried out using graphite monochromized Cu K $\alpha$  radiation (Wavelength 0.154056 nm). Chemical characterization was carried out by X-ray photoelectron spectroscopy (PHI 5600, spectrometer, USA) using AlK $\alpha$  monochromatized radiation. A field-emission scanning electrode microscope (Hitachi FE-SEM S4800, Japan) was used for the morphological characterization of the electrodes. TEM electron microscopy and diffraction patterns were performed on a FEI Tecnai G2 20 S-TWIN with LaB6–cathode, 200 kV accelerating voltage and resolution limit 0.24 nm. The SAED patterns were evaluated by using the software JEMS and incorporating crystal data information from the inorganic crystal structure data base (ICSD). The absorption of the samples were measured using UV/Vis spectrometer (Lambda 950) using an integrating sphere.



**Figure S1.** Current densities of  $Ta_2O_5$  NTs with 1<sup>st</sup>, 2<sup>nd</sup>, and 3<sup>rd</sup> anodization steps. 1<sup>st</sup> and 2<sup>nd</sup> steps anodized at 60 V, and the 3<sup>rd</sup> step anodized at 80 V.



**Figure S2.** SEM top and cross sectional images of  $Ta_2O_5$  NTs after 1<sup>st</sup> anodization step anodized at 60 V for 10 min with various maximum current density: (a1-a3) 0.1 mA cm<sup>-2</sup>, (b1b3) 0.05 mA cm<sup>-2</sup>, (c1-c3) 0.02 mA cm<sup>-2</sup>; (d) current densities as a function of anodization time of  $Ta_2O_5$  NTs.



**Figure S3.** SEM top and cross sectional images of  $Ta_2O_5$  NTs after 1<sup>st</sup> anodization step anodized at various potentials for 5 min with a maximum current density of 0.1 mA cm<sup>-2</sup>: (a1,a2) 80 V, (b1,b2) 60 V, and (c1,c2) 40 V; (d) current densities as a function of anodization time of  $Ta_2O_5$  NTs.

With increasing  $I_{max}$  value (0.02, 0.05, and 0.1 mA cm<sup>-2</sup>) during the 1<sup>st</sup> anodization (Fig. S2), the tube thickness increases from 8 to 16 µm, and is accompanied by a decrease in the thickness of initiation layer, from 450 to 50 nm (the initiation layer is typical for anodic self-organized structures). This initiation layer could be completely removed by increasing the anodization potential to 80 V (at  $I_{max} = 0.1$  mA cm<sup>-2</sup>, see Fig. S3), which is due to the high applied potential, however this leads to bundle formation.



Figure S4. (a,b) Top and (c,d) cross section SEM images of  $Ta_2O_5$  nanotube, (a,c) our work's nanotube layer (0.8 wt.% NH<sub>4</sub>F+13.6 vol% DI water+H<sub>2</sub>SO<sub>4</sub> (98%),  $i_{max}$ =0.1 mA cm<sup>-2</sup>, 60 V); (b,d) traditional nanotube layer (1 vol% HF+4 vol% DI water+H<sub>2</sub>SO<sub>4</sub> (98%), 60 V).

The  $Ta_3N_5$  nanotubes show a more regular and smooth tube structure compared with the  $Ta_2O_5$  nanotubes (as shown in Figure S4).



**Figure S5.** SEM open bottom images of  $Ta_2O_5$  NTs membrane after 3<sup>st</sup> anodization step and etching in 5% HF solution, (a1,a2) part opening bottom; (b1,b2) full opening bottom.



**Figure S6.** Average length of  $Ta_2O_5$  NTs as function of anodization time at 60 V.



Figure S7. Bandgap of nitrided nanotubes at 900 °C, determined from photocurrent measurements



**Figure S8.** (a) Schematic representation of the setup used for flow-through permeation experiments. The chamber on the left is filled with DI water (A), the chamber on the right is filled with Au colloid aqueous solution (10 nm diameter, Au:H<sub>2</sub>O=1:1), and between them a Ta3N5 NT membrane is glued on a holder with an opening; (b) flowing-through efficiency of Au aqueous solution (1:1 Au and H<sub>2</sub>O) (Au/M/water) versus the exposure time under AM 1.5G ( $\lambda$ >420 nm) and dark conditions.

The Au concentration was evaluated spectrophotoscopically, by measuring the intensity of the 521 nm absorption peak that originates from plasmon resonance in the Au colloid.



**Figure S9.** Bead-penetration measurement for bead polystyrene aqueous solution before and after 30 h in dark condition.

Bead-penetration measurement was performed using an aqueous solution containing polystyrene beads with a 600 nm average particle size; there were no leaks or cracks in the NT membrane as the beads solution did not leak inside the other chamber.



**Figure S10.** (a) Degradation efficiency of MB aqueous solution in chamber B; (c) photodegradation efficiency of MB aqueous solution in chamber B versus the exposure time under  $\lambda$ =532 nm and AM 1.5G ( $\lambda$ >420 nm) illumination.



**Figure S11.** (a) Degradation efficiency of MB aqueous solution in chamber B; (b) degradation of MB concentration in chamber A; (c) degradation of MB concentration in chamber A and B (a+b), versus the exposure time under  $\lambda$ =532 nm and AM 1.5G ( $\lambda$ >420 nm) illumination; (d) comparison of degradation efficiency of Ta<sub>3</sub>N<sub>5</sub> NT membrane in dark and AM 1.5G ( $\lambda$ >420 nm) conditions.



**Figure S12.** Photodegradadtion measurement for the Ta3N5 nanotubes immersed in MB solution for hours in dark and under light (532nm).



**Figure S13.** Optical images of solutions used to test the Ta<sub>3</sub>N<sub>5</sub> NT membrane before and after irradiation in various conditions: (a) before irradiation; (b) in dark condition for 41 h; (c) after laser 532 nm irradiation for 30 h; (d) after AM 1.5G ( $\lambda$ >420 nm) irradiation for 22 h.



Figure S14. Photocurrent measurements evaluated to band gap of a typical  $Ta_3N_5$  nanotubular layer immersed in 1 M KOH solution without and with MB.



**Figure S15.** Recyclability of the Ta<sub>3</sub>N<sub>5</sub> NT membrane for the photocatalytic decomposition of MB in AM 1.5G ( $\lambda$ >420 nm) illumination



**Figure S16.** Recyclability test of the Ta<sub>3</sub>N<sub>5</sub> NTs for the photocatalytic decomposition of MB in 532nm or AM 1.5G ( $\lambda$ >420 nm) illumination.

We do not observe a significant difference if 532nm light is used in the recycling test, compared to AM 1.5G in similar conditions (illumination was performed for 6 h for one cycle), as exemplified for Ta<sub>3</sub>N<sub>5</sub> NTs in Fig. S16.



**Figure S17.** XRD patterns of Ta<sub>3</sub>N<sub>5</sub> NT membranes after AM 1.5G ( $\lambda$ >420 nm) irradiation cycling photocatalytic tests.



**Figure S18.** XPS Ta 4p (a), N 1s (b), and O 1s (c) spectra of Ta<sub>3</sub>N<sub>5</sub> NT membranes before and after AM 1.5G ( $\lambda$ >420 nm) irradiation cycling photocatalytic test.

Taking into account the  $\sim 20~\mu m$  tube length of the membrane, one would expect the photocatalytic effect to be confined to the illuminated side, with a penetration depth of a few micrometers (Fig. S19).



**Figure S19.** Degradation efficiency of MB aqueous solution in chamber B versus the exposure time under AM 1.5G ( $\lambda$ >420 nm).