## **Supporting Information on**

## Controlled fabrication of porous double-walled TiO<sub>2</sub> nanotubes via ultraviolet-assisted anodization

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Figure S1. FESEM image (cross-sectional view) of the as-anodized double-walled  $TiO_2$  nanotubes.



**Figure S2.** TEM image of the single-walled  $TiO_2$  nanotubes prepared in the EG electrolyte at 60V without UV illumination: (a) as-anodized and (b) annealed nanotubes.



**Figure S3.** FESEM image of  $TiO_2$  nanotubes formed on the UV-illuminated (a,b) and nonilluminated (c,d) side of a Ti sheet. Scale bars of the SEM images are 100 nm.



Figure S4. XPS survey spectra of the amophous and annealed double-walled TiO<sub>2</sub> nanotubes.



**Figure S5.** High resolution spectra of (a) C1s, (b) F1s and (c) O1s of amorphous single, amorphous double-walled, and annealed double-walled  $TiO_2$  nanotubes (TNTs).



**Figure S6.** FTIR spectra of an as-anodized single-walled  $TiO_2$  nanotubes prepared without UV-illumination and a double-walled  $TiO_2$  nanotubes prepared under UV-illumination.



Figure S7. UV-vis absorption spectra of the single and doubled-wall  $TiO_2$  nanotubes and Ti-sheet.



**Figure S8.** Hole-induced conversion of a single hydroxide bond to double hydroxide bonds through the interactio with a water molecule [21, 25, 26].



Figure S9. pH value of the electrolyte during the anodization with UV-illumination.

## **BET specific surface measurement and pore size distribution:**

The BET measurement show the specific surface area of the single and double-walled  $TiO_2$  nanotubes attached on Ti-substrate is 1.8 m<sup>2</sup>/g and 2.8 m<sup>2</sup>/g, respectively. However, it should be noted that these results have large error and can show only the relative surface area between the two nanotubes. This is because the TiO<sub>2</sub> nanotubes are attached on a Ti metal substrate, and hence, the exact weight of only the TiO<sub>2</sub> nanotubes can't be determined [1]. In order to measure the specific surface area and the pore size distribution of TiO<sub>2</sub> nanotubes accurately and more precisely, TiO<sub>2</sub> nanotubes have to be separated from the substrates [1-3] which is beyond the scope of the present manuscript.

Figure S10a give the  $N_2$  adsorption-desorption isotherm of the single and doubled-wall TiO<sub>2</sub> nanotubes. The isotherm shows the typical behavior of the mesoporous and macroprous morphology [3, 4]. The isotherm exhibit that the double-walled TiO<sub>2</sub> nanotubes have high specific surface area compared to the single-walled TiO<sub>2</sub> nanotubes [4]. Figure S10b displays the Barrett–Joyner–Halenda (BJH) pore-size distributions of the single and doubled-wall TiO<sub>2</sub> nanotubes. The pore size distributions are relatively wide, ranging between 2.6 and 160 nm. The doubled-wall TiO<sub>2</sub> nanotubes exhibit the presence of multi-modal porosity ranging from mesoporous to macroporous morphology, which is consistence with our SEM results.

## References

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**Figure S10.** a) Nitrogen adsorption-desorption isotherm plots, and b) pore size distributions curve of the single and double-walled TiO<sub>2</sub> nanotubes, c) magnified part of ( b).