Supporting Information on

Controlled fabrication of porous double-walled TiO$_2$ nanotubes
via ultraviolet-assisted anodization

Ghafar Ali, Hyun Jin Kim, Jae Joon Kim, and Sung Oh Cho*

Department of Nuclear and Quantum Engineering, Korea Advanced Institute of Science and Technology, Daejeon 305-701 Korea, E-mail: socho@kaist.ac.kr

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 TiO₂ nanotubes formed on the UV-illuminated (a,b) and non-illuminated (c,d) side of a Ti sheet. Scale bars of the SEM images are 100 nm.

**Figure S4.** XPS survey spectra of the amorphous and annealed double-walled TiO₂ 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 interaction 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$^2$/g and 2.8 m$^2$/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$_2$ nanotubes are attached on a Ti metal substrate, and hence, the exact weight of only the TiO$_2$ nanotubes can’t be determined [1]. In order to measure the specific surface area and the pore size distribution of TiO$_2$ nanotubes accurately and more precisely, TiO$_2$ 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$_2$ nanotubes. The isotherm shows the typical behavior of the mesoporous and macroporous morphology [3, 4]. The isotherm exhibit that the double-walled TiO$_2$ nanotubes have high specific surface area compared to the single-walled TiO$_2$ nanotubes [4]. Figure S10b displays the Barrett–Joyner–Halenda (BJH) pore-size distributions of the single and doubled-wall TiO$_2$ nanotubes. The pore size distributions are relatively wide, ranging between 2.6 and 160 nm. The doubled-wall TiO$_2$ nanotubes exhibit the presence of multi-modal porosity ranging from mesoporous to macroporous morphology, which is consistence with our SEM results.

References

Figure S10. a) Nitrogen adsorption-desorption isotherm plots, and b) pore size distributions curve of the single and double-walled TiO$_2$ nanotubes, c) magnified part of (b).