Supplementary Materials

Figure S1. Morphology images of TiO_2 nanotubular arrays prepared by anodizing Ti foil at 20 V for 20 min in 0.5 wt % HF solution, followed by (a) dry in air at 100 °C for 1 h, and (b-d) hydrothermal solid/liquid crystallization (sample was exposed to water) at (b) 130 °C, (c) 160 °C, (d) 180 °C for 4 h. The violent solid/liquid interface reaction damaged the nanotubular architecture.



Figure S2. Cross-sectional image of TiO_2 nanotubular arrays prepared by anodizing Ti foil at 20 V for 20 min in 0.5 wt % HF solution followed by dry in air at 100 °C for 1 h. The length of TiO_2 nanotubes is ~380 nm.



Figure S3. The photocurrent density/time trace curves at +0.2 V (vs. Ag/AgCl) potential when the UV irradiance was switched on (ON) and off (OFF) alternately. The as-anodized TiO₂ nanotubes were treated by (a) thermal anneal at 180 °C and at atmospheric pressure for 4 h, (b) hydrothermal solid/gas reaction at 180 °C for 4 h and (c) thermal anneal at atmospheric pressure at 450 °C for 4 h.



The photocurrent density of low-temperature crystallized TiO_2 nanotubular arrays (curve b) was much lower than that of 450 °C-annealed sample (curve c). This was because the crystallization temperature of 180 °C in our present route was much lower than 450 °C, while the conventional thermal anneal at 180 °C and atmospheric pressure can not realize the anatase crystallization (curve a).