**Supporting information:**

# A facile route to aligned $TiO_2$ nanotube arrays on transparent conducting oxide substrates for dye-sensitized solar cells

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**Details for preparation of ZnO NRAs template by hydrothermal method:**

30-nm-thick ZnO thin film was firstly deposited on the clean transparent conducting F:$SnO_2$ (FTO) glass by sol-gel method to seed the oriented growth of ZnO NR templates. Briefly, 0.75 mol $Zn(CH_3COO)_2 \cdot 2H_2O$ was dissolved in a 100 ml 2-methoxyethanol-monoethanolamine solution at room temperature. The resultant solution was stirred at 60 °C for 0.5 h to yield a clear and homogeneous sol. Then cleaned FTO substrates were slowly immersed into the ZnO sol for 10 s and then withdrawn at 2 mm/s. Subsequently they were preheated at 300 °C for 10 min to form a ZnO gel seed layer. Finally, the preheated substrates were heated from 300 to 550 °C and hold at 550 °C for 1 h to obtain the ZnO seed layers.

The growth solutions for the PPAHT route were prepared by dissolving $Zn(NO_3)_2 \cdot 6H_2O$, $(CH_2)_6N_4$ and polyethylenimine (PEI, branched, low molecular weight) in distilled water with a concentration ratio of 0.02:0.02:0.0005. 150 ml growth solution was first sealed in a glass bottle of maximum volume 250 ml and was then heated to 95 °C till precipitation occurs in the solution. Then the ZnO-seeded FTO substrates were quickly immersed in the turbid growth solution and tilted against the wall of the bottle with ZnO seed layers facing down. Subsequently the bottle was heated to 95 °C without any stirring. For a high growth rate of ZnO NRAs template, the growth solution was refreshed every 12 h (a growth cycle) to supply a sufficient sources, and the desired length is controlled by the number of growth cycle. After growth, the resultant ZnO NR templates were removed from the vials, rinsed thoroughly with ethanol to remove any residual reactants and dried in air.

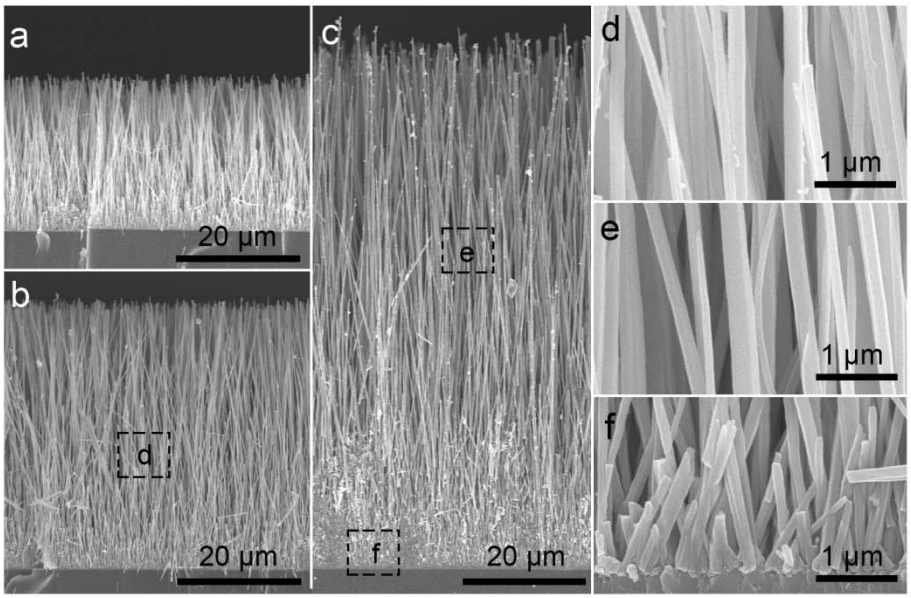


Figure S1 Side-view FESEM images of ZnO NRAs templates with various lengths. Middle-magnification FESEM image of ZnO NRAs with: a) 20 μm, b) 40 μm, and c) 80 μm, respectively. High-magnification FESEM images of ZnO NRAs with: d) 40 μm, and e) 80 μm, as indicated by black rectangles in Figure S1b and S1c, respectively. f) high-magnification FESEM image near the interface between nanorod and FTO film, as indicated by black rectangle in Figure S1d.

Figure S1 shows side-view FESEM images of ZnO NRAs templates with different lengths ranging from 20 to 80 μm by tailoring PPAHT growth cycle number. The middle-magnification FESEM images (Figure S1a, S1b and S1c) suggest that ZnO NRs are nearly perpendicular to the ZnO-seed FTO glass substrates, and the length of ZnO NRAs templates increases linearly with the growth cycle number (and thus the cumulative growth time), with an addition of about 4 μm per cycle (0.33 μm per hour). Figure S1d, S1e shows high-magnification FESEM image of ZnO NRAs templates with 40 and 80 μm in length, respectively, which was captured from Figure S1b and S1c as indicated by black rectangle. An apparent broadening of size distribution was observed and the average diameter of ZnO NRs increases from 150 to 200 nm with increasing the length of ZnO NRs from 40 to 80 μm. Additionally, Figure S1f shows the interface between nanorods and FTO film of ZnO NRAs templates with 80 μm in length, revealing that ZnO NRs directly grow from ZnO-seeded FTO glass substrates.

At the same time, no continuous, dense intermediate layer was formed between nanorods and FTO film, which is generally observed from ZnO NRAs synthesized by traditional hydrothermal method (See Supporting information Figure S1-1), formed by the convergence of the adjacent ZnO NRs. In our case, the convergence of the adjacent ZnO NRs was restrained by two factors: 1) the density of ZnO NRs decreases with a lower concentration of reactive agents resulting from preheating process, 2) the radial growth of nanorods is validly restrained by introducing higher concentration PEI, which is easily adsorbed onto the lateral facets of ZnO NRs due to the electrostatic affinity to cut off the raw reactive supply.

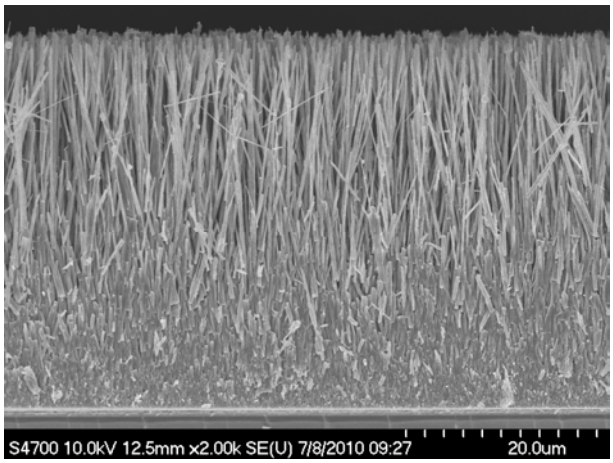


Figure S1-1 Typical FESEM image of ZnO NRAs fabricated by hydrothermal method ( Reference [25] )

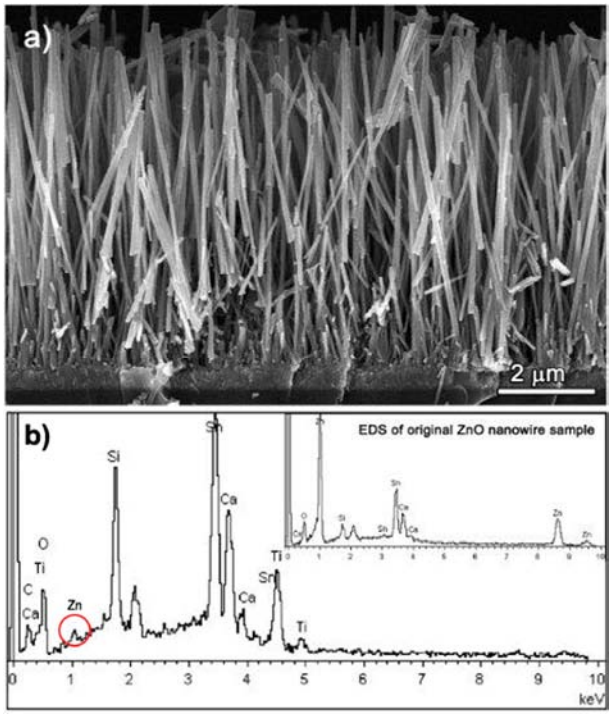


Figure S2 Side-view FESEM image of $TiO_2$ NTAs (a) and its corresponding EDS curve (b). Inset is EDS curve of ZnO NRAs templates

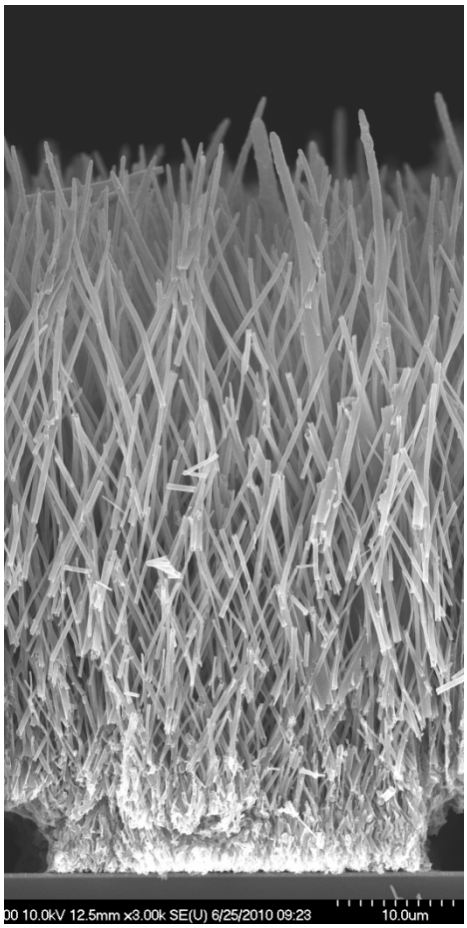


Figure S3 FESEM image of $TiO_2$ NTAs with a large diameter, showing a free-top bundling morphology.

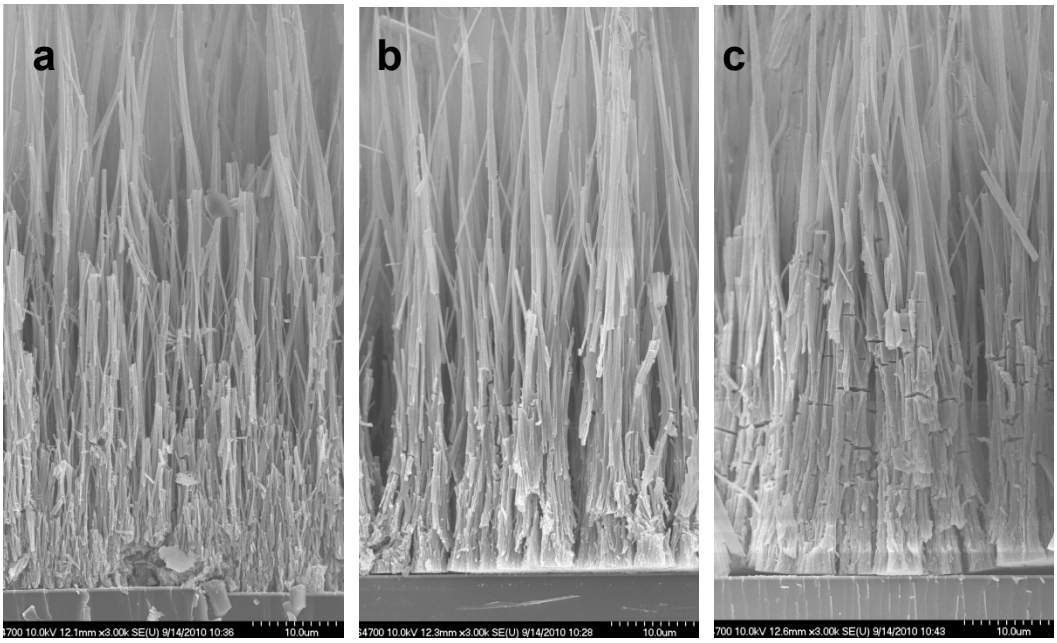


Figure S4 FESEM images of longer $TiO_2$ NTAs with different thickness from various LBL-AR cycle numbers. (a) 10, (b) 20 and (c) 30.