## **Electronic Supplementary Information (ESI)**

## **Crystallographically Preferred Oriented TiO<sub>2</sub> Nanotube Arrays for Efficient Photovoltaic Energy Conversion**

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**Fig. S1.** The FESEM images of the NTAs grown in 2 wt% (a) and 5 wt% (b) water-added electrolytes. The NTA grown using a water content of 2 wt% had a smooth surface, whereas the NTA grown using 5 wt% of water content had a few small ribs. The arrows in (b) indicate the ribs.



**Fig. S2.** (a) XRD patterns of the NTAs (5 wt% of water content) annealed at various temperatures, and (b) peak heights of anatase (101) plane and rutile (110) plane.



**Fig. S3.** Fourier transform infrared spectra of the r-NTA and p-NTA samples before and after annealing at 450°C. The as-grown p-NTA contained fewer hydroxyl groups than the r-NTA as evidenced by the low absorbance peak between 3000 and 3600 cm<sup>-1</sup> associated with the O-H stretching mode.



**Fig. S4**. XRD peak intensity ratio of (004) plane to (200) plane as a function of water content. Inset shows the rocking curves of the NTAs, showing a texture-like characteristic of p-NTAs.

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## Page S6



Fig. S5. TEM image of a thin oxide layer formed between the nanotubes and Ti metal substrate, after

annealing at 450°C for 1 hr. The thickness of the thin oxide layer was about 70 nm.



**Fig. S6.** Thickness (length) of the p-NTA with increasing anodizing time. The thickness was measured by using FESEM.



**Fig. S7.** Incident photon-to-current efficiency (IPCE) of the p-NTA and the NP film-based DSSCs measured under the back-side illumination and the front-side illumination. Most of the UV light is blocked by the counter electrode and the electrolyte, under the back-side illumination.