

Experimental Section

Materials and Reagents: ITO–PEN (Peccell, sheet resistance $13 \Omega \text{ square}^{-1}$, $200 \mu\text{m}$) was used as a transparent conducting substrate. N719 dye purchased from DYESOL LTD was used as photosensitizer. A titanium metal disk (60 mm in diameter and 5 mm in thickness) with a purity of 99.99% was used as the target. High-purity oxygen and argon gases ($>99.99\%$ purity) were used as reactive gases.

Growth of Aligned TiO_2 Nanorod Array Anode: Well-aligned TiO_2 nanorod arrays were prepared on the commercial ITO–PEN substrates by dc reactive magnetron sputtering technique using a common sputtering system (MS500B) equipped with a turbo molecular pumping system. Prior to deposition, the ITO–PEN substrates with dimension of $2 \times 2 \text{ cm}^2$ were thoroughly rinsed by acetone, anhydrous ethanol, and deionized water. After putting the cleaned ITO–PEN substrate on the sputtering chamber, the gas pressure of the chamber was pumped down to $8.0 \times 10^{-4} \text{ Pa}$, followed by flowing oxygen gas (20 sccm) and argon gas (40 sccm). The target–substrate distance was 50 mm. By controlling the sputtering pressure of 0.6 Pa, well-aligned TiO_2 nanorod array was deposited at a sputtering power of 185 W for 5 hours. The deposition rate was controlled at around 100 nm h^{-1} .

Fabrication of TiO_2 Nanoparticle Anode: The low-temperature processed TiO_2 nanoparticle anodes can be prepared according to the following procedures: (1) Under vigorous agitation at room temperature, 100 ml of deionized water was mixed with 10 ml of titanium tetrabutanolate. After 30 min, the mixture was pump-filtrated to obtain dehydrated filter powders. (2) Subsequently, 10 ml of acetic acid and 0.8 ml of nitric acid were added into the above filter powders dropwise in a flask. After an agitation of 15 min at $80 \text{ }^\circ\text{C}$, the volume of the mixture was adjusted to 170 ml by adding deionized water. The reactant was further agitated at $80 \text{ }^\circ\text{C}$ for 15 min in a sealed atmosphere. (3)

Later, the resultant colloid was transferred into a Teflon-lined autoclave and heated at 200 °C for 12 h. (4) A homogeneous mixture consisting of 65 ml of the white colloid and 0.4 g of commercial P25 were made under ultrasonic irradiation for 30 min and (5) then transferred to another Teflon-lined autoclave, which was subsequently heated at 200 °C for 12 h. (6) After removing supernatant liquid, the colloid was mixed with 0.8 g of poly(ethylene glycol) ($M_w = 20,000$) and 1 ml of OP emulsifier and subsequently concentrated at 80 °C. ITO-PEN substrates with a size of $2 \times 2 \text{ cm}^2$ were thoroughly rinsed by deionized water and anhydrous ethanol, and dried by N_2 gas flow. The photoanodes from TiO_2 nanoparticles were fabricated by a spin-coating method. The size of the resultant photoanodes was controlled at $0.5 \times 0.5 \text{ cm}^2$ with an average thickness of 550 nm for TiO_2 layer. The air-dried colloids were calcined in a muffle furnace at 120 °C for 30 min.

Electrochemical Deposition of Pt Counter Electrode: The Pt counter electrode was prepared by a potentiostatic method at a potential of -0.3 V and a deposition time of 300 s. To realize this strategy, a 20 mM H_2PtCl_6 aqueous solution was used as an electrolyte. The deposition of Pt CE was carried out at room temperature and surrounding atmosphere.

Assembly of Flexible DSSC Devices: The anode from well-aligned TiO_2 nanorods or traditional TiO_2 nanoparticles was soaked in a 0.3 mM N719 [cis-di(thiocyanato)-N,N'-bis(2,2'-bipyridyl-4-carboxylic acid-4-tetrabutylammonium carboxylate, purchased from Solaronix, SA, Switzerland] ethanol solution for 24 h to uptake N719 dye for the fabrication of dye-sensitized TiO_2 photoanode. The flexible DSSCs were assembled by sandwiching a dye-sensitized TiO_2 photoanode, a liquid electrolyte containing I^-/I_3^- redox couples, and a Pt counter electrode. A redox electrolyte consisted of 100 mM of tetraethylammonium iodide, 100 mM of tetramethylammonium iodide, 100 mM of tetrabutylammonium iodide, 100 mM of NaI, 100 mM of KI, 100 mM of LiI, 50 mM of I_2 , and 500 mM of 4-tert-butyl-pyridine in 50 ml acetonitrile.

Photovoltaic Measurements: The photovoltaic test of the DSSC was carried out by measuring the photocurrent–voltage (J – V) characteristic curves using the CHI660E Electrochemical Workstation under irradiation of a simulated solar light from a 100 W Xenon arc lamp (XQ-500 W) in ambient atmosphere. The incident light intensity was controlled at 100 mW cm^{-2} (AM1.5 calibrated by a standard silicon solar cell). Each J – V curve was measured at least five times to eliminate experimental error and a compromise J – V curve was employed.

Other Characterizations: The morphologies of the aligned TiO_2 nanorods were captured with a Zeiss Ultra plus field emission scanning electron microscopy (FESEM) and a transmission electron microscopy (TEM, JEM2010, JEOL). Before TEM measurement, the TiO_2 arrays were carefully scraped from the substrate and dispersed in ethanol solution. After ultrasonic dispersion for 5 min, the solution was dipped on a copper screen for TEM observation. The optical absorption spectra were recorded on a UV–vis spectrophotometer (Agilent 8453) at room temperature. Fourier transformed Raman spectroscopic measurements in the ultraviolet light were performed on a Renishaw inVia Reflex Raman Spectrometer. High–resolution gratings were used to give a spectral resolution of 2 cm^{-1} . The spectra were recorded at room temperature from 3200 to 100 cm^{-1} using 16 scans with an exposure time of 1 s per scan. X–ray diffraction (XRD) profiles of the resultant nanocrystallines were recorded on an X–ray powder diffractometer (X’pert MPD Pro, Philips, Netherlands) with $\text{Cu K}\alpha$ radiation ($\lambda = 1.542\text{\AA}$) in the 2θ range from 10 to 70° operating at 40 kV accelerating voltage and 40 mA current). For electrochemical impedance spectroscopy (EIS) measurements, the DSSCs were scanned from 0.1 Hz to 2 MHz at an ac amplitude of 10 mV . Incident photo–to–current conversion efficiency (IPCE) curves were obtained at the short–circuit condition on an IPCE measurement systems (MS260). The light source in this case was a solar simulator (PEC–L11, AM1.5G, Peccell Technologies, Inc.); Light was focused through a

monochromator onto the photovoltaic cell. The monochromator was moved in steps from 400 to 700 nm to generate the IPCE. In order to obtain the IPCE spectra, the anodes were sensitized by N719 dye.

Supporting Figures

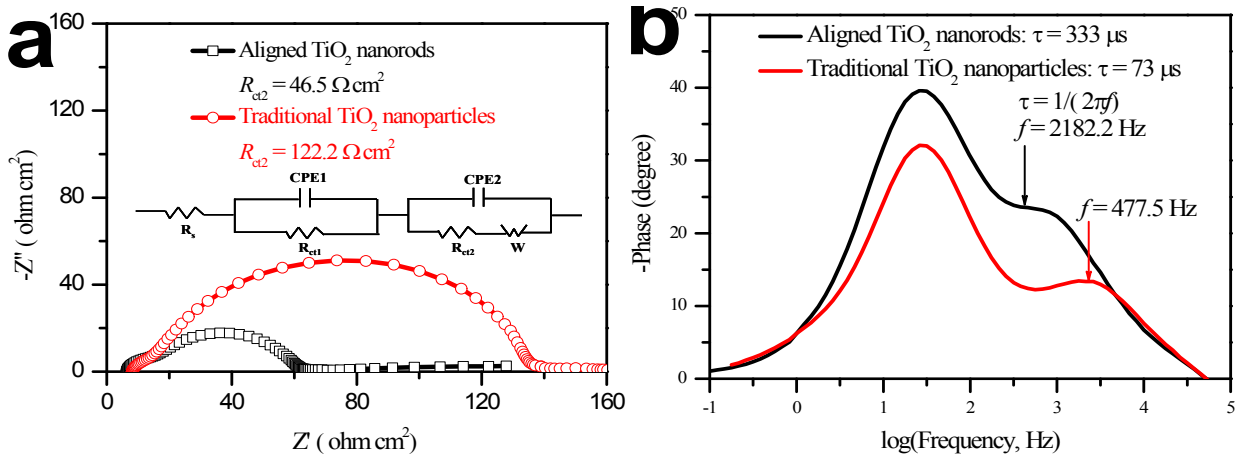


Fig. S1 (a) Bode and (b) EIS spectra for the flexible DSSCs with aligned TiO₂ nanorods and traditional TiO₂ nanoparticles. R_s : sheet resistance; R_{ct1} : charge-transfer resistance at counter electrode/electrolyte interface; R_{ct2} : charge-transfer resistance at TiO₂/dye/electrolyte interface; W : Nernst diffusion impedance corresponding to the diffusion resistance of I⁻/I₃⁻ redox couples; CPE1 and CPE2 are constant phase elements; τ : electron lifetime.