Supporting Information

Flexible nano-TiO$_2$ sheets exhibiting excellent photocatalytic and photovoltaic properties by controlled silane functionalization – Exploring the new prospects of wastewater treatment and flexible DSSCs.

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**Materials used**

All chemicals were purchased and used directly. Titanium (IV) butoxide (Sigma-Aldrich, 97%), ethanol (Merck, 96%), hydrochloric acid (Merck, 37%), deionized water, trimethyloxysilane (Sigma-Aldrich, 95%), toluene (Sigma-Aldrich, 99.8%), N-methylglycine (Sigma-Aldrich, 98%), methylene blue (Sigma-Aldrich, 82%), N-719 dye (Sigma-Aldrich, 95%), and tert-butyl alcohol (Merck, 99.5%). silver nitrate (Sigma-Aldrich, 99%), ethylene diamine tetra acetic acid, EDTA (Alfa-Aesar, 99%), isopropyl alcohol (Alfa-Aesar, 99.5%) tris(2-(1H-pyrazol-1-yl) pyridine) cobalt(III) tri [hexafluorophosphate] or FK 102 Co(III) PF₆ salt (Sigma Aldrich 98%), tris(2-(1H-pyrazol-1-yl)pyridine)cobalt(II) di[hexafluorophosphate] or FK 102 Co(II) PF₆ salt (Sigma Aldrich 98%), tert-butyl pyridine (Merk, 98%), lithium perchlorate (Sigma Aldrich, 99.9%), acetonitrile (Merk, 99.9%), iodine (Sigma Aldrich, 99.8%), potassium iodide (Alfa- Aesar, 99%), PET sheet (G M Polyplast Limited, Mumbai), indium tin oxide (ITO) coated polyethylene terephthalate (PET) sheet (Brand-Technistro, Resistivity < 100 ohms/sq,Transmittance -> 75%).

**Methods**

**Doctor blading technique**: M-TiO₂ powder was mixed with ethanol and ground in a mortar and pestle to obtain a smooth slurry. thin films of M-TiO₂ were prepared by doctor blading 30 µL slurry on an indium tin oxide (ITO) coated polyethylene terephthalate (PET) sheet by fixing 36-micron transparent plain cello self-adhesive tapes on the sides of the ITO sheets.

**Characterization techniques**

**X-Ray Diffraction (XRD)**: Powder X-ray diffraction (XRD) was recorded using a Brucker D8 Advanced X-ray diffractometer equipped with a 2-dimensional Vantec-500 detector. Monochromatized CuKα radiation with characteristic wavelength Kα₁: 1.54056 Å, in the 2θ range from 20-80° (generator's tube voltage 40 KV, tube current 40 mA, scan rate 0.04
degree/minute). The sample was smeared over a low background sample holder (amorphous silica holder) and fixed on the sample stage in a goniometer. The instrument was set with B-B geometry. Coupled theta/two theta mode was used, with a \( \theta \) angle that was half of the 2\( \theta \) angle. As prepared powder samples were loaded and analyzed. Data were collected at room temperature and pressure. Analyses were performed on the collected XRD data using the Bruker XRD search match program EVA™4.3. Crystalline phases were identified using the ICDD-JCPDS powder diffraction database.

The average particle size (D) of the samples was calculated using the Debye-Scherrer equation.

\[
D = \frac{K \lambda}{d \cos \theta} \tag{1}
\]

where K is the particle shape factor, 0.9, \( \lambda \) is the wavelength of X-ray used (0.15 nm), d is the full width at half maximum intensity of the peak (in Rad), and \( \theta = 2\theta/2 \) is the Bragg angle (in degrees) corresponding to the reflection.

The relative contents of the anatase and rutile phases in the M-TiO\(_2\) sample were calculated from the XRD intensity of the characteristic peaks of the phases according to equation 2,\(^{39}\) and were found to be 89.21\% for anatase and 10.78\% for rutile phases, respectively.

\[
W_A = \frac{K_AI_A}{(K_AI_A + I_R)} \tag{2}
\]

where \( W_A \) is the mole fraction of anatase, \( I_A \) and \( I_R \) are the integrated intensities of the characteristic peaks of the anatase and rutile phases in the XRD data, respectively, and \( K_A = 0.886 \).

**Fourier Transform Infra-Red Spectroscopy:** FTIR spectra were recorded using Shimadzu 8400 spectrometer in the range 4000-400 cm\(^{-1}\).
The UV-visible diffuse reflectance spectroscopy: Absorption spectra of the samples were recorded using UV-2450 Shimadzu UV-visible spectrophotometer. The optical band gap of thin films of TiO$_2$ and M-TiO$_2$ was calculated using Tauc’s equation.

\[(\alpha h\nu)^{1/n} = A (h\nu - E_g)\]  

(3)

where $\alpha$ is the light absorption extinction coefficient, $h$ is Planck’s constant, $\nu$ is the light frequency, $A$ is the absorption constant, and $E_g$ is the optical energy bandgap of the material.

Photoluminescence spectroscopy: photoluminescence measurement was carried out using a Shimadzu RF-5301 spectrofluorometer with the samples excited at a wavelength of 320 nm.

Time-correlated single-photon counting (TCSPC): TCSPC was done using a fluorescence spectrometer with lifetime detector (FL3C21) having 330nm LEDy in the air at 298 K. Emission was monitored at 470 nm and split width 29. The instrument response function was monitored using milk powder as a reference.

Raman spectroscopy: Raman spectra were recorded using a JY HR 800 spectrometer in the backscattering geometry with a 458 nm argon-ion laser as the excitation source.

Scanning Electron Microscopy: SEM was taken using JEOL/EO JSM-6390 SEM in a backscattered electron detection mode was obtained with an FEI Magellan 400 XHR FEGSEM microscope at a voltage of 3 kV and spot size 2. Samples were immobilized on a specimen stub using double-sided carbon tape.

High-resolution transmission electron microscopy (HRTEM): TEM images were taken using a JEOL JEM-2100 microscope at an accelerating voltage of 200 kV. The sample was dispersed in isopropyl alcohol solution and drop casted on a carbon coated copper grid (B 200 mesh) and analyzed.
The Brunauer-Emmet-Teller (BET): BET specific surface areas and porous structures of the samples were analyzed by a Quantachrome instrument Nova 1200. All samples were degassed at 140°C before nitrogen adsorption measurements were taken.

Energy Dispersive X-ray Spectroscopy: EDX were recorded at 10 kV and spot size of 4.5

X-ray Photoelectron Spectroscopy: The surface analytical studies were performed using a K-alpha XPS spectrometer (Thermo Fisher Scientific, East Grinstead, UK). All samples were analyzed using a micro-focused, monochromatic Al Kα X-ray source at a power of 180 W (15 kV × 12 mA) and a hemispherical analyzer in fixed analyzer transmission mode (spot size: 30–400 mm) at a pressure of about 1 × 10⁻⁸ mbar. The K-Alpha charge compensation system was employed during analysis, using electrons (8 eV) and low-energy argon ions to prevent any localized charge build-up. The spectra were fitted with CASA XPS.

Electrochemical characterization: Electrochemical measurements were conducted by a Biologic SP-200 electrochemical workstation and EC-Lab software. I-V Measurements were recorded using an Oriel MiniSol LED Solar Simulator, Class ABA, equipped with I sun power (100 mW cm⁻²) and an AM 1.5G filter and calibrated using a silicon reference solar cell.

The photoconversion efficiency of the device was calculated using equation (4).

\[ \eta = \frac{V_{oc} \times J_{sc} \times FF}{P_{in}} \]  

(4)
Figures and tables

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**Figure S14.** PL spectra of samples with different weight percentages of trimethoxy silane (TiO$_2$: trimethoxy silane as 0, 1:1, 1:2, 1:4, and 1:20).
| Sample | Peak intensity at wavelength 470 nm (counts) | Peak intensity at wavelength 420 nm (counts) | Peak intensity ratio | Sample | Peak intensity at wavelength 470 nm (counts) | Peak intensity at wavelength 420 nm (counts) | Peak intensity ratio |
|--------|------------------------------------------|------------------------------------------|-------------------|--------|------------------------------------------|------------------------------------------|-------------------|
| 0      | 5221                                     | 3267                                     | 1.59              | 0      | 5201                                     | 3416                                     | 1.52              |
| 1:1    | 3994                                     | 2760                                     | 1.44              | 1:1    | 943                                      | 631                                      | 1.49              |
| 1:2    | 2098                                     | 1549                                     | 1.35              | 1:2    | 481                                      | 419                                      | 1.14              |
| 1:4    | 2811                                     | 1960                                     | 1.47              | 1:4    | 709                                      | 578                                      | 1.22              |
| 1:20   | 4234                                     | 2760                                     | 1.53              | 1:20   | 1236                                     | 975                                      | 1.26              |

**Table S1.** The ratio of peak intensity (intensity of peak at 470 nm / intensity of peak at 420 nm) from the PL spectra of samples with different weight percentages of trimethoxy silane (TiO$_2$: trimethoxy silane as 0, 1:1, 1:2, 1:4, and 1:20).
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|                | DSSC with TiO$_2$ | DSSC with M-TiO$_2$ |
|----------------|-------------------|----------------------|
| $R_s$ (Ω)      | 26.67             | 26.76                |
| $R_{CE}$ (Ω)   | 23.20             | 30.424               |
| $R_{ct}$ (Ω)   | 562.8             | 710.6                |
| $C_{CE}$ (F)   | 12.07e-6          | 13.08e-6             |
| $C$ (F)        | 3.64e-6           | 3.84e-6              |

Table S2. Values for $R_s$, $R_{CE}$, $R_{ct}$, $C_{CE}$ and $C$ of the DSSCs employing TiO$_2$ and M-TiO$_2$ as photoanodes from the EIS data.

Figure S25. Equivalent circuit of the DSSC.