

## **An Electroless-Plating-Like Solution Deposition Approach for Large-Area Flexible Thin Film of Crystalline Transition Metal Oxides**

### **Supporting information**

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### **Experimental Section**

The PMC precursors were prepared as described below:

peroxo-titanium-complex (PTC) solution,  $\text{TiCl}_4$  was first treated with ammonia to form titanic acid precipitates. The precipitate was then dissolved into 30 wt.%  $\text{H}_2\text{O}_2$  solution, and then the solution was diluted to  $M[\text{Ti}] = 0.125 \text{ mol L}^{-1}$ .

peroxo-vanadium-complex (PVC) solution, a total of 0.23g of vanadium pentoxide powders was first added to 250ml of distilled water to produce a light yellow slurry solution. After the yellow slurry solution was stirred for 15 min, 2.5 mL of 30%  $\text{H}_2\text{O}_2$  was introduced dropwise to the slurry solution and an orange solution formed.

peroxo-molybdenum-complex (PMoC) solution, a total of 0.1g of molybdenum powders was first added to 100ml of distilled water to produce a slurry solution. and 2.5 mL of 30%  $\text{H}_2\text{O}_2$  was introduced drop wise to the slurry solution and an yellow solution formed.

### **Characterization**

X-ray diffraction (XRD) experiments were performed with an XD-2 diffractometer (Purkinje General Instrument Co. Ltd.), using monochromatic  $\text{Cu K}\alpha$  radiation at 36

kV and 30 mA.

Fourier transform infrared (FT-IR) spectra were obtained by using an Excalibur 3100 spectrometer with a resolution of  $4\text{ cm}^{-1}$ . Measurements were performed in the transmission mode in spectroscopic grade KBr pellets for all the powders. Transmission electron microscopy (TEM) studies were performed using Philips CM200/FEG fieldemission-gun transmission electron microscope, operating at 200 kV.

The surface composition and elemental chemical state of the samples were examined by X-ray photoelectron spectroscopy (XPS) using a Model Axis Ultra (Kratos Analytical Ltd.) apparatus.

The surface morphology of the sample was investigated by a scanning electron microscopy (Hitachi Ultra-High-Resolution S-4300).

The ultra-violet photoelectron spectroscopy (UPS) was measured using a Model Axis Ultra (Kratos Analytical Ltd.) apparatus.

Element	Atomic%		
	TiO <sub>2</sub>	V <sub>2</sub> O <sub>5</sub>	MoO <sub>3</sub>
C	44.49	58.65	52.86
O	46.77	35.55	41.59
M	2.96	2.75	3.34
Au	2.22	3.05	3.32

Figure S1 Flexible PET films composition coated with TiO<sub>x</sub>, V<sub>2</sub>O<sub>x</sub>, and MoO<sub>x</sub> obtained by EDS.

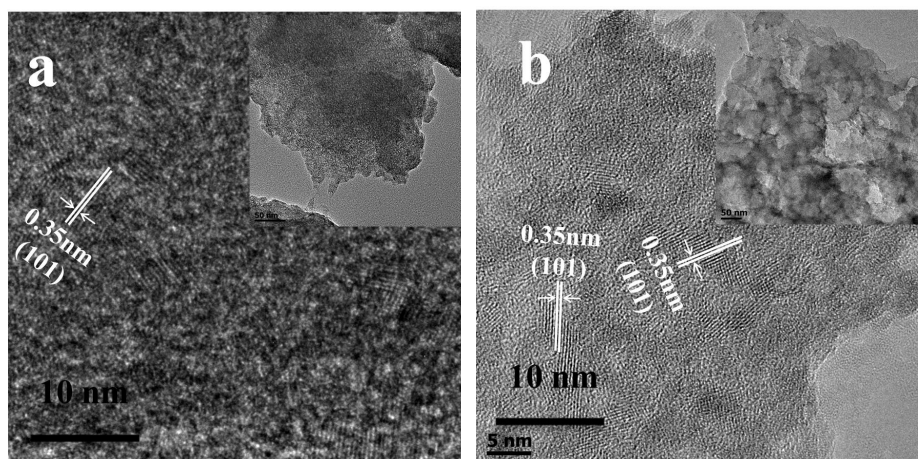


Figure S2 Morphology of the samples that were scraped off the PET films with a sharp knife. TEM images of the as scraped off samples from the EPLSD processed PET films based on the reaction between PTC and (a) EDOT, (b)Pr.

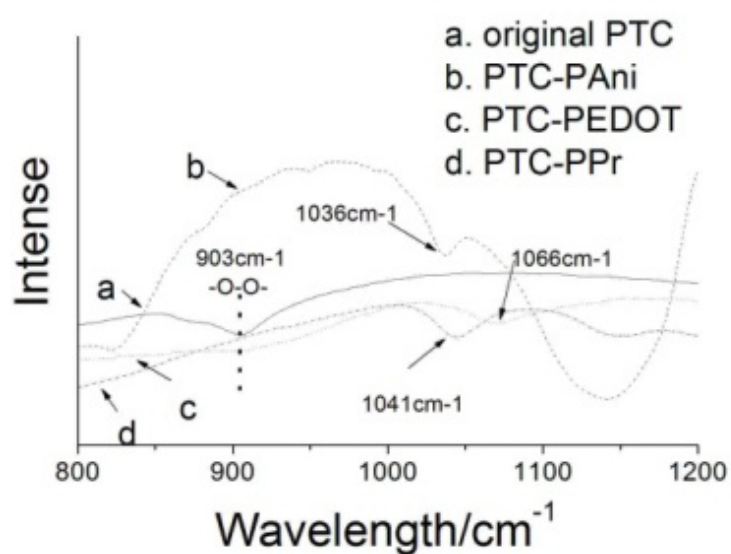


Figure S3 FTIR spectrum taken from the as scraped off samples from the EPLSD processed PET films based on the reaction between PTC and aniline, EDOT and pyrrole.

The FTIR spectra of the as prepared PAni/TiO<sub>2</sub>, PEDOT /TiO<sub>2</sub> and PPr/TiO<sub>2</sub> samples were measured and the results are shown in Figure S2. A characteristic peak

at  $903\text{ cm}^{-1}$  in PTC is attributed to the peroxy bond, which disappears in the spectrum of Polymer/ $\text{TiO}_2$  complex, revealing that the peroxy bond has been broken during the hybrid process. Meanwhile, new bands at  $1036\text{ cm}^{-1}$ ,  $1041\text{ cm}^{-1}$  and  $1066\text{ cm}^{-1}$  can be observed in the PAni/ $\text{TiO}_2$ , PEDOT / $\text{TiO}_2$  and PPr/ $\text{TiO}_2$  composition respectively, which might be assigned to the stretching vibration of Ti-O-C bonds.

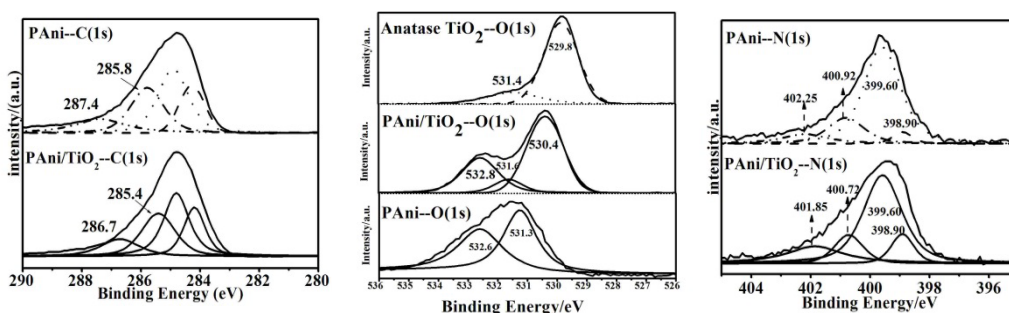


Figure S4 XPS spectra of samples: (a) N (1s) spectrum of pure bulk PAni and PAni/ $\text{TiO}_2$  nanocomposite (AN1/Ti1), (b) O (1s) spectrum of Anatase  $\text{TiO}_2$  and PAni/ $\text{TiO}_2$  (AN1/Ti1), (c) C (1s) spectrum of pure bulk PAni and PAni/ $\text{TiO}_2$  nanocomposite (AN1/Ti1).

XPS was further used to clarify the relationship between the Polyaniline and  $\text{TiO}_2$ . Fig. 4(a) is C (1s) peak, showing that a shifts of the binding energy (BE) of  $\text{C-N}^+/\text{C=N}^+$  and C-O or C=O for PAni/ $\text{TiO}_2$  from 285.81 and 287.48 to 285.41 and 286.74 eV can be observed. Meanwhile, as was contrasted in fig. 4b, except for the residual peak at 532.60 eV for the absorbed  $\text{H}_2\text{O}$  molecular, the BE of O (1s) in PAni/ $\text{TiO}_2$  shifts from 529.84 and 531.41 to 530.40 and 531.62 eV. The N(1s) peak (Fig. 4c) revealed that a variation of BE of 400.92 and 402.25 shifting to 400.72 and 401.85 eV can be characterized. All the above XPS peaks shifting confirmed with the

suggestion that a Ti-O-N-C and Ti-O-C bonding may be generated between PANi and TiO<sub>2</sub> nanoparticles, which could explain the strong adhesion between the metal oxides and the PET substrates.

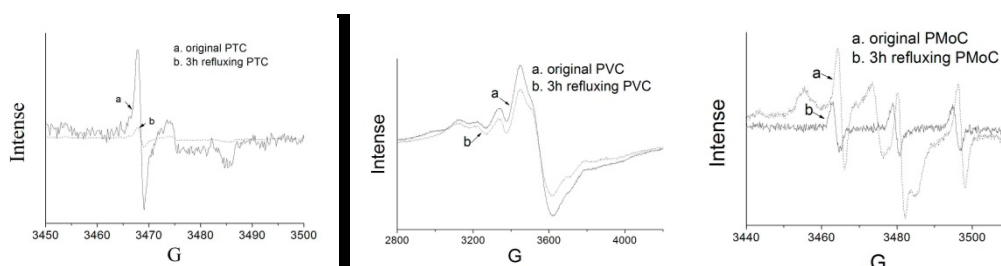


Figure S5. ESR spectrum of the precursor of PVC at room temperature (a) original and (b) after 3h refluxing.

PTC aqueous solution displayed, at room temperature, by ESR a triplet with 'g' value 2.003, 2.009 and 2.020 ( shown in fi. 2a) which, according to the literature, was identified as belonging to the specific interactions between d<sup>0</sup> configuration Ti<sup>4+</sup> and peroxy group in the precursor solutions. Shown in fig. 2b displayed the characteristic ESR spectrum of PVC aqueous solution produced by the coupling of the unpaired electron of the vanadium (IV) with the spin = 7/2 vanadium nucleus and which is consistent with literature report. The spectra obtained by us shown in fig. 2c also indicate the presence of complicated species, according to the observation of literature report, the resonance at g = 1.92 being attributed to Mo (V). When the PMC precursor undergoes a refluxing reaction at 100 ° C for 3 h, a notable signal intense decline of the peak ESR line can be observed for all the PMC solution as was shown in fig. 2, which revealed that the radical concentration was inversed to the refluxing time.

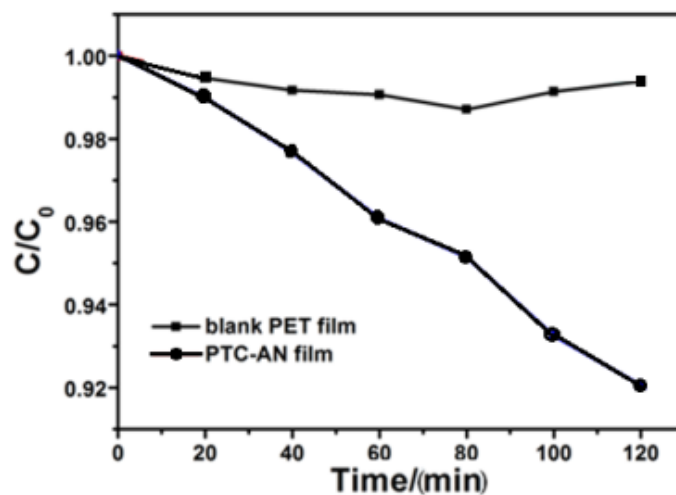


Figure S6. Photocatalytic degradation curves of aqueous MB irradiated under UV light (365 nm) of (a) blank PET film and (b) PBD treated TiO<sub>2</sub> deposited PET film.