

## Supporting Information

### **Two-dimensional polyimide heterojunction for the efficient removal of environmental pollutants under visible-light irradiation**

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### **Experimental section**

#### **1.1 Chemicals and Reagents**

Melamine ( $C_3N_3(NH_2)_3$ ), pyromellitic anhydride ( $C_{10}H_2O_6$ ), 3,4,9,10-decanetetracarboxylic dianhydride ( $C_{24}H_8O_6$ ), sodium sulfate ( $Na_2SO_4$ ), isopropanol

(C<sub>3</sub>H<sub>7</sub>OH), 4-hydroxy-TEMPO, methyl violet (MV), rhodamine B (RhB), triethanolamine, barium sulfate (BaSO<sub>4</sub>), K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> were purchased from Aladdin Chemical Co., Ltd., China. The purity of all reagents is analytical (AR) and not further purified.

## 1.2 Characterization methods

The surface morphology of the photocatalysts has been studied by a JEOL JSM 4800F SEM. Transmission electron microscopy (TEM) and HRTEM images were characterized utilizing a JEM-2100F microscope at an acceleration voltage of 200 kV. X-ray diffraction measurements were carried out on a Bruker AXS D8 Focus by filtered Cu K $\alpha$  radiation ( $\lambda = 1.54056 \text{ \AA}$ ). X-ray photoelectron spectra were obtained applying an ESCALABMKII spectrometer with an Al-K $\alpha$  achromatic X-ray source. The UV-Vis diffuse reflectance spectra (DRS) measurements were received by a UV-2600 UV-Vis spectrophotometer (Shimadzu), and BaSO<sub>4</sub> was employed as a background. The PL spectra were gained on a Hitachi F-7000 spectrophotometer with the excitation wavelength of 385 nm. Fourier transform infrared spectra (FTIR) were derived from Perkin Elmer Fourier transform infrared spectrometer GX. The N<sub>2</sub> adsorption-desorption isotherms were determined at 77 K with a Micromeritics Autosorb IQ instrument.

## 1.3 Photoelectrochemical measurements

Photocurrent measurements were carried out using a CHI700E Electrochemical Workstation in a common three-electrode configuration including a reference electrode, counter electrode and working electrode in a quartz cell. Ag/AgCl electrode and Pt foil were used as the reference electrode and counter electrode, respectively. A 300 W Xe lamp (CEL-HXF300, AULIGHT) was utilized as the light source, and a 0.5 M Na<sub>2</sub>SO<sub>4</sub> solution was employed as the electrolyte. The working electrodes were prepared as the following method: 30 mg of different samples were dispersed in 1 mL of ethanol with sonication for 30 min to obtain slurry. Subsequently, the mixture was

evenly dropped onto a  $1 \times 3 \text{ cm}^2$  FTO glass substrate. Finally, the prepared electrodes were air-dried at room temperature to gain the working electrodes.

#### **1.4 Electrochemical impedance spectroscopy (EIS) measurements**

Electrochemical impedance spectra (EIS) was tested using a CHI700E electrochemistry station in  $0.5 \text{ M Na}_2\text{SO}_4$  under visible-light illumination with the frequency ranging from  $0.01 \text{ Hz}$  to  $10 \text{ kHz}$  at  $-0.2 \text{ V}$ . The EIS data were recorded using a normal three-electrode system, in which samples on FTO glass with an active area of ca.  $2.0 \text{ cm}^2$  were prepared as the working electrode, Pt wire as a counter electrode, and Ag/AgCl as a reference electrode.

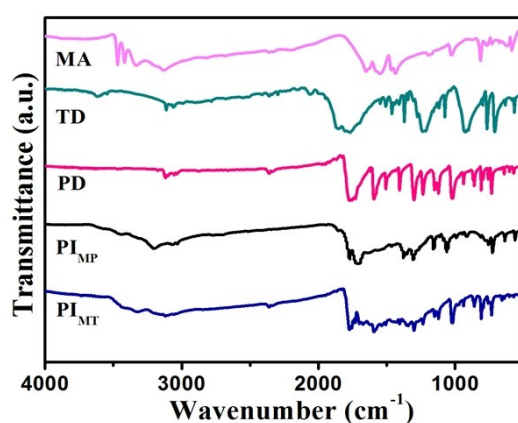
#### **1.5 Photocatalytic tests**

The photocatalytic activities of the as-obtained specimens were evaluated by the photodegradation of RhB, MV and photoreduction of  $\text{K}_2\text{Cr}_2\text{O}_7$ . A glass vessel with a water cooling jacket was applied as reactor, and a  $300 \text{ W Xe}$  lamp was employed as visible-light source with a  $420 \text{ nm}$  cut-off filter. Briefly,  $50 \text{ mg}$  of different samples were dispersed into a solution, which contains  $50 \text{ mL}$  of RhB solution ( $10 \text{ mg} \cdot \text{L}^{-1}$ ),  $50 \text{ mL}$  of MV solution ( $10 \text{ mg} \cdot \text{L}^{-1}$ ),  $40 \text{ mL}$  of  $\text{K}_2\text{Cr}_2\text{O}_7$  solution ( $80 \text{ mg} \cdot \text{L}^{-1}$ ;  $\text{V H}_2\text{O} : \text{V isopropanol} = 1$ ). Before the light irradiation, the mixture was stirred in darkness for a certain time interval until adsorption-desorption equilibrium. Subsequently, the light source was switched on, and further stirring was applied. Afterward,  $1 \text{ mL}$  of the reaction solution was extracted and centrifuged at constant time intervals. The supernatant of RhB, MV and  $\text{K}_2\text{Cr}_2\text{O}_7$  was detected using a Shimadzu UV-2600 UV-Vis spectrophotometer. The degradation efficiency was calculated by  $C/C_0$ , where  $C$  represents the concentration of trace RhB, MV and  $\text{K}_2\text{Cr}_2\text{O}_7$  solution at each irradiated time, and  $C_0$  stands for the concentration before light irradiation.

#### **1.6 Active species trapping experiment**

We conducted the radical-trapping experiments aiming to explore the major active species in the photocatalytic degradation process of RhB. In our study, the 4-hydroxy-

TEMPO, triethanolamine (TEOA), and isopropanol (IPA) were employed as superoxide radical ( $\cdot\text{O}_2^-$ ) scavenger, hole ( $\text{h}^+$ ) scavenger, and hydroxyl radical ( $\cdot\text{OH}$ ) scavenger, respectively. Typically, 50 mg of PI-TD<sub>0.18</sub> sample and different scavenger were dispersed in 50 mL of RhB (10 ppm) aqueous solution and the following process was similar to the RhB degradation test.



**Figure S1.** FTIR of MA, TD, PD, PI<sub>MP</sub> and PI<sub>MT</sub>.

Sample	C/ wt%	N/ wt%	C/N
PI <sub>MP</sub>	51.299	21.285	2.81
PI-TD <sub>0.18</sub>	53.408	19.202	3.25

**Table S1.** Elemental analysis results and C/N atomic ratio of PI<sub>MP</sub> and PI-TD<sub>0.18</sub> samples.

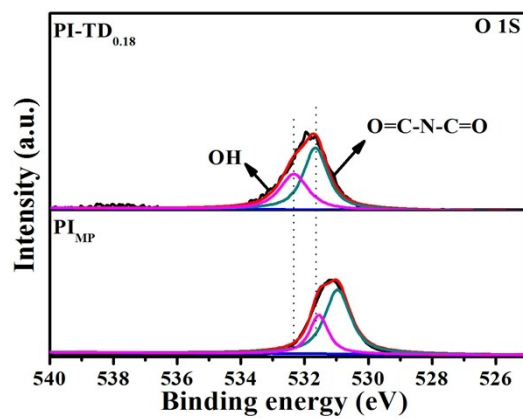


Figure S2. High resolution O1s spectra.

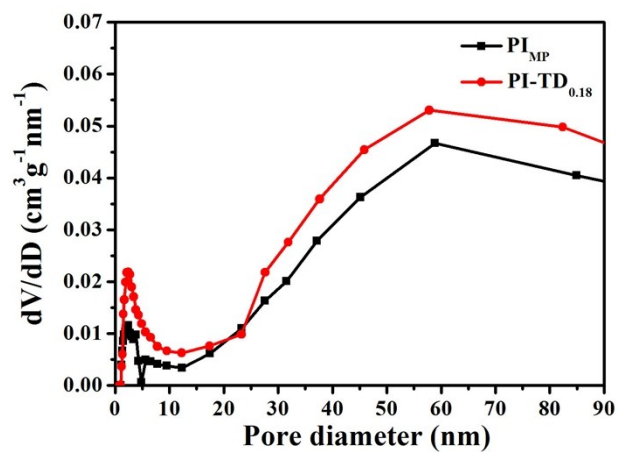
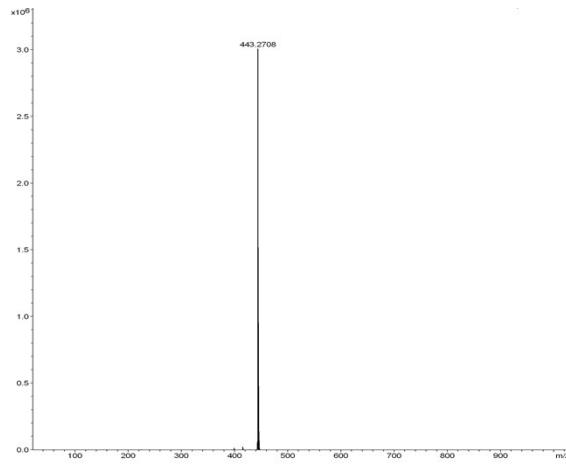
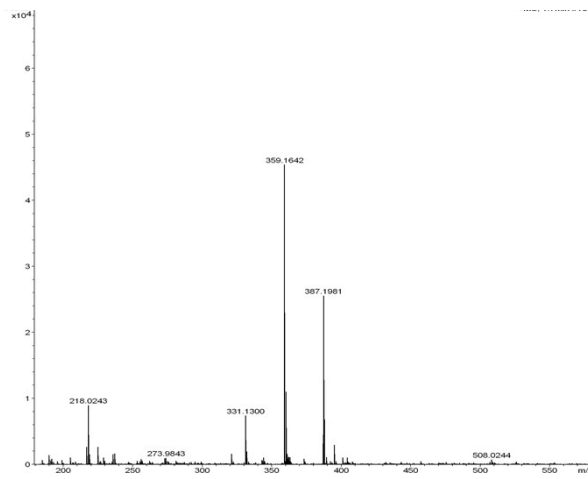


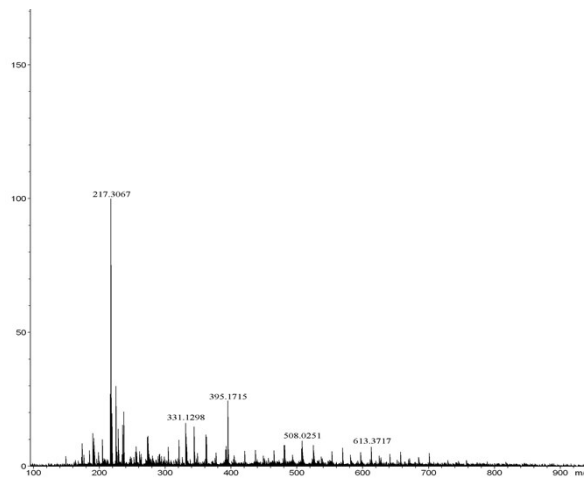
Figure S3. Pore size distribution of PI-MP and PI-TD<sub>0.18</sub> samples.



(a)

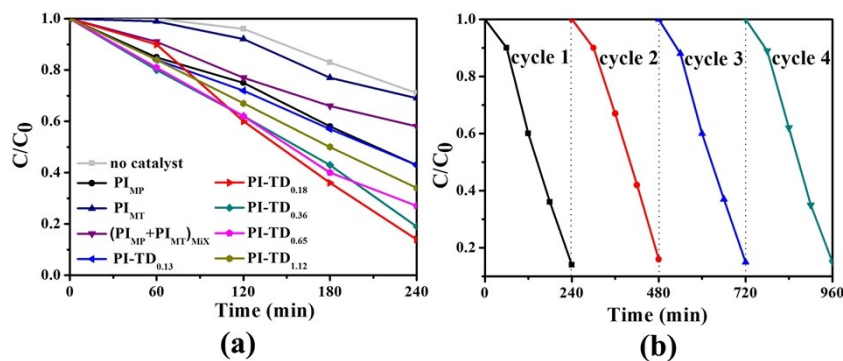


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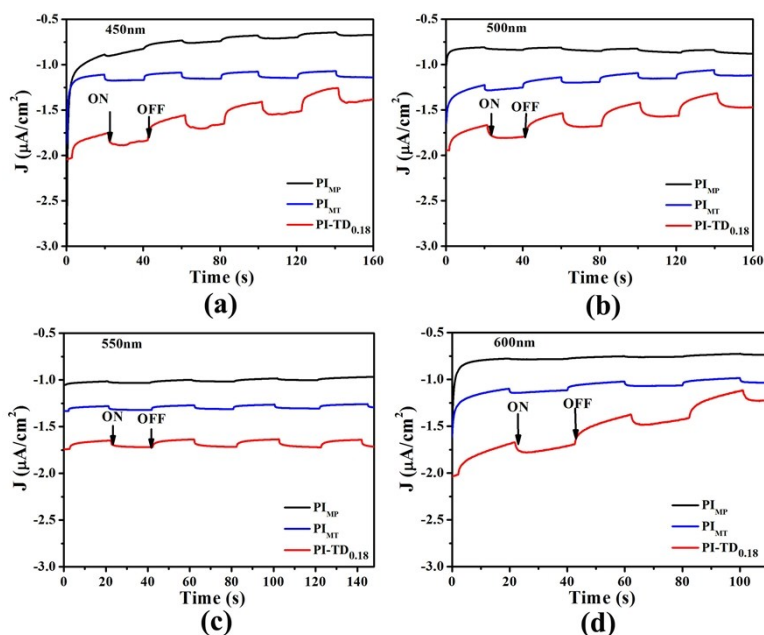


(c)

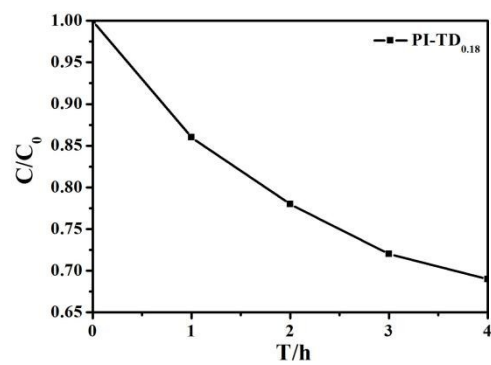
**Figure S4.** The main intermediate products generated during the process of photocatalytic degradation of RhB for (a) 0, (b) 120 and (c) 240 min by PI-TD<sub>0.18</sub>.



**Figure S5.** (a) Photodegradation of methyl violet by different samples under visible-light irradiation ( $\lambda \geq 420$  nm); Recycle experiments of (b) Photodegradation of methyl violet by PI-TD<sub>0.18</sub> ( $\lambda \geq 420$  nm).



**Figure S6.** Transient photocurrents of PI<sub>MP</sub>, PI<sub>MT</sub> and PI-TD<sub>0.18</sub> at different wavelength bands.



**Figure S7.** The profiles of photodegradation of RhB using PI-TD<sub>0.18</sub> as catalysts under 600 nm light irradiation.