

Electronic Supplementary Information

Triptycene-Based Two-dimensional Porous Organic Polymeric Nanosheet

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1. General Information

Materials obtained commercially were used without further purification.

2. Characterizations of 2D-PTNS

2.1 Electron Microscopy

Transmission electron microscopy (TEM) analysis was carried out by Tecani G220 at 200 kV. The samples were prepared by dispersing the product in Methanol with assistance of ultrasonication, followed by dropping the dispersion onto carbon coated copper grids and drying at ambient temperature. Atomic force microscopy (AFM) analysis was carried out by SPM 9700. The samples were prepared by the similar method with TEM, dropping the dispersion onto the mica and drying at ambient temperature.

2.2 IR and Raman Spectroscopy

FT-IR spectra were recorded by VERTEX 70 micro FT-IR spectroscopy. The Raman spectra were obtained by Raman Imaging Microscope System (LabRAM HR800), with excitation laser wavelength at 632.8 nm.

2.3 Thermal Properties Characterization

Thermogravimetric analysis (TGA) analysis was conducted by Pyris1 TGA with heatingrate of 10 °C min⁻¹ in nitrogen atmosphere.

2.4 Nitrogen Adsorption Measurement

Nitrogen adsorption experiments were performed with ASAP 2020 (Micromeritics, USA) at 77 K. The sample was outgassed at 110 °C for 15 hours under vacuum before measurement.

2.5 Gas Adsorption Measurement

Carbon dioxide, methane and ethane adsorption experiments were performed with ASAP 2020 (Micromeritics, USA) at 273 K and 298 K. The sample was outgassed at 110 °C for 15 hours under vacuum before measurement.

2.6 *In situ* Raman spectroscopy

For the *in situ* Raman kinetic experiments, the reaction spectra were recorded using an OPAL-3000 Raman spectrometer from Enwave Optronics, Inc., USA. Data collection was carried out using the Raman Application Center software (Ver. 8.1.3, Enwave Optronics, Inc., USA.), and the collected data was imported to and manipulated by OPUS software (Ver. 7.0.129, Bruker Optik GmbH, Germany).

3. TGA plots of of the 2D-PTNS.

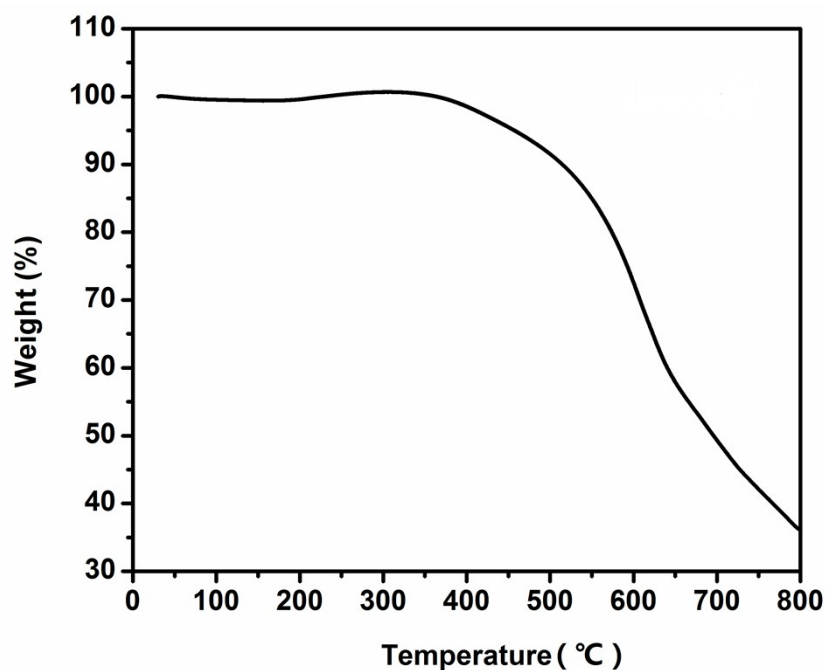


Fig. S1. The TGA plots of the 2D-PTNS.

4. PXRD spectroscopy of the 2D-PTNS.

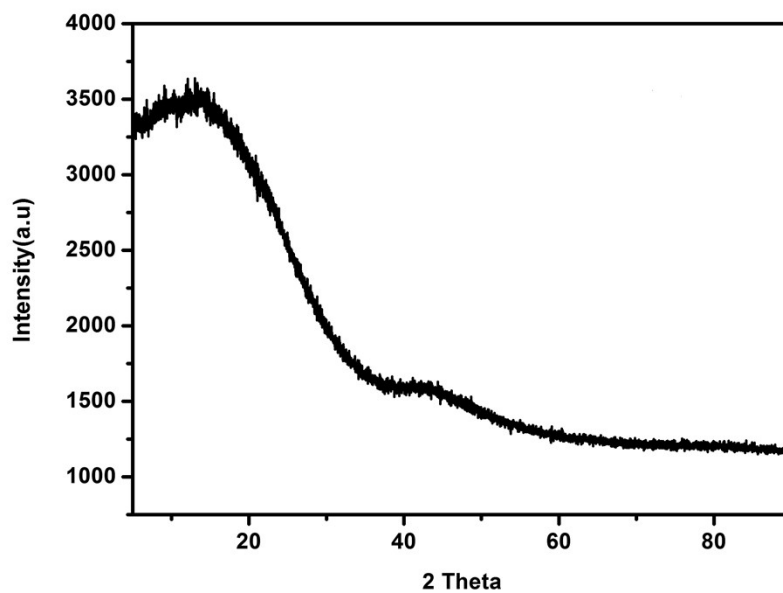


Fig. S2. The PXRD spectroscopy of the 2D-PTNS.

5. Photos of reaction processes of 2D-PTNS

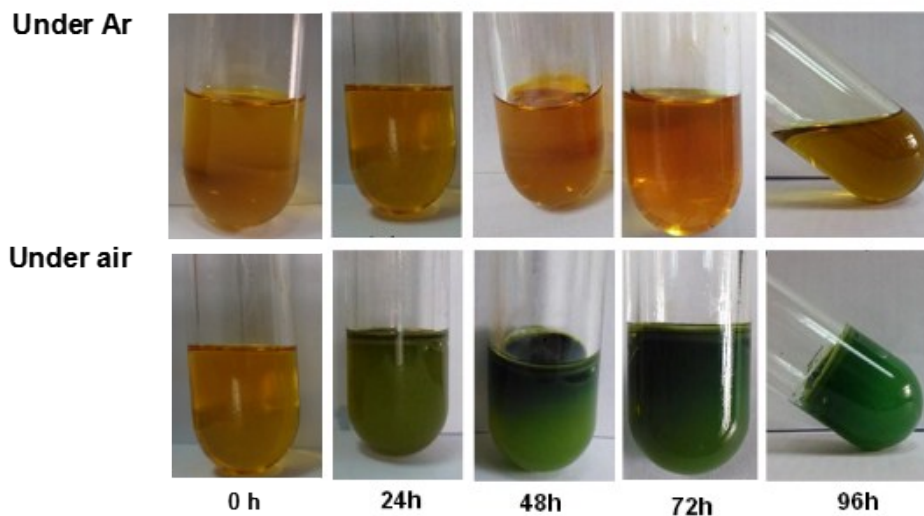


Fig. S3. The photos of reaction processes of 2D-PTNS under argon (top) or air (bottom) atmosphere.

6. *In situ* FT-Raman spectra of the 2D-PTNS

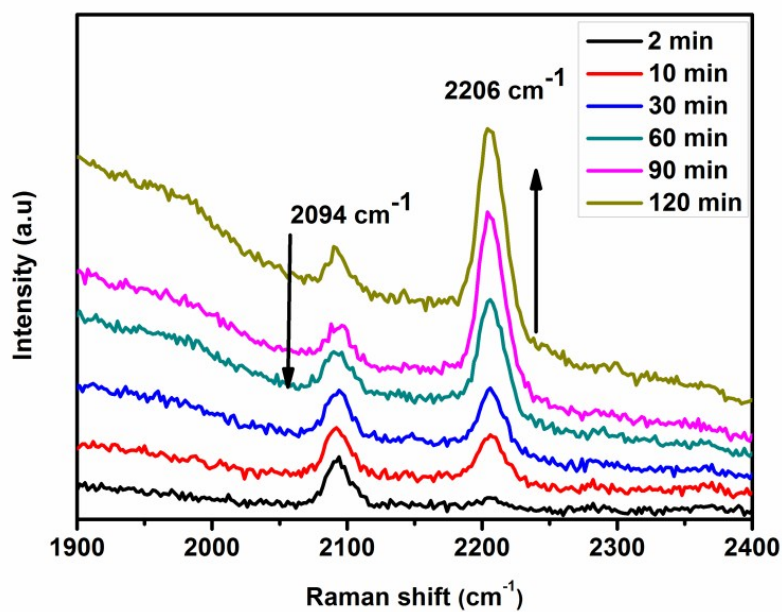


Fig. S4. The *in situ* FT-Raman spectra of the 2D-PTNS.

7. BET surface area plot for 2D-PTNS.

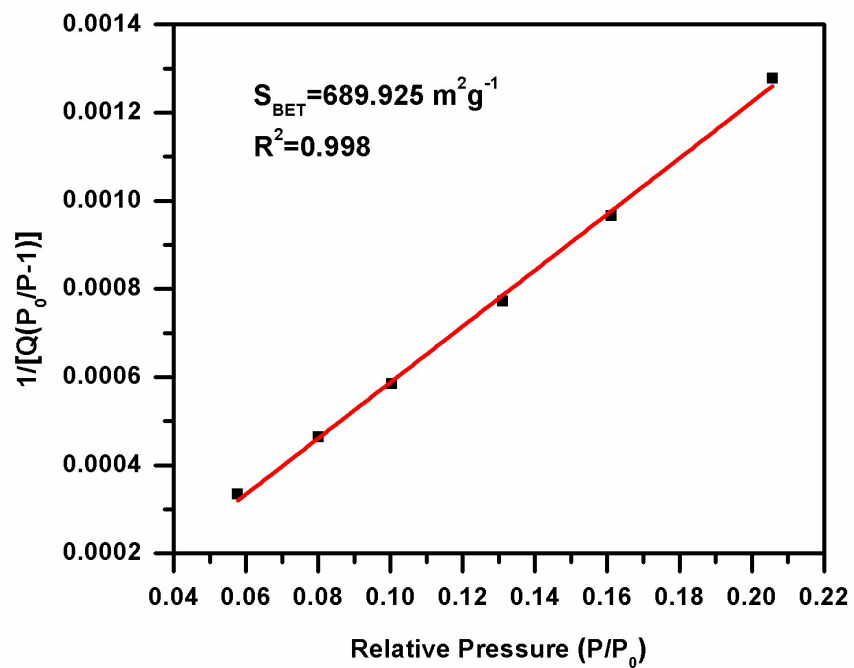


Fig. S5. The Brunauer-Emmett-Teller (BET) surface area of the **2D-PTNS**.

8. The H₂ isotherms of 2D-PTNS.

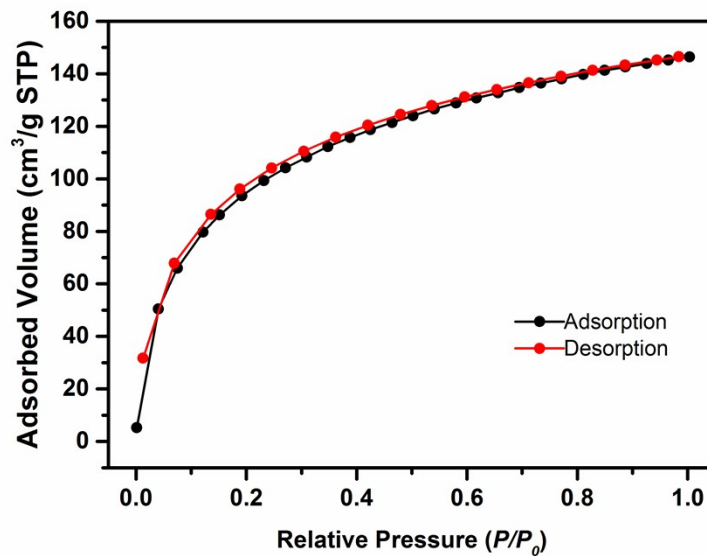


Fig. S6. The H₂ isotherm of **2D-PTNS** measured at 77 K.

9. The Q_{st} of the 2D-PTNS

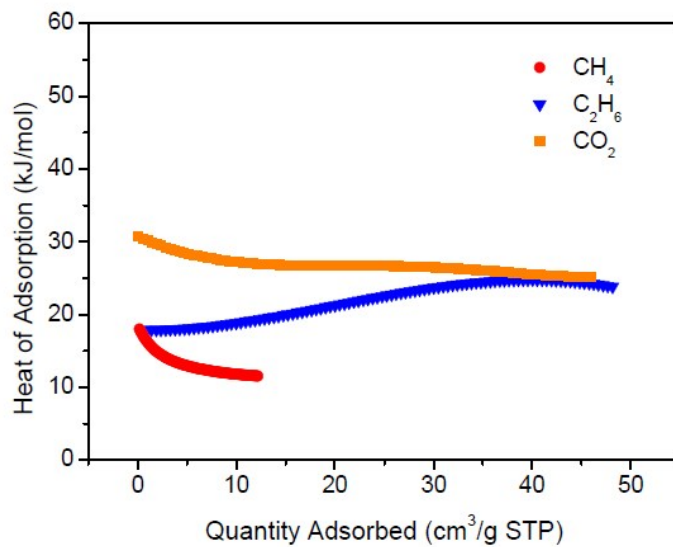


Fig. S7. The heat of adsorption (Q_{st}) of CO₂ (orange), C₂H₆ (blue) and CH₄ (red) of the **2D-PTNS**.

10. Initial gas uptake slopes for CO₂ and N₂ of the 2D-PTNS

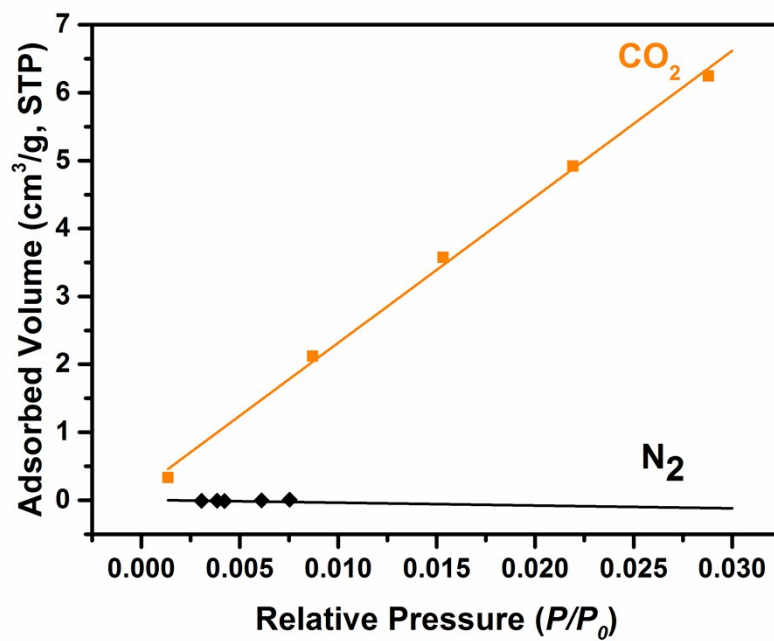


Fig. S8. The Initial gas uptake slopes for CO₂ (orange) and N₂ (black) of the 2D-PTNS at 273K.

11. Initial gas uptake slopes for CO₂ and CH₄ of the 2D-PTNS

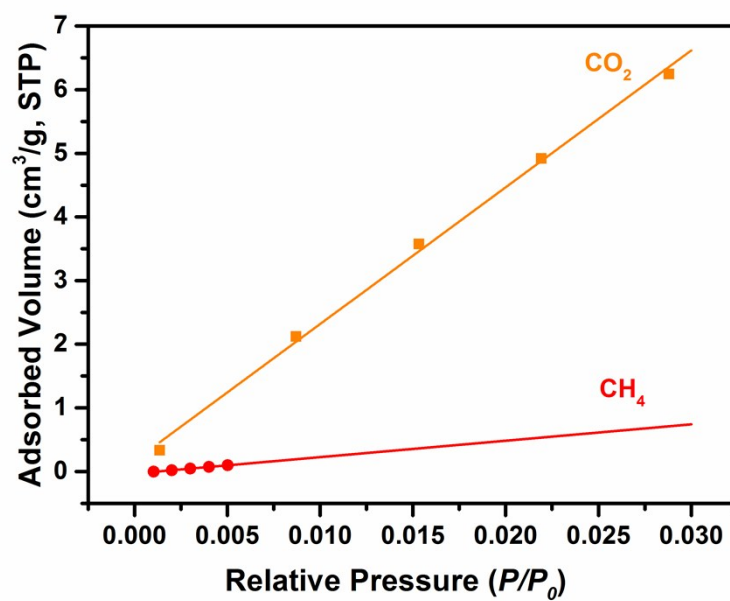


Fig. S9. The Initial gas uptake slopes for CO₂ (orange) and CH₄ (red) of the 2D-PTNS at 273K.

12. Initial gas uptake slopes for C₂H₆ and CH₄ of the 2D-PTNS (273K)

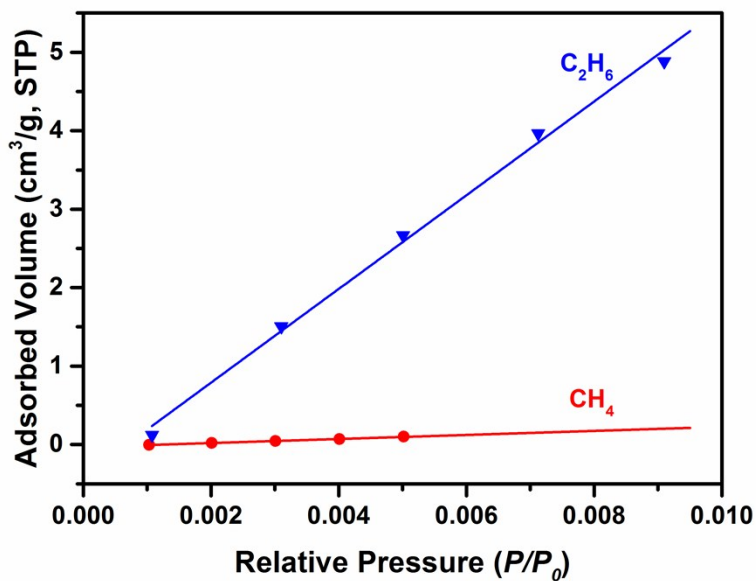


Fig. S10. The Initial gas uptake slopes for C₂H₆ (blue) and CH₄ (red) of the 2D-PTNS at 273K.

13. Initial gas uptake slopes for C₂H₆ and CH₄ of the 2D-PTNS (298K)

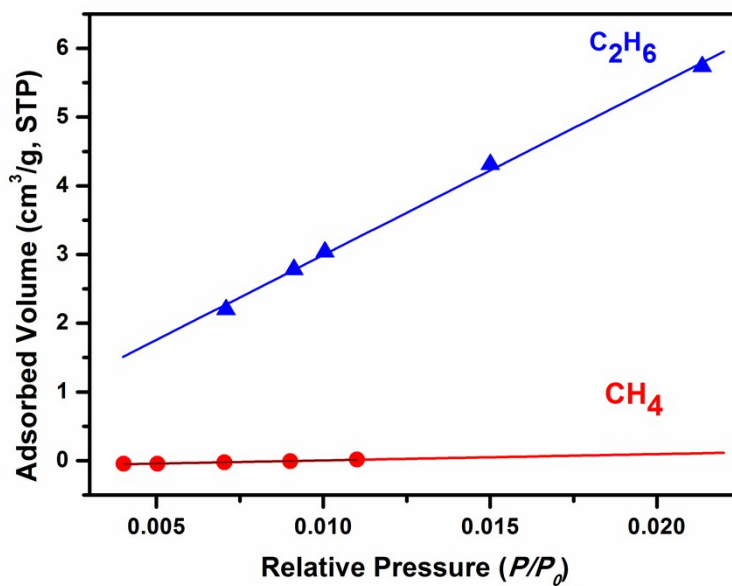


Fig. S11. The Initial gas uptake slopes for C₂H₆ (blue) and CH₄ (red) of the **2D-PTNS** at 298K.