

## Supporting Information

Structural Modification Of Graphene Sheets To Create A Dense Network Of Edge

Stites

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## **1. Preparation of Pt-graphene composites**

### **(1) Preparation of the graphene sheets**

The preparation of graphene involved two steps, (i) oxidation of the starting graphite to synthesize graphite oxide (GO), (ii) thermal expansion/exfoliation of the as-prepared GO to obtain thermally expanded GO (TEGO).

#### **(i) Synthesis of GO**

2 g graphite, 1 g sodium nitrate, and 46 mL of sulfuric acid were mixed and strongly stirred at 0 °C for 15 min in a 500 mL reaction flask immersed in an ice water bath. Then, 6 g potassium permanganate was added slowly to the above solution and cooled for 15 min. After this, the suspended solution was stirred continuously for 1 h, and then 92 mL of water was slowly added to the suspension within 10 min. Subsequently, the suspension was diluted by 280 mL of warm water and treated with 10 mL of H<sub>2</sub>O<sub>2</sub> (30%) to reduce the residual permanganate to soluble manganese ions. Finally, the resulting suspension was filtered, washed with DI water, and dried in a vacuum oven at 60 °C for 24 h to obtain GO.

#### **(ii) Thermal expansion/exfoliation of GO**

The as-prepared GO was thermally expanded to synthesize TEGO by rapidly heating it in a tube furnace. Generally, the as-prepared GO was first loaded in a boat, which was then inserted into a 1m-long quartz tube. After the tube furnace was heated to 1050 °C, argon was introduced and allowed to flow through the tube for 10 min.

The sample boat of GO in the quartz tube was then rapidly moved into the middle heating zone of the furnace and kept there for 30 s, before being quickly removed from the heating zone.

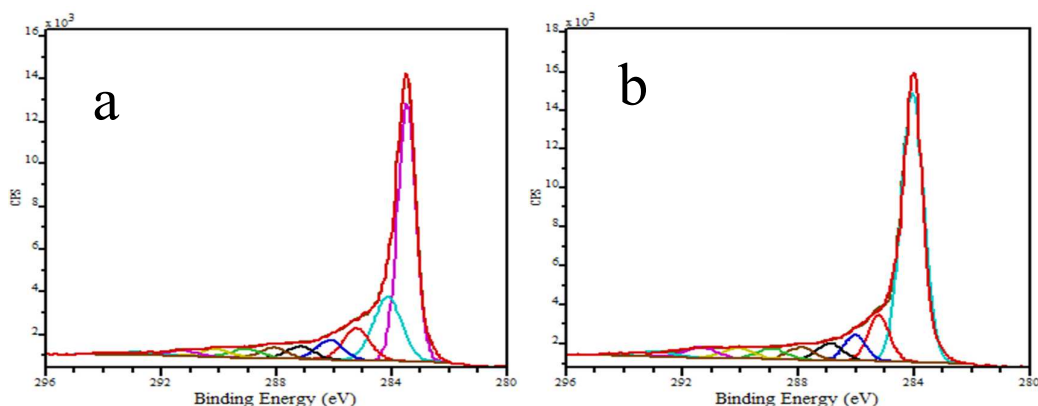
(2) Loading of the Pt nanoparticles onto the surface of the graphene sheets

The TEGO samples were immersed into  $10\text{ g L}^{-1}$  of the platinum precursor  $[\text{Pt}(\text{NH}_3)_4(\text{OH})_2]$  salt solution. The mixture of the TEGO and the platinum precursor was dried in the oven at  $110\text{ }^\circ\text{C}$ . Reduction of the adsorbed platinum precursor to its metallic form was carried out with a hydrogen gas at an elevated temperature. Generally, the preferable temperature of the reducing reaction is in the approximate range of  $200\text{ }^\circ\text{C}$ . The exact platinum loading in these samples was determined using TGA.

(3) Procedure for the measurement of  $\text{CO}_2$  uptake at low-pressure

Low-pressure  $\text{CO}_2$  adsorption measurements (up to 1 atm) were performed on an Autosorb-1 (Quantachrome) volumetric analyzer. Ultrahigh purity grade  $\text{CO}_2$  (99.999%) was used for the adsorption experiments. About 200 mg of samples were used for each measurement. The samples were outgassed to  $10^{-6}$  Torr at 573 K for 24 h prior to the experiment. Helium was used for the estimation of the dead volume, and it was assumed that it was not adsorbed at any studied temperatures.

## 2. XPS data for Pt-graphene sheets



**Figure S1.** XPS of C 1s spectra of a) Pt-graphene sheets and b) Pt-graphene sheets etched at 1000 °C.

**Table S1.** Pt-graphene sheets and b) Pt-graphene sheets etched at 1000 °C.

Sample	C=C	C=C	C-C	C-N	Shake up
Binding energy	283.5	284.1	285.2	287.2	291.3
Pt-graphene	50.0	19.4	9.4	3.8	5.8
Pt-graphene 1000 °C	0	62.9	10.6	4.8	8.8

The C=C  $sp^2$  peak of Pt-graphene sheets appears at about 283.5 eV, which is lower than 284.2-284.6 eV typically observed in graphene. The decrease in binding energy might be caused by an interaction between the graphene and the Pt nanoparticles. After H<sub>2</sub> etching of Pt-graphene sheets, C=C  $sp^2$  peak shifts to higher binding energy of about 284.1 eV. This shift is believed to be related to the formation of defective  $sp^2$  carbon atoms. Another possible cause of this shift is the loss of interaction between Pt and graphene sheets.

### 3. Summary of surface area, CO<sub>2</sub> uptake, and CO<sub>2</sub> adsorption enthalpy of graphene, Pt/Graphene sheets, and Pt/Graphene sheets etched at 1000 °C.

**Table S2** Sample surface area, CO<sub>2</sub> uptake, and CO<sub>2</sub> adsorption enthalpy

Sample	Graphene sheets	Pt/Graphene sheets	Pt/Graphene sheets 1000 °C	ZIF-69 <sup>3</sup>
BET(m <sup>2</sup> /g)	573.0	333.1	396.7	950
CO <sub>2</sub> (cm <sup>3</sup> /m <sup>2</sup> ) <sup>1</sup>	0.066	0.069	0.193	0.073
CO <sub>2</sub> (cm <sup>3</sup> /g) <sup>2</sup>	37.7	23.0	76.3	69
Adsorption enthalpy (kJ/mol)	17.2	20.1	26.3	

Note 1. Sorbent surface area-based; 2. Sorbent mass-based.

3: Banerjee, R.; Phan, A.; Wang, B.; Knobler, C.; Furukawa, H.; O’Keeffe, M.; Yaghi, O.M. Science, 2008, 319, 939.

As shown in Table S2, the BET surface area of the graphene, the as-deposited Pt/graphene, and the etched Pt/graphene composites were 573, 333.1, and 396.7 m<sup>2</sup>/g, respectively. The etched Pt/graphene composite was able to store up to 76.3 cm<sup>3</sup> CO<sub>2</sub> per gram, which is significantly higher than that of the pure graphene and as-deposited Pt/graphene sample. The uptake value of CO<sub>2</sub> at 273 K for the etched Pt/graphene composite increased by 103.4% and 233.4% in comparison with the graphene and Pt/graphene samples, respectively. Yaghi’s group reported a maximum uptake of CO<sub>2</sub> for ZIFs (ZIF-69) to be 69 cm<sup>3</sup>/g at 273 K at ambient pressure, which is

smaller than that of the etched Pt/graphene composite reported here under similar conditions. The CO<sub>2</sub> adsorption enthalpy increased from 17.2 for graphene to 20.1 for Pt-graphene and 26.3 kJ/mol for etched Pt/graphene after heat treated at 1000 °C.