

## Supporting information

### Experiment

#### 1. Material

3-Aminopropyltrimethoxysilane (APTMS), 1, 1, 4, 7, 7 Pentamethyldiethylene-triamine (PMEDTA) were purchased from Alfa Aesar. Phenyltriethoxysilane (PTES), 2-Bromo-2-methylpropionyl bromide were purchased from Acros. Tetraethyl orthosilicate (TEOS) and Methyl orange (MO) were purchased from Sinopharm Chemical Reagent Beijing. 1-Vinyl-3-ethylimidazolium bromide [(ViEtIm)<sup>+</sup>Br<sup>-</sup>] was synthesized<sup>S1</sup>.

#### 2. Characterization

Morphology of the samples was characterized via scanning electron microscopy HITACHI S-4800 operated at 15 kV. The samples for SEM observation were prepared by vacuum sputtering with Pt after being ambient dried. FT-IR spectroscopy was performed after scanning the samples for 32 times using a Bruker EQUINOX 55 spectrometer with the sample/KBr pressed pellets. XPS was performed on X-ray photoelectron spectroscopy ESCALab220i-XL (VG Scientific) using 300W AlK $\alpha$  radiation. Thickness of the samples was measured by AFM with Bruker Multimode 8. Thermogravimetric analysis (TGA) was performed using the PerkinElmer Pyris 1 TGA in air at a scanning rate of 10 °C/min. UV-vis spectroscopy was performed using UV spectrophotometer (TU1901).

#### 3. Experiment

##### 3.1 Synthesis of ATRP Agent Terminated Janus Silica Nanosheets

Amine/Phenyl Janus silica nanosheets were synthesized<sup>S2</sup>. 100 mg of the Janus silica nanosheets was dispersed in 40 mL of 2% (v/v) dry dichloromethane containing triethylamine. 1 mL of 2-bromo-2-methylpropionyl bromide was added slowly within 0.5 h at 0 °C. The reaction was carried out at room temperature for another 12 h. The ATRP agent terminated nanosheets was centrifugated and washed with dichloromethane. The ATRP agent terminated nanosheets were obtained as a light yellow product.

##### 3.2 Synthesis of Polymeric Ionic Liquid functionalized Janus Nanosheets

20 mg of ATRP agent terminated nanosheets, 12  $\mu$ L of PMEDTA, 1 g of [(ViEtIm)<sup>+</sup>Br<sup>-</sup>] were dispersed in 6 mL of methanol. The mixture was degassed by freeze thawing. After 6 mg of Copper bromide (CuBr) was added, the light green mixture was sealed under vacuum. The reaction was performed at 70 °C for 12 h. The product was washed with methanol and centrifugated for three cycles. The polymeric ionic liquids (PILs) functionalized Janus nanosheets were obtained.

##### 3.3 Synthesis of POMs functionalized PILs-Janus nanosheets

After 20 mg of PILs Janus nanosheets was dispersed in 20 mL of water, 60 mg of H<sub>3</sub>PW<sub>12</sub>O<sub>40</sub> was added. The system stood under stirring at room temperature for 1 h.

The  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets were obtained after centrifugation and wash with water for three times.

### 3.4 Catalytic Procedure of Methyl Orange

5 mg of  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets was dispersed in 2 ml of methyl orange aqueous solution ( $50 \text{ mg L}^{-1}$ ). 10  $\mu\text{L}$  of  $\text{H}_2\text{O}_2$  (30 wt.-%) was added under stirring at room temperature. The MO solution was centrifugated from the reaction system at a given interval. In the emulsion system, after 5 mg of  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets was dispersed in 2 ml of aqueous methyl orange solution, 1.4 mL of toluene was added to form a toluene-in-water emulsion at room temperature. The characteristic absorption band at 463 nm of MO is used to evaluate degradation efficiency:  $(C_0-C)/C_0$ .  $C_0$ : original MO concentration, C: MO concentration during the catalytic degradation. The samples were separated from the emulsion system by pipette at a given interval.

### 3.5 Regeneration of the Catalyst

$\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets were centrifugated from the emulsion. After the yellow solid was obtained and washed with DMF, a white solid was obtained.

S1 R. Marcilla, J. A. Blazquez, J. Rodriguez, J. A. Pomposo, D. Mecerreyes, *J. Polym. Sci. Pol. Chem.* **2004**, 42, 208.

S2 F. X. Liang, K. Shen, X. Z. Qu, C. L. Zhang, Q. Wang, J. L. Li, J. G. Liu, Z. Z. Yang, *Angew. Chem. Int. Ed.* **2011**, 50, 2379.

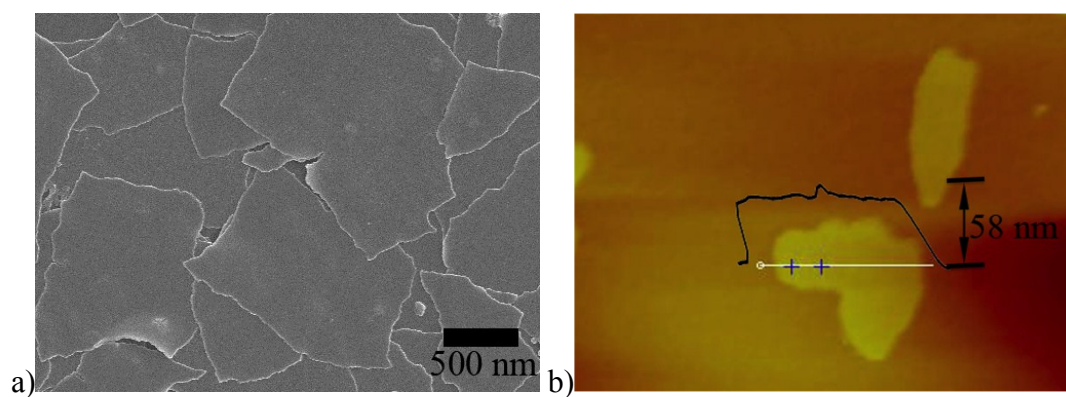


Figure S1. SEM (a) and AFM (a) images of the silica Janus nanosheets.

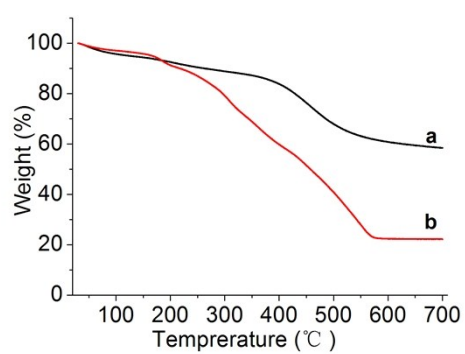


Figure S2. Thermogravimetric analysis (TGA) curves of the silica Janus nanosheets (a), and the Br<sup>-</sup> based PIL Janus nanosheets (b).



Figure S3. Dispersions of the Br<sup>-</sup> based PIL Janus nanosheets in water (left) and toluene (right).

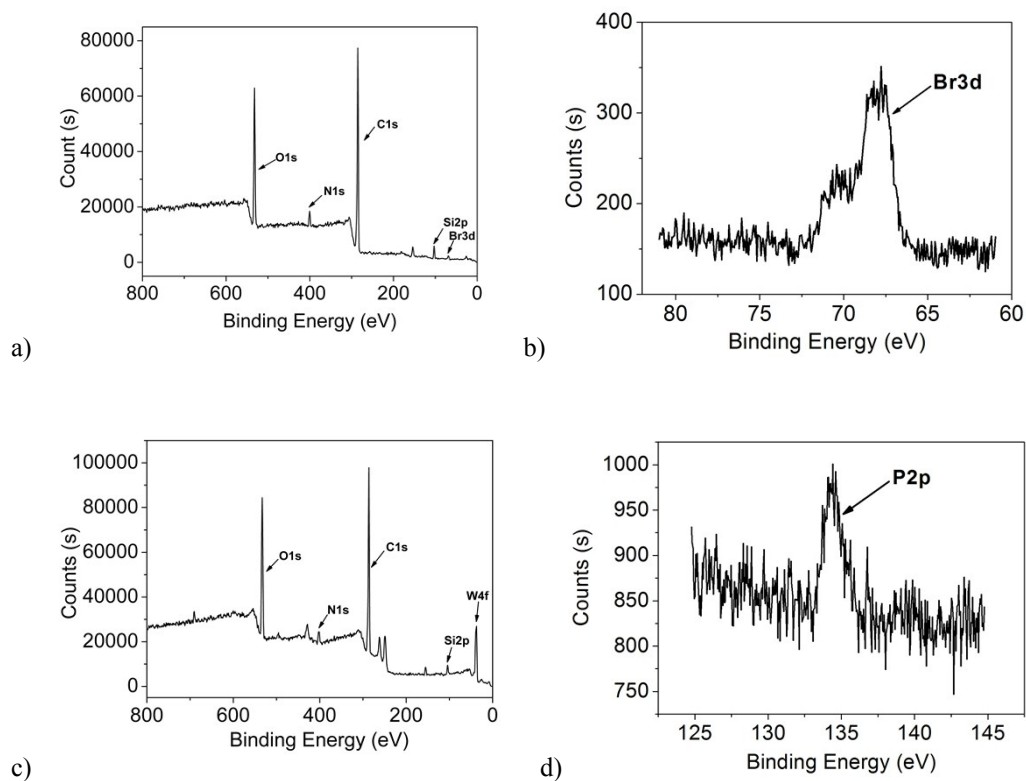


Figure S4. a) XPS and b) Br3d of Br<sup>-</sup> based PILs-Janus nanosheets; c) XPS and d) P2p of PW<sub>12</sub>O<sub>40</sub><sup>3-</sup> based PILs-Janus nanosheets.

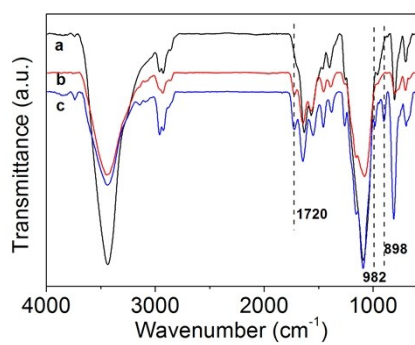


Figure S5. FT-IR spectra of the silica Janus nanosheets (a), the Br<sup>-</sup> based PIL Janus nanosheets (b), and the PW<sub>12</sub>O<sub>40</sub><sup>3-</sup> based PIL Janus nanosheets (c).



Figure S6. Dispersions of the  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets in water (left) and toluene (right).

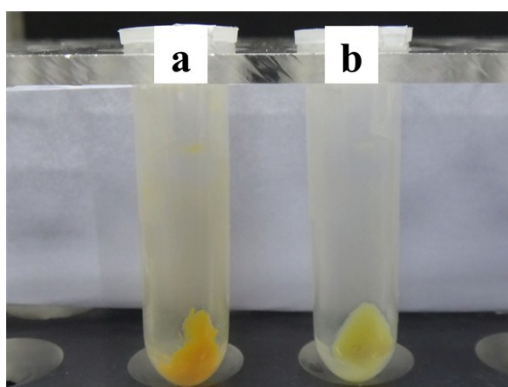


Figure S7. (a) The  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets after separation from the emulsion catalytic degradation system, and (b) from the water phase catalytic degradation system.

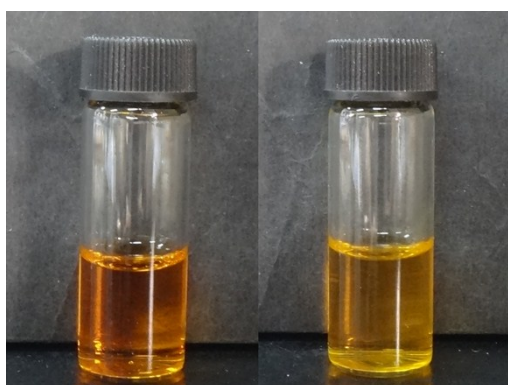


Figure S8 MO aqueous solution (left) and after degradation (right) by  $\text{PW}_{12}\text{O}_{40}^{3-}$  based PIL Janus nanosheets in water phase without the  $\text{H}_2\text{O}_2$ .



Figure S9. The emulsion catalytic system after degradation for one week.