Supplementary Information

Heterogenization of Homogenous Reaction System on Carbon Surface with Ionic liquid as Mediator

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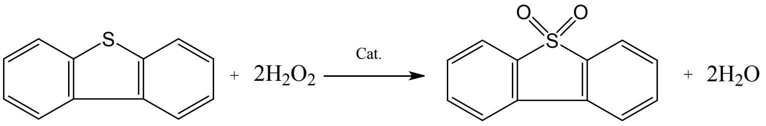
Preparation of OPW/HHT:

In order to obtain a highly dispersed OPW on HHT surface, only 5% mass ratio OPW was designed to give a comparison with OPW-OP/HHT. Incipient-wetness impregnation method was used here. In the prepare process, 50 mg OPW and 950 mg HHT were dispersed with ultrasonic assistant in 50 mL and 100 mL acetonitrile, respectively. Then the OPW solvent was added to HHT liquid phase, and stirred and dried under ultrasonic treatment at 60 °C to near dryness. The obtained "dryness" was dried at 120 °C overnight to get OPW/HHT.

The products of the DBT oxidation by H_2O_2 are the corresponding sulfone $DBTO_2$. The reaction equation is:



Details of calculation methods of DBT conversions, turn over frequency (TOF) and mass-specific activity (MSA) of different catalysts in the oxidizing process:

DBT residue was directly given by GC. The details of calculation method and equations were listed as follow:

$$\text{DBT conversion} = \frac{500\ \text{ppm} - \text{DBT residue}}{500\ \text{ppm}} \cdot 100\%$$

TOF value was based on the DBT conversion <30%, and given in mole of conversion of reactant per mole of OPW in the catalyst per minute. We consider all OPW in the catalyst OPW/HHT and OPW-OP/HHT as active centers due to the high dispersions of OPW molecules on HHT surface, so all OPW was used to calculate their relative TOF value. OPW contents in OPW-OP/HHT were determined by gravimetric determination (GD, a certain amount of OPW-OP/HHT was heated to 1050 ^{o}C maintaining 2 hours to make sure all the tungsten covert into WO_3, then the weight of WO_3 was used to calculate the weight of OPW). The results indicated that OPW, HHT and OP concentration in OPW-OP/HHT were 12.7%, 18.4% and 68.9%, respectively.

$$\mathrm{TOF} = \frac{n(\mathrm{DBT}) \cdot (\mathrm{DBT\ conversion})}{n(\mathrm{\ active\ OPW}) \cdot t}$$

For the dispersion of pure OPW in reaction system is not clear, it is to determine quantity of active centers (OPW which can encounter the oxidant and DBT in the reaction). Here, MSA was introduced to investigate the activity difference between the pristine OPW and supported OPW. Their molar quantities of OPW were used to represent their mass. Therefore, MSA has same unit with TOF.

$$\mathrm{MSA} = \frac{n(\mathrm{DBT}) \cdot (\mathrm{DBT\ conversion})}{n(\mathrm{All\ OPW}) \cdot t}$$

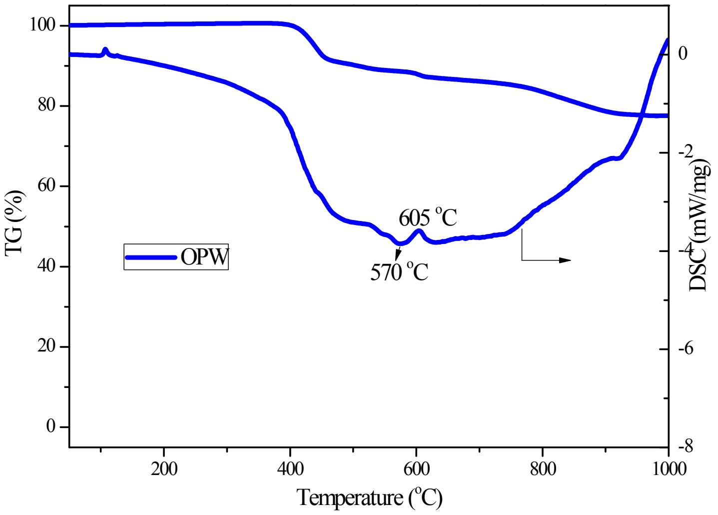


Figure. S1. TG-DSC curves of OPW.

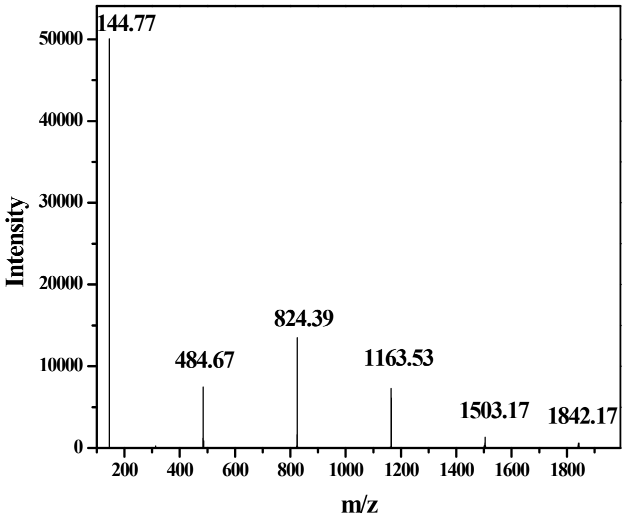


Figure S2. a ESI-MS spectra of the negative ion of OP. Solvent is CH_3CN.

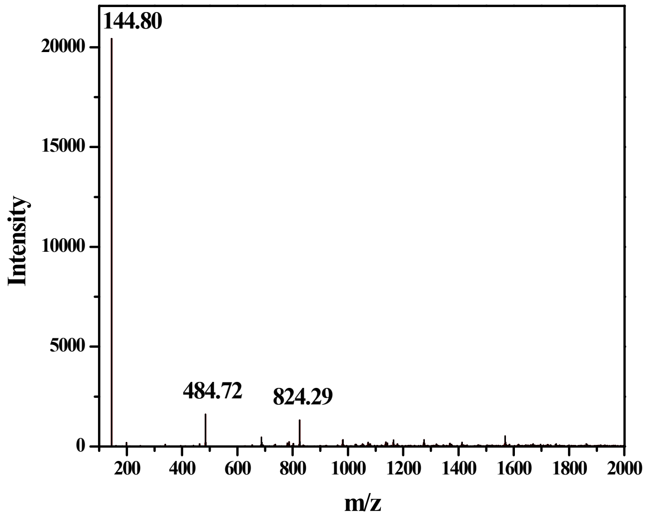


Figure S2. b ESI-MS spectra of the negative ion of the OPW-OP/HHT nanocomposites. Solvent is CH_3CN.

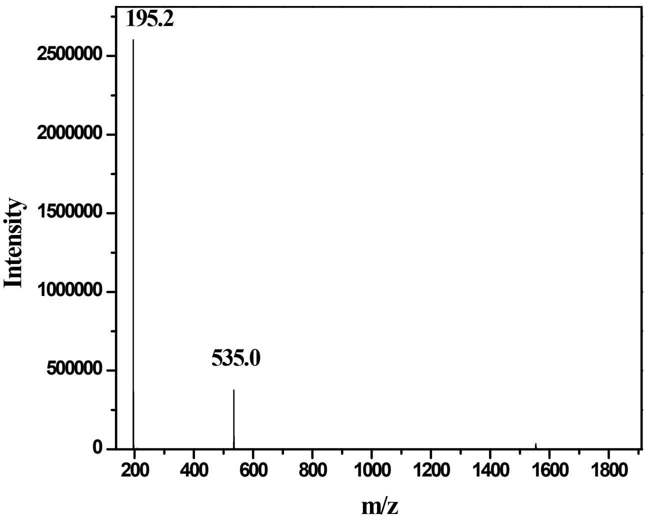


Figure S3. a. ESI-MS spectrum of the OP positive ion. Solvent is CH_3CN.

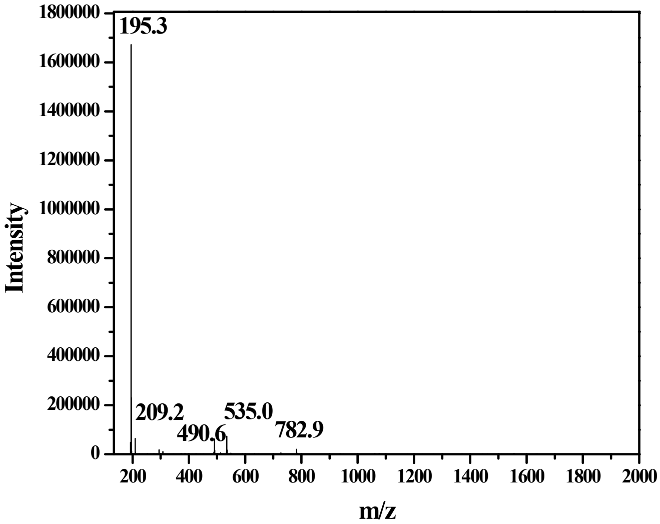


Figure S3. b. ESI-MS spectrum of the OPW-OP/HHT nanocomposites positive ion. Solvent is CH_3CN.

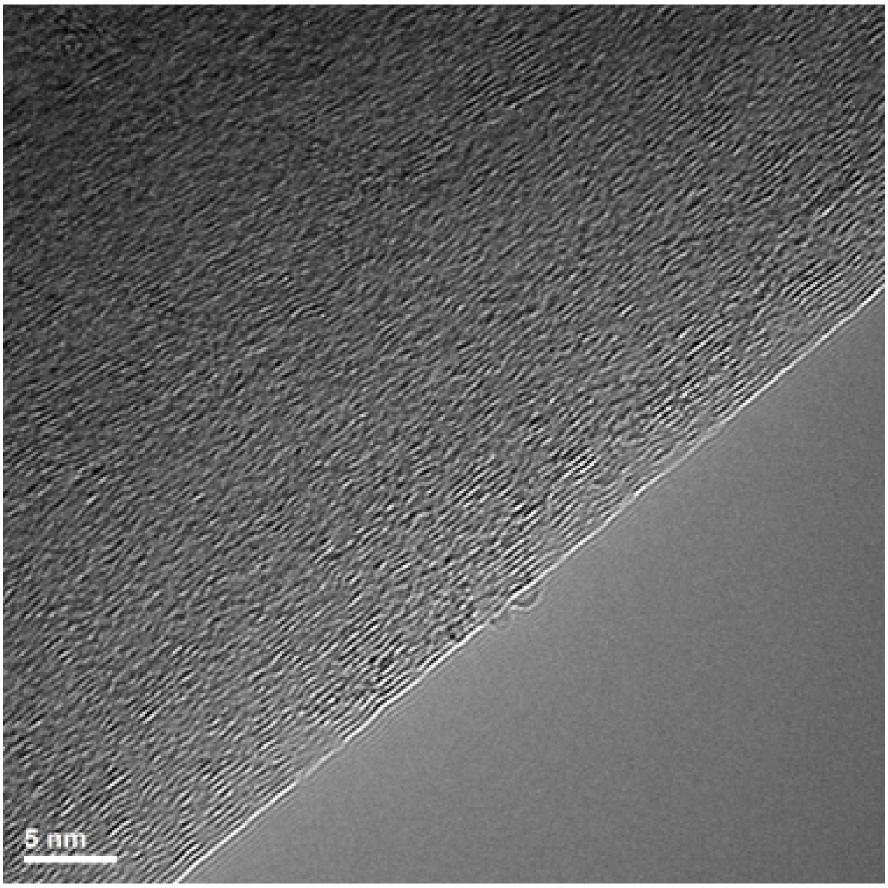


Figure S4. HRTEM image of pristine HHT.

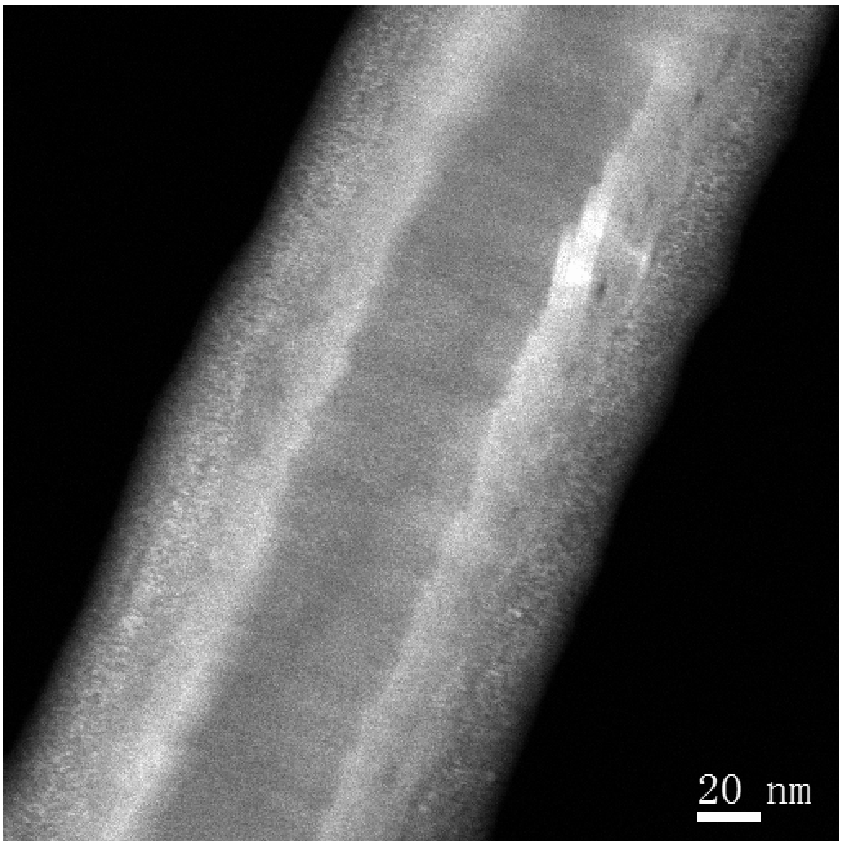


Figure S5. HAADF-STEM of OPW-OL/HHT.

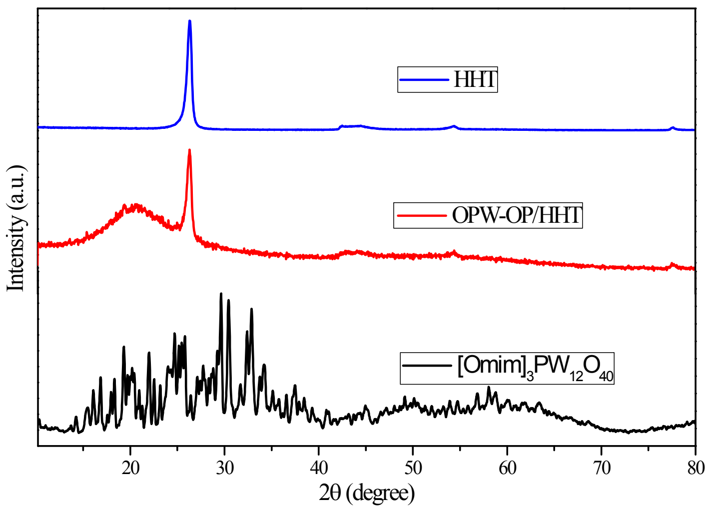


Figure S6. Wide angle XRD patterns of pristine HHT, $[Omim]_3PW_{12}O_{40}$ and OPW-OP/HHT nanocomposites.

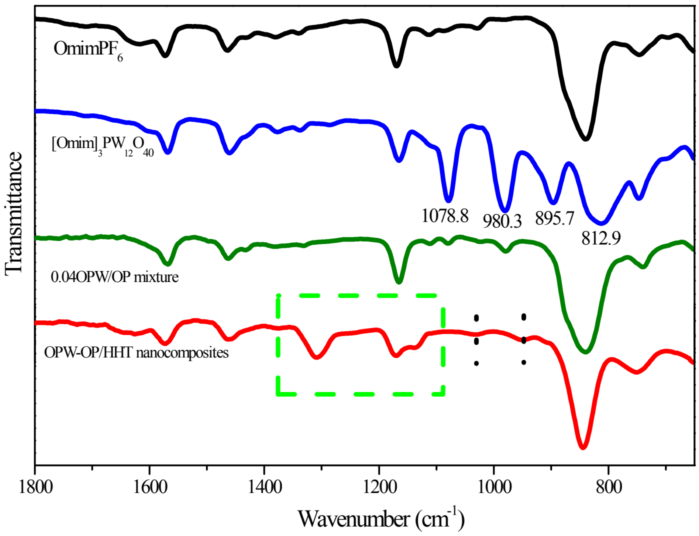


Figure S7. DRIFT spectra of $[Omim]_3[PW_{12}O_{40}]$, $[Omim]PF_6$, OPW/OP mixture and OPW-OP/HHT nanocomposites.

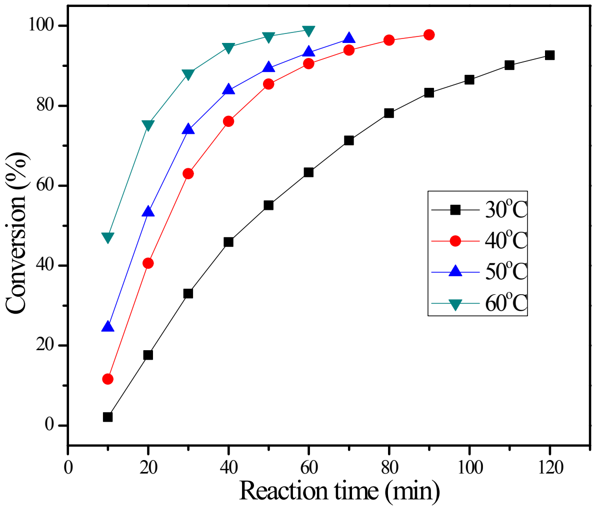


Figure S8. Influence of the temperature and time on the conversion of DBT. Experimental conditions: $V(H_2O_2)$ = 24 μL , 5 mL model oil, *m*(catalyst) = 20 mg.