

## Supplementary materials

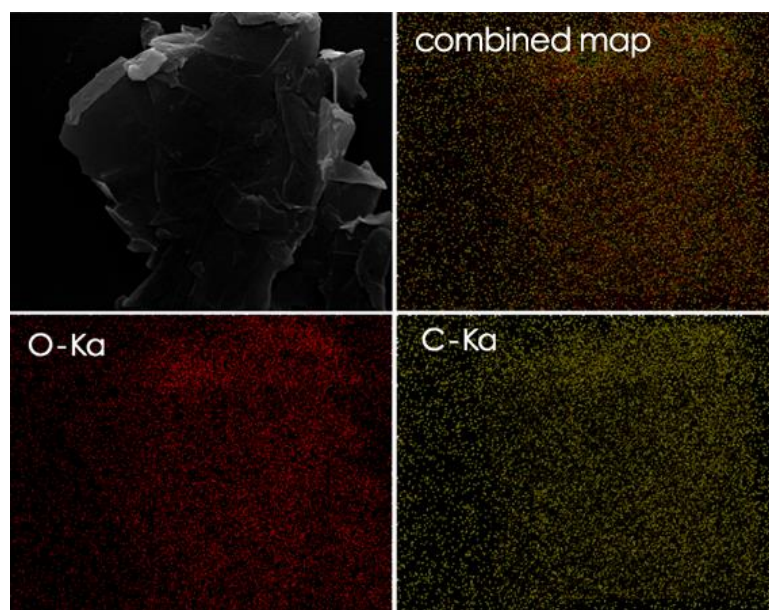
### A Nut-like MOF/Hydroxylated Graphene Hybrid Materials for Adsorptive

#### Desulfurization of Thiophene

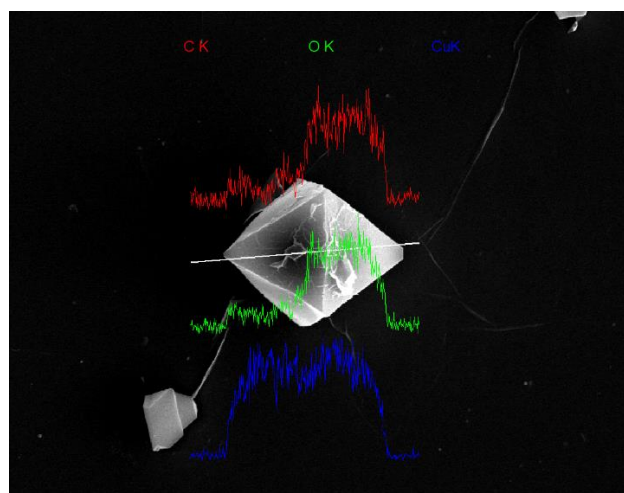
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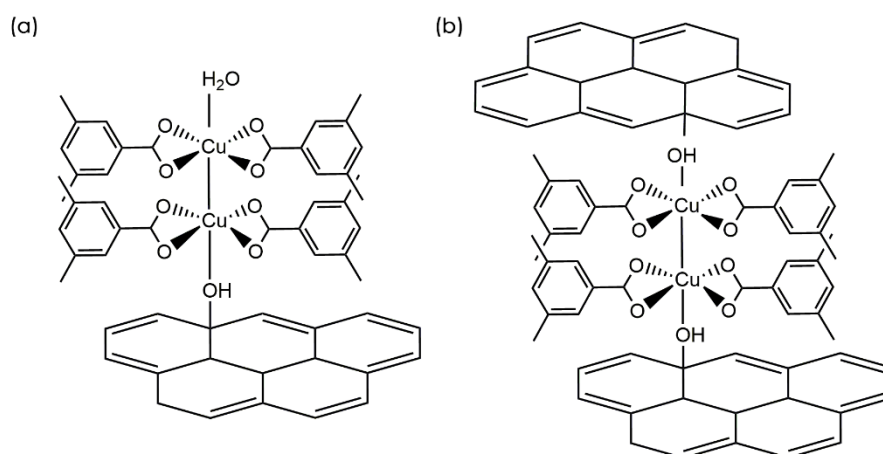
*† These authors contributed equally.*



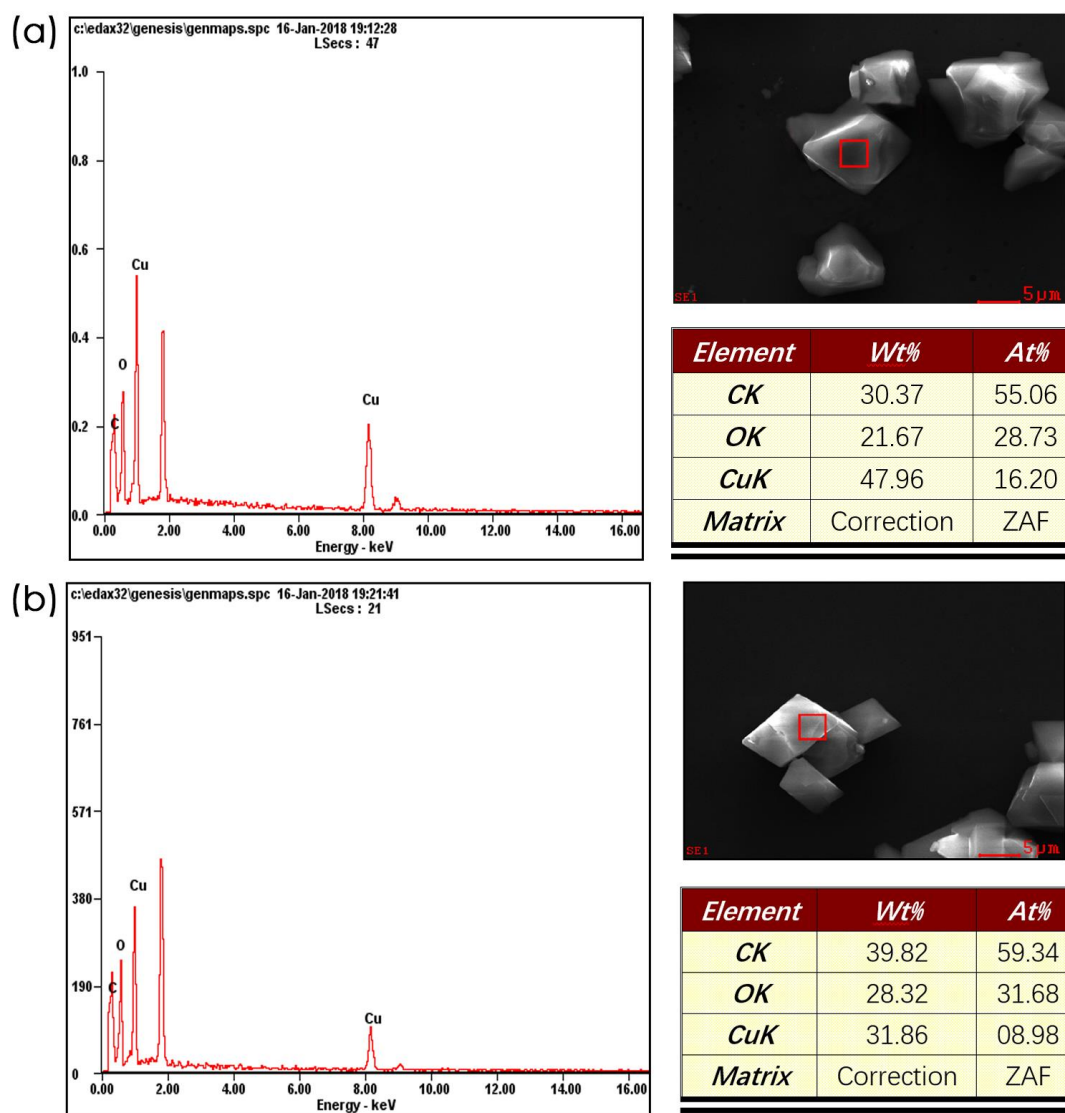
**Fig. S1** The SEM, EDS O-Kα (red) and C- Kα (yellow) images of HG.



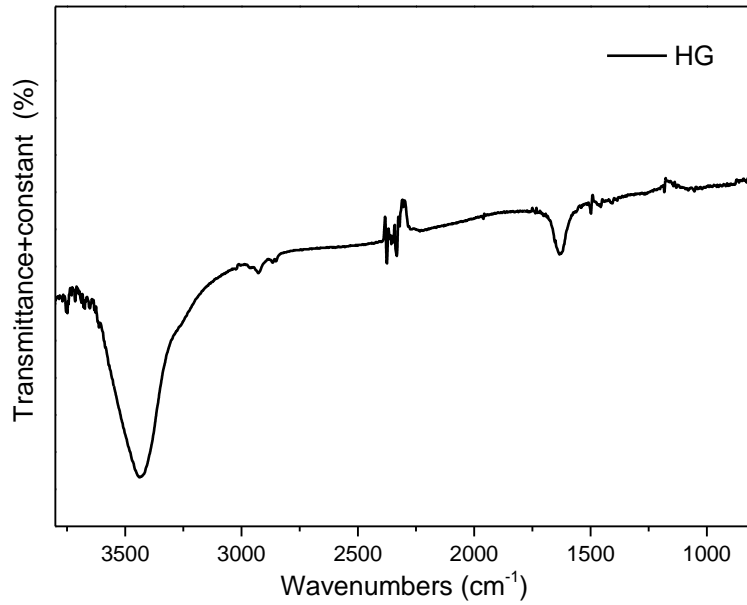
**Fig. S2** EDS line scanning and element content of Cu (blue), O (green) and C (red) for MGr-5 composite.



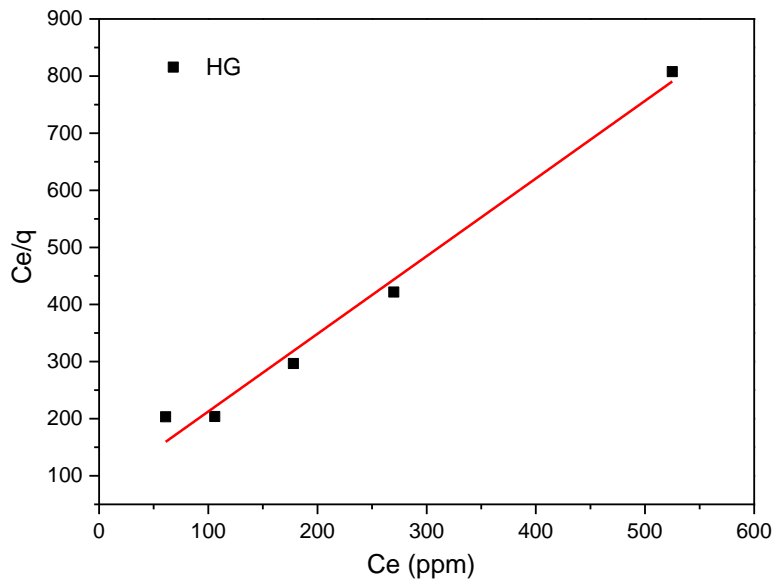
**Fig. S3** The possible interactions between Cu-BTC and hydroxyl groups of HG: (a) the HG dispersed on the surface of Cu-BTC crystal, and (b) the HG inserted inside Cu-BTC crystal to lead to the alternated growth of Cu-BTC and graphene layers.



**Fig. S4** The EDS analyses for Cu-BTC and MGr-5 composite with elemental content of C, O and Cu in surface of material, respectively.



**Fig. S5.** The FT-IR spectrum of HG.



**Fig. S6.** The Langmuir plots for T adsorption by HG

*Calculation of maximum sulfur adsorption capacity (Q<sub>0</sub>):*

The maximum sulfur adsorption capacity of all samples were calculated according to the Langmuir adsorption isotherm. The linear form of Langmuir's isotherm mode was plotted by the following equation[1]:

$$\frac{C_e}{q} = \frac{C_e}{Q_0} + \frac{1}{Q_0 b}$$

Where,

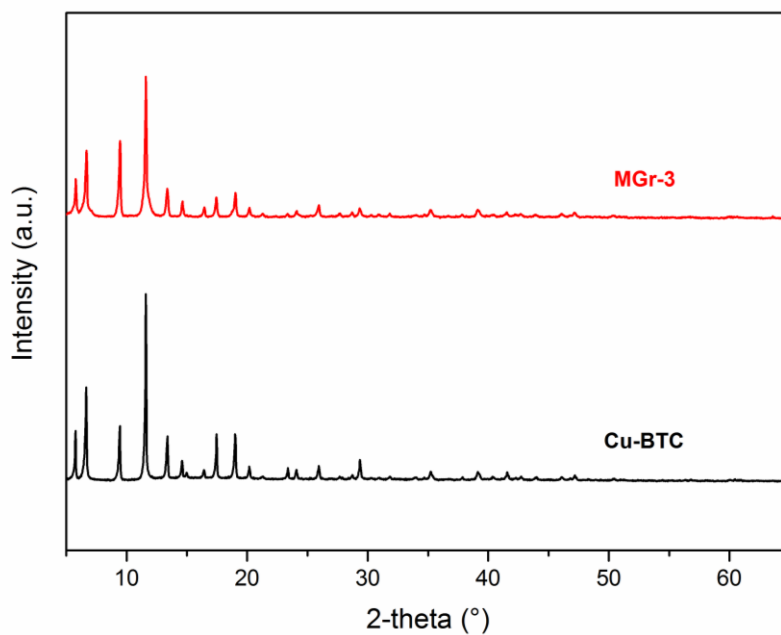
C<sub>e</sub>: equilibrium sulfur concentration in model oils (ppm)

q : adsorption capacity of adsorbent (mg-S/g)

$Q_0$ : Langmuir constant (maximum sulfur adsorption capacity, mg-S/g )

$b$  : Langmuir constant (L/mg)

Thus, the maximum sulfur adsorption capacity can be gained by the reciprocal of the slope of the plot of  $C_e/q$  against  $C_e$ .



**Fig. S7.** The X-ray diffraction patterns of Cu-BTC and MGr-3 composite after fifth regeneration.

1. B.H. Hameed, D.K. Mahmoud, A.L. Ahmad, *Journal of hazardous materials*, 2008, **158** 65-72.