## **Supporting Information**

## Three dimensional MOF-sponge for fast dynamic adsorption

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**Fig. S1** TG analysis (30-800°C) of the commercial sponge after treatment with DMSO. Due to the swell effect of DMSO, weight-loss starting and finishing temperature of the commercial sponge were lowered to 100 °C and 550 °C, and the final weight after TG process was 0.84%.

The DMSO treated commercial sponge was prepared by fully immersing the commercial sponge into DMSO, and absorbing the spare solution by non-woven fabrics. Then the commercial sponge was dried in an air oven at 80 °C for 2 hours. TG process was conducted under an air atmosphere in 30-800°C.

**S2.** Chemical reactions of the decomposition of the polyurethane sponge and the  $Cu_3(BTC)_2$  crystals under air atmosphere during thermogravimetric analysis:

$$\begin{array}{c} O & H \\ \hline O & -C & -N \\ \hline & & & \\ \end{array} + O_2 \longrightarrow CO_2 \uparrow + NO_2 \uparrow + H_2 O \uparrow \\ Cu_3(BTC)_2 + O_2 \longrightarrow CuO + CO_2 \uparrow + H_2 O \uparrow \end{array}$$

**S3.** Calculation of content of the Cu<sub>3</sub>(BTC)<sub>2</sub> crystals in the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge:

According to the TG analysis of the commercial sponge, only 0.88% was left after the thermal decomposition. Such, we could consider that the sponge was completely decomposed. So the residual of the  $Cu_3(BTC)_2$ -sponge after TG process could be considered as the oxidation products of the  $Cu_3(BTC)_2$  crystals, namely, CuO.

$$m_{(\text{Cu}_3(\text{BTC})_2)} = \frac{1}{3} \times \frac{m_{(\text{CuO})}}{M_{(\text{CuO})} / M_{(\text{Cu}_3(\text{BTC})_2)}} = \frac{1}{3} \times \frac{14.84\%}{80 / 606} = 37.47\%$$

Where  $m_{(CuO)}$  was the residual weight of the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge,  $M_{(CuO)}$  and  $M_{(Cu3(BTC)2)}$ were the relative molar mass of CuO and Cu<sub>3</sub>(BTC)<sub>2</sub>. Content of the Cu<sub>3</sub>(BTC)<sub>2</sub> in the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge was 37.47%.



**Fig. S4** SEM image of the pure Cu<sub>3</sub>(BTC)<sub>2</sub> crystals prepared according to the method reported in literature.<sup>[1]</sup>

S5. Chemical reaction occured during the adsorption process of  $N_2$  by the  $Cu_3(BTC)_2$ -sponge.

$$NH_3 + Cu_3(BTC)_2 \rightarrow Cu_3(NH_3)_6(BTC)_2$$

**S6.** Calculation of the NH<sub>3</sub>-adsorption capacity of the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge:

The blue shift of the absorption spectrum showed the adsorption process. The mount of NH<sub>3</sub> needed to alter the pH value of the solution is small that could be neglected during the calculation of the NH<sub>3</sub>-adsorption capacity. The sponge reached adsorption saturated after 3 min, as shown in Figure 3b). Flow speed of the gas mixture was 10 mL/min (gas pressure was an atmosphere). Mass of the commercial sponge used for the adsorption was 140 mg. So the purification capacity of the commercial sponge could be calculated as follows:

$$Q_{(sponge)} = 10 \times 3 \div 0.14 = 214.3 \frac{mL}{g}$$
 (5% NH<sub>3</sub>/N<sub>2</sub> mixture)  
 $Q_{(sponge)} = 10 \times 1.25 \div 0.14 = 89 \frac{mL}{g}$  (10% NH<sub>3</sub>/N<sub>2</sub> mixture)

For the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge:

$$Q_{(Cu_3(BTC)_2 - sponge)} = 10 \times 37.5 \div 0.14 = 2678.6 \frac{mL}{g} \quad (5\% \text{ NH}_3/\text{N}_2 \text{ mixture})$$
$$Q_{(Cu_3(BTC)_2 - sponge)} = 10 \times 16 \div 0.14 = 1142.9 \frac{mL}{g} \quad (10\% \text{ NH}_3/\text{N}_2 \text{ mixture})$$

Note that the adsorption process was conducted under an air atmosphere, the weight of the NH<sub>3</sub> adsorbed by the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge could be calculated:

$$W_{(Cu_3(BTC)_2-sponge)} = 2678.6 \times 10^{-3} L \times 5\% \div 22.4 L / mol \times 17g / mol = 101.6mg / g$$

 $(5\% \text{ NH}_3/\text{N}_2 \text{ mixture})$ 

$$W_{(Cu_3(BTC)_2 - sponge)} = 1142.9 \times 10^{-3} L \times 10\% \div 22.4L / mol \times 17g / mol = 86.7mg / g$$
(10% NH<sub>3</sub>/N<sub>2</sub> mixture)

Adsorbents	Adsorption mode	Adsorption capacity	Ref.
		(mg/g)	
Activated carbon	static	3.74	Ref. <sup>[2]</sup>
Activated alumina	static	52.7	Ref. <sup>[3]</sup>
Charcoal	static	89.6	Ref. <sup>[4]</sup>
Silica Gel	static	106.25	Ref. <sup>[4]</sup>
Natural Zeolites	static	100.3	Ref. <sup>[4]</sup>
MOF-199 film	static	290	Ref. <sup>[5]</sup>
MOF-199	dynamic	87	Ref. <sup>[6]</sup>
Cu <sub>3</sub> (BTC) <sub>2</sub> -sponge	dynamic	101.6	This work

Table S7. Comparison of NH<sub>3</sub> adsorption capacity of different adsorbents.

**S8.** Calculation of the Rhodamine B-adsorption capacity of the commercial sponge and the Cu<sub>3</sub>(BTC)<sub>2</sub>-sponge:

$$M_{(\text{sponge})} = \int R_e \cdot c \cdot d_v = 0.083mg$$
$$Q_{(sponge)} = \frac{M_{(sponge)}}{m_{(sponge)}} = \frac{0.083}{140} = 0.59 \frac{mg}{g}$$
$$M_{(\text{Cu}_3(\text{BTC})_2\text{-sponge})} = \int R_e \cdot c \cdot d_v = 1.231mg$$
$$Q_{(Cu_3(BTC)_2\text{-sponge})} = \frac{M_{(Cu_3(BTC)_2\text{-sponge})}}{m_{(Cu_3(BTC)_2\text{-sponge})}} = \frac{1.231}{140} = 8.8 \frac{mg}{g}$$

Where  $M_{(\text{sponge})}$  and  $M_{(\text{Cu3(BTC})2)}$  were the total RB-adsorption amount of the commercial sponge and the Cu<sub>3</sub>(BTC)<sub>2</sub>,  $Q_{(\text{sponge})}$  and  $Q_{(\text{Cu3(BTC})2)}$  were the adsorption capacity of the commercial sponge and the Cu<sub>3</sub>(BTC)<sub>2</sub>, respectively; *R*e was the real-time removal efficiency during the adsorption process, *c* was the initial concentration of the Rhodamine B solution,  $m_{(\text{Cu3(BTC)}2)}$  and  $m_{(\text{sponge})}$  were the weight of the Cu<sub>3</sub>(BTC)<sub>2</sub> and the commercial sponge used for Rhodamine B adsorption, respectively, which was 140 mg in the experiments.

**Table S9.** Comparison of Rhodamine B adsorption capacity of different adsorbents in

 relatively low concentration.

Adsorbents	Adsorption mode	Initial	Adsorption	Reference
		concentration	capacity	
		(mg/L)	(mg/g)	
Activated	Static adsorption with	10	5	Ref. <sup>[7]</sup>
carbon	long-time-shaking			
ALMCM-	Static adsorption with	3.85	41	Ref. <sup>[8]</sup>
41	long-time-shaking			
Natural	Static adsorption with	4.8	9.34	Ref. <sup>[9]</sup>
Zeolites	long-time-shaking			
Neem	Static adsorption with	12	2.35	Ref. <sup>[10]</sup>
sawdust	long-time-shaking			
Red mud	Static adsorption with	2	2.5	Ref. <sup>[11]</sup>
	long-time-shaking			
Cu <sub>3</sub> (BTC) <sub>2</sub> -	Dynamically flow	2	8.8	In this
sponge	through adsorbents			work

**S10.** Preparation process of the ZIF-8-sponge.

ZIF-8-sponge was prepared by simple method according to literature.<sup>[12]</sup> Briefly, commercial sponge was cut into small pieces and rinsed with ethanol for three times. Then the sponge pieces was dried under nitrogen flow. ZIF-8 precursor was prepared by mixing 5 mL Zn(NO<sub>3</sub>)<sub>2</sub> methanol solution (25mM) and 5 mL 2-methylimidazole methanol solution (50 mM). The sponge was immersed into the mixture solution for 30 minutes at room temperature. Then the sponge was washed with methanol and dried under nitrogen flow. To deposit enough ZIF-8 crystals onto the sponge skeleton, the whole process was repeated with fresh mixture solution for 10 times. Macroscopic morphology, crystallinity and microscopic structure of the as-prepared ZIF-8-sponge was shown in Figure S12. Comparison with the PXRD of ZIF-8, see Fig. S15, ESI<sup>†</sup>.



**Fig. S11** Macroscopic morphology, crystallinity and microstructure characterization of the prepared ZIF-8-sponge. a) Optical images of the commercial sponge (left) and the ZIF-8-sponge (right). b) Powder X-ray diffraction analysis of the ZIF-8-sponge. c,d) SEM and magnified SEM images of the ZIF-8-sponge.

**S12.** Pore distribution of the MOF-sponge.

There are two kinds of pores in as-prepared sample. The large pores in the sponge structure contribute the high permeability of the MOF-sponge, and the micropores in MOF crystals have great meaning for the adsorption capacity of the MOF-sponge. Characteristic pore size distributions of the MOFs and the sponge were shown in Fig. S13.



Fig. S13 Size distribution of the micropores in MOFs (up) and the sponge (down).



**Fig. S14** Comparison of the XRD pattern between the as-prepared  $Cu_3(BTC)_2$ -sponge and the  $Cu_3(BTC)_2$  powder stimulated from the single crystal data of the  $Cu_3(BTC)_2$  from CCDC.



**Fig. S15** Comparison of the XRD pattern between the as-prepared ZIF-8-sponge and the ZIF-8 powder stimulated from the single crystal data of the ZIF-8 from CCDC.

**S16.** Effect of the EtOH in the precursor solution.

There are two aspects of roles of the EtOH in precursor. Firstly, DMSO was use as the solvent to dissolve H<sub>3</sub>BTC and copper nitrate. It could prevent the nucleation of HKUST-1 crystals in solution, so that the precursor could keep stable at room temperature. (*Adv. Mater., 2010, 22, 2685*) To accelerate the HKUST-1 crystallization, ethanol was added into the precursor as a cosolvent, for that the ethanol molecules could partially or completely replace the DMSO ones around the building blocks and thus promote the nucleation reaction. (*Adv. Funct. Mater., 2011, 21, 1442*) Secondly, as the surface tension of the polyurethane (about 30 mN/m, Table S18) was lower than DMSO (43.6 mN/m, Table S18), it was hard for the solution to penetrate and achieve the well contact with the PU-sponge. (Fig. S17) Adding EtOH (22.5 mN/m, Table S18) into the precursor would lower the surface tension of the solution, which contributed to the spreading and well contact of the solution with the sponge surface, as shown in Fig. S17.



**Fig. S17** Contact state of the precursor solution without (a and b) and with (c and d) EtOH. Adding EtOH will lower the surface tension of the precursor, and the solution is much easier to penetrate and contact with the inner of the sponge.

Table S18 Surface tension of DMSO, EtOH and polyurethane.				
	DMCO	E4OU		

material	DMSO	EtOH	polyurethane
Surface tension (mN/m)	43.6	22.4	about 30



**Fig. 19** Comparison of the structure of the MOF-sponge prepared with (a and b) and without (c and d) EtOH in the precursor solution. As shown in Fig. 19 a and b), surface of the MOF-sponge prepared with EtOH in precursor solution were covered with a compact layer of MOF-crystals. In contrast, parts of the sponge surface (in the blue dash boxes) were not coated with MOF-crystals without EtOH in precursor solution (Fig. 19 c and d).



**Fig. S20** Adsorption-regeneration cycles of the MOF-sponge. Regeneration efficiencies of five cycles were 86.5%, 80.3%, 77%, 74.6% and 60.4%, respectively.



Fig. S21  $N_2$  adsorption isotherm of the MOF crystals, with BET surface area of 518.8

m  $\frac{3}{2}$ g and t-plot micropore volume of 0.23 cm  $\frac{3}{2}$ g.

**Video S1.** Dynamical purification process of the Rhodamine B solution using commercial sponge. Rhodamine B solution (2 mg/L) was controlled to flowed through the commercial sponge (140 mg). A quartz colorimetric dish was placed at the terminal of the tube to collect the treated solution for adsorption efficiency characterization.

**Video S2.** Dynamical purification process of the Rhodamine B solution using MOFsponge. Rhodamine B solution (2 mg/L) was controlled to flowed through the MOFsponge (140 mg). A quartz colorimetric dish was placed at the terminal of the tube to collect the treated solution for adsorption efficiency characterization.

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