## **Electronic Supplementary Information**

## Construction of flexible metal-organic framework (MOF) papers through MOF growth on filter paper and their selective dye capture

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## General Methods

All chemicals and solvents were obtained from commercial sources and were used as received. Filter papers (Grade 2 90mm diameter) were purchased from Advantec and used as received. The scanning electron microscopy (SEM) images were acquired using a JOEL JSM-6701F fieldemission instrument. Energy dispersive X-ray (EDX) spectra were obtained using a Hitachi SU 1510 SEM equipped with a Horiba EMAX Energy E-250 EDS system. X-ray diffraction patterns (XRD) were measured using a Rigaku Ultima IV equipped with a graphite monochromated Cu K $\alpha$ radiation source (40 kV, 40 mA). The IR spectra of solid samples were obtained using a Jasco FT/IR 4200 spectrometer using an attenuated total reflection (ATR) module. Thermogravimetric analysis (TGA) measurements were conducted using a Shimadzu TGA-50 under a nitrogen atmosphere at a heating rate of 5 °C min<sup>-1</sup>. Zeta-potentials were measured in water with pH values ranging from 4 to 9 using a Malvern Nano-ZS Zetasizer. Flat zeta-potential was measured using an Otsuka Electronics ELS-2000ZS. UV-Vis absorption spectra were obtained by using a Shimadzu UV-1650PC spectrophotometer in quartz cells (10 × 4 mm light path).

*Preparation of the carboxymethylated filter paper (CMFP).* In order to acquire suitable anchoring sites for the growth of ZIFs on the filter paper, the filter paper was carboxymethylated by treatment with sodium chloroacetate in the presence of sodium hydroxide catalyst. A piece of filter paper was submerged in a 1.0 M solution of sodium chloroacetate prepared in 15% (w/v) sodium hydroxide, and stirred for 1 h. The resulting carboxymethylated filter paper (CMFP) was washed vigorously with a large amount of water to remove any unreacted residues. Subsequently, it was allowed to dry overnight in air at room temperature. ATR-FTIR for CMFP (cm<sup>-1</sup>): 3352 (br), 2893 (m), 1650 (w), 1597 (w), 1425 (w), 1368 (w), 1311 (w), 1260 (w), 1231 (w), 1198 (w), 1157 (s), 1097 (s), 1053 (s), 1017 (s), 991 (s), 896 (w), 808 (w), 708 (w).

*Preparation of ZIF-8.* A precursor solution for ZIF-8 was prepared by mixing 2-methylimidazole (2-MeIM, 414.6mg, 5.05mmol) and Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (371.9mg, 1.25mmol) in 50 mL of methanol and kept at room temperature for 12 h. The resulting rhombic dodecahedral particles were isolated and washed with methanol several times via centrifugation-redispersion cycles. Each successive supernatant was decanted and replaced with fresh solvent. ATR-FTIR for ZIF-8 (cm<sup>-1</sup>): 3135 (w), 2930 (w), 1584 (w), 1425 (s), 1383 (w), 1310 (s), 1179 (m), 1146 (s),1091 (w), 994 (s), 955 (w), 759 (s).

*Preparation of ZIF-67.* A precursor solution for ZIF-67 was prepared by mixing 2-MeIM (1034.5mg, 12.6mmol) and Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (363.8mg, 1.25mmol) in 50ml of methanol and kept at room temperature for 12 h. The resulting rhombic dodecahedral particles were isolated and washed with methanol several times via centrifugation-redispersion cycles. Each successive supernatant was decanted and replaced with fresh solvent. ATR-FTIR for ZIF-67 (cm<sup>-1</sup>): 2360 (w), 1578 (w), 1423 (s), 1382 (w), 1305 (s), 1174 (w), 1142 (s), 992 (s), 755 (s).

*Preparation of CMFP/ZIF-8.* A precursor solution for ZIF-8 was prepared by mixing 2methylimidazole (2-MeIM, 414.6 mg, 5.05 mmol) and Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (371.9 mg, 1.25 mmol) in 50 mL of methanol, and CMFP was immersed in the resulting solution. The final precursor solution containing CMFP was placed at room temperature for 12 h. After this time, the resulting CMFP/ZIF-8 was thoroughly rinsed with methanol to remove any unreacted precursors and ZIF-8 particles that were not chemically attached to the paper. CMFP/ZIF-8 was then dried overnight in air at room temperature. ATR-FTIR for CMFP/ZIF-8 (cm<sup>-1</sup>): 3339 (br), 2894 (m), 1643 (w), 1573 (w), 1456 (w), 1426 (w), 1335 (w), 1368 (w), 1311 (w), 1228 (w), 1198 (w), 1159 (w), 1147 (w), 1050 (s), 1020 (s), 993 (s), 892 (w), 823 (w), 759 (w). *Preparation of CMFP/ZIF-67.* A precursor solution for ZIF-67 was prepared by mixing 2-MeIM (1034.5mg, 12.6mmol) and Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (363.8mg, 1.25mmol) in 50 mL of methanol, and CMFP was immersed in the resulting solution. The final precursor solution containing CMFP was placed at room temperature for 12 h. After this time, the resulting CMFP/ZIF-67 was thoroughly rinsed with methanol to remove any unreacted precursors and ZIF-67 particles that were not chemically attached to the paper. CMFP/ZIF-67 was then dried overnight in air at room temperature. ATR-FTIR for CMFP/ZIF-67 (cm<sup>-1</sup>): 3339 (br), 2894 (m), 1646 (w), 1583 (w), 1450 (w), 1423 (w), 1365 (w), 1307 (w), 1228 (w), 1199 (w), 1159 (w), 1143 (w), 1051 (s), 1019 (s), 993 (s), 894 (w), 754 (w).

*Organic dye capture.* CMFP or CMFP/ZIFs was folded and inserted into a conical glass funnel supported with a ring stand, and a vial was placed beneath the funnel. An aqueous solution of the organic dye (MO<sup>-</sup>, MB<sup>+</sup>, IC<sup>-</sup>, R6G<sup>+</sup>, or mixture of them; 3 mL, 5 ppm) was slowly poured into the funnel and the filtrate collected in the vial was analyzed using a UV-Vis spectrophotometer.

*Recyclability test.* A recyclability test was carried out to investigate whether CMFP/ZIF-8 could be reused for dye capture. After the completion of the first dye capture experiment, the used CMFP/ZIF-8 was thoroughly washed with methanol to remove all of the captured organic dyes. It was then dried overnight in air at room temperature. Subsequently, it was subjected to the dye capture and regeneration process for three times.



**Fig. S1** SEM images of (a) unmodified filter paper and (b) CMFP. (c) IR spectra and (d) PXRD patterns of unmodified filter paper (top) and CMFP (bottom).



**Fig. S2** TGA curves of (a) CMFP, (b) CMFP/ZIF-8, and (c) CMFP/ZIF-67. The relative amounts of ZIF nanoparticles present on the CMFP surface were calculated from the TGA curves of CMFP/ZIFs.



Fig. S3 Zeta-potentials of the (a) ZIF-8 and (b) ZIF-67 particles in water at various pH.



**Fig. S4** Zeta-potential measurement of CMFP in water. The negative flat zeta-potential value (-19.32 mV) was also measured using an Otsuka Electronics ELS-2000ZS.



**Fig. S5** Selective organic dye capture. (a) A photograph showing the filtration of an aqueous organic dye solution using CMFP/ZIF-67. (b) Methyl orange ( $MO^-$ ) capture via filtration of the  $MO^-$  solution on CMFP or CMFP/ZIF-67. (c) Methylene blue ( $MB^+$ ) capture via filtration of the  $MB^+$  solution on CMFP or CMFP/ZIF-67. UV-Vis spectra of the filtrates were acquired after the separation. Insets in (b) and (c) show the color change after filtration through CMFP or CMFP/ZIF-67.



**Fig. S6** (a) and (b) SEM images of the CMFP/ZIF-8 that was regenerated after a washing process. (c) PXRD pattern of the regenerated CMFP/ZIF-8.



**Fig. S7** Selective organic dye capture. (a) Indigo carmine (IC<sup>-</sup>) capture via filtration of the IC<sup>-</sup> solution using CMFP, CMFP/ZIF-8, or CMFP/ZIF-67. (b) Rhodamine 6G (R6G<sup>+</sup>) capture via filtration of the R6G<sup>+</sup> solution using CMFP, CMFP/ZIF-8, or CMFP/ZIF-67. (c) Effective capture of IC<sup>-</sup> via filtration of an aqueous mixture of IC<sup>-</sup> and R6G<sup>+</sup> using CMFP/ZIF-8. UV-Vis spectra of the filtrate were acquired after the separation. Insets in (a–c) show the change in color after filtration through CMFP, CMFP/ZIF-8, or CMFP/ZIF-67.