## **Supporting Information for**

## Binder-Free mechanochemical metal-organic framework nanocrystal coatings



Fig. S1 (a) SEM image and (b, c) the corresponding EDS mapping of the used ZrO<sub>2</sub> ball surface.



Fig. S2 XPS spectra (Co 2p) of Co-ZIF-62 (black) and used ZrO<sub>2</sub> ball after the Co-ZIF-62 synthesis (red).



Fig. S3 XRD patterns of the simulated Co-ZIF-62 and the  $ZrO_2$  balls after ball milling. These diffraction peaks are attributed to the  $ZrO_2$ .



Fig. S4 SEM image of the presynthesised ZIF-8 crystals.



Fig. S5 (a) Optical photos of the mechanochemical coating process, and (b) carbon cloth (CC) before and after coated with ZIF-8.



Fig. S6 SEM images of carbon cloth before (a, b) and after (c, d) coating with ZIF-8.



Fig. S7 (a)  $N_2$  sorption isotherm at 77K (adsorption: closed symbols; desorption: open symbols) and (b) pore size distribution of ZIF-8-CC



Fig. S8 Raman spectra of the carbon cloth after ball milling in different environments (bottom: before ball milling; middle: after ball milling in air; top: after ball milling in argon)



Fig. S9 SEM images of (a)ZIF-67 and (b)  $a_{\rm m}ZIF\text{-}67$ 



Fig. S10 TEM images of (a) ZIF-67, (b)  $a_m ZIF-67,$  (c) Co-ZIF-4, and (d)  $a_g Co-ZIF-4$ 



Fig. S11 SEM images (a) ZIF-67-CC, (b)  $a_m$ ZIF-67-CC, (c) Co-ZIF-4-CC, and (d)  $a_g$ Co-ZIF-4-CC



Fig. S12 SEM secondary electron images of substrates before coating procedure: (a, b) PVDF, (c, d) Ni foam, (e, f) Ti foil, (g, h) FTO.



Fig. S13 ZIF-8 coating on PVDF *via* the one-pot technique. SEM images of ZIF-8 coating on PVDF prepared with (a, b) precursor: 0.005g ZnO and 0.01g 2-methylimidazole and (c, d) precursor: 0.01g ZnO and 0.02g 2-methylimidazole.



Fig. S14 (a) Optical photo and (b) SEM image of the ZIF-8 coating on the nickel foam.



Fig. S15 (a) Optical photo and (b) SEM image of the ZIF-8 coating on a titanium foil. (c) CO<sub>2</sub> adsorption-desorption isotherms of ZIF-8-Ti at 273 K (adsorption: closed symbols; desorption: open symbols). (d) Pore size distribution of ZIF-8-Ti.



Fig. S16 Ultraviolet-visible (UV-Vis) absorption spectra for ZIF-8, ZnO, Ti foil and ZIF-8-Ti



Fig. S17 Optical photos of the ZIF-8 coating on carbon cloth prepared by a large ball milling jar.

![](_page_11_Figure_0.jpeg)

Fig. S18 (a-c) Optical photos of the dual side MOF coatings (ZIF-8 and Co-ZIF-62) on a porous Matrimid support. (d) SEM image and the cooresponding (e) EDS mapping of the cross-section of porous Matrimid with dual side MOF coating (Co in green and Zn in blue).

![](_page_12_Figure_0.jpeg)

Fig. S19 (a) Photograph of the FTO taped on the inner wall of the ball milling jar. (b) SEM image of ZIF-8-FTO. (c-d) XPS spectra of ZIF-8-FTO (c: Sn 3d; d: Zn 2p).

	Area (cm²)	Total amount of Zn (mg)	Density of ZIF-8 on CC (mg cm <sup>-2</sup> )
ZIF-8-CC	4.0	1.48	1.31
ZIF-8-CC after 100 cycles bending	4.0	1.40	1.24

Table S1 Loading of ZIF-8 on ZIF-8-CC samples before and after 100 cycles bending according to the ICP-OES results.

Table S2 Loading of MOFs on ZIF-67-CC, a<sub>m</sub>ZIF67-CC, Co-ZIF-4-CC and a<sub>g</sub>Co-ZIF-4-CC according to the ICP-OES results.

	Area (cm²)	Total amount of Co (mg)	Density of catalyst on CC (mg cm <sup>-2</sup> )
ZIF-67-CC as-synthesised	4.0	0.947	0.897
ZIF-67-CC after rinsing	4.0	0.035	0.032
a <sub>m</sub> ZIF67-CC as-synthesised	4.0	0.966	0.914
a <sub>m</sub> ZIF67-CC after rinsing	4.0	0.033	0.031
Co-ZIF-4-CC	4.0	1.472	1.218
a <sub>g</sub> Co-ZIF-4-CC	4.0	1.476	1.222