

## Supporting Information

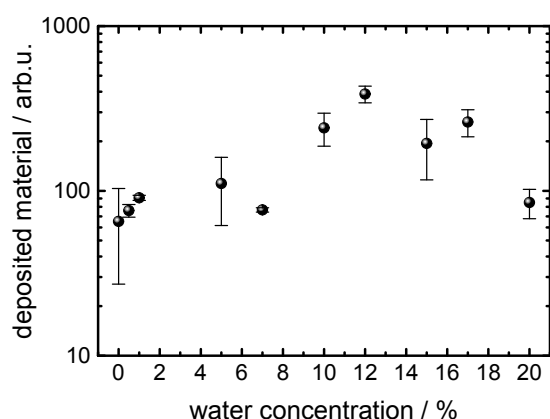


Figure SI 1: Ellipsometry results of films prepared with varying water concentration. At small water concentrations (0-2%), the amount of deposited film, corresponding to the film thickness by assuming a dielectric constant, is slightly smaller than the amount deposited from solutions with 10-17% water. The difference is up to a factor 5, however, tremendous changes, as observed for the crystallinity (Fig. 3), were not observed.

The measurements were performed with a spectroscopic ellipsometer Woollam M-44 with a fixed angle of 70° and a wavelength range of 400-800 nm. The analysis was performed using the VASE software provided by the manufacturer.

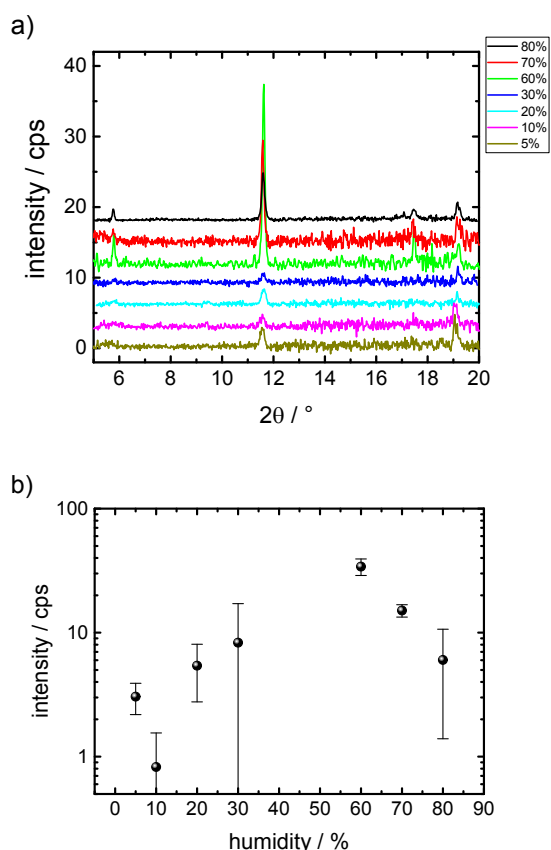


Figure SI 2: Impact of water humidity in the surrounding atmosphere during the dip-coating process. a) X-ray diffractograms of HKUST-1 SURMOF films prepared in 40 synthesis cycles with pure ethanol as solvent in an atmosphere with controlled humidity, i.e. between 5% and 80%, see inset. b) Intensity of the (222) XRD HKUST-1-SURMOF reflexes versus humidity, i.e. relative water vapor pressure.

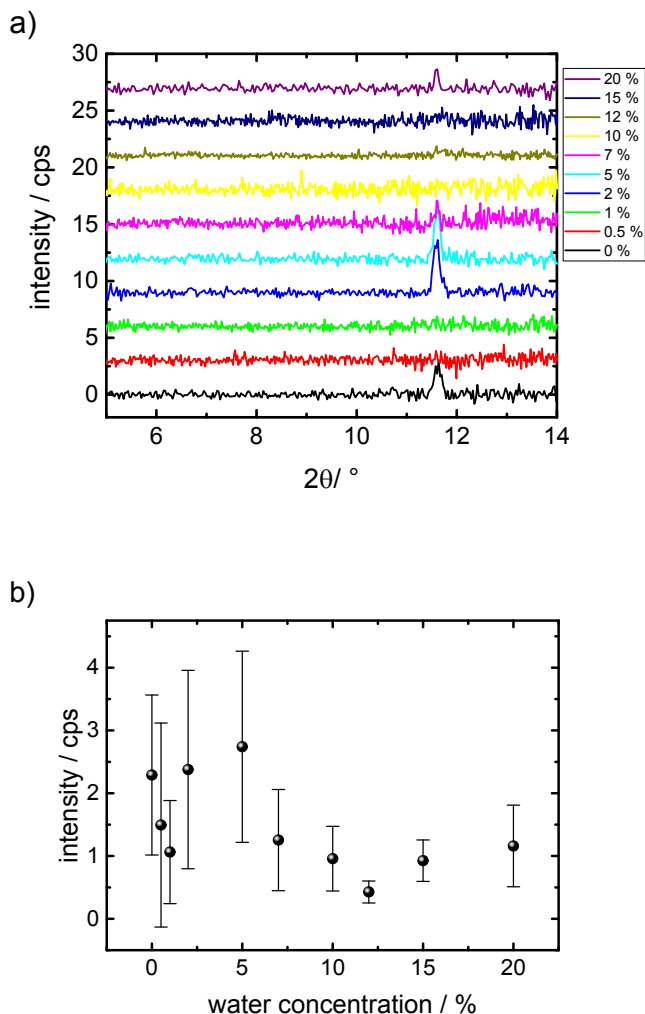


Figure SI3: Impact of water amount in synthesis solutions during spray-coating-process. a) X-ray diffractograms of HKUST-1 SURMOF films prepared in 40 spray cycles with different amount of water in the linker solution, see legend. b) Intensity of the (222) XRD HKUST-1-SURMOF peak versus water concentration in linker solution.

For the layer-by-layer materials deposition using the spray-method, the metal solution (0.5 mM copper acetate in ethanol) is sprayed for 25 s on the substrate, followed by a 30s break and rinsing the sample with ethanol to remove the unreacted metal nodes (for roughly 3s). Then, the organic linker solution (0.1 mM BTC in ethanol) is sprayed on the sample, followed by the break and rinsing process. The process is repeated for many cycles. More details for the spray process can be found in ref. <sup>1</sup>.

During the spray process, the humidity in the lab air was between 45-50%.

## References

- 1 a) H. K. Arslan, O. Shekhah, J. Wohlgemuth, M. Franzreb, R. A. Fischer and C. Wöll, *Adv. Funct. Mater.*, 2011, **21**, 4228–4231; b) S. Hurre, S. Friebe, J. Wohlgemuth, C. Wöll, J. Caro and L. Heinke, *Chemistry (Weinheim an der Bergstrasse, Germany)*, 2017, **23**, 2294–2298.