† Electronic Supplementary Information

Advancement of sorption-based heat transformation by a metal coating of highly-stable, hydrophilic aluminium fumarate MOF

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Nitrogen sorption isotherm for bulk µp-AF



Figure S1: Nitrogen adsorption isotherm of bulk aluminum fumarate (filled squares: adsorption, empty squares: desorption). Acquired at 77 K, after 24 h of vacuum degassing at 120 °C.

PXRD comparison



Figure S2: Powder X-ray diffractograms for μ p-AF from patent literature (a), for the prepared bulk sample in dry (b) and wet (c) condition, and the coated sheet *l* (d). All diffractograms are normalized to the highest reflection.



Figure S3: Measured c_p values (black) and interpolation (red), which was used for the conductivity measurements.



Figure S4: FT-IR spectrum (ATR) of bulk µp-AF.

In-situ PXRD: Individual diffractograms



Figure S5: Waterfall plots of the individual diffractograms acquired during adsorption (a) and desorption (b). Cu-K α radiation, 0.02°/step, 1.0 s/step.

PXRD of top and bottom layer

Part of the top layer was manually removed, ground in an agate mortar and examined by PXRD analysis, and the substrate with the remaining bottom layer was also examined by PXRD.



Figure S6: PXRD (Cu-K α) of the top and bottom layers of the thermal gradient coating. Peaks marked with an asterisk (*) are due to the Al substrate under the bottom layer.

Desorption isobar of the coated sheet



Figure S7: Representative desorption isobar of the μ p-AF coated sheet of aluminum fumarate, after application of the protective lacquer and after completing 4500 ad-/desorption cycles with water vapor.

Packing diagram of MIL-53



Figure S8: Section of the packing diagram of $[Al(BDC)(\mu-OH)]$, MIL-53 with a flexible, 'breathing' network adapting to guest molecules. Each benzene-1,4-dicarboxylate ligand bridges between four Al atoms. The hydroxido-bridging occurs along the metal chains in the *b* direction. The channels can contain guest molecules in the as-synthesized structure (MIL-53as) (a, guest molecules not shown) or be empty after a thermal guest removal in the identical activated hightemperature, large pore structure MIL-53ht. Cooling down to room temperature with adsorption of water from air then transforms the structure into the low-temperature, narrow pore form MIL-53lt (b). Hydrogen atoms are not shown (CSD-Refcodes SABVOH and SABWAU).