Supporting Information:

S1. Experimental procedures

Preparation of magnetic nanoparticles: To a solution of $FeCl_3 \cdot 6\ H_2O$ (5.2 g, 19.2 mmol) and $FeCl_2 \cdot 4\ H_2O$ (3.825 g, 19.2 mmol) in ultrapure water (25 ml), concentrated hydrochloric acid was added (0.85 ml). The solution was homogenized for 10 min under sonication and afterwards added drop wise to 250 ml of 1.5 M NaOH under vigorous stirring. The resulting dispersion is centrifuged at 4000 rpm for 30 min. The supernatant solution is discarded and the precipitate washed with 100 ml of ultrapure water under sonication. The centrifugation and washing steps are repeated three times. The resultant precipitate is finally dispersed in 500 ml of 0.01 M hydrochloric acid and stored in a tightly sealed glass bottle.

Preparation of AlOH(bpdc) (DUT-5, M-DUT-5): Biphenyl-4,4'-dicarboxylic acid (H_2bpdc, 0.8 g, 3.3 mmol) and $Al(NO_3)_3 \cdot 9\ H_2O$ (1.88 g, 5 mmol) are solubilized in 60 ml of dimethylformamide each. The solutions are mixed and homogenized for 30 min under sonication and afterwards heated for 24 h at 180 °C in a 250 ml Teflon lined stainless steel autoclave. The product is separated from the reaction media using centrifugation and washed three times with 100 ml of ethanol followed by overnight drying at 80 °C in a cabinet desiccator and activation at 200 °C under vacuum. To obtain magnetically functionalized MOF particles, up to 40 ml of the above mentioned dispersion are centrifuged for 30 min at 5000 rpm. The resultant precipitate is then dispersed in the H_2bpdc solution using 10 min of sonication. After mixing and homogenizing, the dicarboxylic acid/nanoparticle dispersion and the aluminium containing solution are heat treated and washed as mentioned above. Prior to the drying steps, magnetic and possibly occurring non-magnetic products are separated using a permanent magnet that is placed on the side of the centrifugation vessel.

Preparation of AlOH(ndc) (DUT-4, M-DUT-4): 2,6-naphthalenedicarboxylic acid (H_2ndc, 0.8 g, 3,7 mmol) and $Al(NO_3)_3 \cdot 9\ H_2O$ (1.3 g, 3.5 mmol) are dissolved in 80 ml and 40 ml of dimethylformamide, respectively. The solutions are homogenized by sonication for 10 min, mixed and heat treated in a 250 ml Teflon lined stainless steel autoclave for 24 h at 110 °C. Magnetic functionalization, washing and drying steps are performed as described for AlOH(bpdc).

Preparation of Cu_3btc_2 (HKUST-1, M-HKUST-1): A solution of trimesic acid (H_3btc, 0.5 g, 2.4 mmol) in 15 ml dimethylformamide is added to a solution of $Cu(OAc)_2 \cdot H_2O$ (0.475 g, 2.4 mmol) in 15 ml dimethylformamide. The mixture is refluxed for 12 h under vigorous stirring to prevent gel formation. A magnetic functionalization is achieved if the centrifuged precipitate of up to 15 ml of the iron oxide dispersion is dispersed in the trimesic acid solution prior to the reaction. The resultant products are washed with ethanol for 24 h in a Soxhlet extractor and dried overnight at 80 °C in a cabinet desiccator. Activation was performed at 120 °C under vacuum for at least 12 h.

S2. X-ray powder diffraction

Powder diffraction patterns were measured with Cu-Kα radiation at room temperature, using a Siemens D5000 diffractometer in reflexion geometry.

HKUST-1 ($Cu_3(btc)_2$)

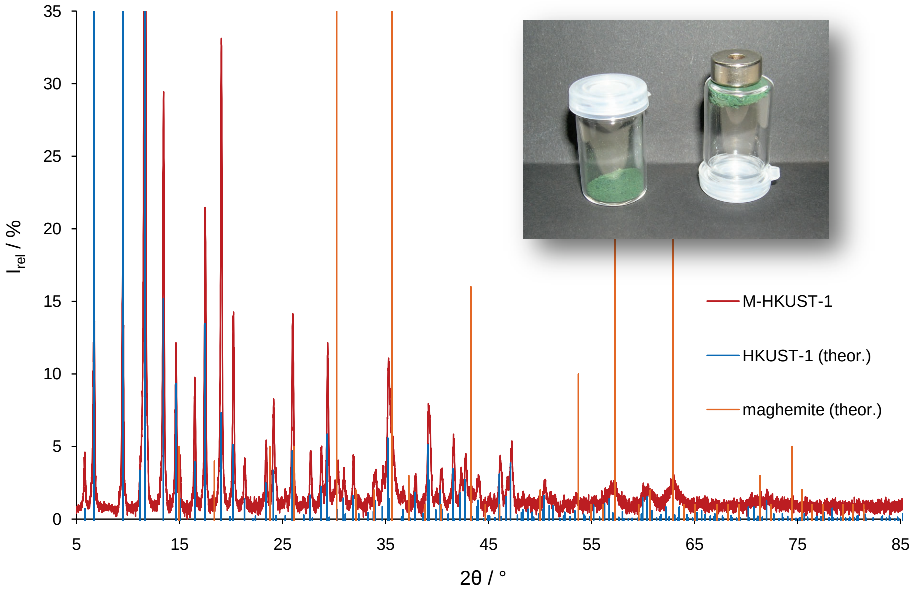

Figure S1: X-ray powder diffraction pattern of M-HKUST-1 composite (red) with theoretical peak positions of HKUST-1 (blue) and maghemite (orange).

DUT-4 (AlOH(ndc))

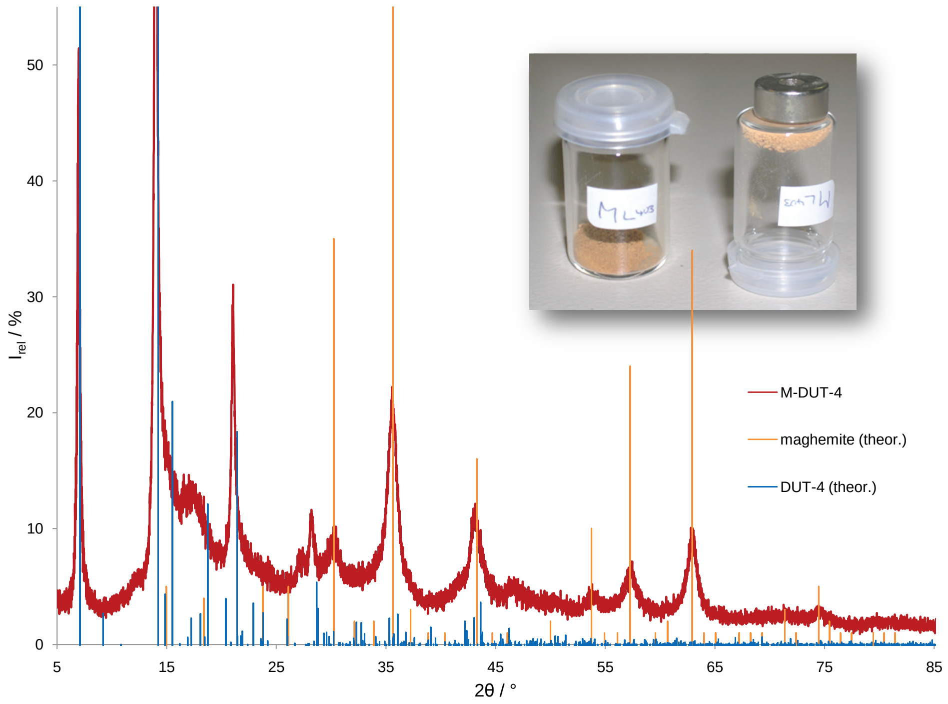

Figure S2: X-ray powder diffraction pattern of M-DUT-4 composite (red) with theoretical peak positions of DUT-4 (blue) and maghemite (orange).

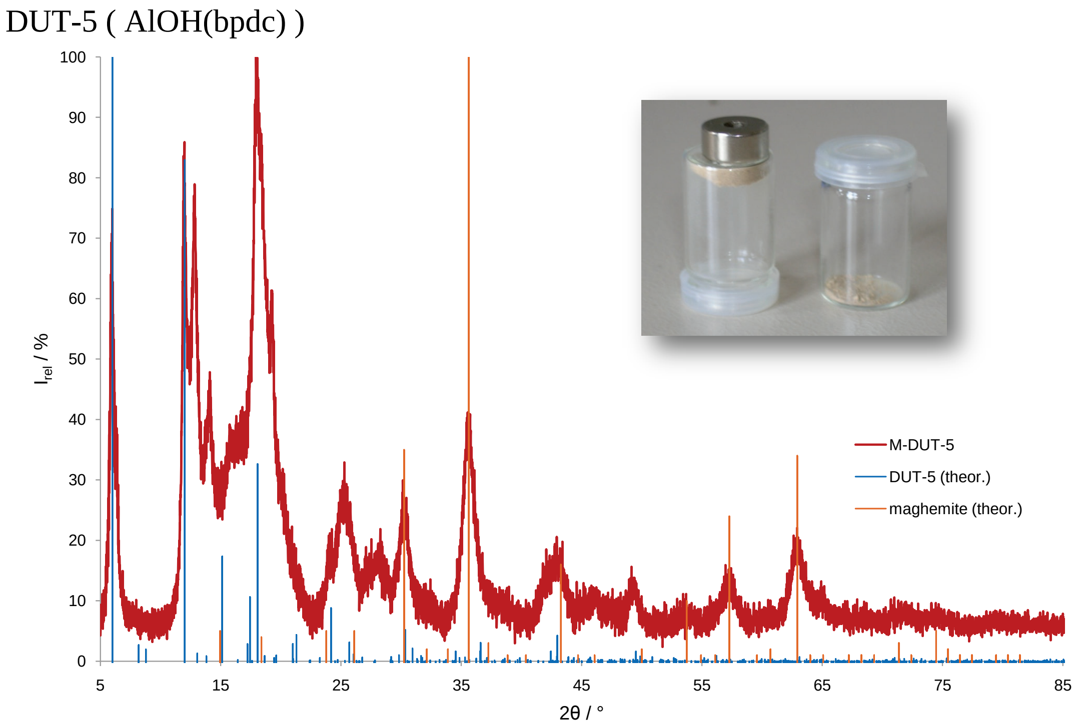

Figure S3: X-ray powder diffraction pattern of M-DUT-5 composite (red) with theoretical peak positions of DUT-5 (blue) and maghemite (orange).

S3. Magnetization measurements

Magnetic measurements were performed using a vibrating sample magnetometer by Quantum Design (Physical Property Measurement System with VSM option). The powdered samples were filled into gelatin capsules and sealed with two-component adhesive. The sealed capsules were fixed in a small plastic tube and mounted onto the instruments sample holder. All measurements have been performed at room temperature.

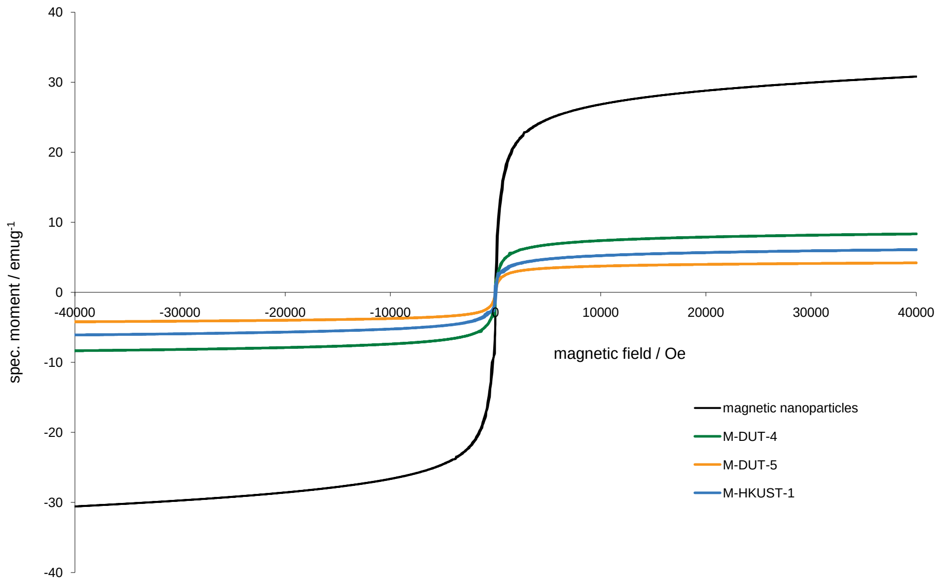

Figure S4: Magnetization curves of magnetic MOF composites and magnetite nanoparticles that were used for functionalization.

S4. Nitrogen physisorption measurements

Nitrogen adsorption/desorption isotherms were recorded at 77 K on a Quadrasorb by Quantachrome Instruments. Between 50 and 100 mg of the powdered sample were weighed into the measuring cell and activated at least 24 h at 120 °C under vacuum. Low pressure measurements down to 10^{-6} atm were measured on an Autosorb 1C by Quantachrome Instruments.

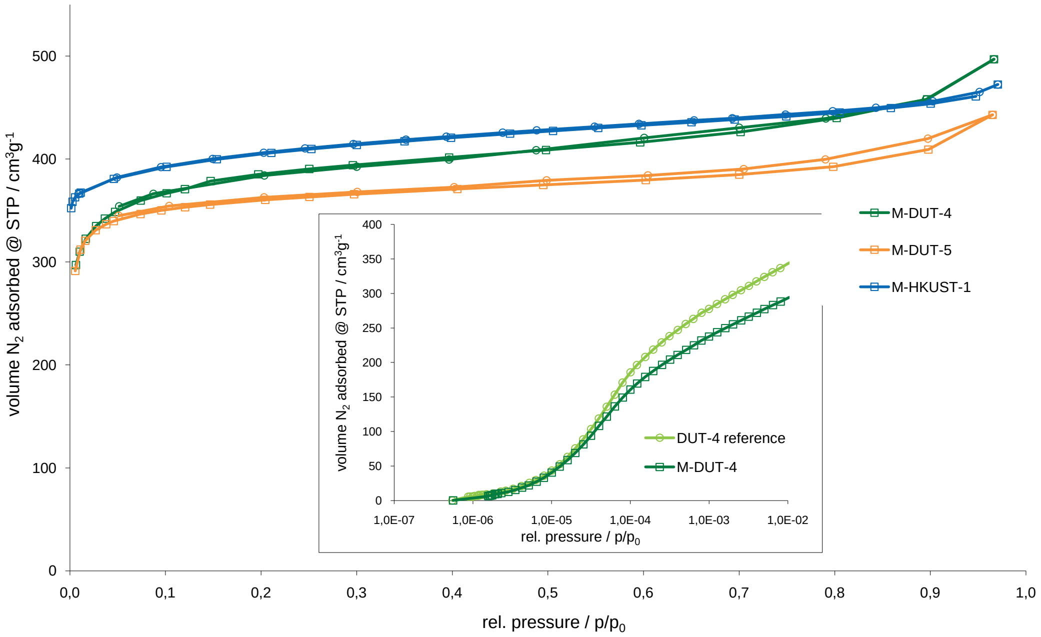

Figure S5: Nitrogen physisorption isotherms of magnetic MOF composites measured at 77 K. The inset shows the adsorption behavior of M-DUT-4 in comparison to a nonmagnetic reference at low pressures.

S5. Scanning electron microscopy

SEM micrographs of powdered samples on carbon pads were recorded using a DSM-982 Gemini by Zeiss. Prior to imaging the samples were sputtered with a thin layer of gold to improve conductivity and image quality.

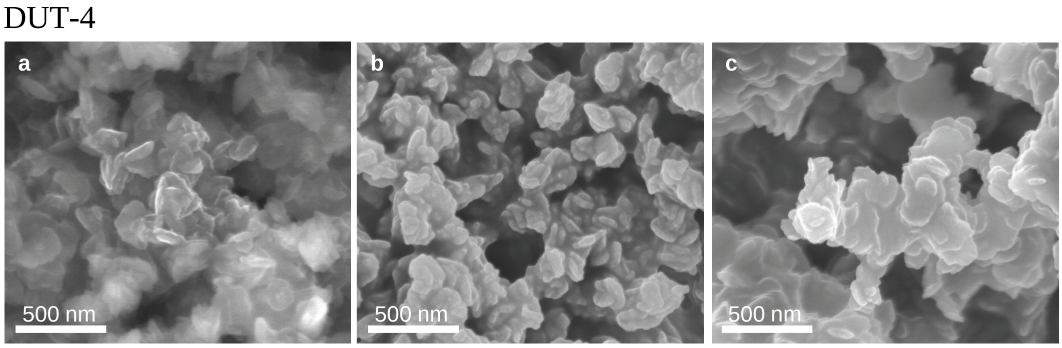

Figure S6: Scanning electron micrographs of magnetic M-DUT-4 samples (a,b) and a nonmagnetic reference sample (c).

DUT-5

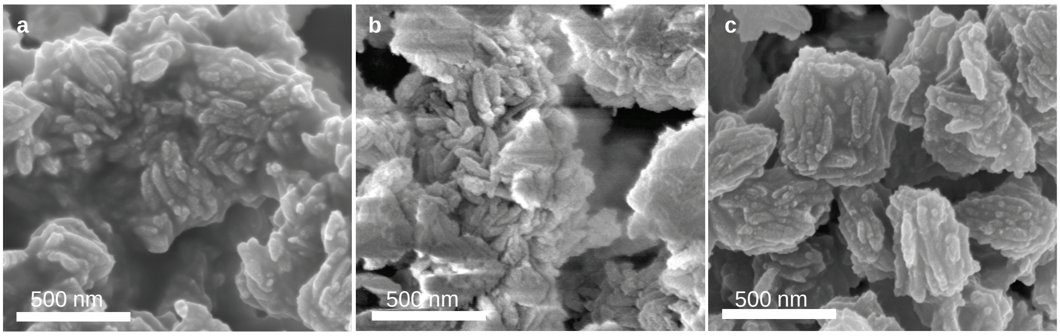

Figure S7: Scanning electron micrographs of M-DUT-5 samples (a,b) and a nonmagnetic reference sample (c).

S6. Dynamic light scattering

DLS measurements clearly indicate that the magnetic particles tend to aggregate in a solution of polycarboxylic acids. Figure S9 shows the DLS data of dispersion of magnetic nanoparticles in a DMF solution containing 2,6-naphthalenedicarboxylic acid. The main particle size shifts from about 20 nm to 40 nm and finally 60 nm in dispersion 5, 10 and 30 minutes after a 10-minute ultrasonic treatment, respectively.

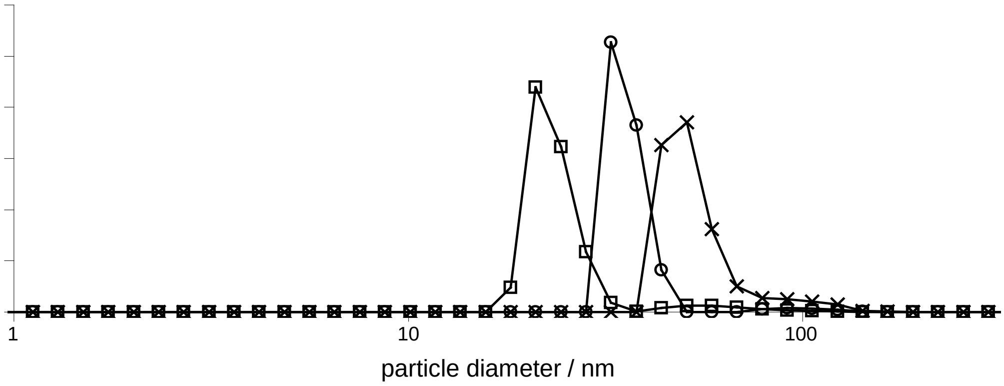

Figure S8: Particle size distribution of a magnetic nanoparticle dispersion in DMF solution containing H_2ndc, 5 (□), 10 (○) and 30 minutes (×) after a ten-minute ultrasonic treatment.

S7. Cyanosilylation of benzaldehyde, filtration test

In a glovebox, 40 mg pre-activated AlOH(ndc) (DUT-4) or Fe_3O_4@AlOH(ndc) (M-DUT-4), respectively, were transferred to the reaction flask. After connection to the vacuum line, the flask was flushed with argon and 17 ml *n*-heptane (containing *n*-octane and *n*-hexadecane as internal standards) were introduced. After addition of 425 mg (4 mmol) freshly distilled benzaldehyde, 794 mg (8 mmol) trimethylsilylcyanide (TMSCN) were added. The reaction mixture was stirred at 40°C and the reaction was monitored using a SHIMADZU GCMS QP5000 equipped with a non-polar BPX5 column (5 % Phenylpolysilphenylene-siloxane) from SGE. For the filtration test, 3 ml of the reaction mixture were filtered off after approx. 4 hours reaction time using a permanent magnet for the separation of the catalyst and were transferred into an argon flushed flask held at 40°C with a syringe. Stirring was continued and the composition of the reaction mixture was determined using GC-MS analysis. Since the crystal structure of DUT-4 should not exhibit Lewis acidic centers, the catalytic activity is attributed to a large number of defects in the microcrystalline material.

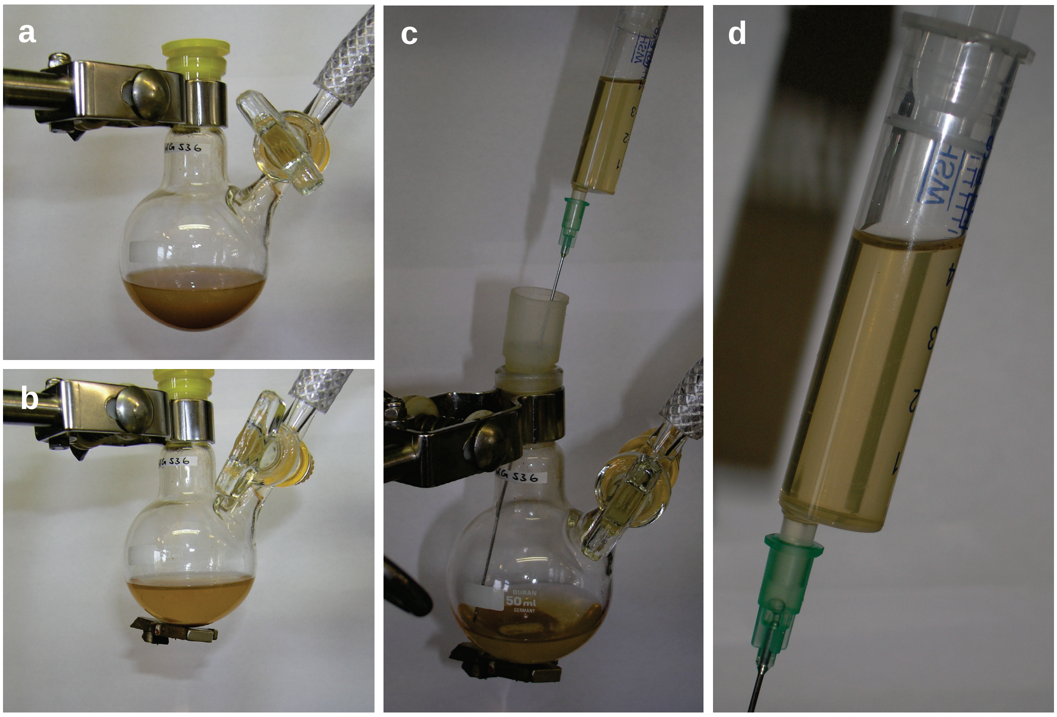

Figure S9: Photographs of the magnetic separation of M-DUT-4 during cyanosilylation. The pictures show the turbid reaction mixture after 5 min without stirring (a) and the less turbid mixture 30 sec after placement of a strong permanent magnet (b). A clear supernatant solution is obtained after less than 2 min (c), which a part of was separated using a syringe without any filter (d) and stored under reaction conditions for the heterogeneity test.

S8. Magnetic heating

For the magnetic heating experiments a TruHeat HF5005 high frequency generator by Hüttinger Elektronik was used. The inductor used was a water-cooled copper coil with 11 turns on a length of 105 mm and a diameter of 42.5 mm. For measurement an open topped vessel with a dispersion of the desired particles is placed in the inductor. Whilst application of the AC magnetic field the temperature is monitored using a pyrometer that was placed above the inductor and focused on the dispersion surface. To obtain the heating curves of the samples a reference measurement of the pure solvent (equivalent volume) is subtracted from the data.

S9. Ibuprofen loading and release

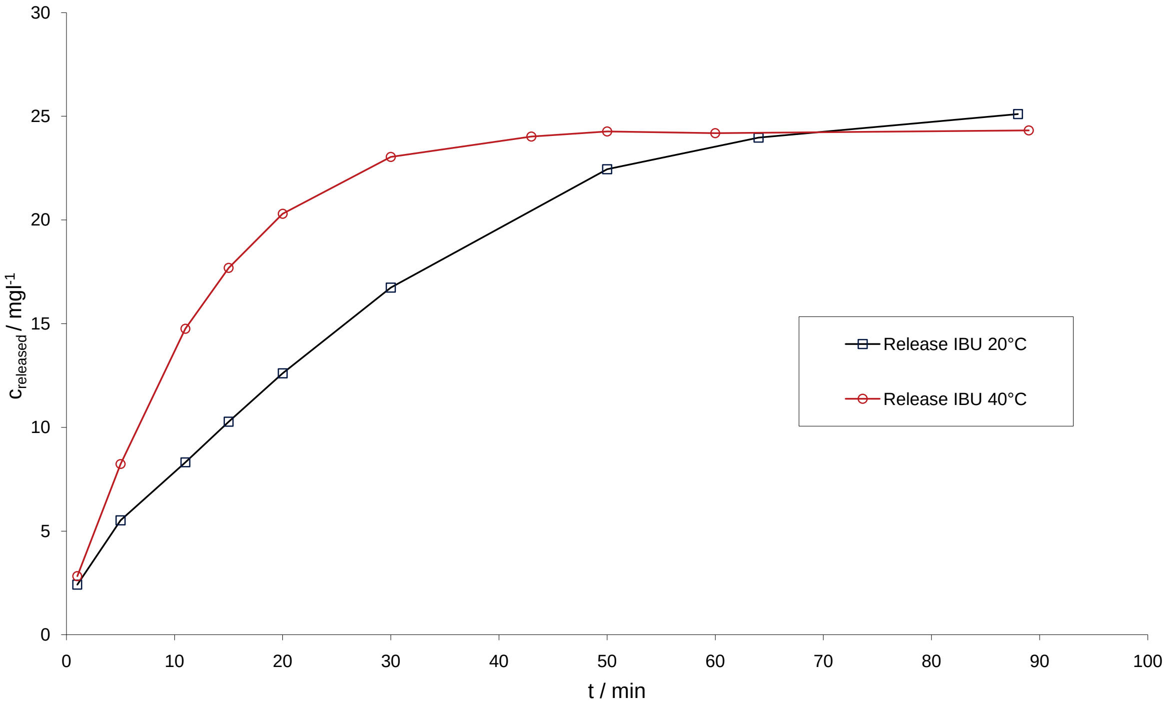

Figure S10: Release curves of Ibuprofen loaded M-HKUST-1 samples at 20 °C(□) and 40 °C (○).

For drug loading 100 mg of activated M-HKUST-1 is added to a solution of 15 mg Ibuprofen in heptane (100 ml). The wet infiltration procedure is performed using a rotary evaporator. For release tests the loaded samples (theoretical loading 14,3 % wt.) are then stirred in 50 ml of ethanol at various temperatures. At specific time intervals 1 ml of the ethanolic solution is withdrawn and filtered using a syringe filter and added to 9 ml of pure ethanol. The thus obtained solution is characterized using UV-VIS spectroscopy. The concentration of Ibuprofen in the solution is calculated using a calibration line obtained from extinctions of ethanolic Ibuprofen solutions (1-50 mg l^{-1}) at 224 nm.

S10. Mössbauer spectroscopy

The Mössbauer absorption spectrum measured at room temperature shows a quasi-symmetrical set of 6 peaks which leads to the assumption that mainly Fe^{3+} is present in the sample. This is consistent with the data from X-ray powder diffraction, were only maghemite, but no magnetite peaks are observed.

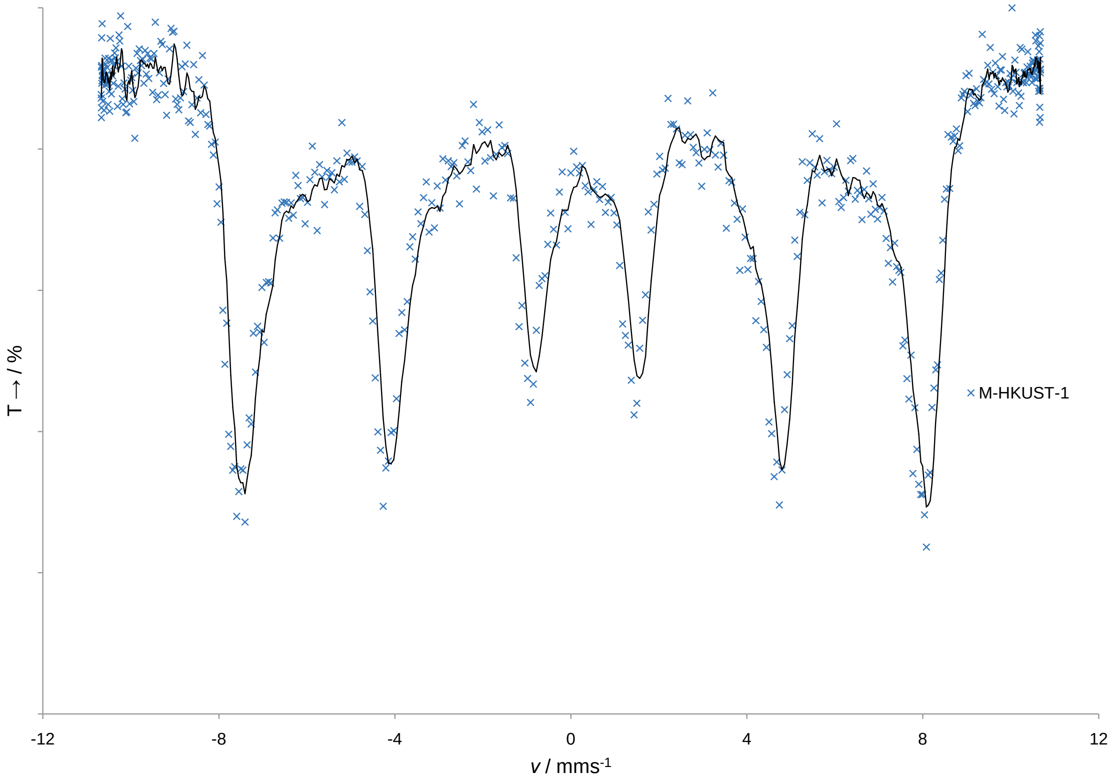

Figure S11: Mössbauer absorption spectrum of M-HKUST-1 at room temperature.