Supporting Information

CMPO-functionalized MIL-101(Cr) as highly selective uranium adsorbent

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ABSTRACT: The carbamoylmethylphosphine oxide (CMPO) functionalized MIL-101(Cr) was investigated as a potential uranium scavenger. This metal−organic framework-based adsorbent shows very high selectivity toward uranium, as well as thorium, in competition with various rare earth metals. Furthermore, it showed rapid adsorption kinetics, in both batch conditions and a dynamic (column) setup. The adsorbent is fully regenerable, using oxalate solution. Fast elution kinetics in the column setup were observed during the regeneration. In addition, reusability studies were performed under dynamic conditions. Five consecutive adsorption/desorption cycles were carried out, showing a consistent 100 % recovery, at pH 4, using 0.1 M oxalate solution as an effective stripping agent. Additionally, the successive use over various adsorption/desorption cycles with constant performance proves the high stability of this adsorbent in an acidic, aqueous environment.

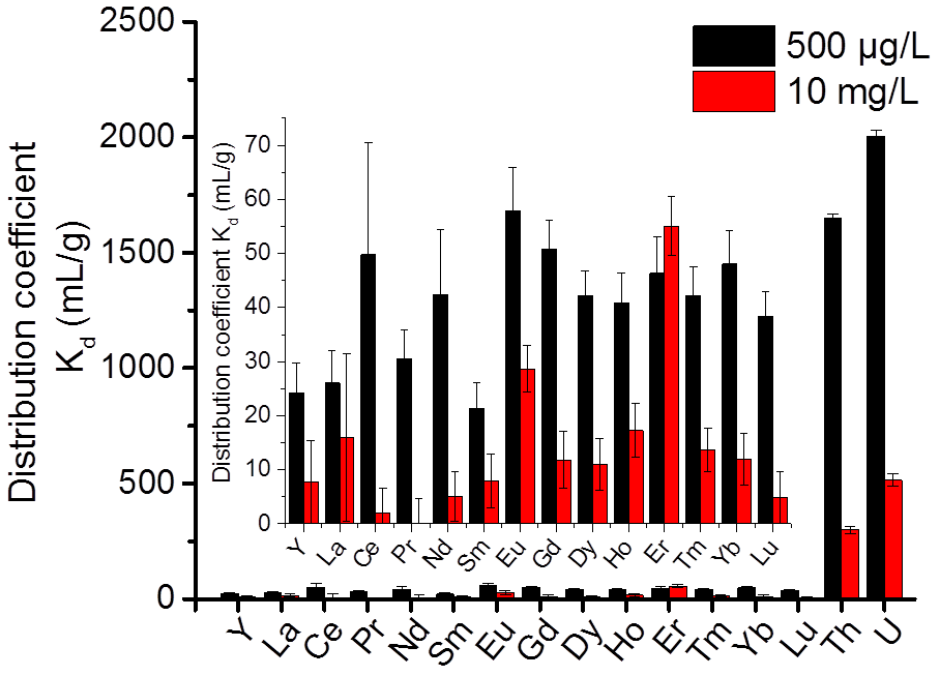

Figure S1. Selectivity results of CMPO@MIL-101(Cr). Zoom on the competing REE elements included.

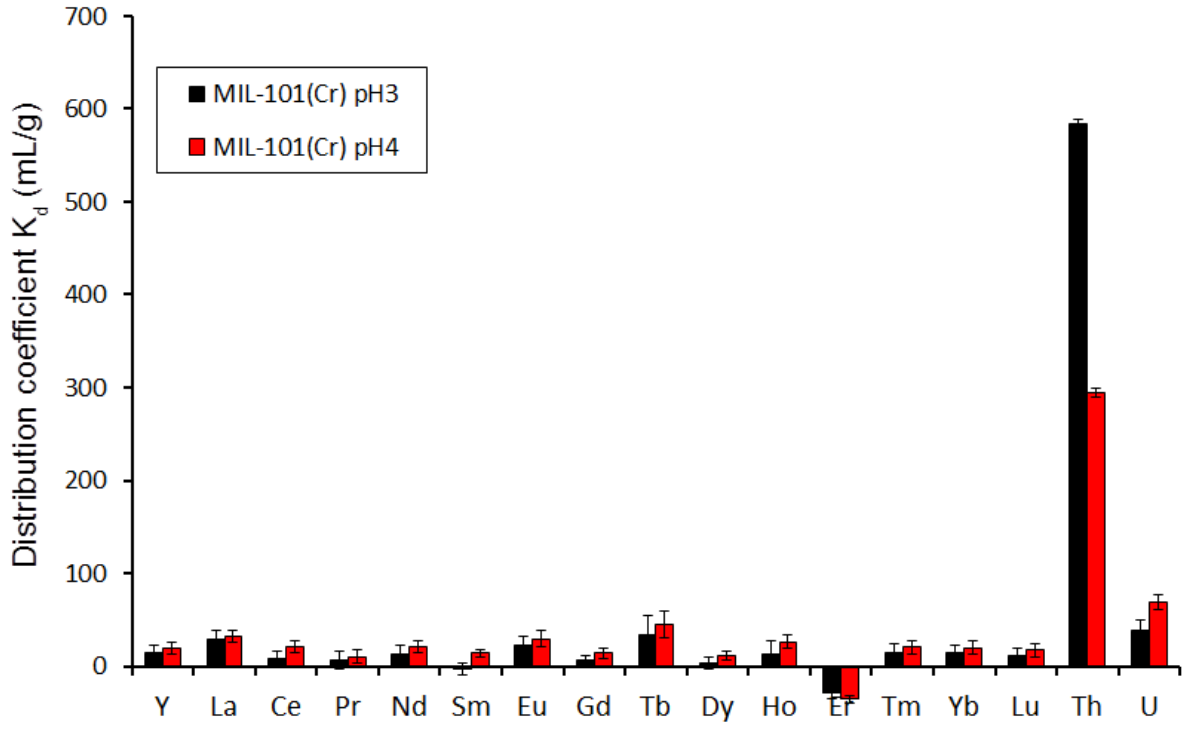

Figure S2. Selectivity results of pure MIL-101(Cr) at pH 3 and pH 4. C_0: 10 mg/L

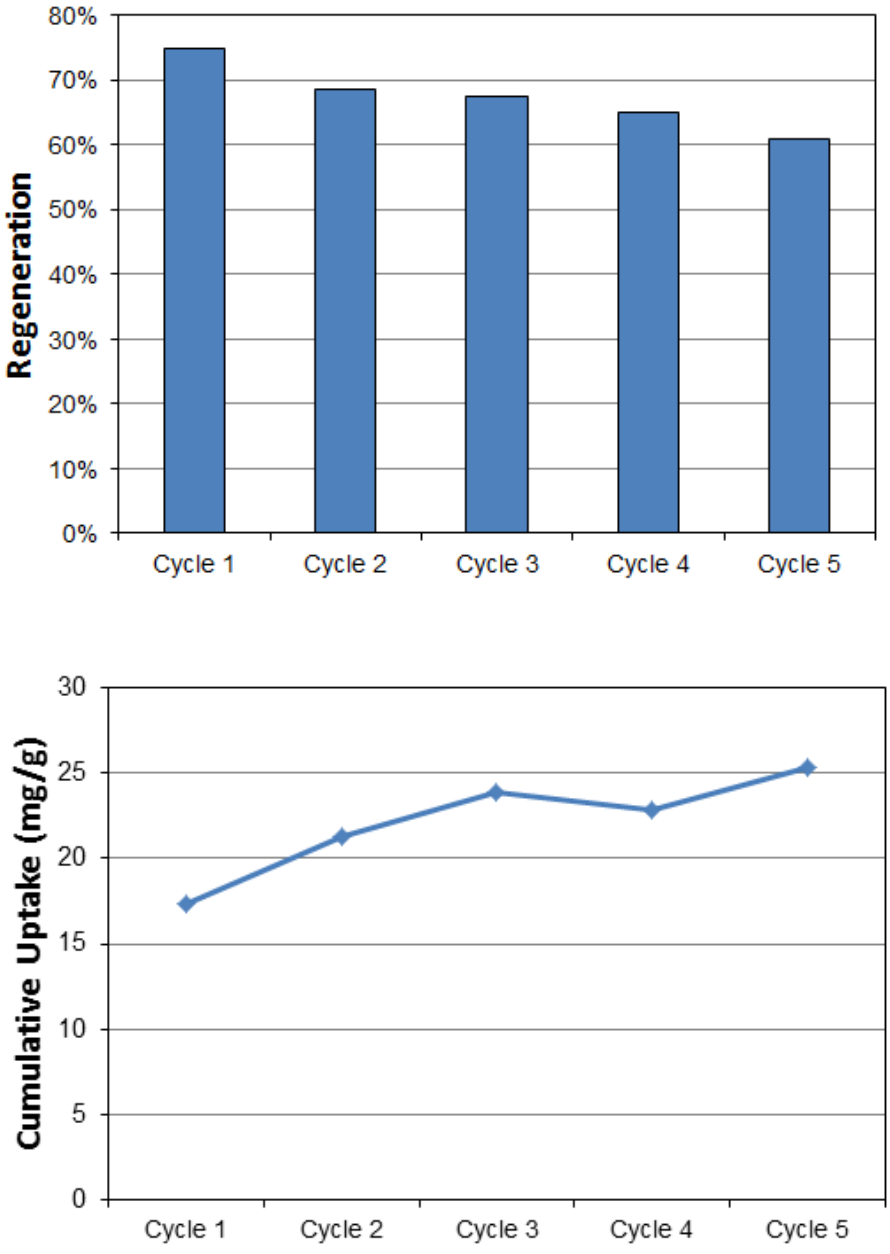

Figure S3. (Top) Uranium regeneration results for CMPO@MIL-101(Cr) using 0.1 M oxalate solution (pH 2) via column setup. (Bottom) Cumulative uranium uptake over five consecutive adsorption/desorption cycles with 0.1 M oxalate solution (pH 2) as stripping agent. (Experimental conditions in main article).

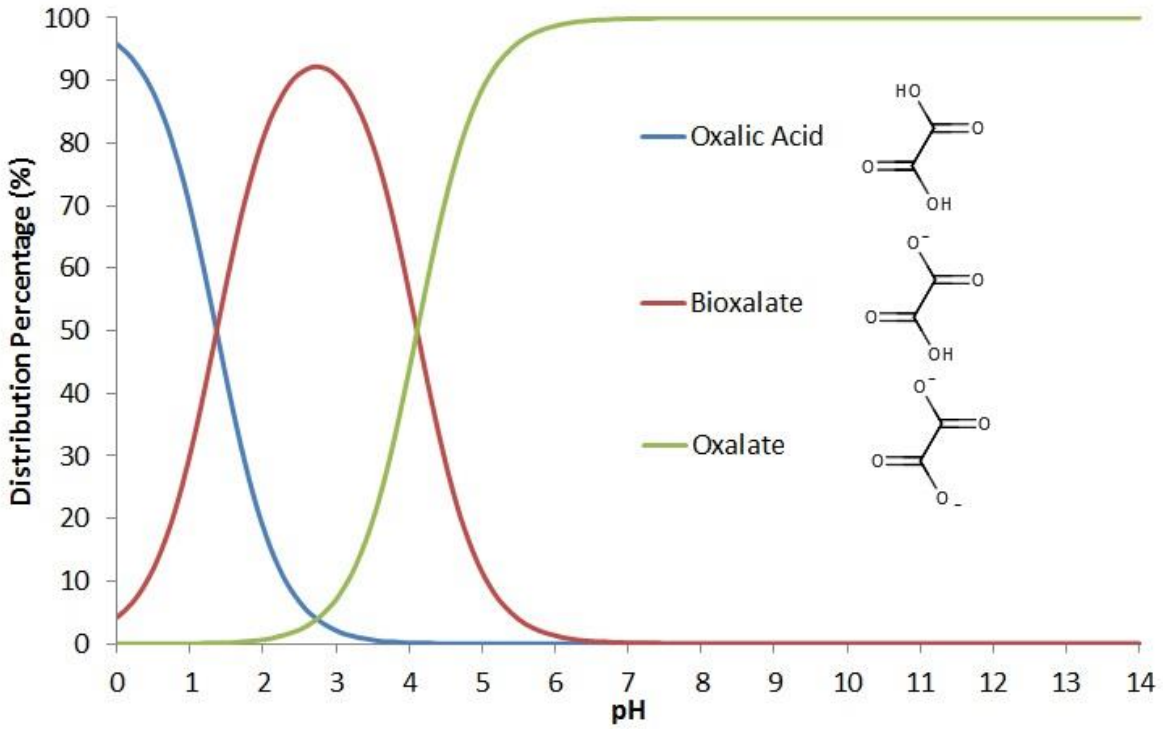

Figure S4. Speciation diagram for the oxalate system in water as a function of pH. The y-axis gives the fraction of each species present.

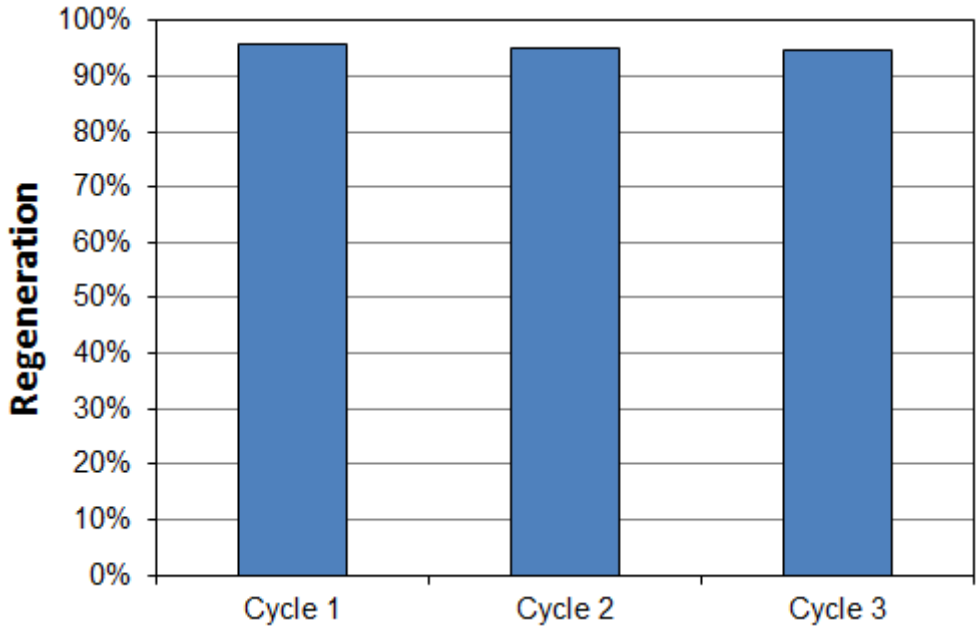

Figure S5. Uranium regeneration results for CMPO@MIL-101(Cr) using an increased 1 M oxalate solution (pH 2) via column setup. (Experimental conditions in main article).

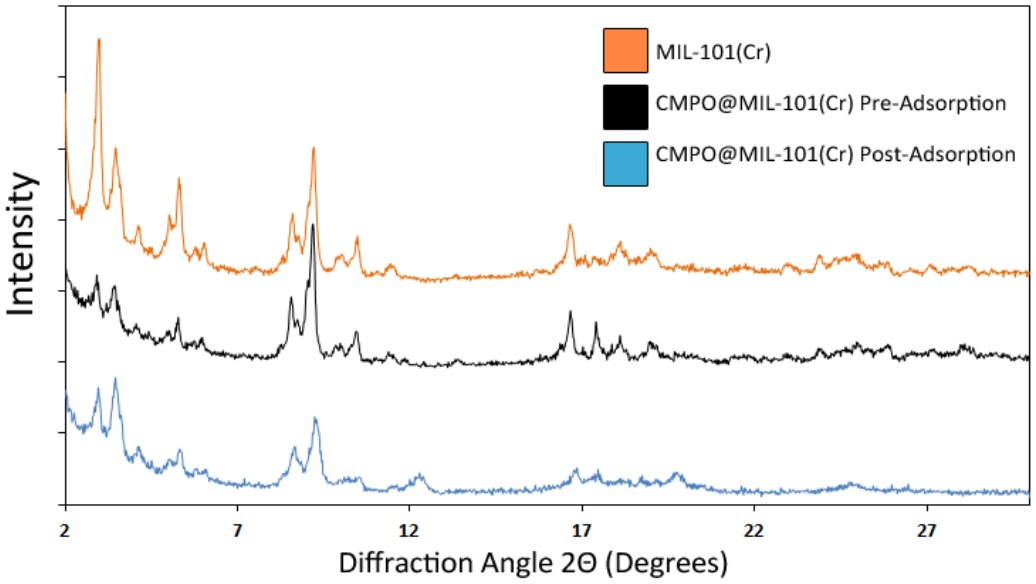

Figure S6. Powder X-ray Diffraction comparison of pristine MIL-101(Cr) (orange) and CMPO@MIL-101(Cr) pre- (black) and post-adsorption/desorption experiments (blue).

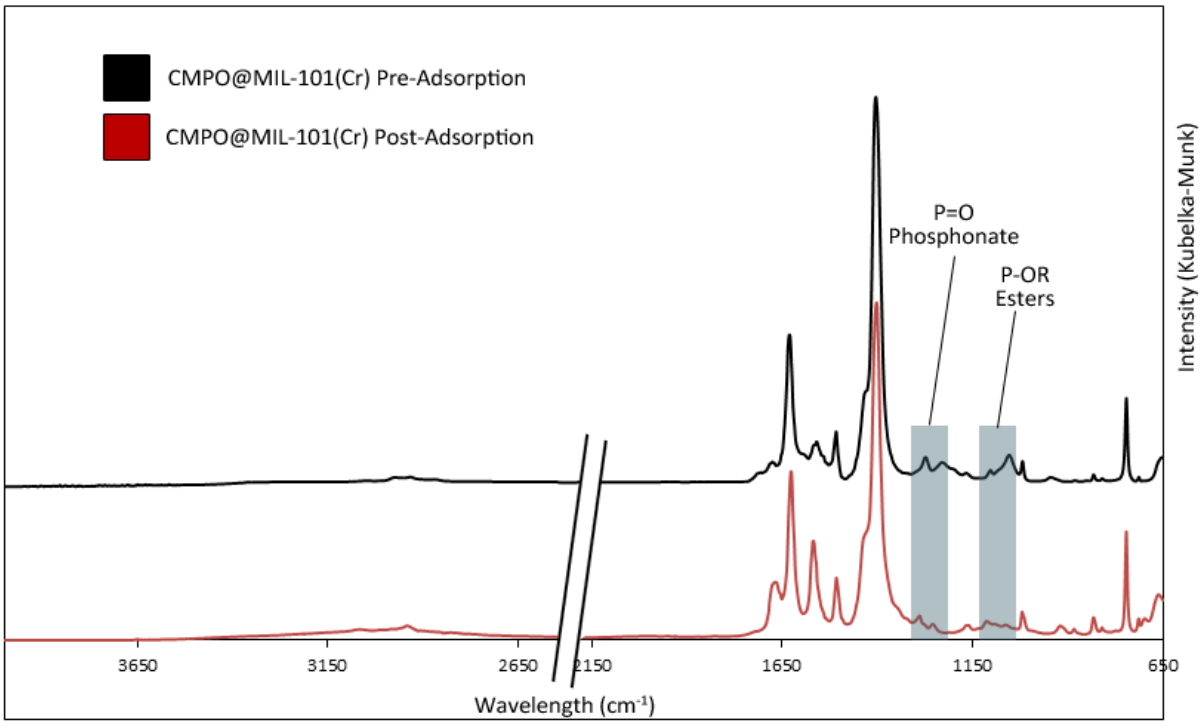

Figure S7. FTIR comparison (DRIFTS) of CMPO@MIL-101(Cr) pre- (black) and post-adsorption/desorption experiments (red). The zone of interest (i.e., corresponding to ligand vibrations) is highlighted and annotated.

Table S1. Numerical N_2-adsorption data and leaching analysis for CMPO@MIL-101(Cr) prior/post column adsorption studies.

CMPO@MIL-101(Cr)	S_L[a] (m^2/g)	S_{BET}[b] (m^2/g)	Pore Volume (cm^3/g)	Leaching (wt. %)[c]
Prior adsorption studies	1336	1014	0.59	-
Post adsorption studies	1780	1310	0.68	15 %

[a] Langmuir model. [b] Calculated between p/p_0 0.05 – 0.3. [c] As determined by XRF.

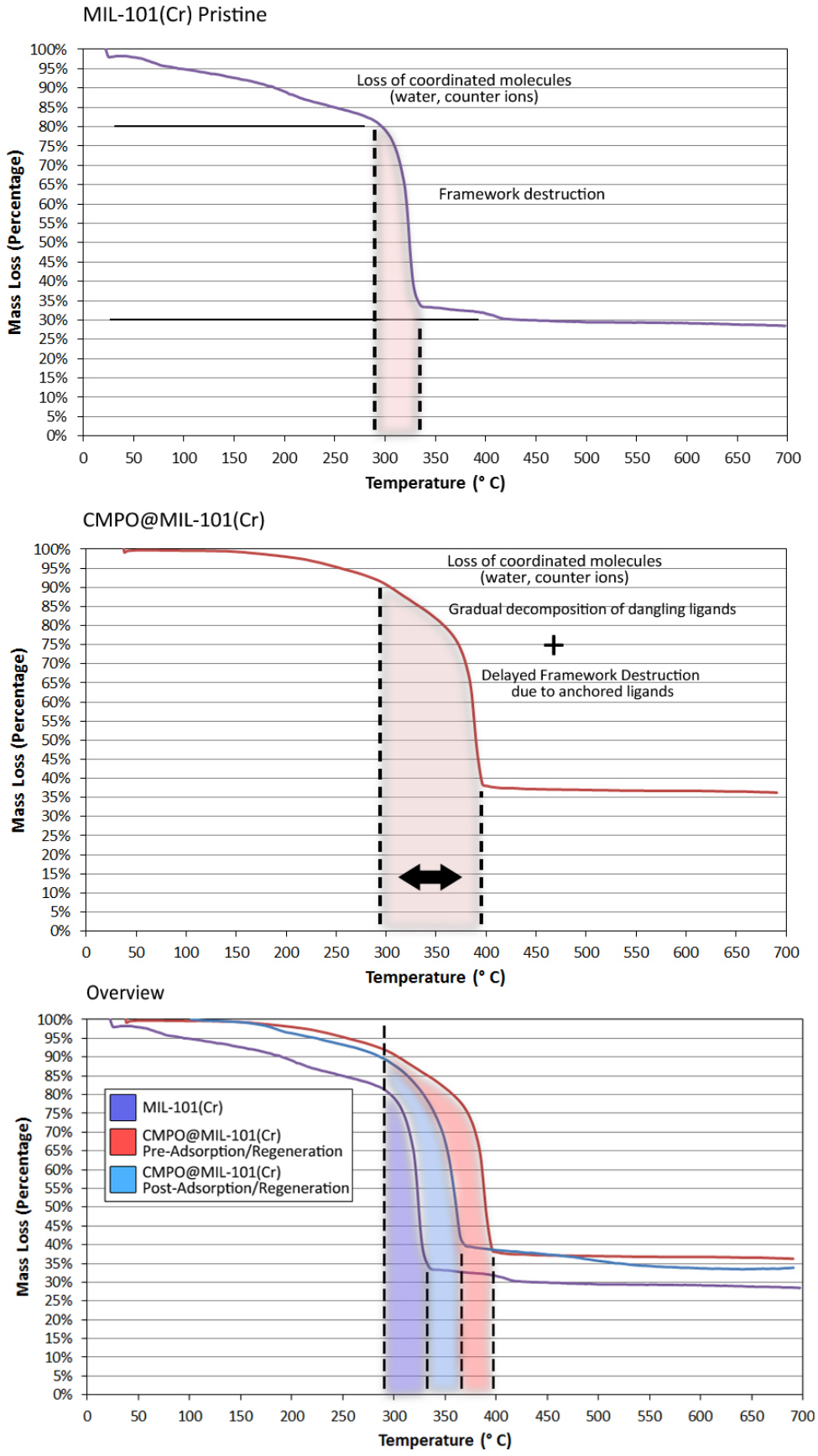

Figure S8. Comparison of Thermogravimetric analysis results of CMPO@MIL-101(Cr), pre- (red) and post-adsorption/desorption experiments (blue).