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Electronic Supplementary Information

Organically modified clay with potassium copper hexacyanoferrate for enhanced Cs⁺ adsorption capacity and selective recovery by flotation

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1. Flotation setup:



Figure S1. Schematic representation of the laboratory-scale flotation cell. The flotation column inner diameter is 65 mm and the foam was retrieved through a side-arm located 65 mm from the base of the flotation column.

2. TGA data:

Based on the TGA data an estimate of the HCF content in the composites was determined by the mass ratio of the combustion residue (*w* at 1,000 °C) to the initial mass of montmorillonite (x), KCuHCF (y), and organo-clays (z) (*w* at 100 °C):

$$x = \frac{w_{Mont,1000^{\circ}C}}{w_{Mont,100^{\circ}C}}; \ y = \frac{w_{HCF,1000^{\circ}C}}{w_{HCF,100^{\circ}C}}; \ z = \frac{W_{Organoclay,1000^{\circ}C}}{W_{Organoclay,100^{\circ}C}}$$
(S1)

The three parameters were determined from the TGA curves of the single bulk components. It was assumed that the mass increase of the composite compared to the corresponding organo-clay originated only from the KCuHCF (abbreviated as HCF). The HCF fraction in the composite is given by *k* and hence the fraction of the supporting organo-clay matrix is (1 - k). On the basis of the initial $(W_{comp,1000} \circ_C)$ and the residual $(W_{comp,1000} \circ_C)$ masses of composite from the TGA curve, a mass



Figure S2. a) TGA curves of montmorillonite, organic components (EHDA-Br and DT), organo-clays (Mont-EH-DT) and composites (Mont-HCF); b) TGA and DSC curve of bulk KCuHCF; c) TGA curves of organo-clays and composites with varied DT contents.

equivalence equation can be readily obtained as follows:

$$k \bullet W_{comp,100^{\circ}C} = W_{comp,100^{\circ}C} - W_{comp,100^{\circ}C} (1-k) z \not \downarrow y$$
(S2)

The left hand side of Eq. S2 is the initial amount of HCF in the composite and the right hand side represents the value estimated from the HCF residue in the composite at 1,000 °C by subtracting the residues of organo-clay from the total composite residues. This estimation was based on an assumption that the components in the composites experienced identical decomposition kinetics (same mass loss ratio) to that in the bulk system.

Likewise, the organic loading in the composite, l in %, can be calculated to be

$$l = (1 - k)(1 - z / x)$$
(S3)

with (1-z/x) being the fraction of organic components in the organo-clay

 $\frac{w_{organoclay,100^{\circ}C} - w_{organoclay,1000^{\circ}C} / x}{w_{organoclay,100^{\circ}C}}.$

3. Langmuir adsorption isotherm for organo-clay adsorbents:



Figure S3. Adsorption isotherms of organo-clays (Mont-EH-DT) with varying DT contents compared to pristine montmorillonite. Solid lines are fittings of the Langmuir model.

4. Element analysis as measured by XPS:

Label _	Atomic fraction (wt%)			C/S:	Es/C:	C/E-
	Si	С	Ν	. Cu/Si	Fe/81	Cu/re
Montmorillonite	19.93	23.53	0.18	0	0	-
Mont-HCF(DT0.1)	19.38	19.24	2.32	0.018	0.012	1.46
Mont-HCF(DT0.8)	18.25	19.05	3.83	0.020	0.018	1.12
Mont-HCF(DT8)	14.91	39.32	2.71	0.014	0.011	1.24
Mont-HCF(DT50)	6.43	55.36	6.52	0.076	0.065	1.17
KCuHCF	0	63.66	19.27	-	-	1.28

Table S1. Surface composition of organo-clay-HCF adsorbents determined by XPS.



Figure S4 a) XPS survey of Mont-HCF composites before and after Cs uptake ($C_0 = 5$ mM). From bottom to top: before Cs adsorption for DT0.8, after Cs adsorption for DT0.8, DT8 and DT50. b) K 2*p* and c) Cs 3*d* spectra of Mont-HCF(DT0.8) before and after Cs⁺ uptake ($C_0 = 5$ mM).

5. SEM-EDX of Cs contaminated clay-HCF composite adsorbents:



Figure S5. SEM image and EDX map of Mont-HCF(DT8) (a-c) and Mont-HCF(DT50) (d-f) after Cs⁺ uptake ($C_0 = 5$ mM).

6. Particle size distribution:



Figure S6. Particle size distribution of Mont-HCF composites.