Electronic Supplementary Material (ESI) for RSC Advances.

Layer Charge Robust Delamination of Organo-Clays

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Supporting Information

Content:

1. Supporting Data

PXRD of Clay Minerals.

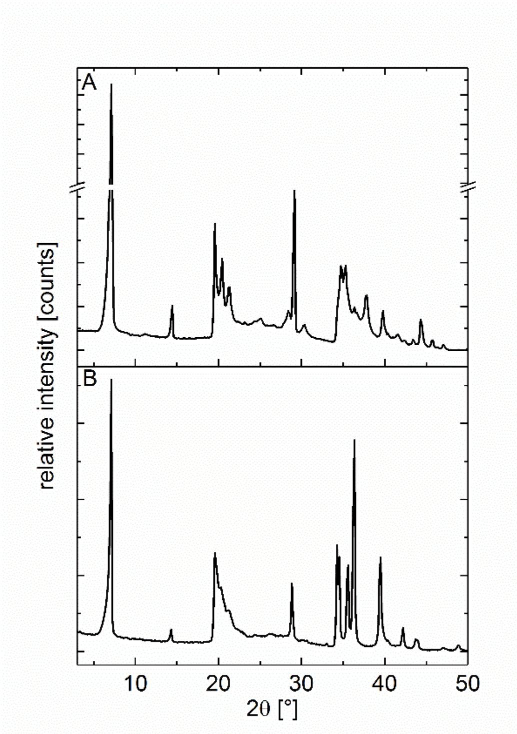

Fig. S1: PXRDs of Hec (A) and Verm (B) indicate a crystalline swollen phase with one layer of water molecules in the interlayer space (d_{001} = 12.5 Å). The patterns can be indexed in C2/m (No. 12) and least square refined lattice parameters for Hec (a = 5.20(1) Å, b = 9.10(1) Å, c = 12.48(4) Å, γ = 95.44(4)°) and Verm (a = 5.26(1) Å, b = 9.07(2) Å, c = 12.48(4) Å, γ = 95.22(4)°) can be derived without unidexed lines.

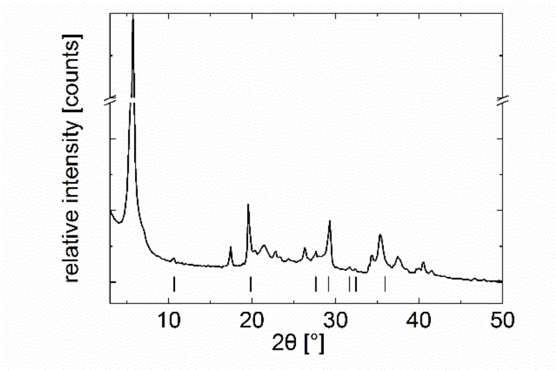

Fig. S2: PXRD of as synthesized Hec_{pri}, Proto-amphibol (PDF2-database number 00-013-0409, the most intense reflexions are marked by ticks) is found as a sidephase.

Layer Charge Determination.

Tab. S1: Layer charges corresponding to dense packings of mono- or bilayers for n-alkylammonium chains ($C_nH_{2n+1}NH_3^+$) of different length n. The layer charge that results in a dense monolayer or a dense bilayer arrangement is given as follows, assuming a typical *a,b* area of 47.6 Å^2:[1]

chain lenght n	monolayer [p.f.u.]	bilayer [p.f.u.]
5	0.56	1.12
6	0.50	0.99
7	0.44	0.89
8	0.40	0.80
9	0.37	0.73
10	0.34	0.67
11	0.31	0.62
12	0.29	0.58
13	0.27	0.54
14	0.25	0.51
15	0.24	0.48

Meglumine-clays: CHN-analysis and EDX.

The completeness of exchange was determined by CHN- and by EDX-analysis for the remaining sodium-content. Interlayer cation content was calculated by carbon content. The amount of organo-cations per gram clay is calculated from the determined wt% of carbon in the organo-clay; the amount of carbon atoms per Meglumine-cation (7) and the molar mass of carbon:

$$CEC_{Meglu-clay} = \frac{wt\%\, C}{7 \cdot 12.011 g/mol}$$

$CEC_{Meglu\text{-}clay}$ was compared to the expected maximum Meglumine-content in mmol / 100 g ($CEC_{Meglu\text{-}max.}$) as calculated from the amount of exchangeable Na^{+}-ions n per 100 g. The amount n is given in mval as determined by the $BaCl_2$-method, n_{verm} = 185 mval (per 100 g), n_{Hec} = 129 mval (per 100 g), n_{LCR1} = 103 mval (per 100 g), n_{LCR2} = 75 mval (per 100 g) and $n_{Hec\text{-}Pri}$ = 116 mval (per 100 g). Furthermore, the molar masses M of the former interlayer ions of the clays and Meglumine-cations are needed.

$$CEC_{Meglu-max} = \frac{n}{100\, g - M(former\ interlayer\ ions) \cdot n + M(Meglumine) \cdot n}$$

The completeness of organo-exchange is calculated from the ratio $CEC_{Meglu\text{-}clay}$ / $CEC_{Meglu\text{-}max}$ and is listed in Table S2.

Tab. S2: CHN-analysis of Meglumine-clays.

	Verm	Hec	LCR1	LCR2	Hec_{pri}
wt% C	8.769	11.580	7.598	5.809	7.470
$CEC_{Meglu\text{-}clay}$ [mmol/100g]	138	104	90	69	89
$CEC_{Meglu\text{-}max}$ [mmol/100g]	140	105	87	66	97
deviation	-2%	-1%	+4%	+5%	-8%

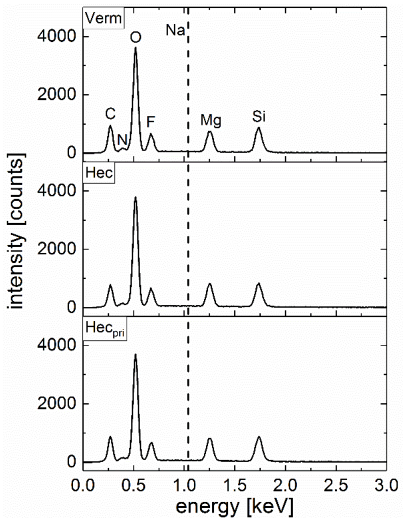

Fig. S3. The EDX-measurements of the Na-clays (Verm, Hec and Hec_{pri}) do not show remaining Na (position of Na-K_{α} shown by dashed line) in dried Meglumine-clay (separation of Na-containing side phases like Protoamphibole was carried out by sedimentation for the case of delaminated Hec_{pri} prior to sample preparation).

Volume Fraction of Gels.

First of all, the weight-fraction of the clay lamellae in the dry Meglumine-clay sample is calculated as follows:

$wt\%_{lamellae} = 1 - wt\%_{Meglumine}$

The weight fraction of Meglumine is determined by CHN-analysis.

The volume fraction ϕ is calculated using the density of the lamellae (2.7 gcm^{-3}) and water (1.0 gcm^{-3}):

$$\phi = \frac{wt\%_{lamellae} \cdot m_{clay} / \rho_{lamellae}}{wt\%_{lamellae} \cdot m_{clay} / \rho_{lamellae} + m_{water} / \rho_{water}}$$

The expected d-spacing is calculated using the thickness (t = 9.6 Å) of a clay lamella:

$d = t/\phi$

Hec_{pri}: Birefringence.

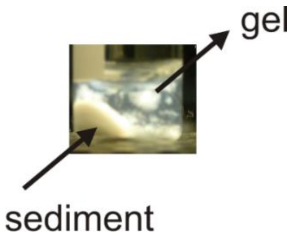

Fig. S4. Suspension of Hec_{Pri} between crossed polarizers. The supernatant gel (containing < 5 wt% of the total solid content) forms a birefringent, lyotropic suspension between crossed polarizers.

Glucosamine-clays.

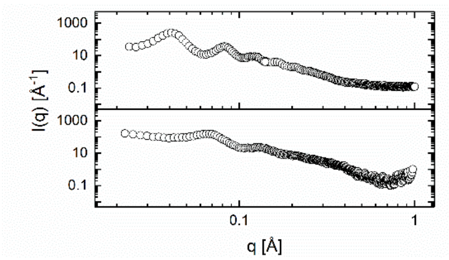

Fig. S5. SAXS-patterns of Glucosamine-clays: Verm (154 Å, 6.1 vol%, expected: 157 Å) and LCR2 (97 Å, 9.3 vol%, expected: 103 Å).

2. Supporting References

(1) Mermut, A. R.; Lagaly, G. Baseline Studies of the Clay Minerals Society Source Clays: Layer-charge Determination and Characteristics of those Minerals Containing 2:1 Layers. *Clays Clay Miner.* **2001,** *49*, 393-397.