

## Electronic Supplementary Information: One-step synthesis and AFM Imaging of hydrophobic LDH mono-layers

Gang Hu, Nan Wang, Dermot O'Hare\* and Jason Davis

### *Experimental*

Synthesis of LDHs:

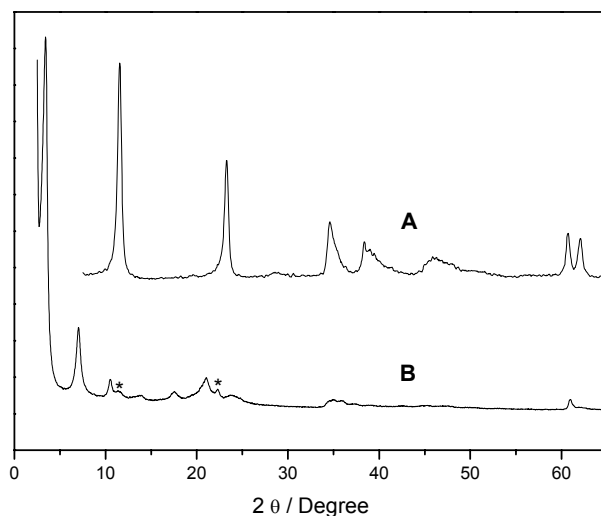
18.43g  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and 9.00g  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  were dissolved in 30ml de-ionized water to make Solution A. 6.97g  $\text{NaNO}_3$  and 4.8g  $\text{NaOH}$  were dissolved in 30 ml de-ionized water to make Solution B. A 4 M  $\text{NaOH}$  solution was also prepared to adjust the pH value of the reaction system. 1.73g Solution A was added dropwisely into 50ml isooctane containing 0.72g sodium dodecyl sulfate and 0.76g 1-butanol to make a clear system. 1.25g Solution B and 0.47g  $\text{NaOH}$  solution were similarly dispersed to make a clear reverse microemulsion system, which was added dropwisely into the former system afterwards. The mixture turned to be slightly translucent and became milky when heated at 75 °C in oil bath for 24 hours. All the above mentioned mixing and reaction processes were carried out with continuous magnetic stirring. Gel-like solids were separated by centrifugation and washed with acetone.

For comparison, pristine nitrate  $\text{Mg}_2\text{Al}$ -LDH was also synthesized using the same recipe through a co-precipitation method. The as-synthesized nitrate LDHs were mixed with NaDDS (1:3 in weight) in water and heated at 75 °C with stirring for 72 hrs to get DDS-intercalated  $\text{Mg}_2\text{Al}$ -LDH.

Characterisation:

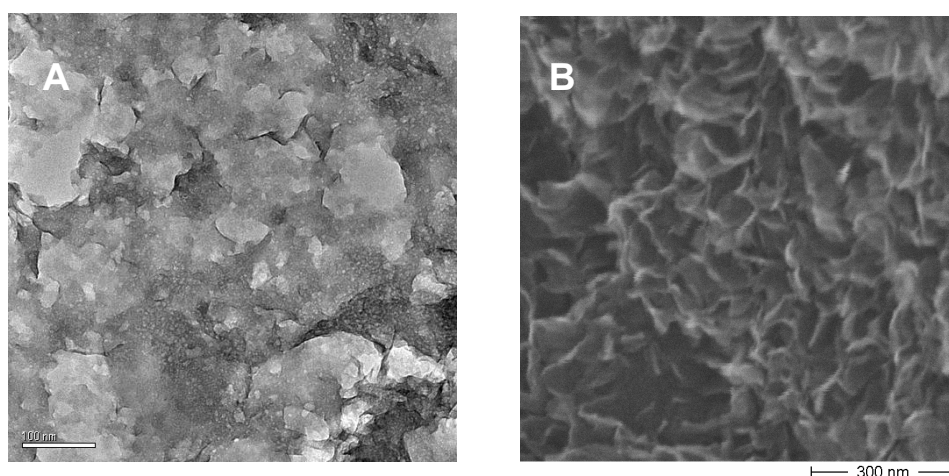
The samples were characterized by Inductively Coupled Plasma (ICP) atomic emission spectroscopy for elemental analysis, powder X-ray diffraction (XRD, Philips PANalytical X'pert pro diffractometer with  $\text{Cu}_{K\alpha}$  radiation,  $\lambda=1.5406$  nm, 40 kV, 40 mA), scanning electron microscopy (SEM, JSM840F with a field emission gun, samples were dispersed in ethanol and spread on aluminum tubs by natural drying in air and coated with platinum before test), transmission electron microscopy (TEM, JEOL 4000EX operating at 400kV, samples were dispersed in ethanol and loaded onto copper grids coated with Formvar), atomic force microscopy (AFM, Veeco Digital Instruments Multimode Nanoscope (IV) with 5611JV (100 $\mu\text{m}$ ) piezoelectric scanner, all data obtained by tapping mode at room temperature with a 30-50 % humidity). The AFM specimen was prepared as follows: Approximately 10 mg LDH was suspended in 200ml ethanol with sonication and deposited onto freshly cleaved Highly Oriented Pyrolytic Graphite (HOPG, 10\*10 \*3mm, Agar, G3389) by spin-coating (4000rpm, 20 s) to make a dry sample for further observation.

Z peizo calibration was carried out by imaging atomic steps on highly orientated pyrolytic graphite (HOPG), 5 nm gold nanoparticles and a 100 nm calibration grid (Mikromasch). Across this range measured height was within 9% of that expected.

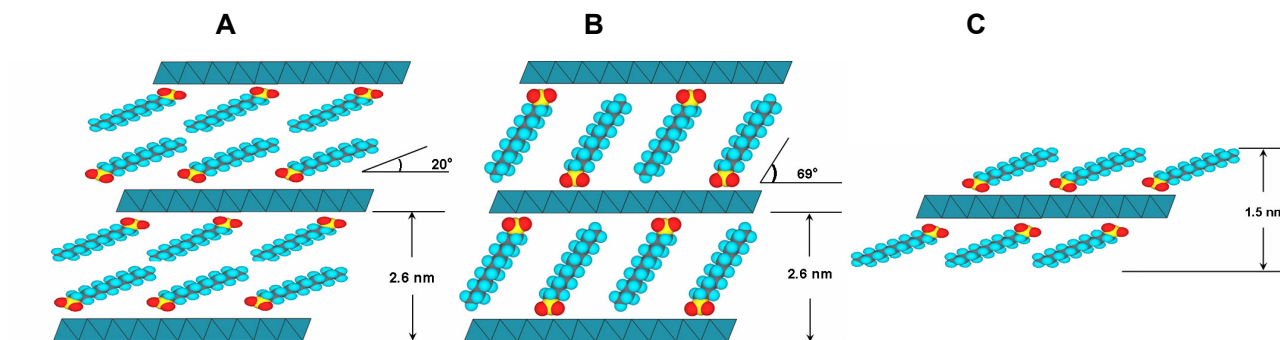


Sample A		Sample B	
2θ / degree	<i>hkl</i>	2θ / degree	<i>Hkl</i>
11.54	003	3.41	003
23.27	006	7.01	006
34.57	012	10.46	009
38.40	015	13.81	0012
45.85	018	17.51	0015
60.71	110	21.07	0018
62.04	113	23.80	0021
		34.62	012
		60.83	110

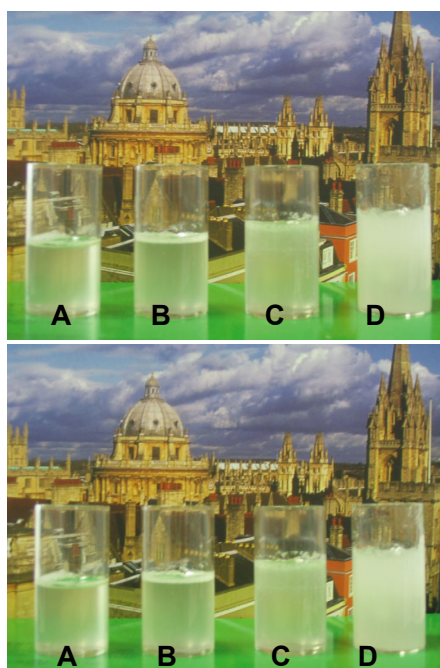
**Figure 1S.** Prestine  $\text{Mg}_2\text{Al}(\text{NO}_3)\text{-LDH}$  was synthesized with the same recipe in the Experimental section by a traditional co-precipitation method. This sample was mixed with sodium dodecyl sulfate in water with mixing at 75 °C for 72 hrs to get  $\text{Mg}_2\text{Al}(\text{C}_{12}\text{H}_{25}\text{SO}_4)\text{-LDH}$ . The figure above shows the XRD patterns of the two samples and the peaks have been indexed with a rhombohedral unit cell with  $a = b = 2d_{110} = 3.04 \text{ \AA}$  and  $c = 3d_{003} = 77.88 \text{ \AA}$  for DDS form and  $7.80 \text{ \AA}$  for  $\text{NO}_3^-$  form respectively. And the intercalated  $\text{Mg}_2\text{Al}(\text{C}_{12}\text{H}_{25}\text{SO}_4)\text{-LDH}$  is more crystalline than the one prepared in the reverse microemulsions and has a better resolved XRD pattern. Two peaks marked with \* come from the remaining starting materials.



**Figure 2S.** TEM A) and SEM image B) of the platelets synthesized in the reverse microemulsion system. Severe aggregation occurred during sample preparation and no single platelet can be discerned.



**Figure 3S.** Structural models of DDS intercalated LDHs with A) a bilayer arrangement and B) an interpenetrating pseudo monolayer arrangement of the DDS chains between two adjacent layers. Model C) shows a mono-layer LDH surfacially modified with charge-balancing DDS groups.



**Figure 4S.** HEMA with loadings (wt%) of LDH nano-layers; (A) 0.5%; (B) 1.0%; (C) 3% and (D) 8%. The bottom picture shows the dispersions are still homogeneous after 5 hrs.