

ELECTRONIC SUPPLEMENTARY INFORMATION

Synthesis and Characterisation of Layered Double Hydroxide Dispersions in Organic Solvents

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1. General Details

1.1 Fourier transform infrared (FTIR) spectroscopy

FTIR spectra were recorded on a Biorad FTS-6000 FRIT Spectrometer equipped with a high performance DuraSamp1IR II diamond accessory of attenuated total reflection (ATR) mode in the range of 400 - 4000 cm^{-1} with 40 scans at 4 cm^{-1} resolution.

1.2 Powder X-ray diffraction (PXRD)

PXRD patterns were recorded on a PANalytical X'Pert Pro instrument in reflection mode with Cu K α radiation ($\lambda = 1.542 \text{ \AA}$). The accelerating voltage was set at 40 kV with 40 mA current at 0.01 $^\circ \text{ s}^{-1}$ from 3 to 70 $^\circ$ with a slit size of 1 $^\circ$. Powder samples were loaded on stainless steel sample holders.

1.3 Surface area analysis

Surface area analysis was carried out by Dr. Ashley Shepherd (University of Oxford). The gas adsorption isotherm for nitrogen adsorption onto the LDH surface was measured using a CE Instruments Sorptomatic 1990. The surface area was then calculated using the Brunauer–Emmett–Teller (BET) method. Before each measurement, LDH samples were first degassed overnight at 110 $^\circ\text{C}$.

1.4 Thermogravimetric analysis (TGA)

TGA was carried out using a Perkin Elmer TGA7 Thermogravimetric Analyser. Approximately 10 mg of sample was heated in a platinum pan in the furnace. Initially the temperature was held at 30 $^\circ\text{C}$ for 5 minutes and then was increased to 800 $^\circ\text{C}$ at a rate of 5 $^\circ\text{C}$ per minute. The sample was held at 800 $^\circ\text{C}$ for five minutes.

1.5 Transmission electron microscopy (TEM)

TEM images were acquired at Harwell Science and Innovation Campus. A small amount of solid LDH was dispersed in ethanol using sonication and then cast onto copper grids coated with Formvar film. The samples were viewed using a JEOL 2100 microscope with an accelerating voltage of 200 kV.

1.6 Ultraviolet-visible (UV-Vis) spectroscopy

Measurements were taken on a Cary Series UV-Vis Spectrometer. The dispersion was shaken and a small amount pipetted into a quartz cuvette. Three measurements were taken: of the sample immediately after it was pipetted into the cuvette, and of the upper and lower parts of the dispersion once it had settled. The wavelength range used was from 800 to 200 nm.

2. Observations for dispersions

Table S1.

| Washing Solvent | Dispersion Solvent | Time take to phase separate | Lower layer description |
|-----------------|--------------------|-----------------------------|--|
| 2-propanol | Ethyl benzene | 24 hours | Translucent. |
| Acetone | Ethyl benzene | 1 hour | White solid. |
| Acetonitrile | Ethyl benzene | 1 hour | Pearlescent, thick. |
| Dioxane | Ethyl benzene | 1 minute | Translucent, pearlescent. |
| DMF | Ethyl benzene | 24 hours | Translucent, pearlescent. |
| DMSO | Ethyl benzene | 20 seconds | White solid. |
| Ethanol | Ethyl benzene | 1 minute | White solid. |
| EA | Ethyl benzene | 1 hour | White solid. |
| EG | Ethyl benzene | 1 minute | White solid. |
| Methanol | Ethyl benzene | 6 hours | Pearlescent, but with some large gel-like aggregates. |
| THF | Ethyl benzene | 24 hours | Pearlescent, but with some large gel-like aggregates. |
| 1-propanol | <i>o</i> -xylene | 1 hour | Thick, pearlescent, translucent, almost opaque. |
| 2-propanol | <i>o</i> -xylene | 1 minute | Thick, pearlescent, translucent, almost opaque. Some aggregates. |
| Acetonitrile | <i>o</i> -xylene | 1 hour | Translucent. |
| Dioxane | <i>o</i> -xylene | 24 hours | Translucent. |
| DMF | <i>o</i> -xylene | 10 seconds | White solid. |
| DMSO | <i>o</i> -xylene | 1 minute | White solid. |
| Ethanol | <i>o</i> -xylene | 1 minute | White solid. |
| EA | <i>o</i> -xylene | 6 hours | Translucent, pearlescent. |
| Methanol | <i>o</i> -xylene | 1 hour | Translucent. |
| THF | <i>o</i> -xylene | 1 hour | White solid. |
| 1-propanol | <i>p</i> -xylene | 1 hour | White solid. |
| 2-propanol | <i>p</i> -xylene | 1 hour | White solid. |

| Washing Solvent | Dispersion Solvent | Time take to phase separate | Lower layer description |
|-----------------|--------------------|-----------------------------|------------------------------------|
| Acetone | <i>p</i> -xylene | 6 hours | Translucent, pearlescent. |
| Dioxane | <i>p</i> -xylene | 1 hour | Translucent. |
| DMF | <i>p</i> -xylene | 24 hours | White solid. |
| DMSO | <i>p</i> -xylene | 1 hour | White solid. |
| Ethanol | <i>p</i> -xylene | 6 hours | Translucent. |
| EA | <i>p</i> -xylene | 1 minute | White solid. |
| EG | <i>p</i> -xylene | 10 seconds | White solid. |
| THF | <i>p</i> -xylene | 1 hour | White solid. |
| 1-propanol | Toluene | 1 hour | Translucent. |
| Acetone | Toluene | 24 hours | Translucent, pearlescent. |
| Acetonitrile | Toluene | 24 hours | Translucent, almost transparent. |
| Dioxane | Toluene | 6 hors | Translucent, with some aggregates. |
| DMF | Toluene | 1 hour | White solid. |
| DMSO | Toluene | 1 minute | White solid. |
| Ethanol | Toluene | 6 hours | Translucent. |
| EA | Toluene | 1 hour | White, gel like solid layer. |
| EG | Toluene | 1 minute | White solid. |
| Methanol | Toluene | 1 hour | Translucent, pearlescent. |
| THF | Toluene | 1 hour | White, gel like solid layer. |

3. Characterisation

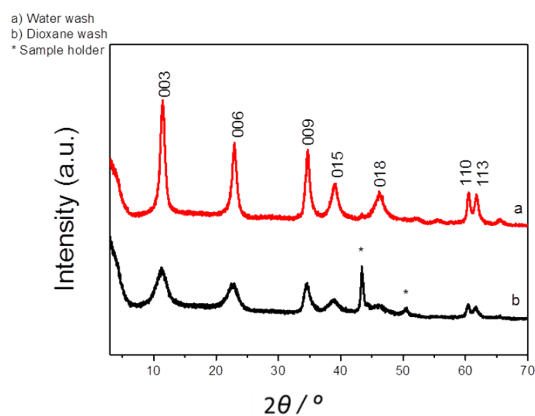


Fig. S1 Powder X-ray diffraction patterns of dioxane-washed and water-washed MgAlCO₃-LDH.

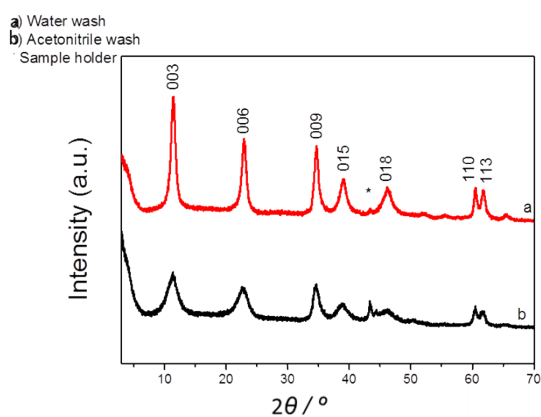


Fig. S2 Powder X-ray diffraction patterns of acetonitrile-washed and water-washed MgAlCO₃-LDH.

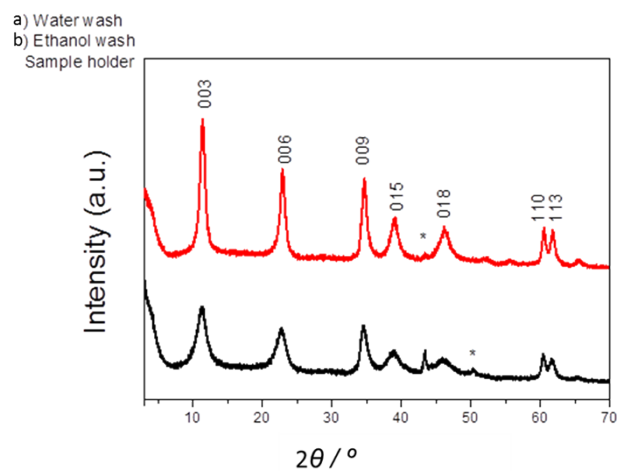


Fig. S3 Powder X-ray diffraction patterns of ethanol-washed and water-washed MgAlCO₃-LDH.

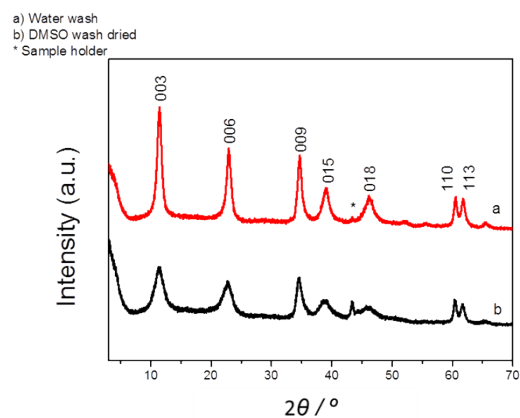


Fig. S4 Powder X-ray diffraction patterns of DMSO-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

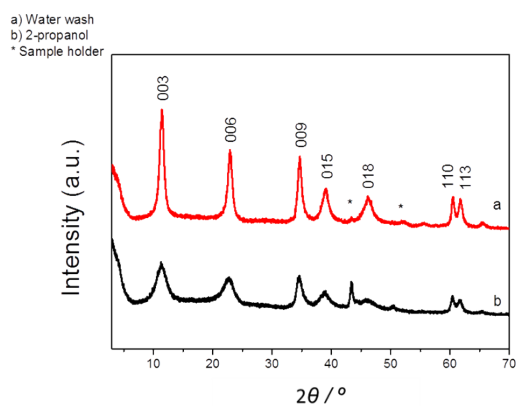


Fig. S5 Powder X-ray diffraction patterns of 2-propanol-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

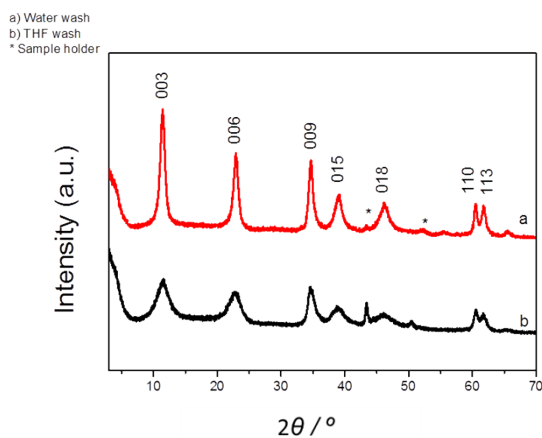


Fig. S6 Powder X-ray diffraction patterns of THF-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

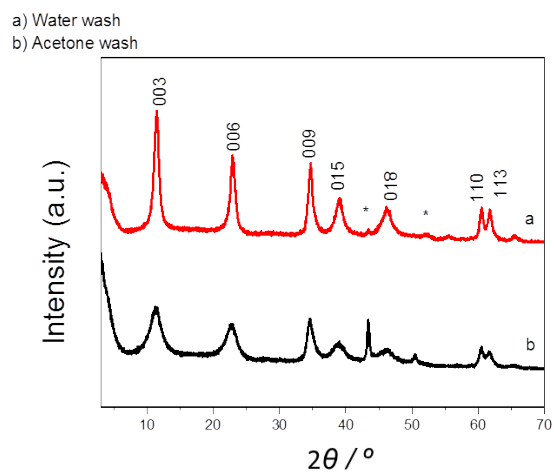


Fig. S7 Powder X-ray diffraction patterns of acetone-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

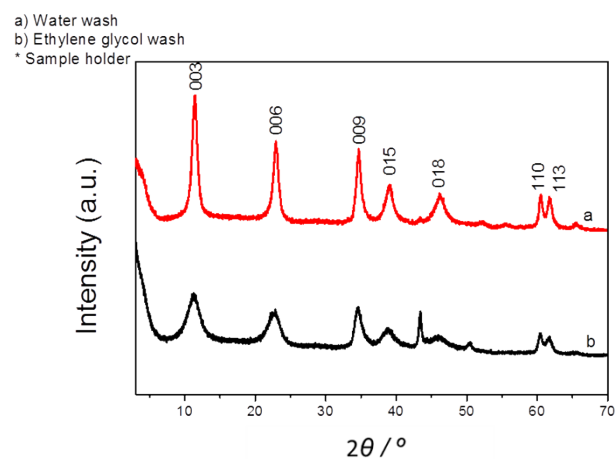


Fig. S8 Powder X-ray diffraction patterns of ethylene glycol-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

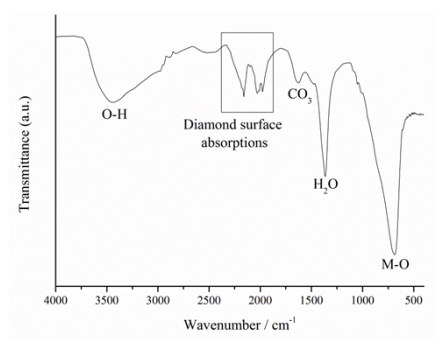


Fig. S9 FTIR spectrum of ethanol-washed $\text{MgAlCO}_3\text{-LDH}$.

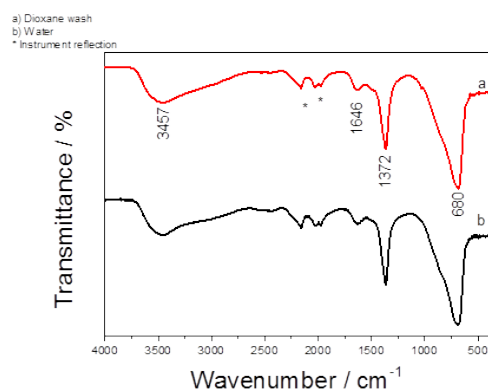


Fig. S10 FTIR spectrum of dioxane-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

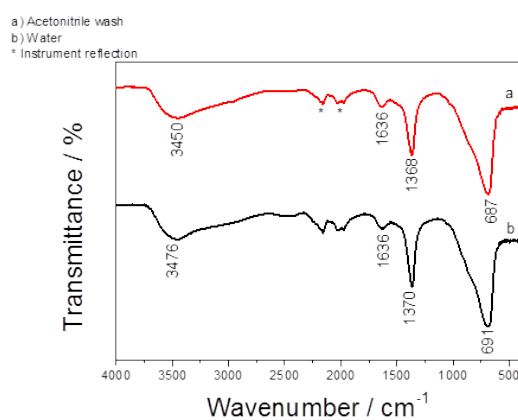


Fig. S11 FTIR spectrum of acetonitrile-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

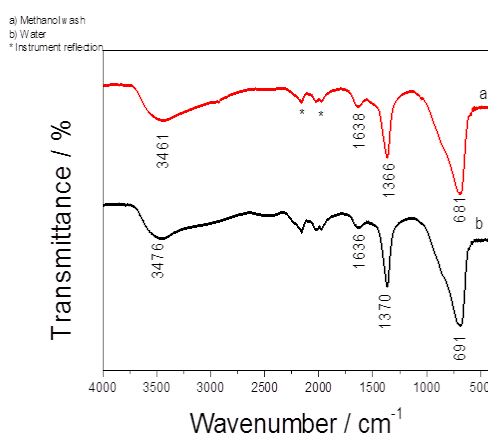


Fig. S12 FTIR spectrum of methanol-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

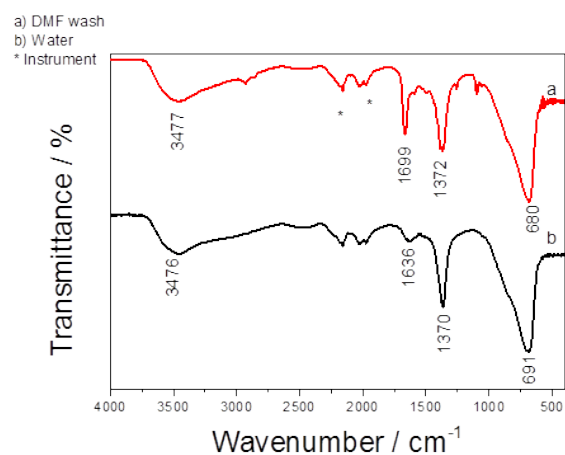


Fig. S13 FTIR spectrum of DMF-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

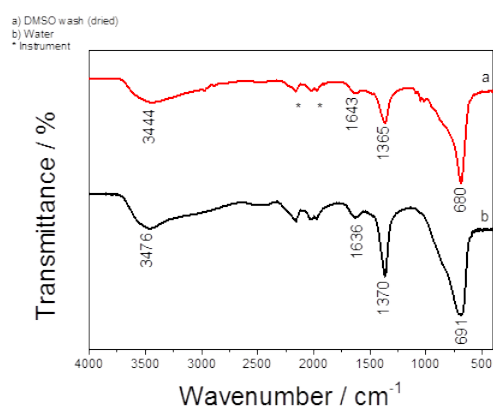


Fig. S14 FTIR spectrum of DMSO-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

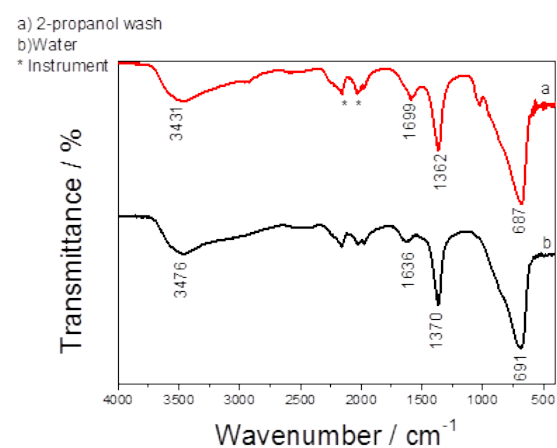


Fig. S15 FTIR spectrum of 2-propanol-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

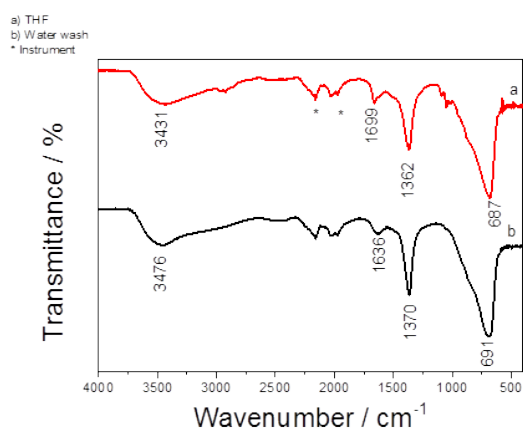


Fig. S16 FTIR spectrum of THF-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

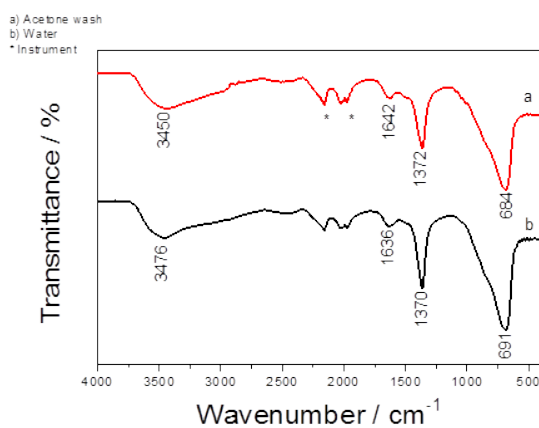


Fig. S17 FTIR spectrum of acetone-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$

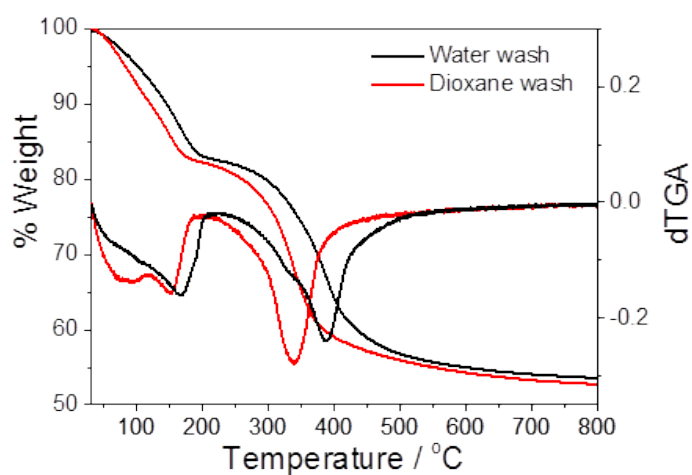


Fig. S18 TGA and dTGA curves for dioxane-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

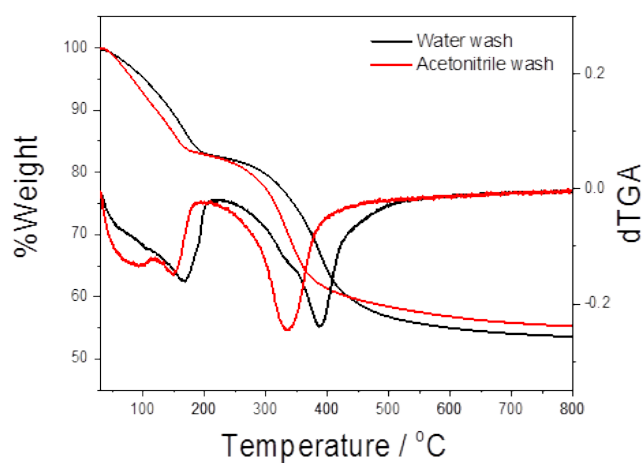


Fig. S19 TGA and dTGA curves for acetonitrile-washed and water-washed MgAlCO₃-LDH.

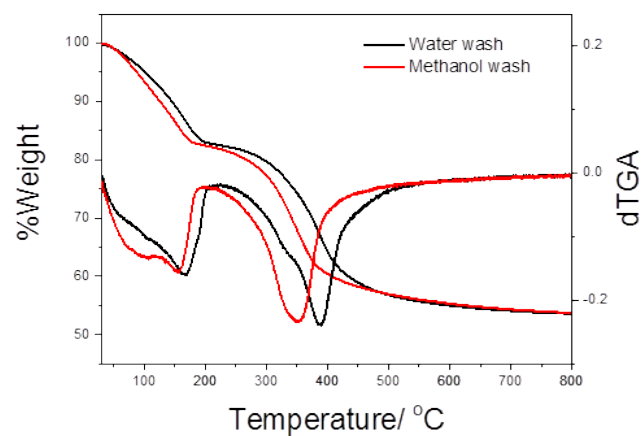


Fig. S20 TGA and dTGA curves for methanol-washed and water-washed MgAlCO₃-LDH.

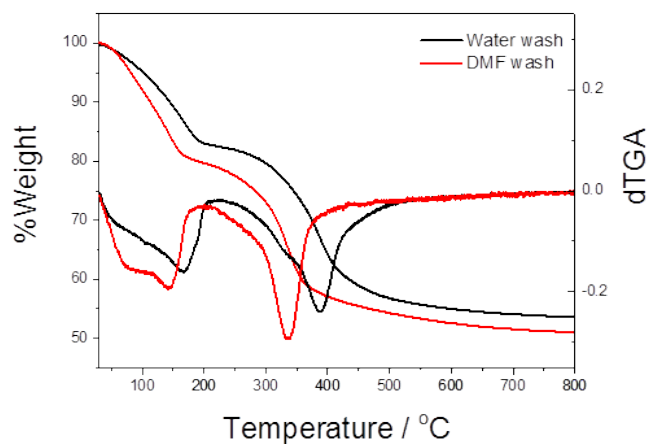


Fig. S21 TGA and dTGA curves for DMF-washed and water-washed MgAlCO₃-LDH.

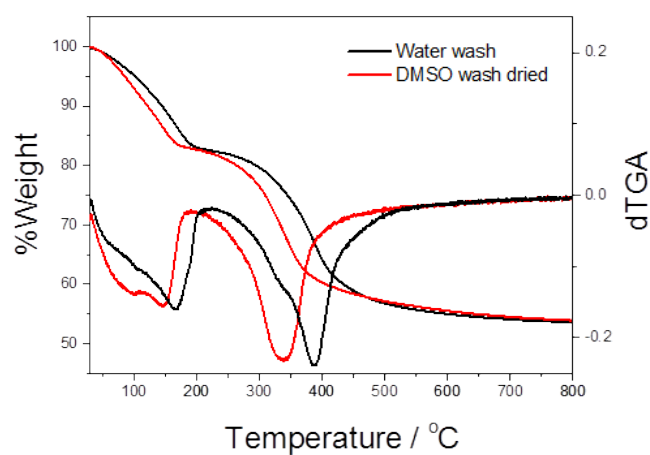


Fig. S22 TGA and dTGA curves for DMSO-washed and water-washed MgAlCO₃-LDH.

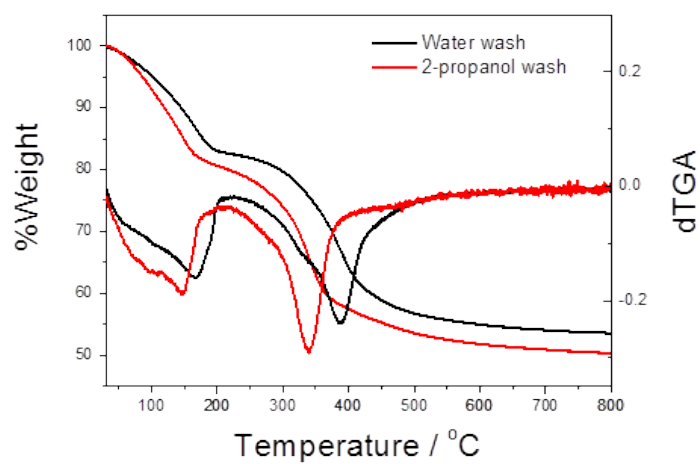


Fig. S23 TGA and dTGA curves for 2-propanol-washed and water-washed MgAlCO₃-LDH.

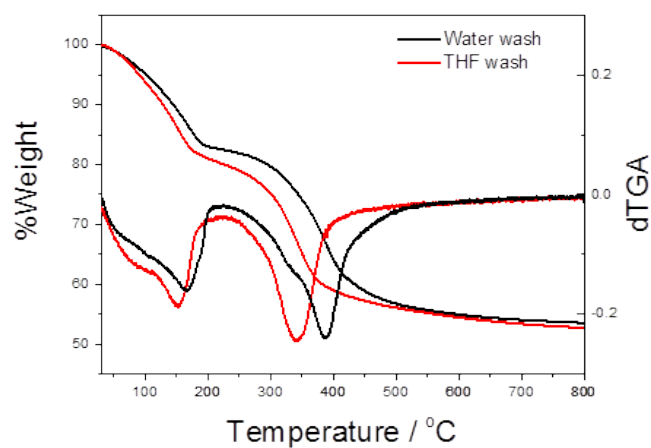


Fig. S24 TGA and dTGA curves for THF-washed and water-washed MgAlCO₃-LDH.

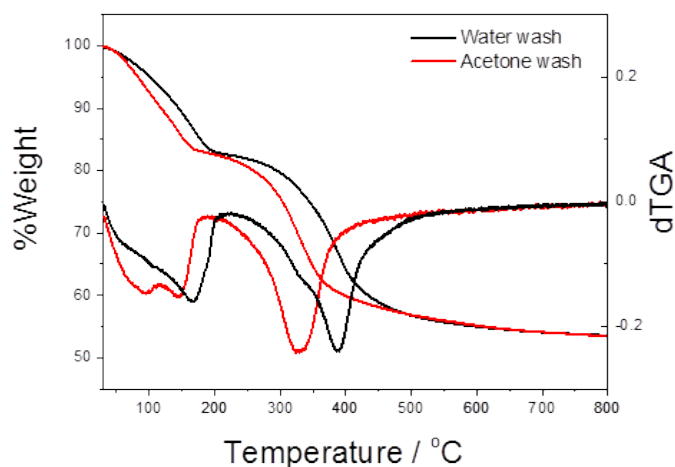


Fig. S25 TGA and dTGA curves for acetone-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

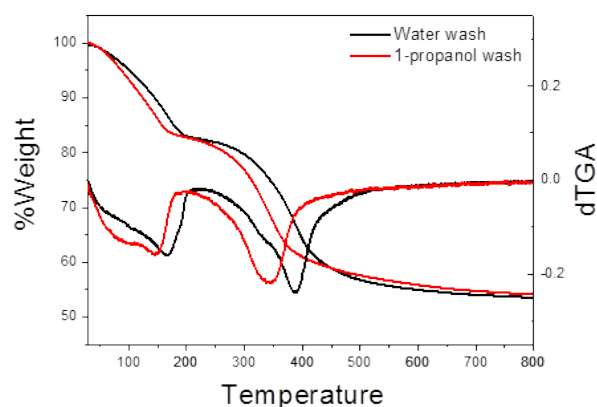


Fig. S26 TGA and dTGA curves for 1-propanol-washed and water-washed $\text{MgAlCO}_3\text{-LDH}$.

Table S2 Summary of the chemical formula of the AMO-LDHs using the thermal analysis data

| AMO Solvent | Formula of LDH |
|--------------|---|
| Water | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.78(\text{H}_2\text{O})$ |
| Dioxane | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.28(\text{H}_2\text{O}) \cdot 0.07(\text{Dioxane})$ |
| Acetonitrile | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.27(\text{H}_2\text{O}) \cdot 0.15(\text{Acetonitrile})$ |
| Methanol | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.29(\text{H}_2\text{O}) \cdot 0.20(\text{Methanol})$ |
| Ethanol | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.30(\text{H}_2\text{O}) \cdot 0.13(\text{Ethanol})$ |
| DMF | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.41(\text{H}_2\text{O}) \cdot 0.09(\text{DMF})$ |
| DMSO | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.32(\text{H}_2\text{O}) \cdot 0.06(\text{DMSO})$ |
| 2-propanol | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.40(\text{H}_2\text{O}) \cdot 0.07(2\text{-propanol})$ |
| THF | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.43(\text{H}_2\text{O}) \cdot 0.06(\text{THF})$ |
| 1-propanol | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.31(\text{H}_2\text{O}) \cdot 0.06(1\text{-propanol})$ |
| Acetone | $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_2(\text{CO}_3)_{0.125} \cdot 0.26(\text{H}_2\text{O}) \cdot 0.08(\text{Acetone})$ |

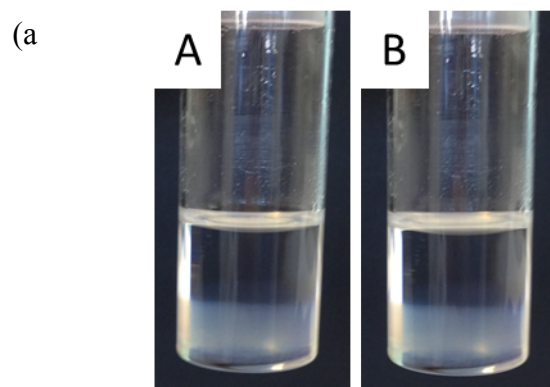


Fig. S27 Photographs of Time Dependence Study after (a) 1 hour and (b) 24 hours.

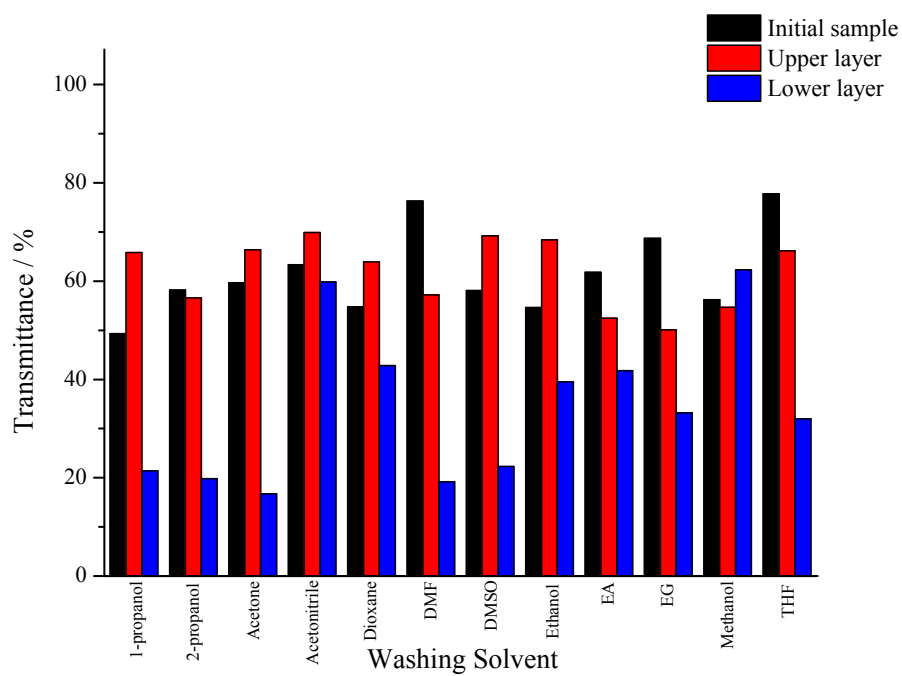


Fig. S28 T% measured for MgAlCO₃-LDH dispersed in *p*-xylene.

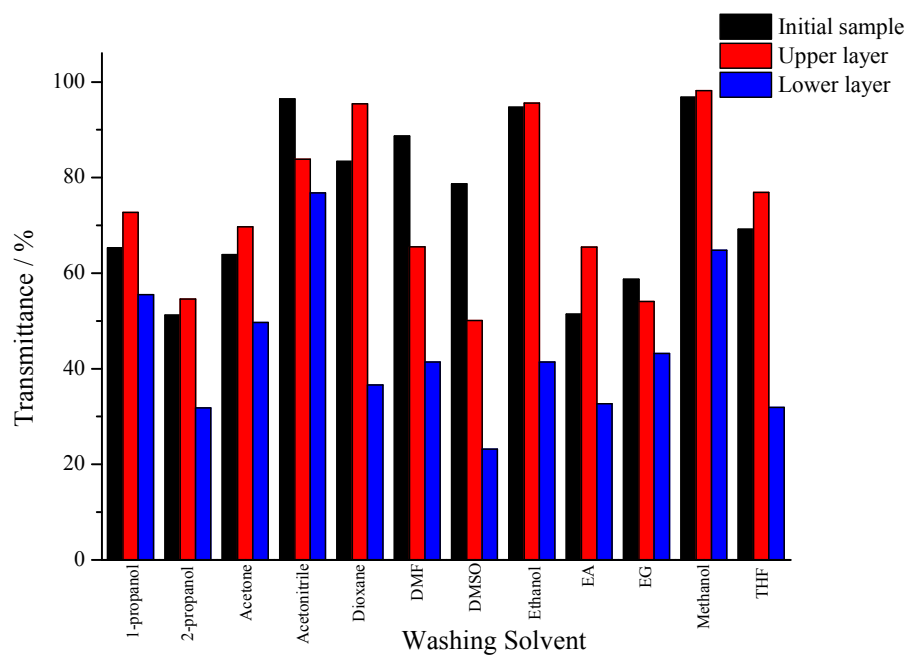


Fig. S29 T% measured for MgAlCO₃-LDH dispersed in toluene.

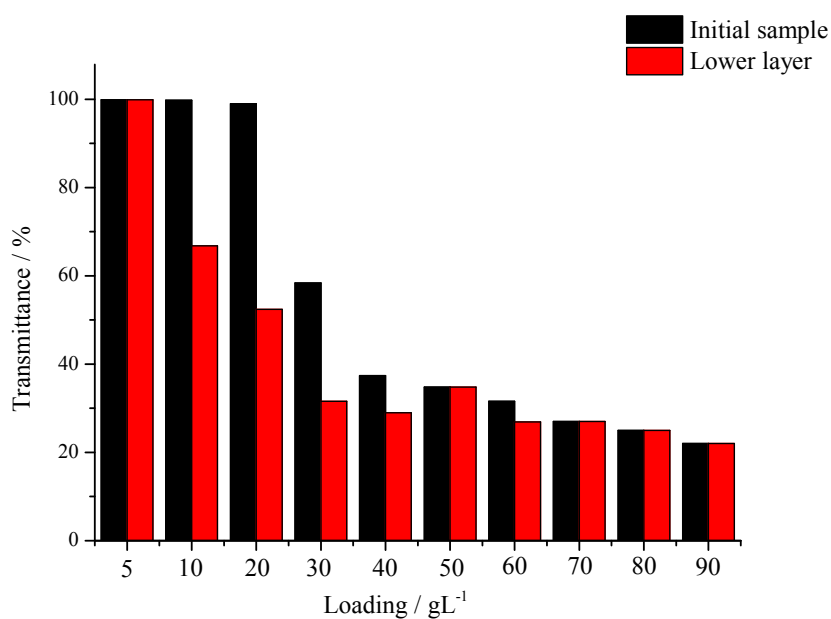


Fig. S30 Bar chart showing effect of increasing loading for methanol-washed LDH dispersed in *o*-xylene

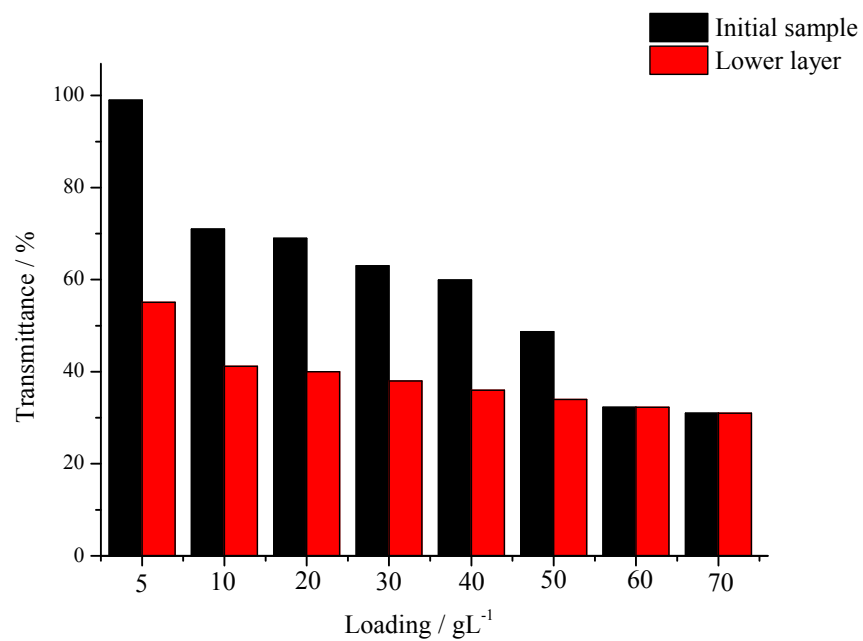


Fig. S31 Bar chart showing effect of increasing loading for methanol-washed LDH dispersed in *p*-xylene