

SUPPLEMENTARY DATA

Silica@Layered Double Hydroxide Core-shell Hybrid Materials

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1. General details

The specific surface area of the samples was determined by the Brunauer-Emmett-Teller (BET) method using the N₂ adsorption and desorption isotherms collected at 77 K on a Micromeritics TriStar II 3030 instrument. The samples were degassed at 110 °C overnight before analysis.

Fourier transform infrared (FTIR) spectra were collected on a ThermoScientific Nicolet iS5 spectrometer fitted with an iD3 ATR (attenuated total reflection) accessory. The instrument was set to record 40 scans between 4000 – 700 cm⁻¹ at 4 cm⁻¹ resolution.

Powder X-ray diffraction data were collected on a PANAnalytical X'Pert Pro Diffractometer in scanning mode using Cu K α radiation ($\alpha_1 = 1.540598 \text{ \AA}$, $\alpha_2 = 1.54426 \text{ \AA}$, weighted average = 1.541847 \AA). Scans were recorded between $2\theta = 3.0 - 80^\circ$, with step size 0.017° and scan step time 26.67 s. The samples were mounted on stainless steel holders that gave rise to peaks at 43.36, 44.29, and 50.51° (these did not interfere with the analysis).

Scanning electron microscopy (SEM) was performed at the Harwell Research Complex on a JSM-6610LV microscope with an accelerating voltage of 200 kV. Samples were prepared by dispersion in ethanol using sonication, cast onto carbon tabs and sputter coated with 10 nm Au using a Quorum Q150T ES.

Thermogravimetric analyses (TGA) was carried out on a Mettler Toledo TGA/DSC 1 system. Samples (10 mg) were heated in an alumina crucible from 25 to 800 °C (at 5°C min^{-1}) under a stream of N₂ flowing at $100 \text{ cm}^3 \text{ min}^{-1}$.

Transmission electron spectroscopy (TEM) images were acquired at the Harwell Research Complex on a JEOL 2100 microscope with an accelerating voltage of 200 kV. Samples were prepared by dispersion in ethanol using sonication and cast onto copper grids coated with carbon mesh.

Solid state ²⁷Al DPMAS solid-state nuclear magnetic resonance (NMR) spectra were collected by Dr Nicholas Rees (University of Oxford) at 104.2 MHz (9.4 T) on a Bruker Avance III HD spectrometer. In order to obtain quantitative MAS spectra, a single pulse excitation was applied using a short pulse length (0.23 μs). 7000 scans were acquired with a 0.1 s delay and a MAS rate of 15 kHz using 4 mm O.D zirconia rotors. The ²⁷Al chemical shift is externally referenced to an aqueous solution of Al(NO₃)₃.

2. Supporting Figures

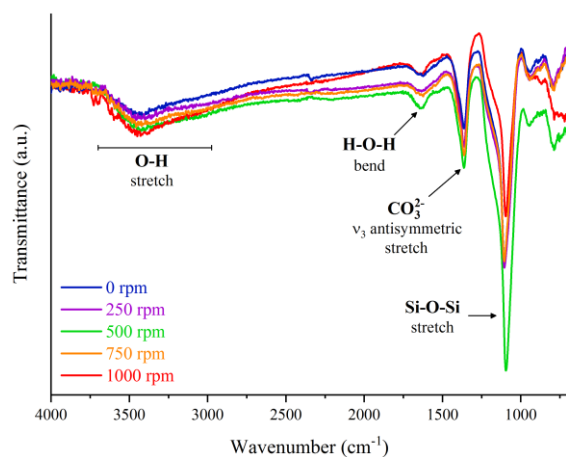


Fig. S1 FTIR spectra of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised with a metal addition of 0.43 mmol h^{-1} and different stirring speeds.

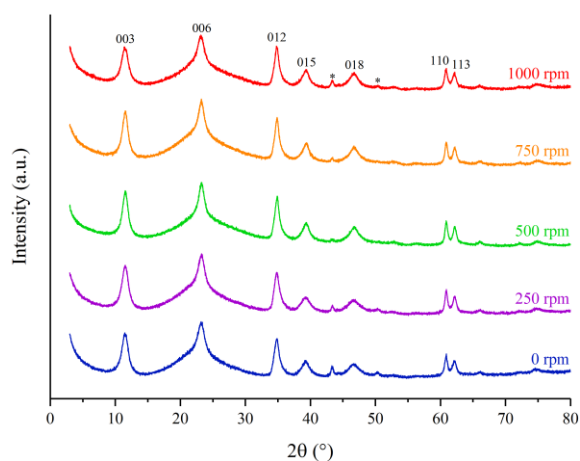


Fig. S2 Powder XRD diffractograms of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised with a metal addition rate of 0.43 mmol h^{-1} and different stirring speeds.

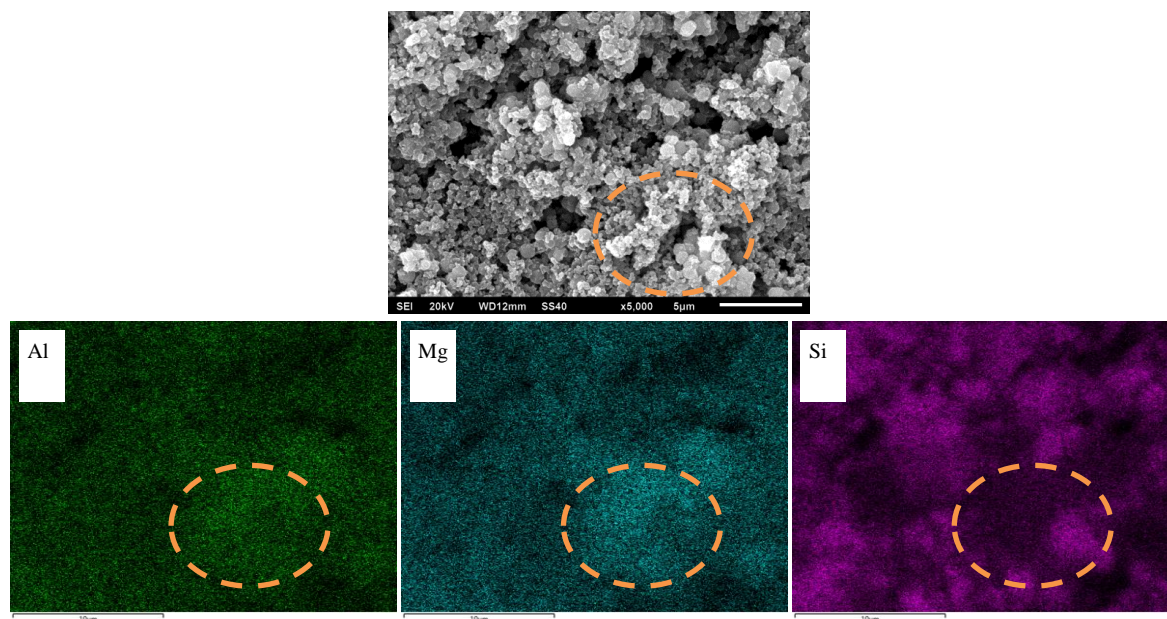


Fig. S3 SEM and SEM-EDS images of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised using a metal addition rate of 0.11 mmol h^{-1} and without stirring (e.g. 0 rpm).

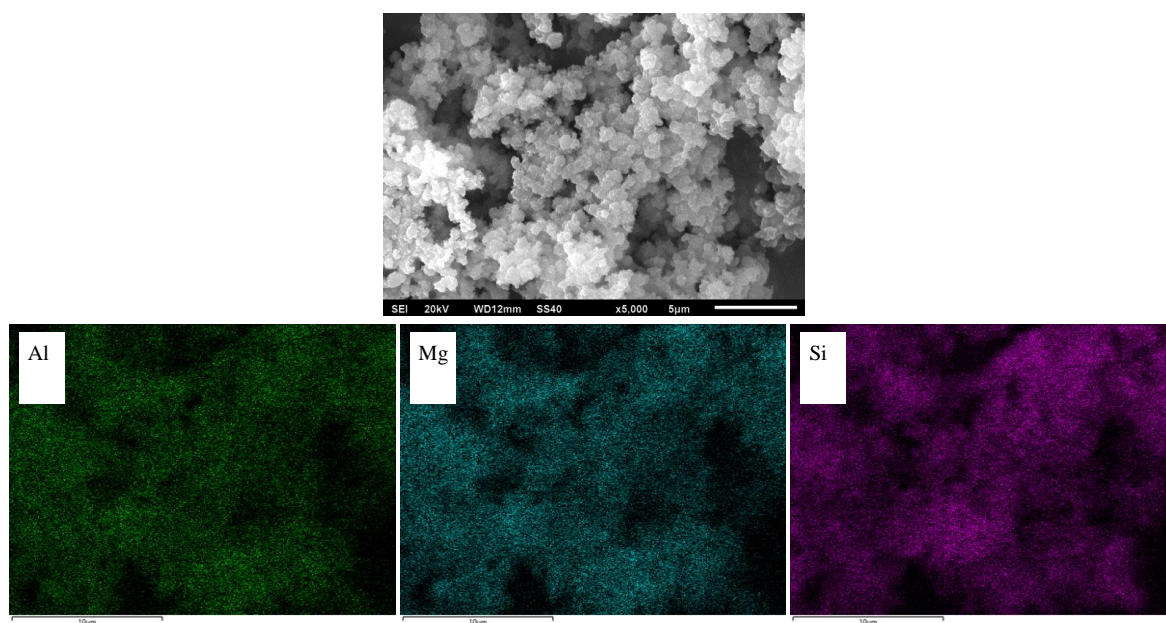


Fig. S4 SEM and SEM-EDS images of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised using a metal addition rate of 0.11 mmol h^{-1} and a stirring speed of 500 rpm.

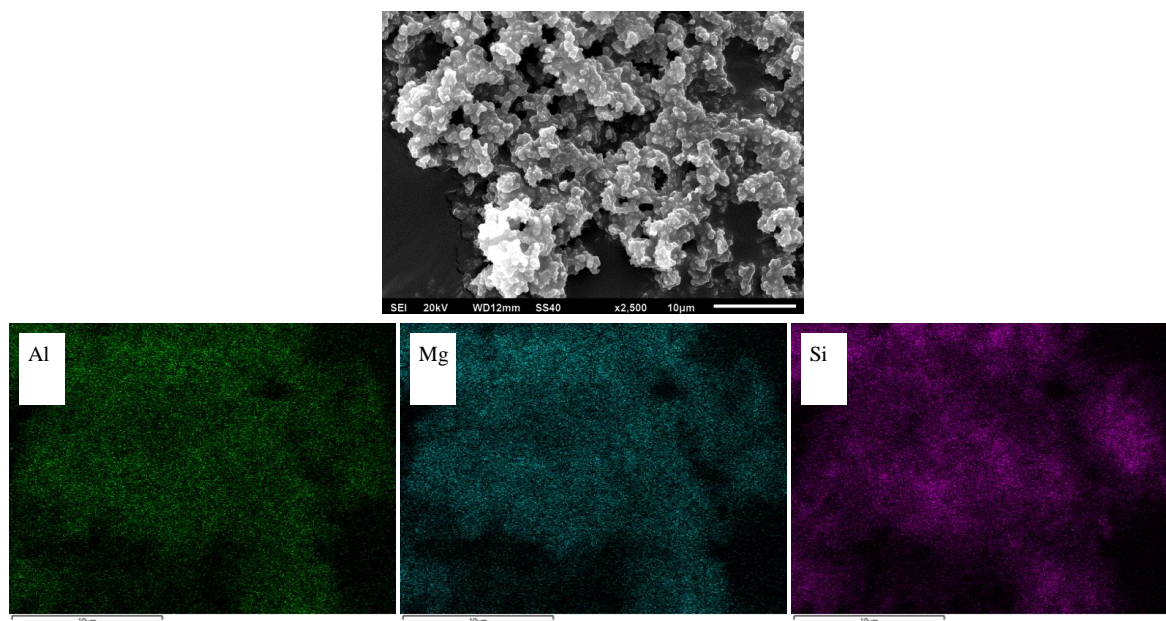


Fig. S5 SEM and SEM-EDS images of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised using a metal addition rate of 0.11 mmol h^{-1} and a stirring speed of 1000 rpm.

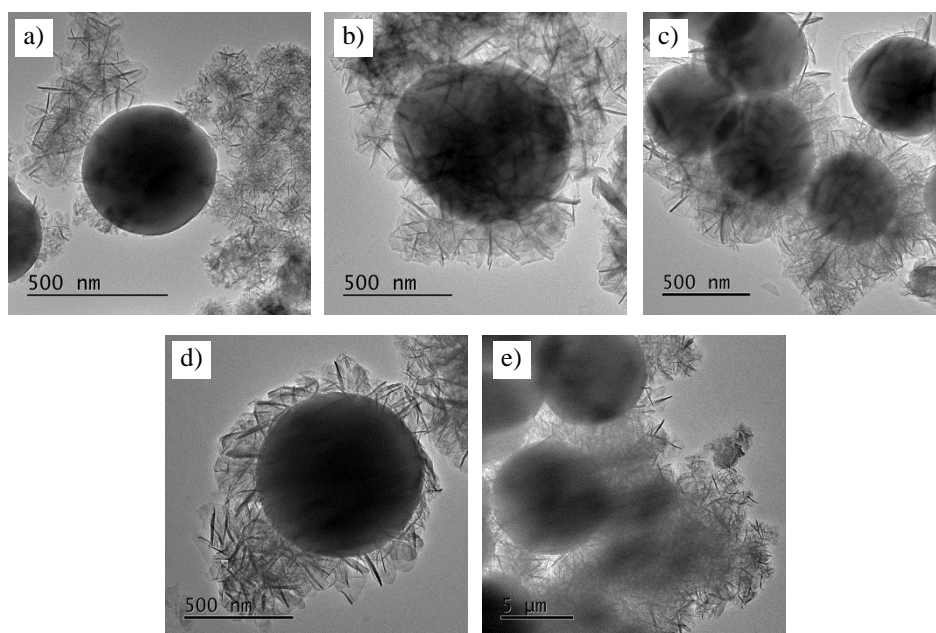


Fig. S6 TEM images of $\text{SiO}_2@\text{Mg}_2\text{Al-CO}_3\text{-AMO-LDHs}$ synthesised with metal addition rate 0.11 mmol h^{-1} at different stirring speeds: a) 0, b) 250, c) 500, d) 750, and e) 1000 rpm.

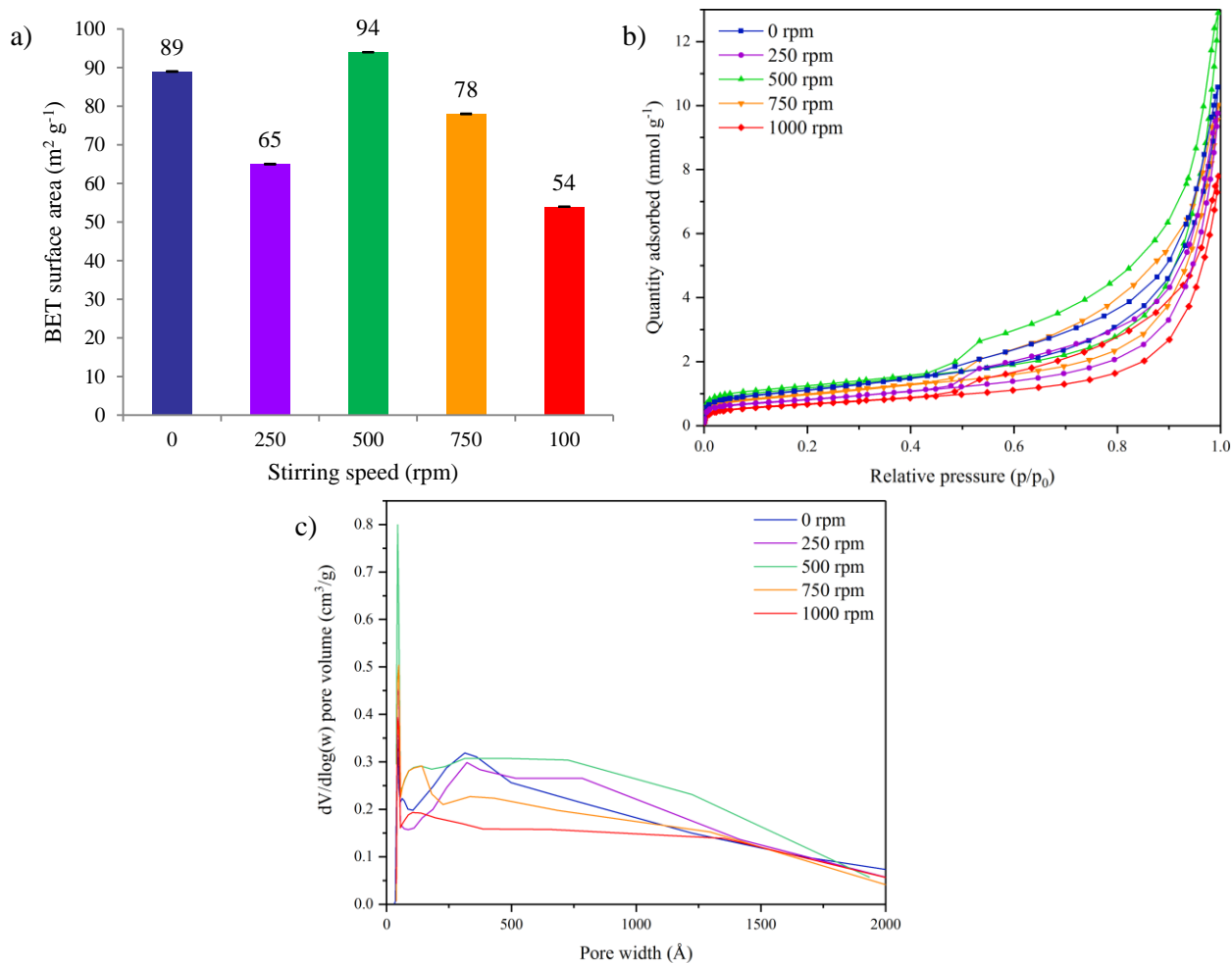


Fig. S7 a) Bar chart showing the BET areas of $\text{SiO}_2@Mg_2Al\text{-CO}_3\text{-AMO-LDHs}$ synthesised as stirring speeds between 0 – 1000 rpm at a fixed metal addition rate of 0.11 mmol h^{-1} , b) the corresponding N_2 sorption isotherms, and c) the corresponding BJH (desorption) pore size distributions.

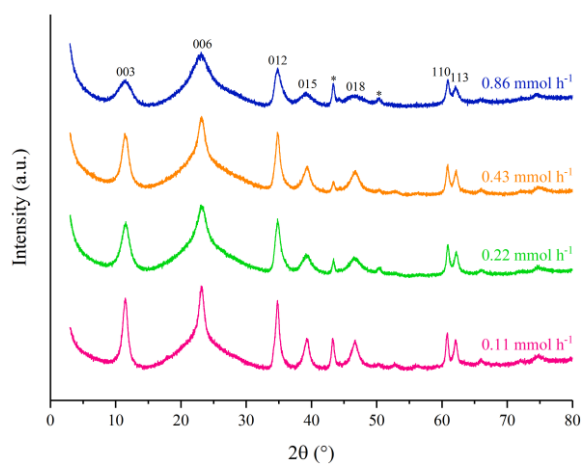


Fig. S8 Powder XRD diffractograms of $\text{SiO}_2@Mg_2Al\text{-CO}_3\text{-AMO-LDHs}$ synthesised with different metal addition rates and at a stirring speed of 1000 rpm.

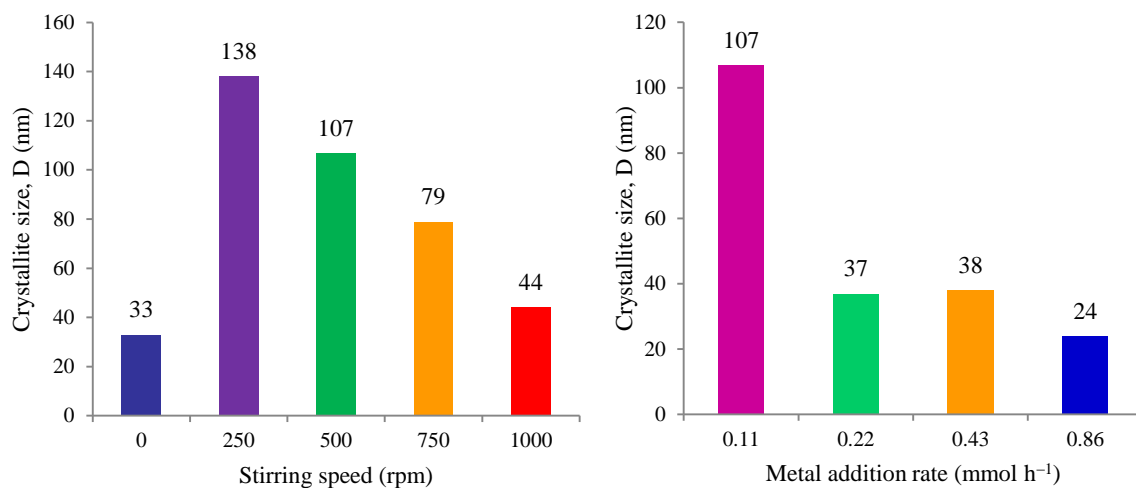


Fig. S9 Crystallite sizes, calculated by the Scherrer equation using the (110) reflection, as a function of a) stirring speed at a constant metal addition rate of 0.11 mmol h⁻¹ and b) metal addition rate at a constant stirring speed of 500 rpm.

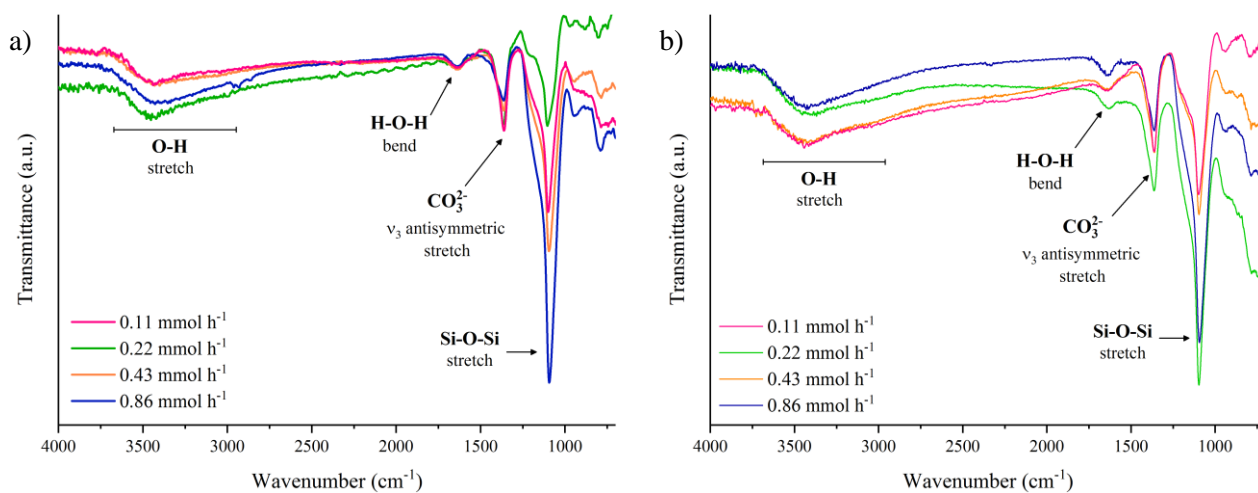


Fig. S10 FTIR spectra of SiO₂@Mg₂Al-CO₃-AMO-LDHs synthesised with different metal addition rates and at stirring speeds of a) 500 and b) 1000 rpm.

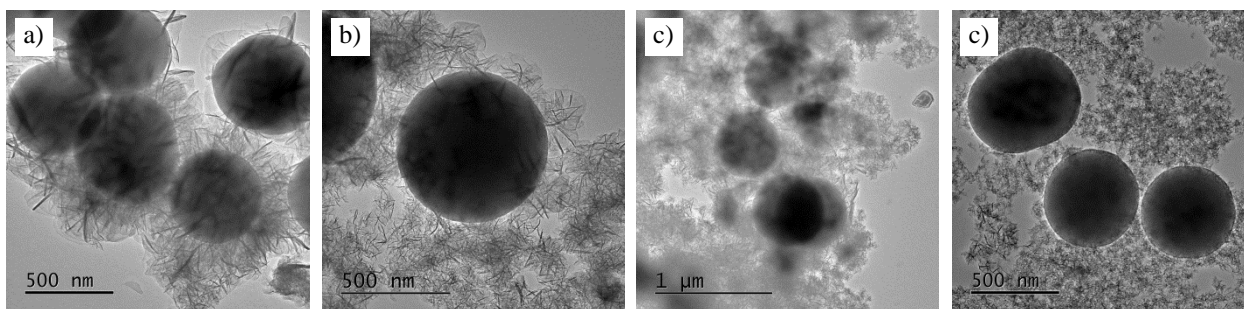


Fig. S11 TEM images of SiO₂@Mg₂Al-CO₃-AMO-LDHs synthesised using various metal addition rates: a) 0.11, b) 0.22, c) 0.43, and d) 0.84 mmol h⁻¹. The stirring speed was kept constant at 500 rpm.

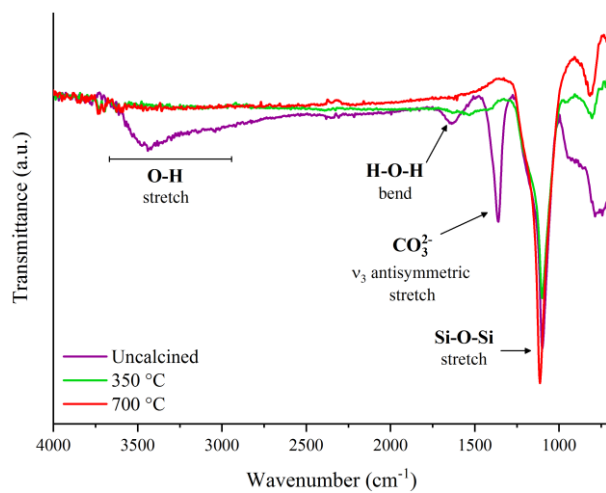


Fig. S12 FTIR spectra of $\text{SiO}_2@Mg_2Al\text{-CO}_3\text{-AMO-LDH}$, synthesised with a metal addition rate of 0.11 mmol h^{-1} and a stirring speed of 500 rpm, and its calcined products.

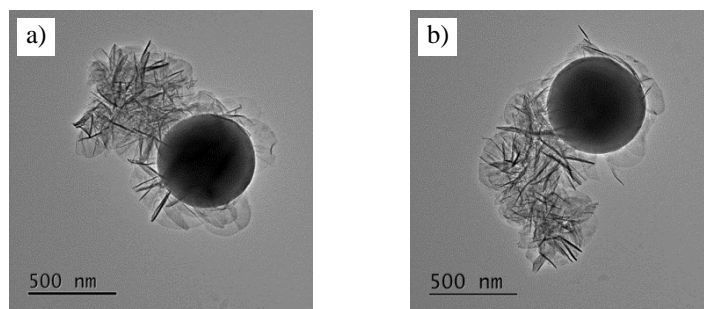


Fig. S13 TEM images of $\text{SiO}_2@Mg_2Al\text{-CO}_3\text{-AMO-LDH}$, synthesised with a metal addition rate of 0.11 mmol h^{-1} and a stirring speed of 500 rpm, calcined at a) 350 and b) 700 °C.

3. Supporting Tables

Table S1 Chemical formulae of the core-shell materials derived from TGA and dTGA.

Sample name ^a	Formula calculated
0.11-500	SiO ₂ @[Mg _{0.67} Al _{0.33} (OH) ₂ (CO ₃) _{0.17} · 0.30(H ₂ O) · 0.12(EtOH)] _{0.59}
0.86-500	SiO ₂ @[Mg _{0.67} Al _{0.33} (OH) ₂ (CO ₃) _{0.17} · 0.23(H ₂ O) · 0.16(EtOH)] _{0.63}
0.86-1000	SiO ₂ @[Mg _{0.67} Al _{0.33} (OH) ₂ (CO ₃) _{0.17} · 0.21(H ₂ O) · 0.18(EtOH)] _{0.63}

^aSamples are labelled with reference to the metal addition rate and stirring speed: e.g. "0.11-500" refers to a SiO₂@Mg₂Al-CO₃-AMO-LDH synthesised with a metal addition rate of 0.11 mmol h⁻¹ and stirring speed of 500 rpm.

Table S2 Crystallite sizes of SiO₂@Mg₂Al-CO₃-AMO-LDHs calculated by the Scherrer equation using the (110) Bragg reflections. The metal addition rate was 0.11 mmol h⁻¹.

Stirring speed (rpm)	2θ(°)	cos θ	FWHM (°)	d ₁₁₀ (Å)	D = 0.89λ/βcosθ (nm)
0	60.96	0.59	0.40	1.5	33
250	60.89	0.57	0.10	1.5	138
500	60.85	0.55	0.13	1.5	107
750	60.84	0.54	0.18	1.5	78
1000	60.81	0.53	0.33	1.5	44

λ = 1.541847 Å, β = FWHM in radians

Table S3 N₂ BET areas of SiO₂@Mg₂Al-CO₃-AMO-LDHs synthesised with a metal addition rate of 0.11 mmol h⁻¹.

Stirring speed (rpm)	BET area (m ² /g)	Error ^a
0	89	±0.09
250	65	±0.04
500	94	±0.06
750	78	±0.06
1000	54	±0.10

^aMathematical error associated with the fit of the data to the BET equation.

Table S4 Ratio of tetrahedral (T_d) and octahedral (O_h) aluminium species in SiO₂@Mg₂Al-CO₃-AMO-LDHs synthesised with metal addition rate 0.43 mmol h⁻¹ calculated from solid state ²⁷Al DPMAS NMR.

Stirring speed (rpm)	T _d Al/O _h Al ratio
500	1:8
750	1:11

Table S5 N₂ BET areas of SiO₂@Mg₂Al-CO₃-AMO-LDHs.

Metal addition rate (mmol h ⁻¹)	Stirring speed (rpm)	BET area (m ² /g)	Error ^a
0.11	500	94	±0.06
0.86	500	100	±0.22
0.11	1000	54	±0.10
0.43	1000	58	±0.16

^aMathematical error associated with the fit of the data to the BET equation.