Electronic Supplementary Information (ESI)

Dendritic Silica@AMO-LDH hybrids

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1. Experimental details

1.1 Synthesis of SiO_2@MgAl-LDH

1.1.1 Synthesis of silica nanoparticles

A silica sphere, with 500 nm particle size, was used as the core for the synthesis of AMO-SiO_2@MgAl-LDH. The detailed synthetic procedure followed the Stöber method[1]: 9.15 mL of tetraethyl orthosilicate (TEOS) was added to a mixed solution of 10 mL ammonia (35 wt%), deionised water and ethanol (30 and 50 mL, respectively). The white suspension was left stirring vigorously for 17 h. The final solid was washed with ethanol four times and then washed with water two times, followed by drying under vacuum overnight.

1.1.2 Synthesis of SiO_2@MgAl-LDH

The SiO_2@MgAl-LDH particles were synthesised by co-precipitation method; silica spheres (100 mg) were dispersed in deionised water (20 mL) using an ultrasound treatment, for 30 minutes. Then the anion salt Na_2CO_3 (0.96 mmol, Sigma-Aldrich) was added to the solution and further treated by ultrasound for 5 minutes; the final solution was named A. Then, the metal precursor solution (19.2 mL) containing $Mg(NO_3)_2 \bullet 6H_2O$ (1.08 mmol, Sigma-Aldrich) and $Al(NO_3)_3 \bullet 9H_2O$ (0.36 mmol, Sigma-Aldrich) was added in solution A at the rate of 60 mL/h with vigorous stirring. The pH of the reaction solution was controlled at pH 10 by dropwise of 1 M NaOH. After ageing for 60 minutes with stirring at room temperature, the obtained solid was collected after centrifugation (5000 rpm for 5 minutes) and then re-dispersed in deionised water (40 mL) and stir for 1 h, the washing was repeated twice. The final pH was around 7.

1.2 Synthesis of SiO_2@AMO-MgAl-LDH

AMO-method[2] was used on the SiO_2@MgAl-LDH. Before final isolation of SiO_2@MgAl-LDH, the solid was rinsed with acetone or ethanol and disperse in acetone or ethanol (40 mL) again, then left to stir for a certain amount of time. The suspension was centrifuged and dry in vacuum oven. Then, the AMO-SiO_2@MgAl-LDH was obtained as a fluffy white power.

2. Characterisation

The morphology of the samples described in this work was observed using Transmission Electron Microscopy (TEM) analysis performed on a JEOL 2100 microscope with an accelerating voltage of 200 kV.

Powder X-ray diffraction (XRD) data were collected on a PAN Analytical X'Pert Pro diffracto meter at 40 kV and 40 mA using Cu Kα radiation ($\alpha1 = 1.54057$ Å, $\alpha2 = 1.54433$ Å, weighted average = 1.54178 Å). The reflections at $2\theta = 43 - 44^\circ$ and 50° are produced by the XRD sample holder. Thermogravimetric analysis (TGA) measurements were collected using a Netzsch STA 409 PC instrument. The sample (10 - 20 mg) was heated in a corundum crucible between 30 and 500 °C at a heating rate of 5 °C/min under a flowing stream of nitrogen. Differential thermogravimetric analysis (DTG) is obtained from the 1st derivative of TGA data. Brunauer–Emmett–Teller (BET) specific surface areas were measured from the N_2 adsorption and desorption isotherms at 77 K collected from a Micromeritic TriStar. We can observe that all samples have shown a narrow peak at 3.8 nm which is a fake peak derived from the desorption data at P/P_0 around 0.42. When N_2 at 77 K, a steep region of desorption branch can be observed at $P/P_0 = 0.42$. This steep is contributed by adsorbate (N_2) but not the adsorbent. This kind of misinterpretation is due to the "tensile strength effect".[3] The solid state NMR spectroscopy (^{13}C and ^{27}Al) was recorded on a Varian Chemagnetics CMX Infinity 200 (4.7 T) by Dr. Nicholas H. Rees (University of Oxford). Samples were packed in 7.5 mm zirconia rotors. A double resonance MAS probe was used for all measurements and a MAS rate of 4 kHz for ^{13}C, whereas MAS rate of 6 kHz was used for ^{27}Al. ^{27}Al MAS NMR spectra were acquired with a single pulse excitation applied using a short pulse length (0.7 μs). Each spectrum resulted from 2000 scans separated by 1 s delay. The ^{27}Al chemical shifts are referenced to an aqueous solution of $Al(NO_3)_3$ ($\delta = 0$ ppm).

The bulk density and tap density of AMO-LDHs and C-LDHs were measured by both standard test method (ASTM D7481-09). The content of actual Mg and Al was measured by inductively coupled plasma (ICP-MS).

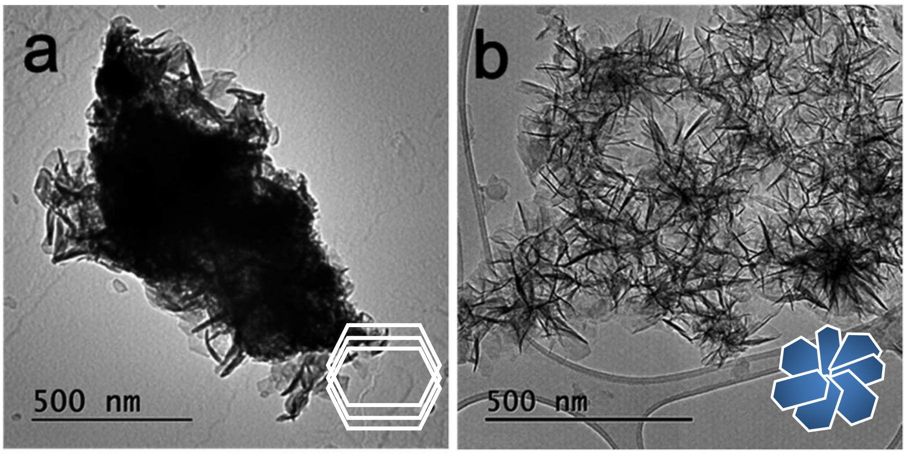

Fig. S1 TEM image of water washed Mg_3Al-CO_3-10 and AMO-Mg_3Al-CO_3-10.[2]

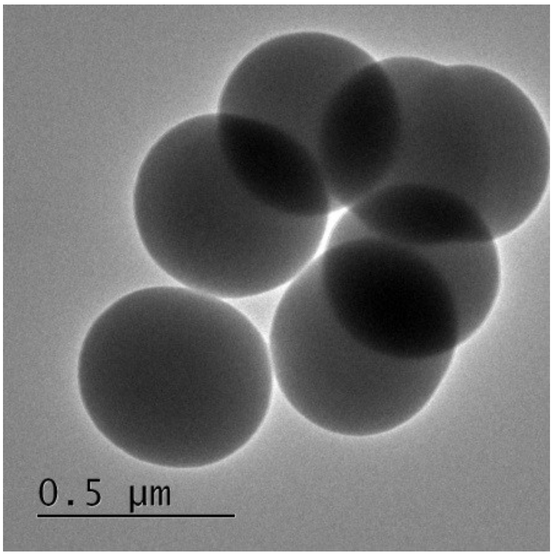

Fig. S2 TEM image of uniformed silica sphere with 500 nm diameter used as the core in this research.

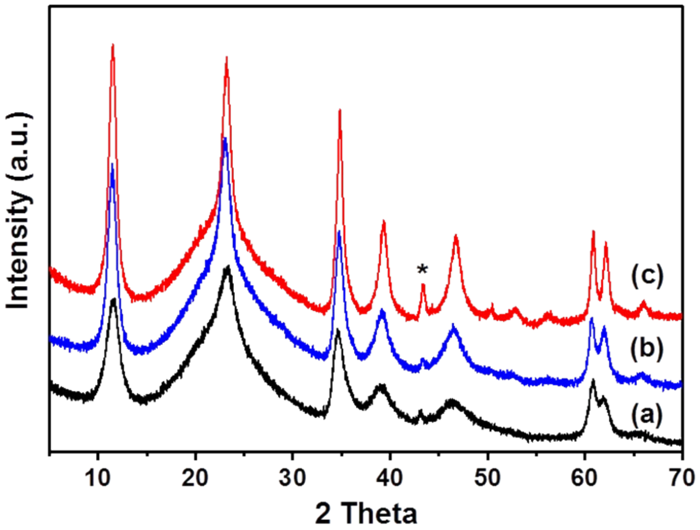

Fig. S3 XRD patterns of SiO_2@LDHs samples prepared with Mg:Al ratios (a) 2:1; (b) 3:1; (c) 4:1. (*) are Bragg reflections from the Al sample holder.

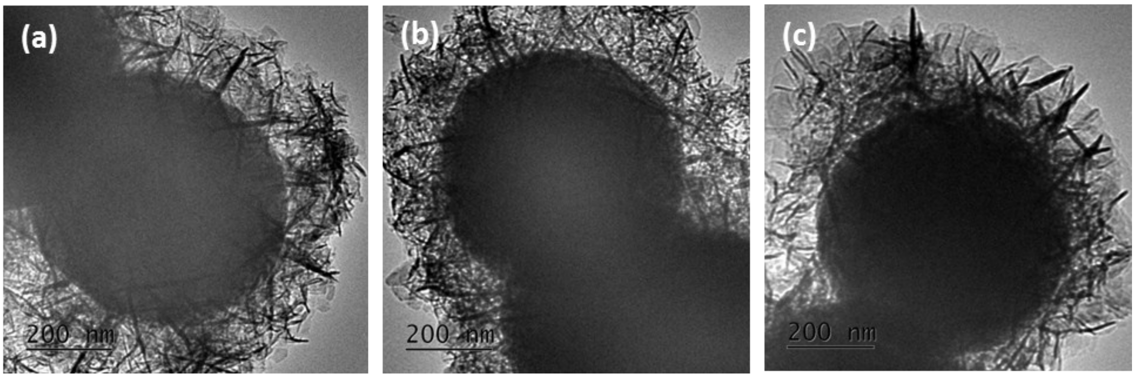

Fig. S4 TEM images of SiO_2@AMO-Mg_4Al-CO_3-LDH after acetone dispersion for 20 h a) before calcination; b) after calcination at 400 °C for 6 h and c) rehydration in H_2O/CO_3^{2-} solution for 10 h at room temperature.

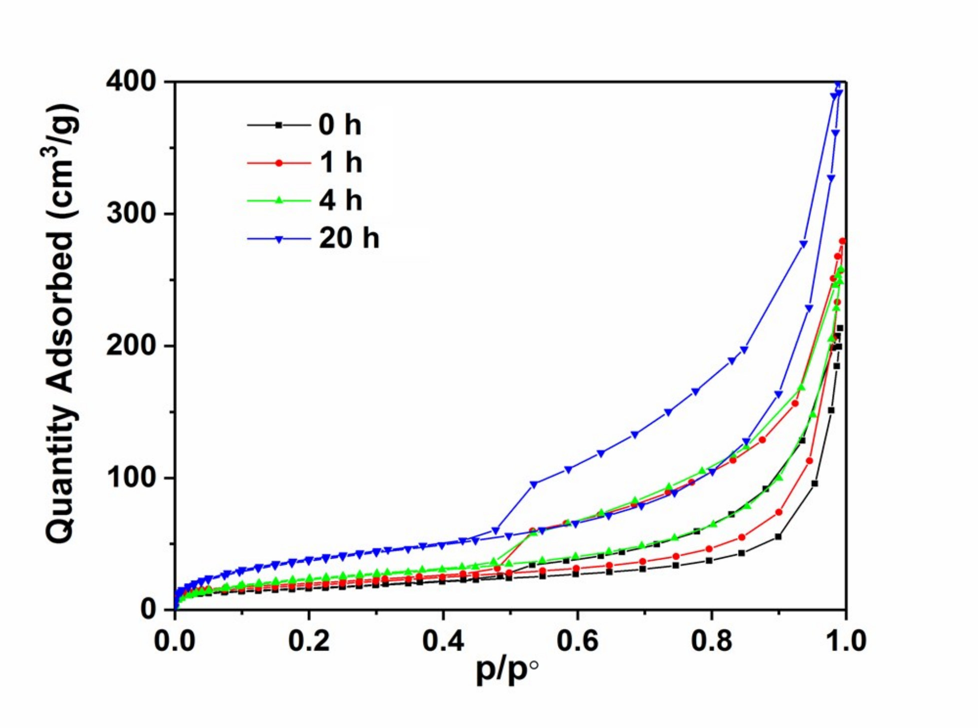

Fig. S5 N_2 adsorption and desorption isotherm of SiO_2@Mg_4Al-LDH posted treated with various acetone dispersion time.

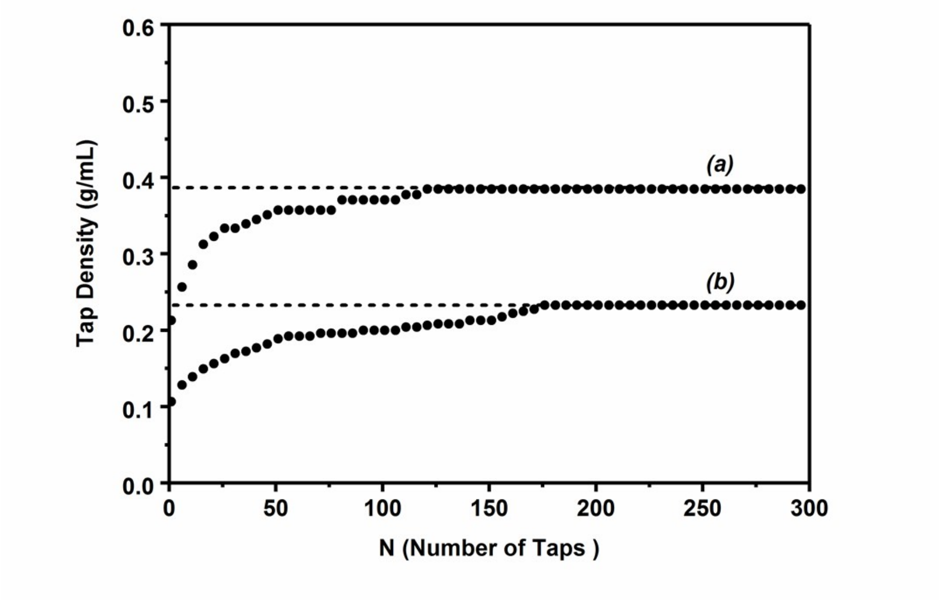

Fig. S6 Density measurement for (a) SiO_2@Mg_4Al-CO_3-LDH and (b) SiO_2@AMO-Mg_4Al-CO_3-LDH after AMOST post treatment for 20 h using acetone as the AMO-solvent. (1 g of samples after 300 taps; Tap density = m/v).

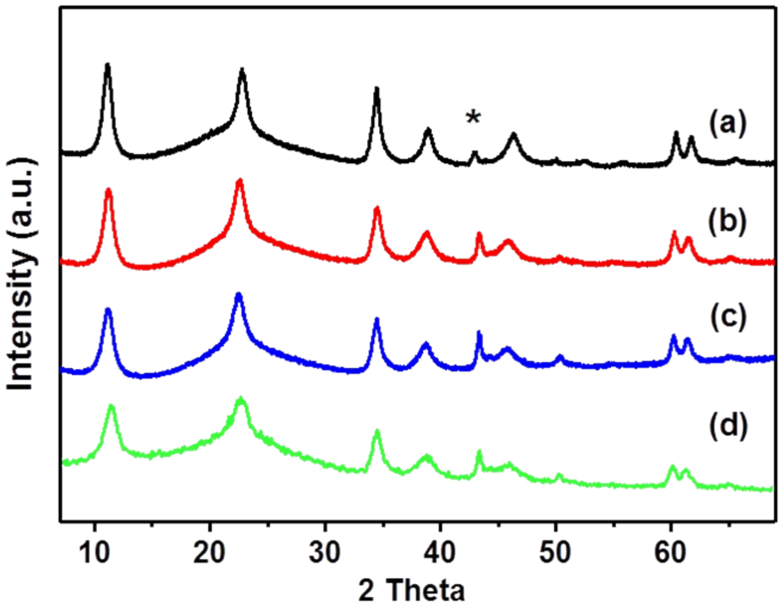

Fig. S7 XRD patterns for $SiO_2@Mg_4Al–CO_3$-10 with AMOST post treatment in acetone for (a) 0, (b) 1, (c) 4 and (d) 20 h. (*) are Bragg reflections from the Al sample holder.

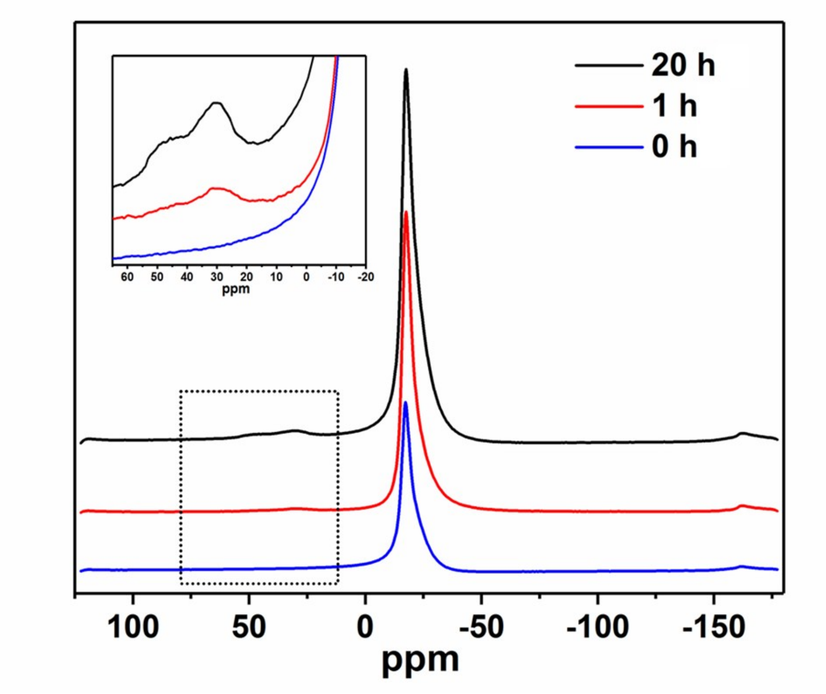

Fig. S8 Solid state ^{27}Al NMR of $SiO_2@Mg_4Al$-LDH with AMOST post treatment in acetone for (a) 0 h, (b) 1 h and (c) 20 h.

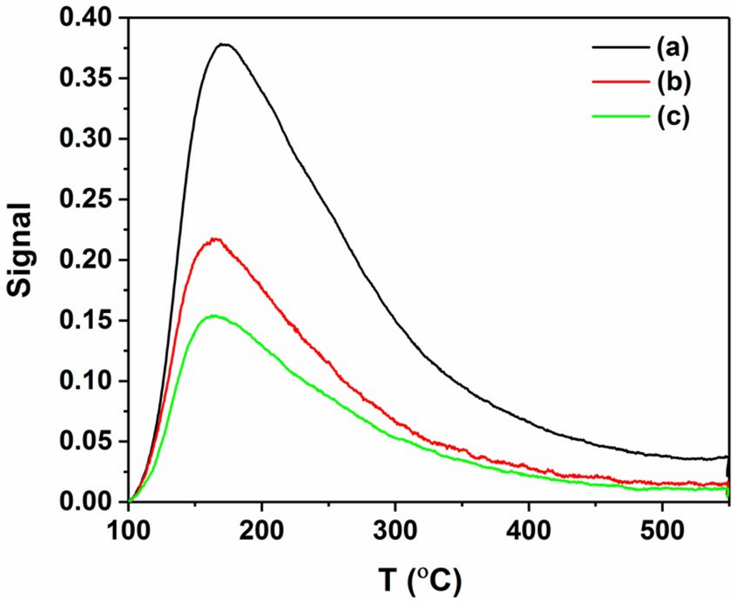

Fig. S9 TPD-CO_2 of SiO_2@Mg_4Al-LDH with AMOST post treatment in acetone for (a) 0 h, (b) 1 h and (c) 20 h.

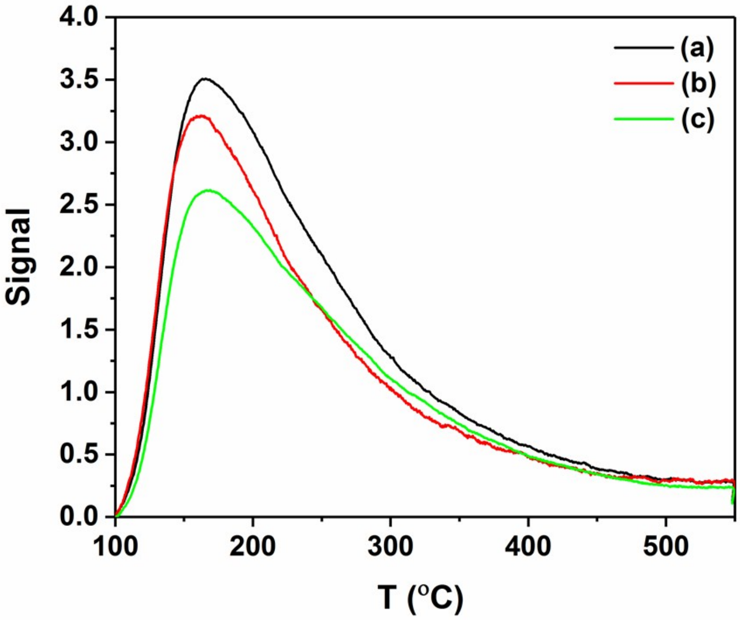

Fig. S10 TPD-CO_2 of Mg_4Al-LDH with AMOST post treatment in acetone for (a) 0 h, (b) 1 h and (c) 20 h.

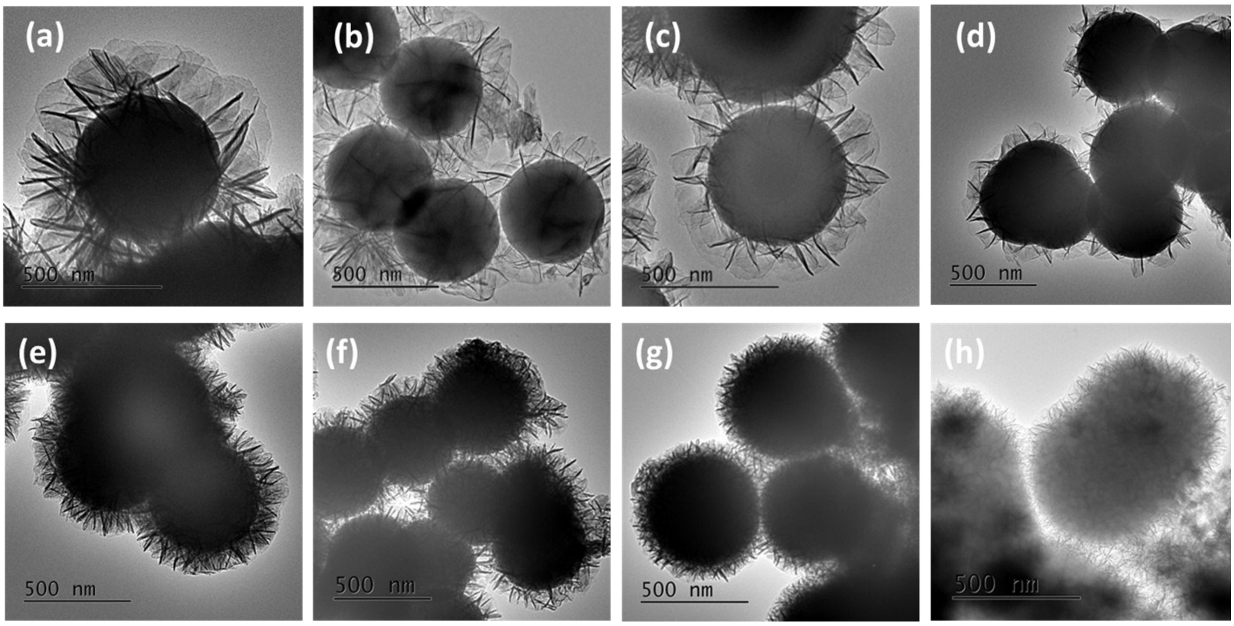

Fig. S11 TEM images of (a) SiO_2@Mg_3Al-CO_3-LDH doping with 10% Fe and after AMOST post treatment product (b), (c), (d) with different Fe doping amount of 10%, 60%, 100%; (e) SiO_2@ Mg_3Al-CO_3-LDH doping with 10% Ni, and after AMOST post treatment product (f), (g), (h) with different Ni doping amount of 10%, 60%, 100%; (AMOST post treatment in acetone for 4 h).

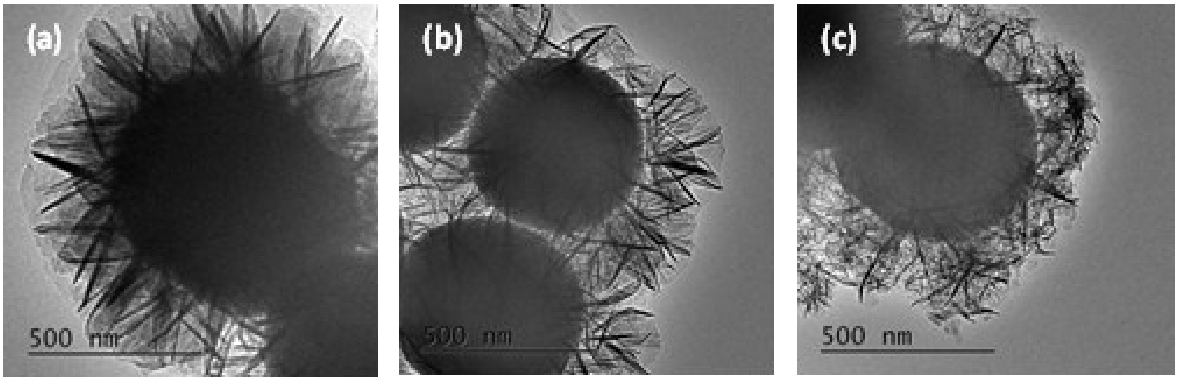

Fig. S12 TEM images of (a) water dispersed in SiO_2@Mg_4Al-CO_3-LDH and SiO_2@AMO-Mg_4Al-CO_3-LDH with AMOST post treatment in (b) ethanol for 20 h and (c) acetone for 20 h.

Table S1 Crystallinity data for SiO_2@Mg_4Al–CO_3-10 posted treated with acetone for various duration.

Acetone dispersion time (h)	d_{003} nm	d_{110} nm	a^{1} nm	c^{1} nm	D_{003}^{2} nm
0	0.789	0.153	0.306	2.367	8.7
1	0.788	0.153	0.306	2.364	7.9
4	0.786	0.153	0.306	2.358	6.2
20	0.778	0.153	0.306	2.334	5.7

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[1] $a = 2*d_{110}$, $c = 3*d_{003}$;

[2] *D003* is the crystalline domain length (CDL) in *c* direction, calculated according to Scherrer equation : $D_{hkl} = 0.89\lambda/(\beta\cos\theta)$, λ=0.154 nm, θ is the Bragg diffraction angle (deg.), and β is the FWHM (rad.) of the diffraction peaks.

Table S2 Element analysis for SiO_2@LDHs and SiO_2@AMO-LDHs.

Sample	Mg/Al ratio[1]	H[2] wt%	C[2] wt%	Mix metal oxide[3] wt%	Chemical Formula of SiO_2@AMO-LDHs
SiO_2@ Mg_4Al-CO_3-LDH	3.3 : 1	2.69	1.41	70.3	$[SiO_2]_{0\cdot 65}@[Mg_{3\cdot 3}Al(OH)_{8\cdot 6}(CO_3)_{0\cdot 5}\cdot(H_2O)_{1\cdot 3}]_{0\cdot 35}$
SiO_2@AMO-Mg_4Al-CO_3-LDH	3.2 : 1	2.27	1.56	69.8	$[SiO_2]_{0\cdot 65}@[Mg_{3.2}Al(OH)_{8.4}(CO_3)_{0\cdot 5}\cdot(H_2O)_{0.2}\cdot(C_3H_6O)_{0\cdot 01}]_{0\cdot 35}$

[1] Mg/Al ratio was obtained from ICP.

[2] Hydrogen and carbon content were collected from elemental analysis CHN.

[3] H_2O+AMO solvent and mix metal oxide were obtained from TGA.

Table S3 TPD-CO_2 analysis of SiO_2@Mg_4Al-LDH posted treated with acetone for (a) 0 h, (b) 1 h and (c) 20 h.

Sample	Acetone dispersion time (h)	Total evolved CO_2 (mmol/g, after calibration)
Mg_4Al-CO_3-LDH	**0**	**2.39**
	1	**2.04**
	20	**1.80**
SiO_2@Mg_4Al-CO_3-LDH	**0**	**2.58**
	1	**1.40**
	20	**1.01**

Ref:

[1]. W. Stöber, A. Fink and E.Bohn, J. *Colloid Interface Sci*., 1968, 26, 62–69.
[2]. C. Chen, M. Yang, Q. Wang, J.-C. Buffet and D. O'Hare, *J. Mater. Chem. A*, 2014, **2**, 15102.
[3]. J. C. Groen, L. A. Peffer and J. P´erez-Ram´ırez, *Micropor. Mesopor. Mater*, 2003, 60, 1–17. 23