

Supplementary Data

Optimising the Acid-Base Ratio of Mg-Al Layered Double Oxides to Enhance CO₂ Capture Performance: The Critical Role of Calcination Conditions

D. W. Justin Leung, Katherine Laney, Philip Kenyon, Nicholas H. Rees, Jean-Charles Buffet, Chunping Chen* and Dermot O'Hare*

Chemistry Research Laboratory, Department of Chemistry, University of Oxford, 12 Mansfield Road, OX1 3TA, UK

Corresponding author: chunping.chen@chem.ox.ac.uk, dermot.ohare@chem.ox.ac.uk

1. XRD and TGA of LDHs/LDOs samples

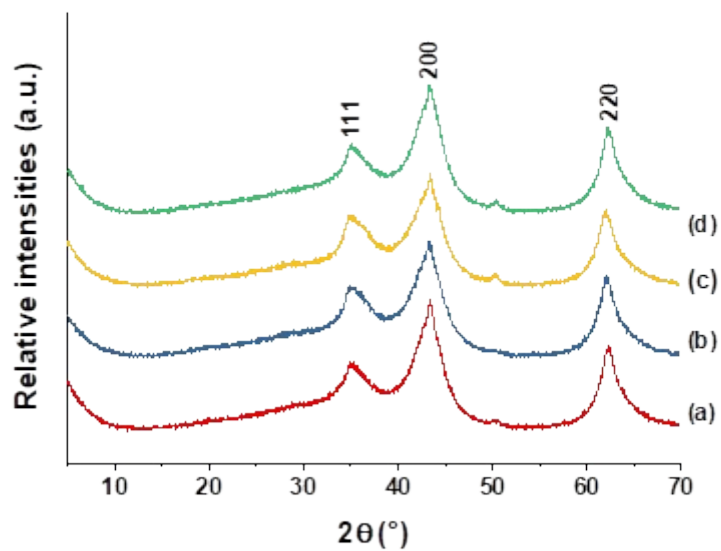


Figure S1- XRD of (a) 400-2h-5, (b) 400-2h-20, (c) 400-2h-50 and (d) 400-2h-100.

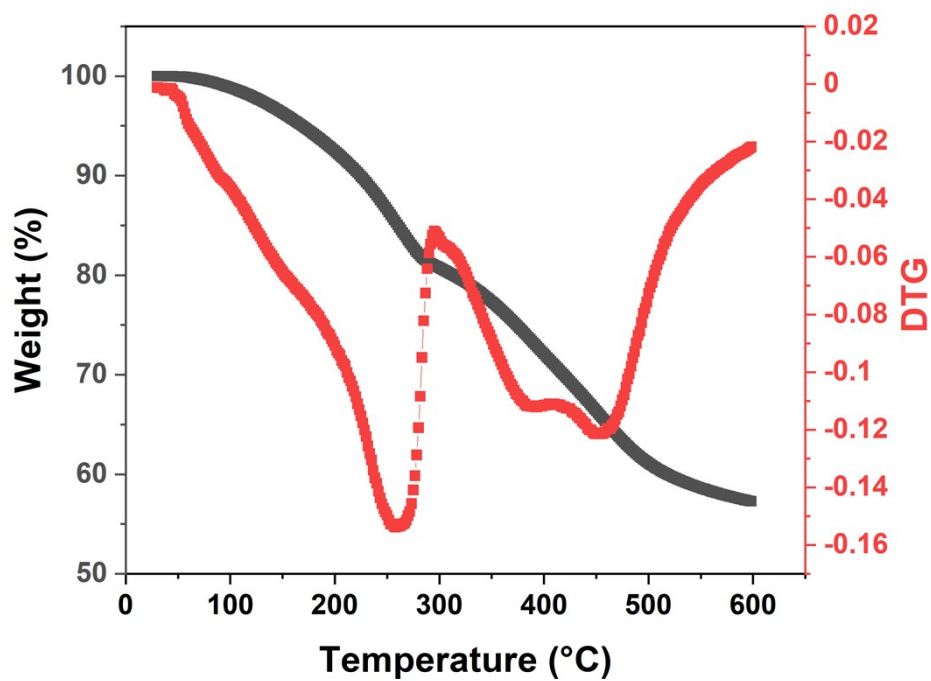


Figure S2 – TGA and DTG plot of $\text{Mg}_2\text{Al W LDHs}$

2. CO₂ capture per surface area

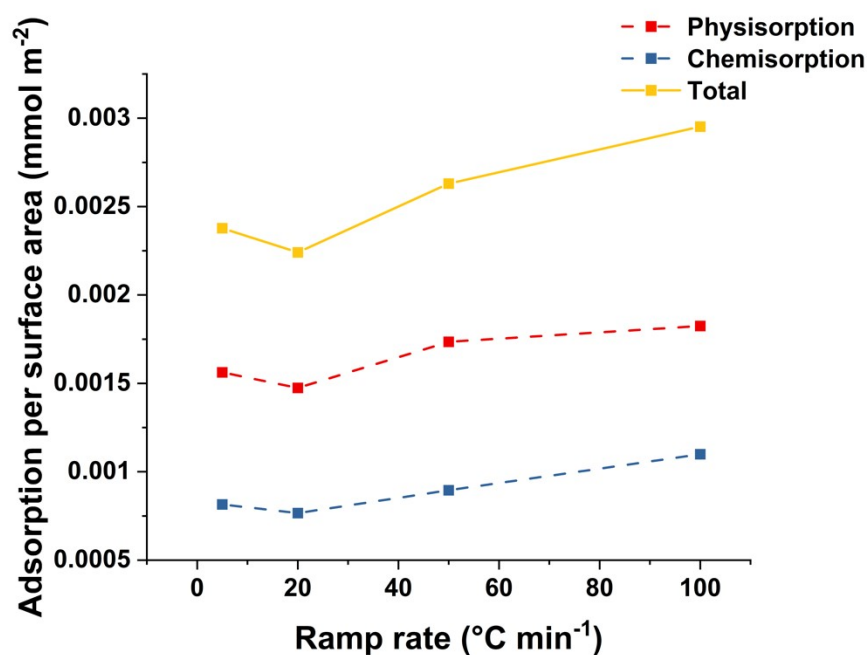


Figure S3 - CO₂ adsorption per surface for 400-2h-X samples at 40 °C and 100 kPa.

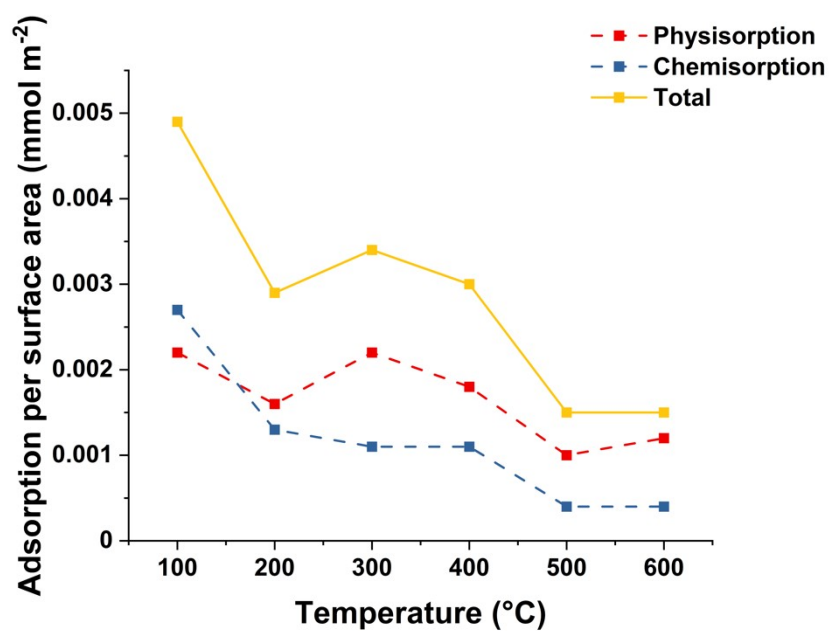


Figure S4 - CO₂ adsorption per surface area for X-2h-100 samples at 40 °C and 100 kPa.

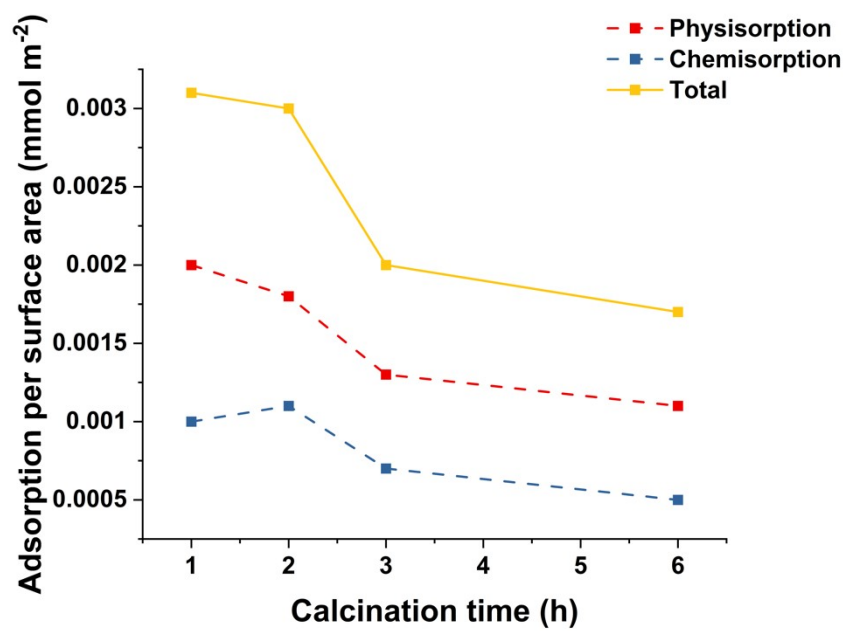


Figure S5 - CO₂ adsorption per surface area for 400-X-100 samples at 40 °C and 100 kPa.

3. Hydroxyl numbers

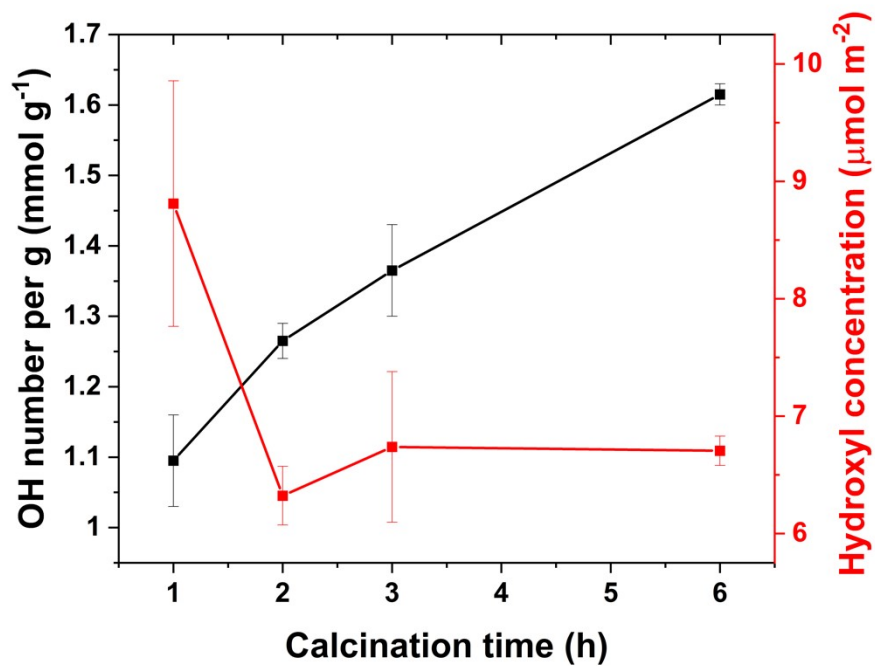
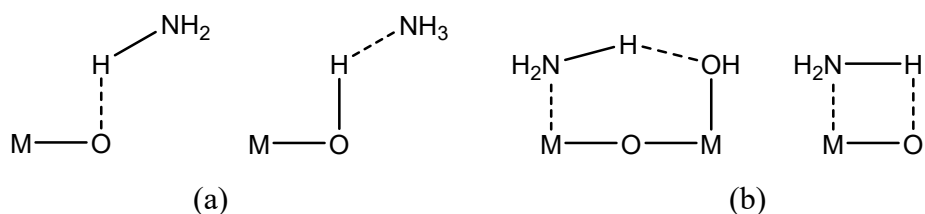
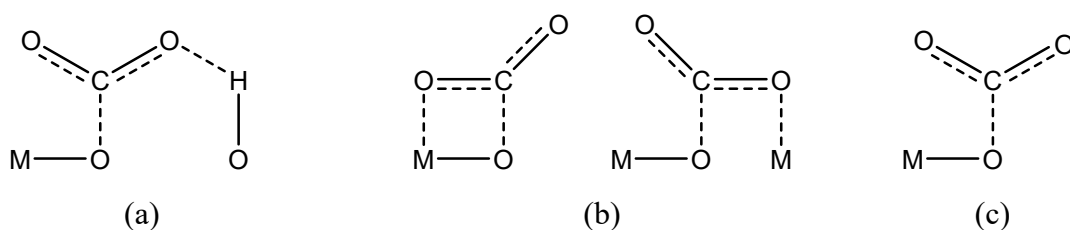


Figure S6 - Hydroxyl number of 400-X-100 samples.

4. CO₂ and NH₃ bonding modes and deconvolution of TPD data



Scheme S1 - NH₃ bonding modes.



Scheme S2 - (a) bicarbonate, (b) bidentate and bridging and (c) monodentate CO₂ bonding modes.

Table S1 - Deconvolution of TPD data.

	NH ₃				CO ₂			
	Peak	Weight (%)	Peak	Weight (%)	Peak	Weight (%)	Peak	Weight (%)
400-2h-5	149	15	230	85	157	33	222	67
400-2h-20	162	41	231	59	162	30	232	70
400-2h-50	159	44	225	56	157	32	223	68
400-2h-100	163	44	214	56	160	35	219	65
400-1h-100	162	45	212	55	157	35	213	65
400-3h-100	162	46	216	54	159	35	219	65
400-6h-100	160	44	212	56	158	37	214	63

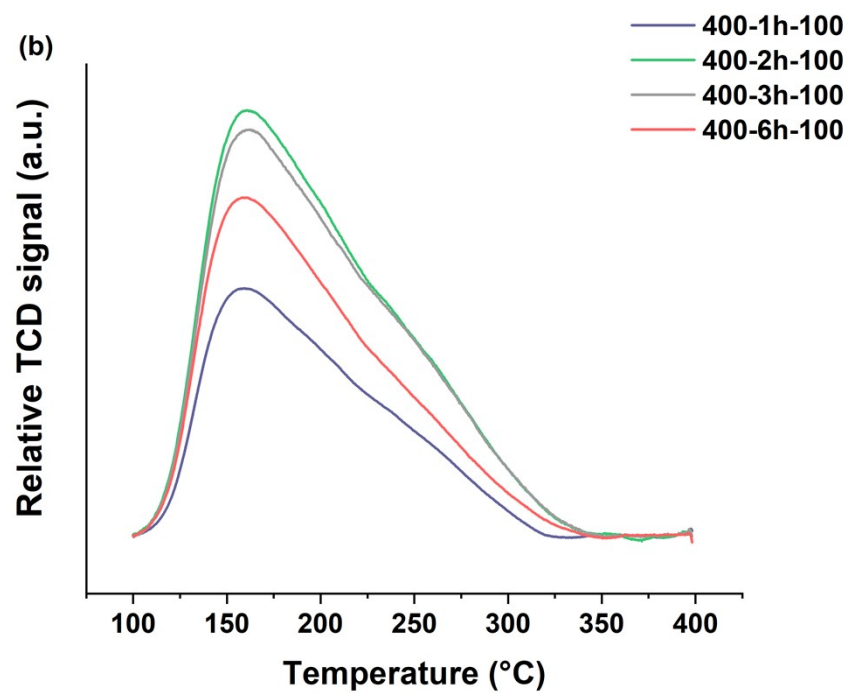
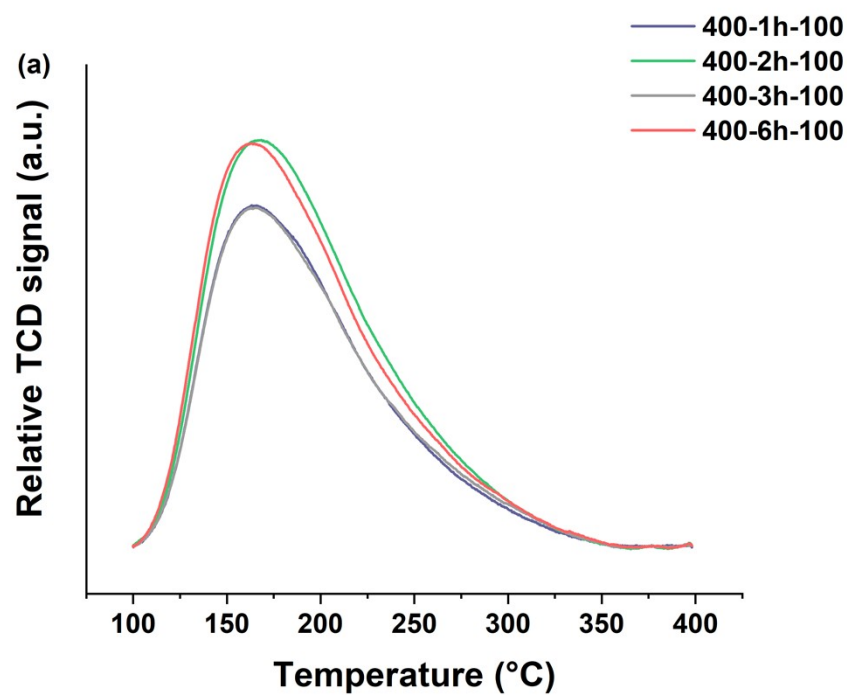


Figure S7 - (a) NH₃ and (b) CO₂ TPD of 400-X-100 samples.

5. SEM images

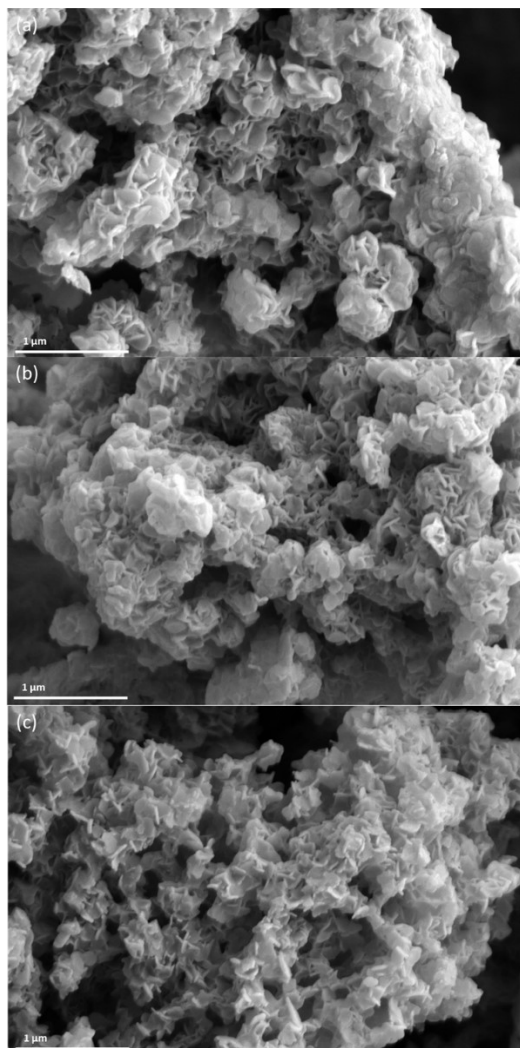


Figure S8 - SEM images of a) 400-2h-5, b) 400-2h-50 and c) 400-2h-100.

6. NMR

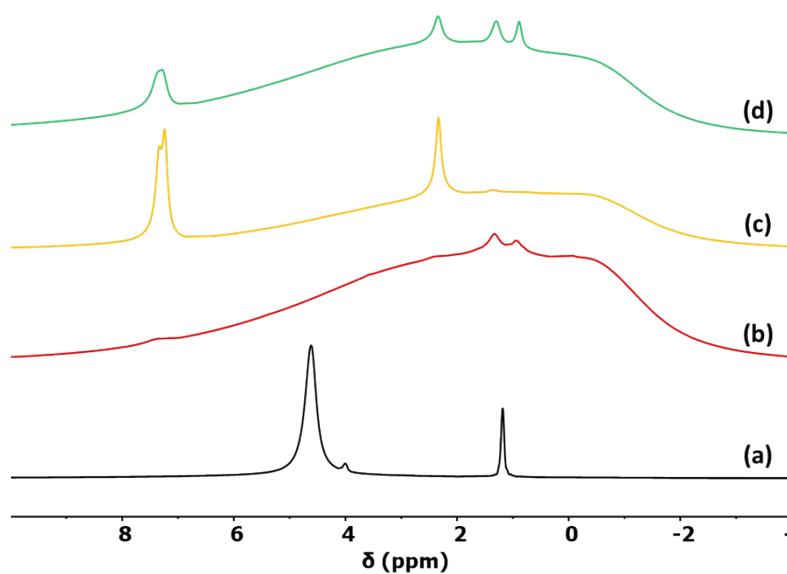


Figure S9 - ^1H ssNMR of (a) Mg_2Al LDH, (b) 400-2h-5, (c) 400-2h-50 and (d) 400-2h-100. Numbers denote relative population of each coordination environment.

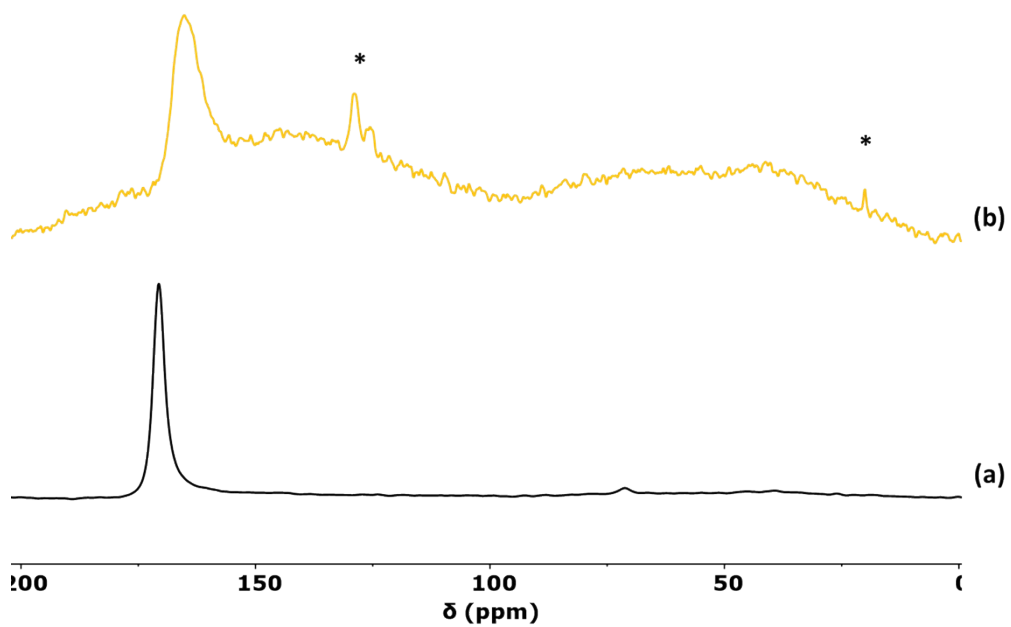


Figure S10 - ^{13}C dpmas ssNMR of (a) Mg_2Al LDH and (b) 400-2h-50. (*) denotes peaks from toluene.