Electronic Supplementary Material (ESI) for RSC Advances.

Low-temperature gas-solid carbonation of magnesia and magnesium hydroxide promoted by non-immersive contact with water

James Highfield, Jason Chen, Mojtaba Haghighatlari, Jacob Åbacka, Ron Zevenhoven

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Table S1 Physicochemical properties of $Mg(OH)_2$ extracted from Finnish serpentinite by ÅA method[Σ]

$Mg(OH)_2$ source[♀]	Sulphur Content		BET Surface Area[‡]
	EA[*] (wt.%)	XRF[†] (wt.%)	N_2 /77K (m^2g^{-1})
Aldrich > 99%	0.00	0.00	8.1
Aldrich > 95%	0.00	0.00	15.7
Ext. as supplied	2.72	3.65	13.3
Ext. cold wash x1	1.02	1.37	22.6
Ext. cold wash x2	0.95	1.29	28.2
Ext. hot wash x1	0.84	0.90	27.0
Ext. hot wash x2	0.68	0.80	29.6
Dead Sea Periclase	0.08	0.05	3.6

[Σ] Extraction method developed at Åbo Akademi based on ammonium sulphate flux yielding aq. Mg^{2+}/SO_4^{2-}, followed by precipitation of $Mg(OH)_2$ in aqueous NH_3(pre-separated from iron impurities).
[♀] In all cases only a single diffraction pattern of (hexagonal) brucite was seen by XRD (Bruker D8)
[*] S analysis by Thermo Scientific Flash 2000 CHNS-O Elemental Analyzer
[†] S analysis by Bruker S4 Explorer X-ray fluorescence spectrometer with "standardless" method
[‡] After overnight evacuation at 200 °C

Fig. S1 Snapshots of high-pressure thermogravimetric analyser (HPTGA, mod. 150S; *TA Instruments*)

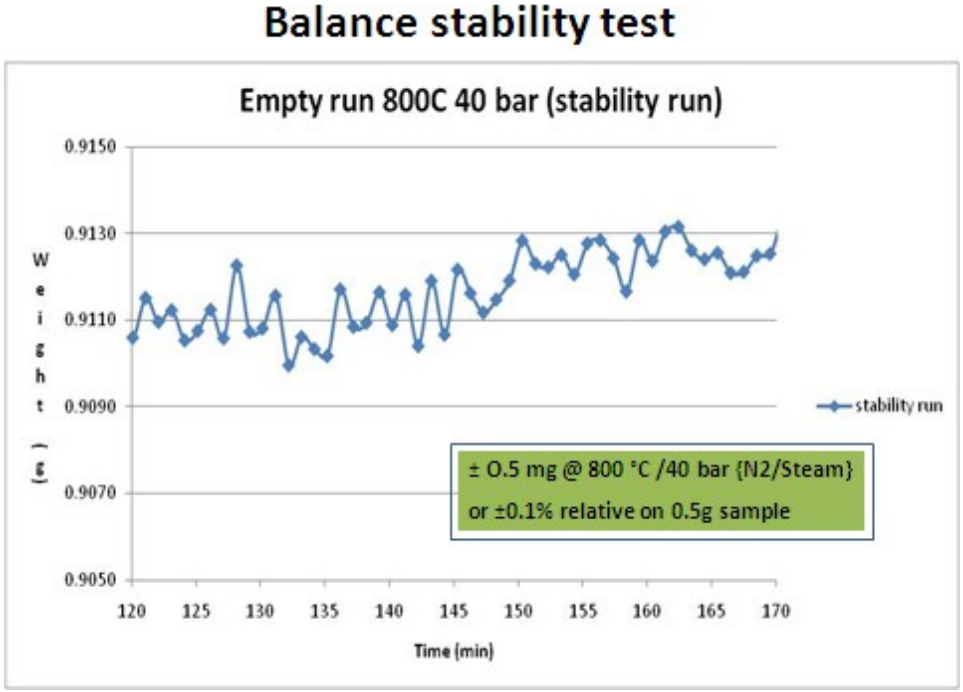

Fig. S2 Stability test on the magnetic suspension balance (*Rubotherm AG*) - empty crucible at 800 °C under 10% steam/N_2 (150 ml/min) at 40 bar

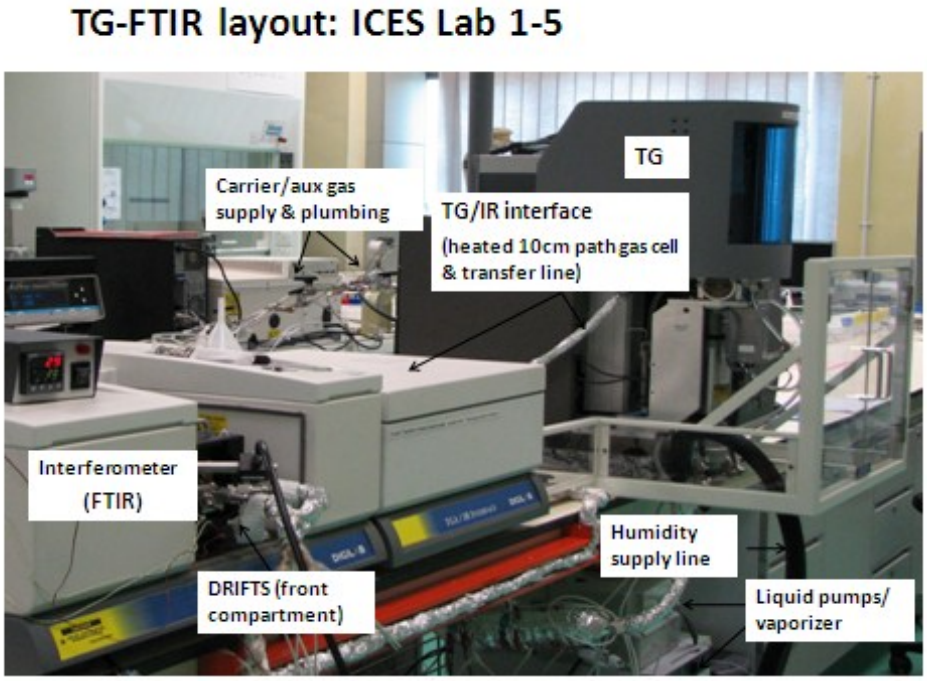

Fig. S3 Laboratory snapshot of TGA (Setaram *Setsys 12*) coupled to FTIR (*Excalibur FTS-3000*, Digilab). Humidity is supplied by from a Setaram *Wetsys* unit (not shown) housed under the TGA-FTIR.

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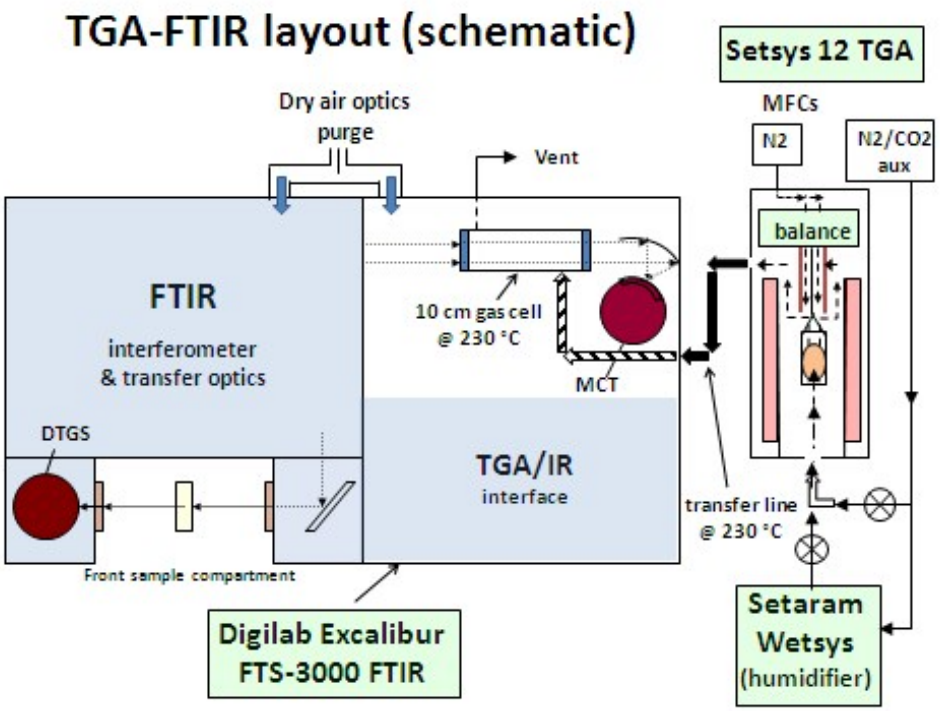

Fig. S4. Flow scheme of the TGA-FTIR for dry or humid gas supply

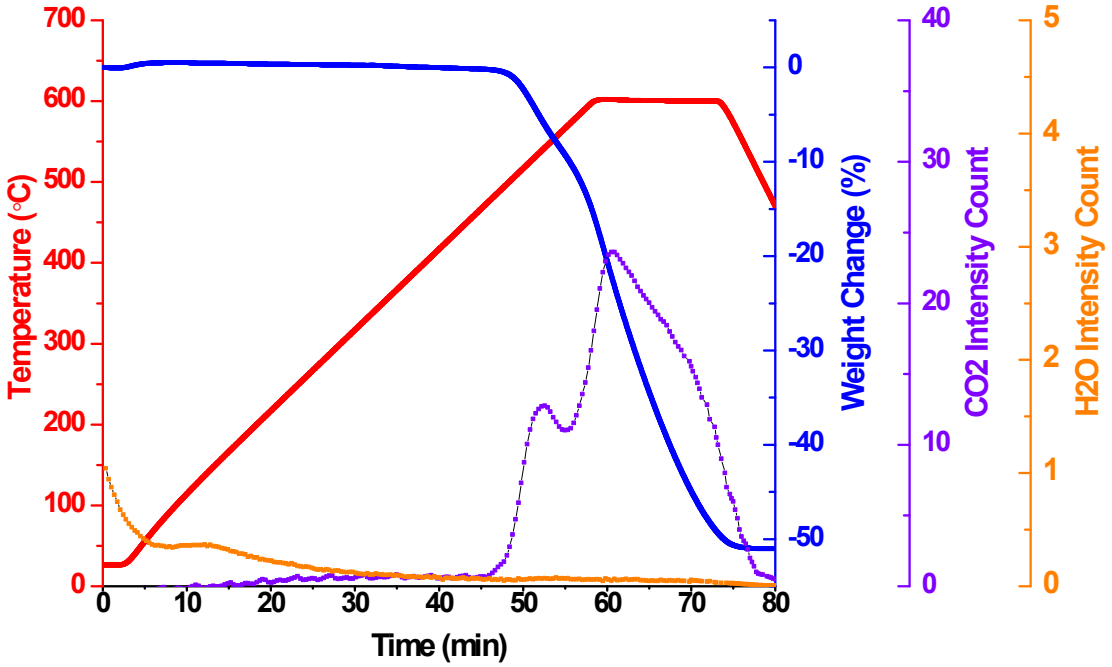

Fig. S5 TGA-FTIR profile for magnesite ($MgCO_3$) obtained from $Mg(OH)_2$ (Aldrich > 95%) by carbonation in the high-pressure thermogravimetric analyser

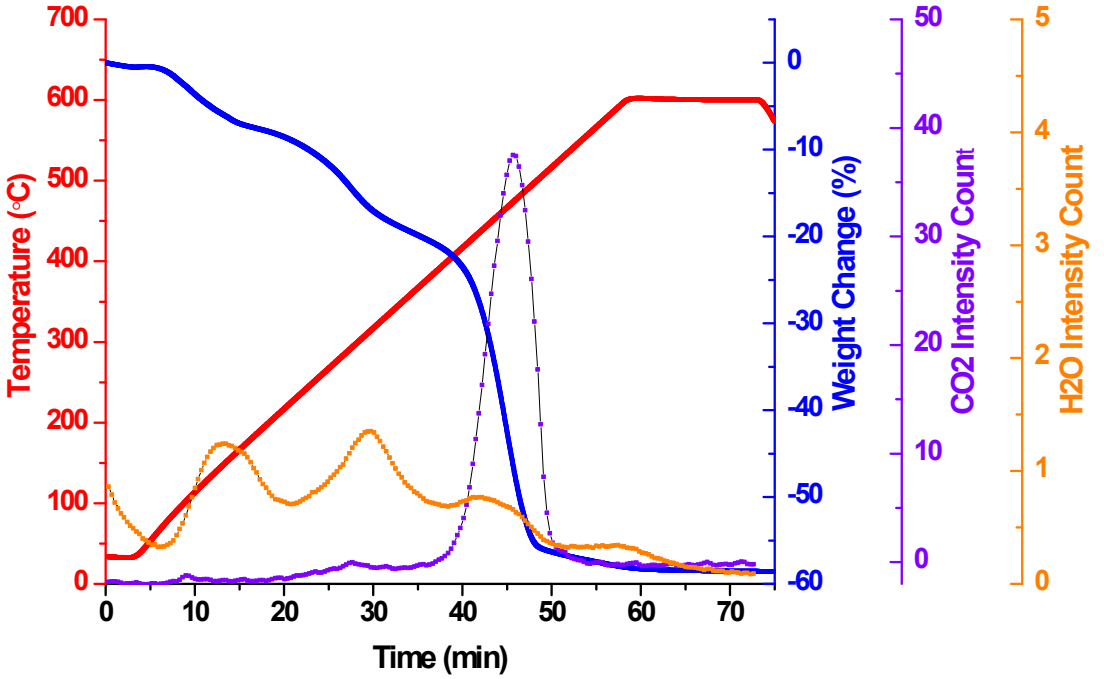

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Fig. S6 TGA-FTIR profile for hydromagnesite [$Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$] obtained from $Mg(OH)_2$ (Aldrich > 95%) by carbonation in the high-pressure thermogravimetric analyser

Fig. S7 Snapshots of autoclave (Parr mod. 4523 Bench Top Reactor, 1L capacity) adapted for batch-mode CO_2 absorption into $MgO/Mg(OH)_2$ promoted by saturated steam: 1. Reactor pot; 2. Sample holder; 3.Oven (with thermocouple); 4. Temperature gauge; 5. Discharge valve (vent); 6. CO_2 feed; 7. N_2 feed; 8. Pressure gauge; 9. Safety release valve (50 bar); 10. Temperature controller.

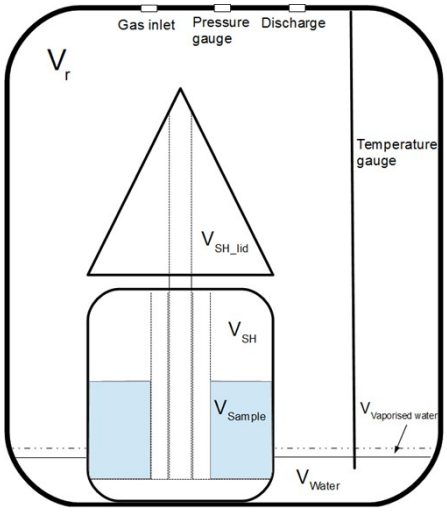

Fig. S8 Schematic layout of autoclave kept at 100% RH with sample held above liquid water reservoir in custom-made holder allowing good access by gas/vapour.

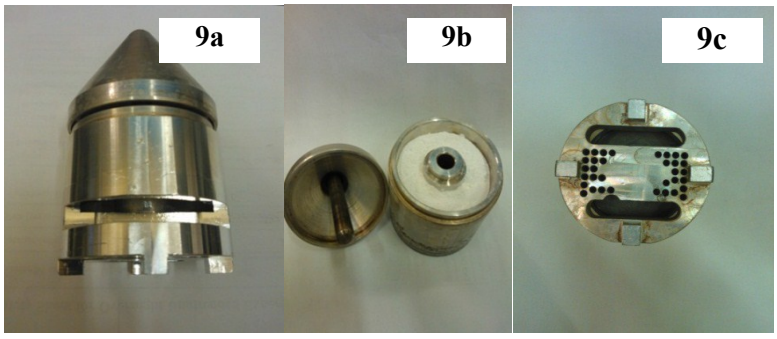

Fig. S9 Custom made two-piece stainless steel sample holder: **a.** side view; **b.** conical lid demounted; **c.** underside view showing vents/perforations.

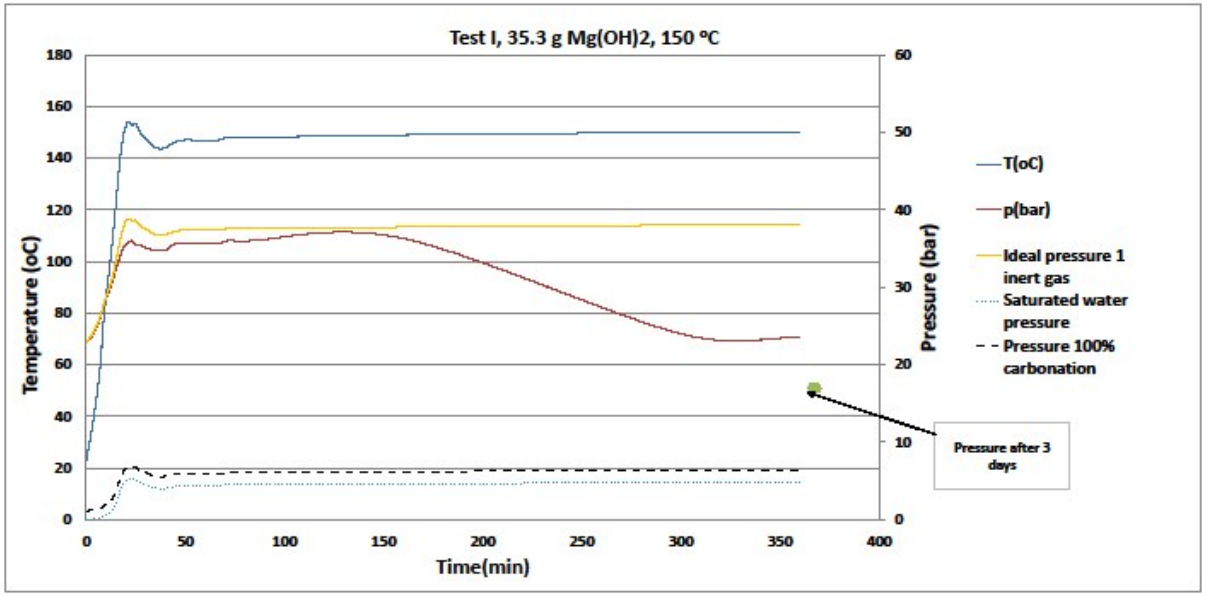

Fig. S10 Typical pressure vs. time curve for a large (>35g) sample of $Mg(OH)_2$ [Aldrich >95%] loaded into the autoclave and rapidly heated to 150 °C to give a total initial pressure of ~40 bar [35 bar CO_2, 5 bar H_2O (svp)]

Table S2 Direct carbonation of Finnish serpentinite in humid CO_2 in the high-pressure thermogravimetric analyzer

Preheat	Conditions				XRD	CO_2 level	Degree of
(°C)/N_2	T (°C),	P_{CO2} (bar),	RH (%)	phases‡	I_{mag}‡‡ / $[I_{mag} + I_{liz}]$	TG-FTIR (wt%)	Carbonation (%)
300	150	40	83	Mag, Ant	0.43	7.8	22.0
500	150	40	83	Mag, Ant	0.44	10.2	28.8
300	150	40	126	Mag, Ant	0.22	4.9	13.8
300	150	40	168	Mag, Ant	0.23	4.9	13.8
300	165	40	86	Mag, Ant	0.18	5.0	14.1
300	180	40	80	Mag, Liz, Ant	0.40	9.3	26.3
300	180	40	100	Mag, Ant	0.12	4.7	13.3
500	180	40	100	Mag, Ant	0.27	5.1	14.4
300	200	40	51	Mag, Ant	0.47	9.2	26.0
300	200	40	64	Mag, Ant	0.15	4.6	13.0
500	200	40	64	Mag, Ant	0.37	3.2	9.0

‡‡ mag = magnesite ($MgCO_3$), liz = lizardite, Ant = Antigorite

‡ Index [h k l], 2θ: Magnesite = [1 0 4], 32.63°, Lizardite = [1 1 1], 35.91°, Antigorite = [2 0 1], 35.45°

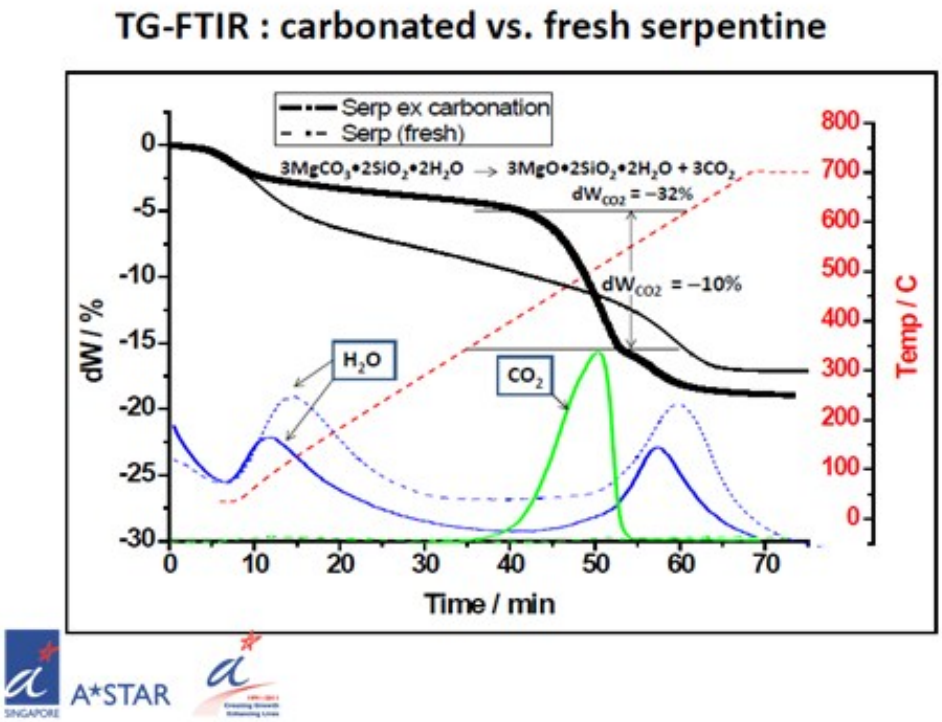

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Fig. S11 TGA-FTIR profile of Finnish serpentinite (vs.fresh control) carbonated under humid CO_2 in the high-pressure thermogravimetric analyser (HPTGA)