Electronic Supplemental Information for:

## Dolomite: A Low Cost Thermochemical Energy Storage Material

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**Figure S1.** XRD analysis of (a) as supplied Richgro dolomite (Rich-A) and (b) Watheroo dolomite (Wath-A). \* = dolomite, ! = calcite, \$ = quartz, # = andradite, ^ = NaCl, @ = magnesite.  $\lambda$  = CuK $\alpha_{1,2}$ .



**Figure S2.** XRD analysis of thermally outgassed (a) Watheroo dolomite at 1000 °C (Wath-B), (b) Richgro dolomite at 1000 °C (Rich-B) and (c) synthetic dolomite at 650 °C (Synth-B). ! = CaO, @ = MgO, \$ = quartz, \* = larnite, - = Ca<sub>9</sub>(Al<sub>2</sub>O<sub>6</sub>)<sub>3</sub>, ^ = NaCl, # = andradite, + = gehlenite. Flowing Ar.  $\lambda$  = CuK $\alpha_{1,2}$ .



**Figure S3.** XRD analysis of carbonated (a) Watheroo dolomite (Wath-C) and (b) synthetic dolomite (Synth-C) reacted at 500 °C and 50 bar CO<sub>2</sub>. ! = calcite, \$ = quartz, \* = larnite, @ = MgO,  $- = Ca_9(Al_2O_6)_3$ .  $\lambda = CuK\alpha_{1,2}$ .



**Figure S4.** XRD analysis of carbonated synthetic dolomite and catalyst (Synth-C-cat) reacted at 500 °C and 50 bar CO<sub>2</sub>. + = Dolomite,  $^{\text{A}}$  = NaCl, - = CaMg<sub>2</sub>Cl<sub>6</sub>(H<sub>2</sub>O)<sub>12</sub>.  $\lambda$  = 0.8263076 Å.



**Figure S5.** Carbon dioxide pressure cycling results for catalysed (NaCl:MgCl<sub>2</sub>) synthetic dolomite at 525 °C (Synth-D-cat,  $CO_2$  absorption pressure 35 bar, desorption pressure 2 bar).

**Table S1.** Molar quantity of  $CO_2$  absorbed and desorbed during Carbon dioxide pressure cycling for catalysed (NaCl:MgCl<sub>2</sub>) Watheroo dolomite (Wath-B-cat) and catalysed (NaCl:MgCl<sub>2</sub>) synthetic dolomite (Synth-D-cat). Uncertainties vary between absorption ( $\pm$  0.01 mol) and desorption ( $\pm$  0.1 mol) for the Wath-B-cat sample due to a larger reservoir volume being employed during the desorption measurement. Uncertainties for Synth-D-cat are  $\pm$  0.01 mol.

	Molar quantity of CO <sub>2</sub>			
Cycle	Wath-B-cat	Synth-D-cat		
1 - Absorption	1.09	1.77		
- Desorption	-0.3	-0.02		
2 - Absorption	0.46	0.16		
- Desorption	-0.4	-0.12		
3 - Absorption	0.47	0.18		
- Desorption	-0.5	-0.14		
4 - Absorption	0.45	0.16		
- Desorption	-0.5	-0.14		
5 - Absorption	0.43	0.17		
- Desorption	-0.5	-0.13		
6 - Absorption	0.47	0.17		
- Desorption	-0.5	-0.14		
7 - Absorption	0.45	0.17		
- Desorption	-0.5	-0.14		
8 - Absorption	0.57	0.16		
- Desorption	-0.5	-0.13		
9 - Absorption	0.54	0.16		
- Desorption	-0.5	-0.14		
10 - Absorption	0.60	0.16		
- Desorption	-0.5	-0.13		
11 - Absorption	0.55	0.22		
- Desorption		-0.15		
12 - Absorption		0.22		
- Desorption		-0.16		
13 - Absorption		0.22		
- Desorption		-0.15		
14 - Absorption		0.23		
- Desorption		-0.14		



**Figure S6.** XRD analysis of cycled (a) Watheroo dolomite (Wath-D-cat) and (b) synthetic dolomite (Synth-D-cat) measured after absorption of CO<sub>2</sub>. + = Dolomite, ! = calcite, @ = MgO, ^ = NaCl, \$ = quartz, # = andradite, - = CaCl<sub>2</sub>.  $\lambda$  = CuK $\alpha_{1,2}$ . CO<sub>2</sub> absorption pressure 35 bar, desorption pressure 2 bar.

Table S2. Brunauer–Emmett–Teller (BET) surface area, cumulative pore volume and average pore size of the
synthetic and mined dolomite samples from the Barrett–Joyner–Halenda (BJH) method.

Sample name	BET surface area (m²/g)	BJH adsorption cumulative volume of pores (cm <sup>3</sup> /g)	BJH adsorption average pore width (nm)
Rich-A	16.90 ± 0.16	0.0155	4.5
Richgro dolomite			
Wath-A	16.78 ± 0.16	0.0146	4.6
Watheroo dolomite			
Synth-B	$0.98 \pm 0.01$	0.0014	7.5
Synthetic CaO/MgO			
Rich-B	4.95 ± 0.03	0.0047	4.7
Calcined Richgro			
(from Rich-A)			
Wath-B	6.19 ± 0.02	0.0061	4.7
Calcined Watheroo			
(from Wath-A)			