

## Supplementary Information for Reviewers

# ***Elucidating the Differences in the Carbon Mineralization Behaviors of Calcium and Magnesium-bearing Alumino-Silicates and Magnesium Silicates for CO<sub>2</sub> Storage***

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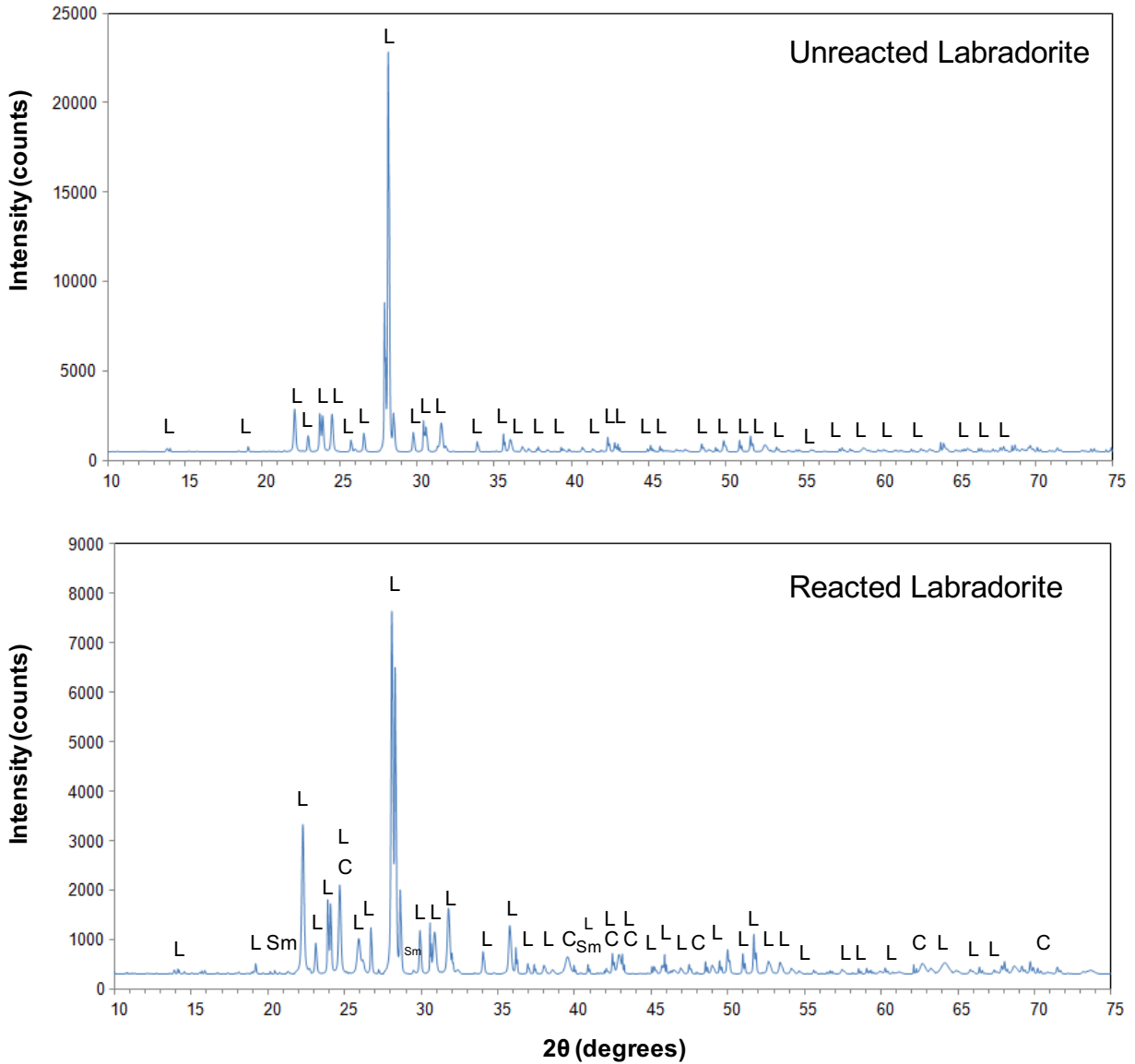
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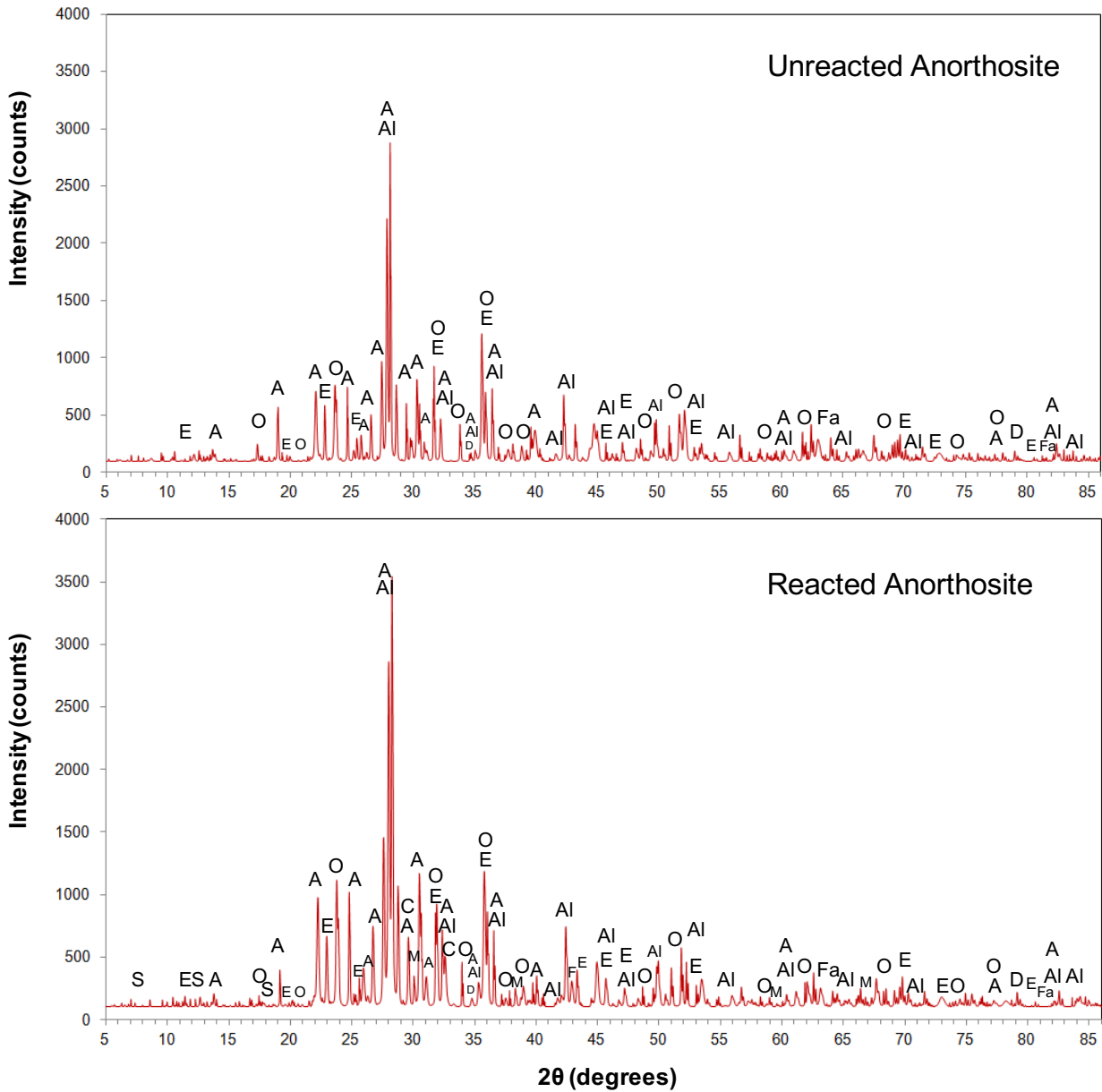
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**Table S1.** Summary of the amount of CO<sub>2</sub> stored as solid carbonate (wt%) in olivine[1], labradorite, anorthosite and basalt reacted at varying reaction times, temperatures, and CO<sub>2</sub> partial pressures in the presence of various chemical additives. The solid (mineral/rock) to liquid ratio was 15 wt% and a stirring speed of 800 rpm was maintained throughout the reaction. Extents of carbonation are reported as an average of TCA estimates for labradorite, anorthosite and basalt, and as an average of both TCA and TGA for olivine [1] (Reported values are the average and standard deviation (1 sigma) for three runs of Total Carbon Analysis (TCA))

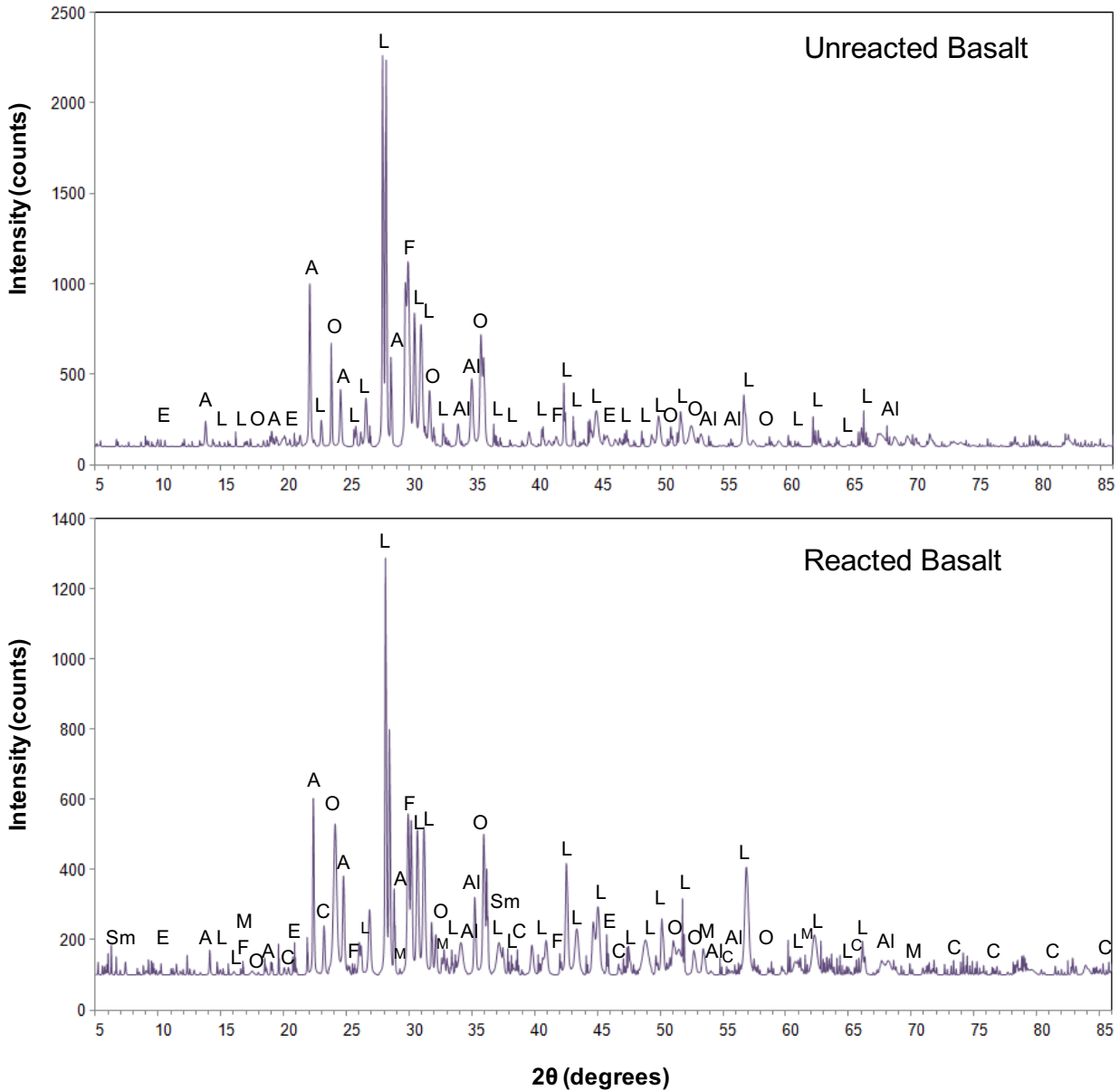
	CO <sub>2</sub> in reacted minerals (wt%)		
	Labradorite	Anorthosite	Basalt
<b><i>Effect of Reaction Time (185 °C, P<sub>CO2</sub> = 139 atm, 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub>)</i></b>			
1 hour	0.89 ± 0.05	1.59 ± 0.10	0.97 ± 0.07
3 hours	2.84 ± 0.03	4.26 ± 0.10	1.00 ± 0.01
5 hours	3.66 ± 0.01	4.26 ± 0.09	1.14 ± 0.06
<b><i>Effect of Partial Pressure of CO<sub>2</sub> (185 °C, 3 hours, 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub>)</i></b>			
64 atm	2.76 ± 0.03	3.98 ± 0.05	0.95 ± 0.03
89 atm	2.82 ± 0.08	4.33 ± 0.12	0.94 ± 0.03
139 atm	2.84 ± 0.03	4.26 ± 0.10	1.00 ± 0.01
164 atm	3.16 ± 0.06	4.11 ± 0.05	1.15 ± 0.07
<b><i>Effect of Temperature (P<sub>CO2</sub> = 139 atm, 3 hours, 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub>)</i></b>			
90 °C	1.14 ± 0.03	1.33 ± 0.07	0.64 ± 0.02
125 °C	1.16 ± 0.01	1.57 ± 0.07	0.83 ± 0.04
150 °C	2.08 ± 0.01	2.32 ± 0.08	0.89 ± 0.04
185 °C	2.84 ± 0.03	4.26 ± 0.10	1.00 ± 0.01
<b><i>Effect of [NaHCO<sub>3</sub>] (185 °C, P<sub>CO2</sub> = 139 atm, 3 hours)</i></b>			
Deionized Water	0.40 ± 0.02	1.74 ± 0.03	0.41 ± 0.04
0.48 M NaHCO <sub>3</sub>	0.69 ± 0.01	1.86 ± 0.01	1.20 ± 0.02
0.64 M NaHCO <sub>3</sub>	0.95 ± 0.03	2.34 ± 0.08	1.21 ± 0.01
1.0 M NaHCO <sub>3</sub>	2.72 ± 0.06	5.61 ± 0.02	1.65 ± 0.08
<b><i>Effect of [NaCl] (185 °C, P<sub>CO2</sub> = 139 atm, 3 hours)</i></b>			
Deionized Water	0.40 ± 0.02	1.74 ± 0.03	0.41 ± 0.04
0.5 M NaCl	0.26 ± 0.06	1.63 ± 0.04	0.49 ± 0.01
1.0 M NaCl	0.20 ± 0.04	1.92 ± 0.10	0.44 ± 0.22



**Figure S1.** Phase transformation of labradorite via carbonation reaction at 185 °C and  $P_{\text{CO}_2} = 139$  atm in 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub> for 3 hours with 15 wt% slurry density and a stirring speed of 800 rpm. The phases identified are: C = Calcite (CaCO<sub>3</sub>), L = Labradorite ((Ca<sub>0.6</sub>Na<sub>0.4</sub>)(Al<sub>1.6</sub>Si<sub>2.4</sub>)O<sub>8</sub>), and Sm = Smectite ((Na, Ca)<sub>0.3</sub>(Al, Mg)<sub>2</sub>(Si<sub>4</sub>O<sub>10</sub>)(OH)<sub>2</sub> · nH<sub>2</sub>O).



**Figure S2.** Phase transformation of anorthosite via carbonation reaction at 185 °C and  $P_{\text{CO}_2} = 139$  atm in 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub> for 3 hours with 15 wt% slurry density and a stirring speed of 800 rpm. The phases identified are: A = Anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>), Al = Albite (NaAlSi<sub>3</sub>O<sub>8</sub>), C = Calcite (CaCO<sub>3</sub>), D = Diopside (MgCaSi<sub>2</sub>O<sub>6</sub>), E = Enstatite (MgSiO<sub>3</sub>), Fa = Fayalite (Fe<sub>2</sub>SiO<sub>4</sub>), M = Magnesite (MgCO<sub>3</sub>), Olivine ((Mg,Fe)<sub>2</sub>SiO<sub>4</sub>), and S = Al-bearing Sepiolite (Mg<sub>4</sub>Al<sub>2</sub>Si<sub>10</sub>O<sub>27.15</sub>H<sub>2</sub>O).



**Figure S3.** Phase transformation of basalt via carbonation reaction at 185 °C and  $P_{CO_2} = 139$  atm in 1.0 M NaCl + 0.64 M NaHCO<sub>3</sub> for 3 hours with 15 wt% slurry density and a stirring speed of 800 rpm. The phases identified are: A = Anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>), Al = Albite (NaAlSi<sub>3</sub>O<sub>8</sub>), C = Calcite (CaCO<sub>3</sub>), E = Enstatite (MgSiO<sub>3</sub>), F = Forsterite (Mg<sub>2</sub>SiO<sub>4</sub>), L = Labradorite ((Ca,Na)(Al,Si)<sub>4</sub>O<sub>8</sub>), M = Magnesite (MgCO<sub>3</sub>), Sm = Smectite ((Na, Ca)<sub>0.3</sub>(Al, Mg)<sub>2</sub>(Si<sub>4</sub>O<sub>10</sub>)(OH)<sub>2</sub>·nH<sub>2</sub>O), and S = Al-bearing Sepiolite (Mg<sub>4</sub>Al<sub>2</sub>Si<sub>10</sub>O<sub>27</sub>·1.5H<sub>2</sub>O), and Olivine ((Mg, Fe)<sub>2</sub>SiO<sub>4</sub>).