

## SUPPORTING INFORMATION

### Effects of CO<sub>2</sub> and Temperature on the Structure and Chemistry of C-(A)-S-H Investigated by Raman Spectroscopy

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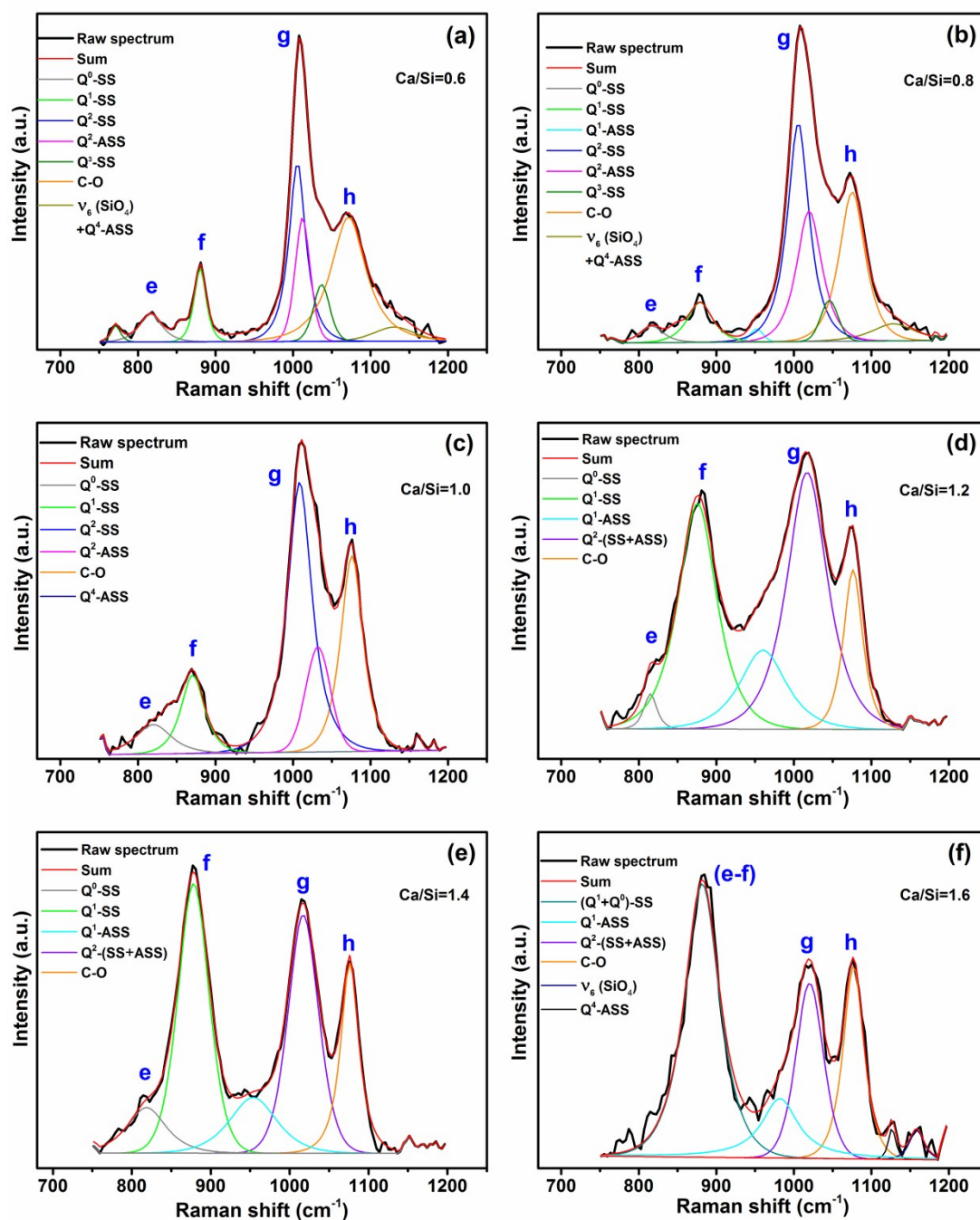


Figure S1. Deconvoluted peaks of C-S-H samples with Ca/Si molar ratio of a) 0.6, b) 0.8, c) 1.0, d) 1.2, e) 1.4 and f) 1.6 in the range of 750-1200 cm<sup>-1</sup>. All the samples have been equilibrated at 20 °C.

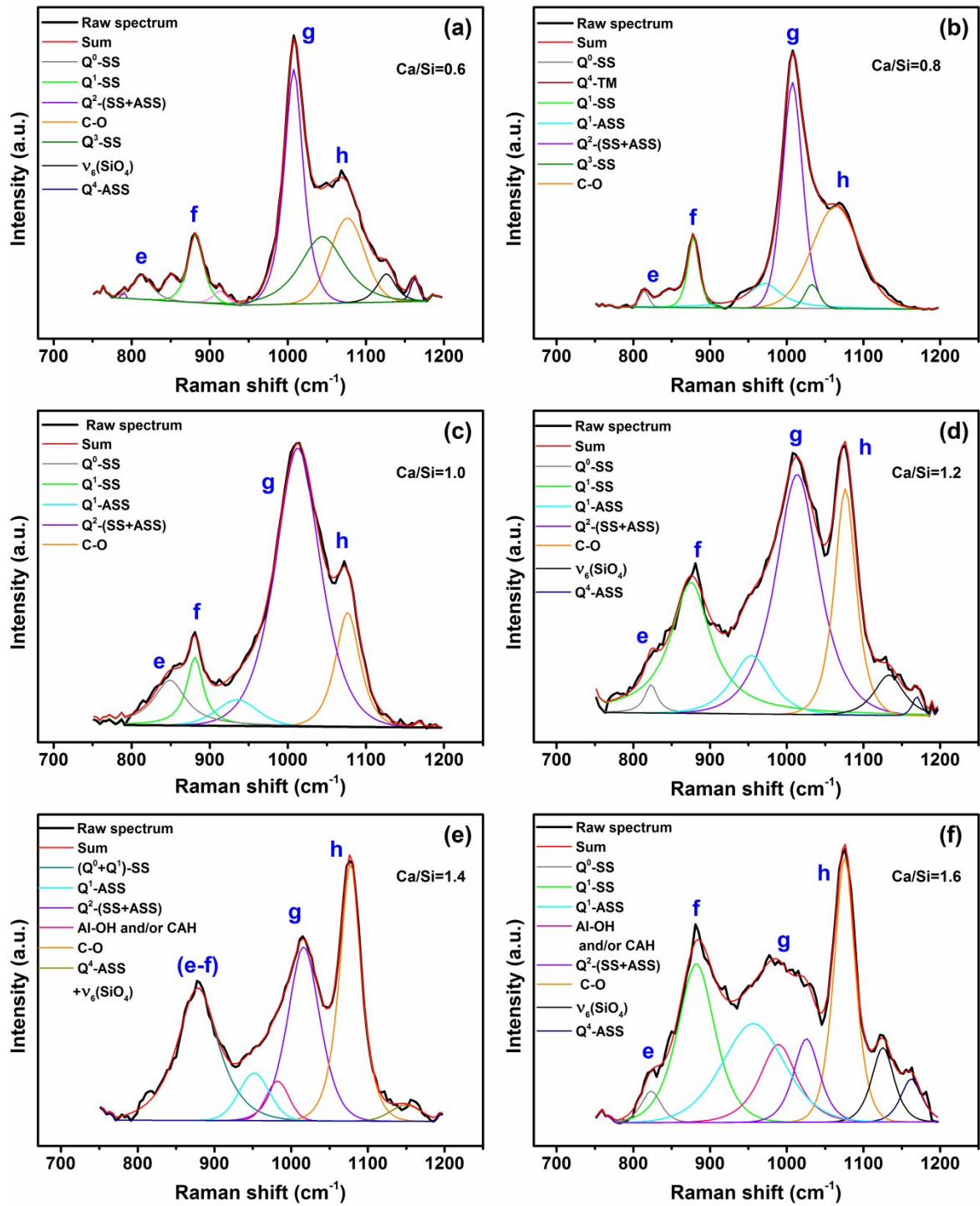


Figure S2. Deconvoluted peaks of C-A-S-H samples (Al/Si=0.05) with Ca/Si molar ratio of a) 0.6, b) 0.8, c) 1.0, d) 1.2, e) 1.4 and f) 1.6 in the range of 750-1200  $\text{cm}^{-1}$ . All the samples have been equilibrated at 20 °C.

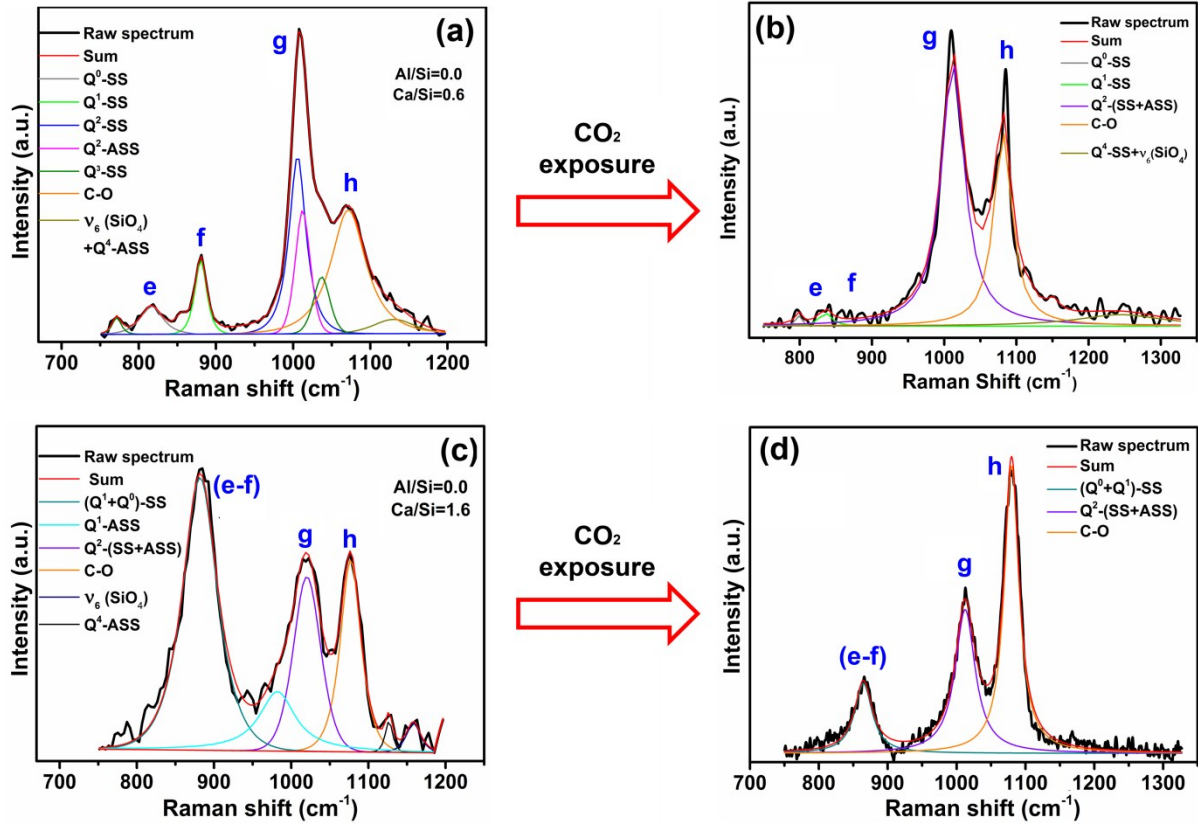


Figure S3. Effect of CO<sub>2</sub> uptake on Al-free samples for the low Ca/Si molar ratio. a) Before, b) after 10 days exposure; for high Ca/Si molar ratio, c) before and d) after CO<sub>2</sub> uptake. The images have been extracted from Figure 4a at the range of 750 -1200 cm<sup>-1</sup>.

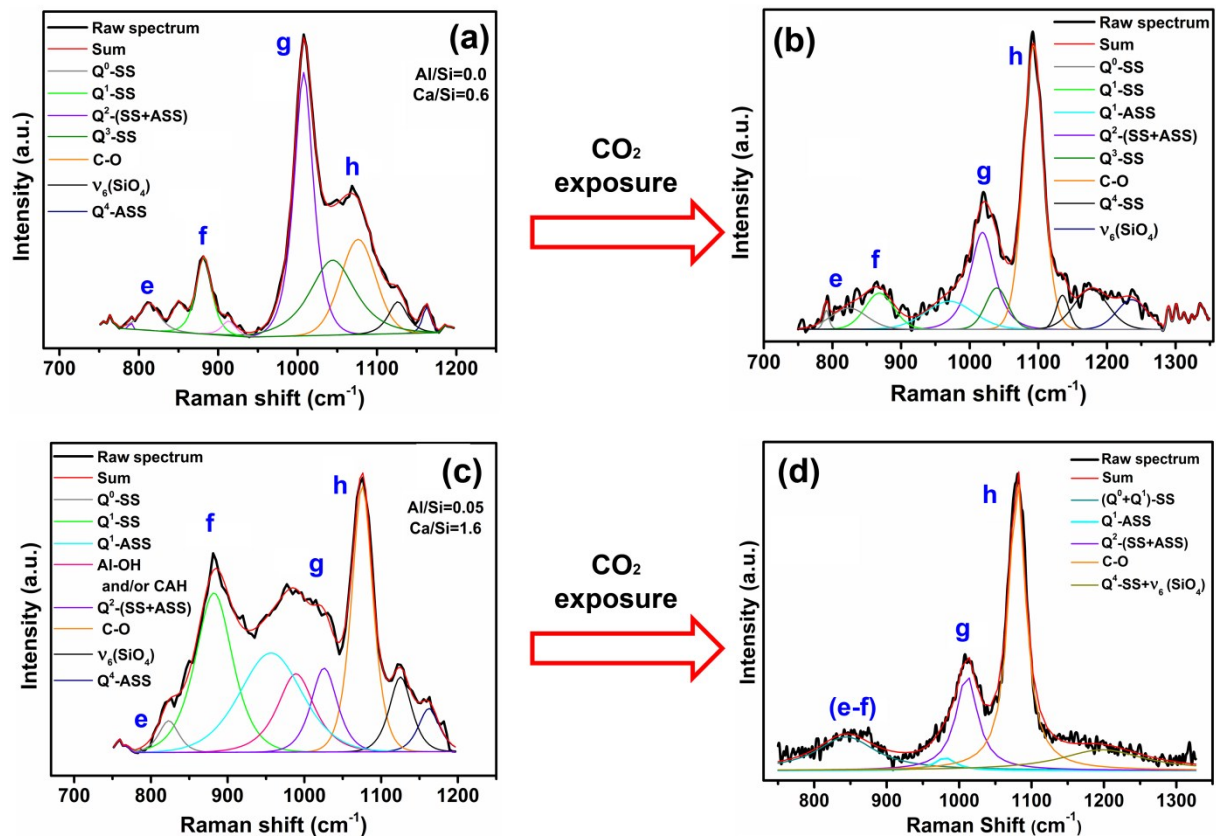


Figure S4. Effect of CO<sub>2</sub> uptake on Al-containing samples (Al/Si=0.05) for the low Ca/Si molar ratio a) before, b) after 10 days exposure; for high Ca/Si molar ratio c) before and d) after CO<sub>2</sub> uptake. The images have been extracted from Figure 4b at the range of 750 -1200 cm<sup>-1</sup>.

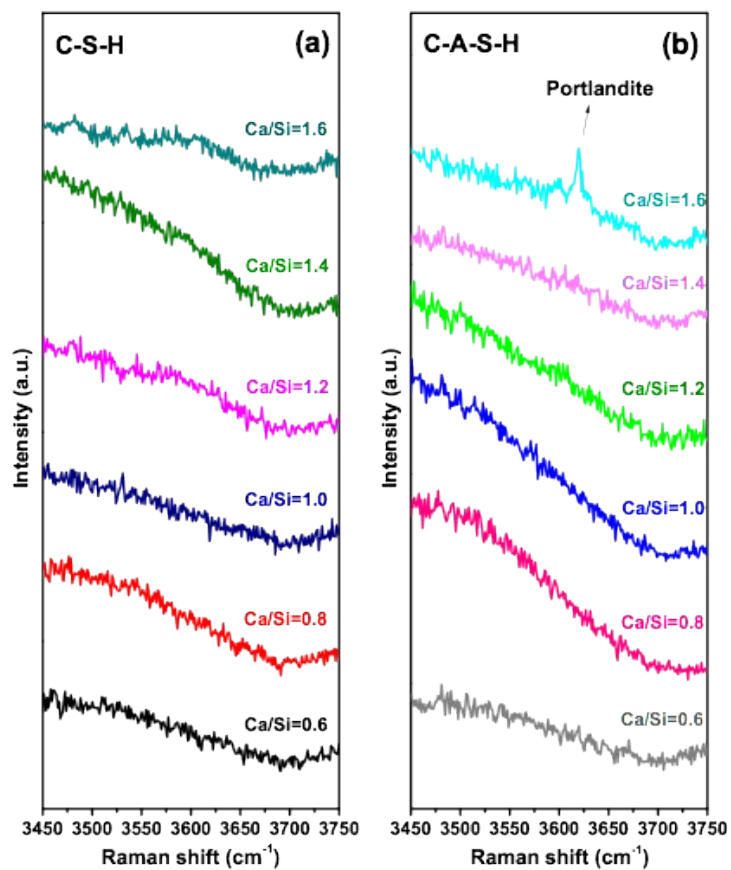


Figure S5. Raman spectra of C-S-H and C-A-S-H ( $\text{Al/Si} = 0.05$ ) samples with Ca/Si molar ratios from 0.6 to 1.6. All samples were synthesized at  $20^\circ\text{C}$ .

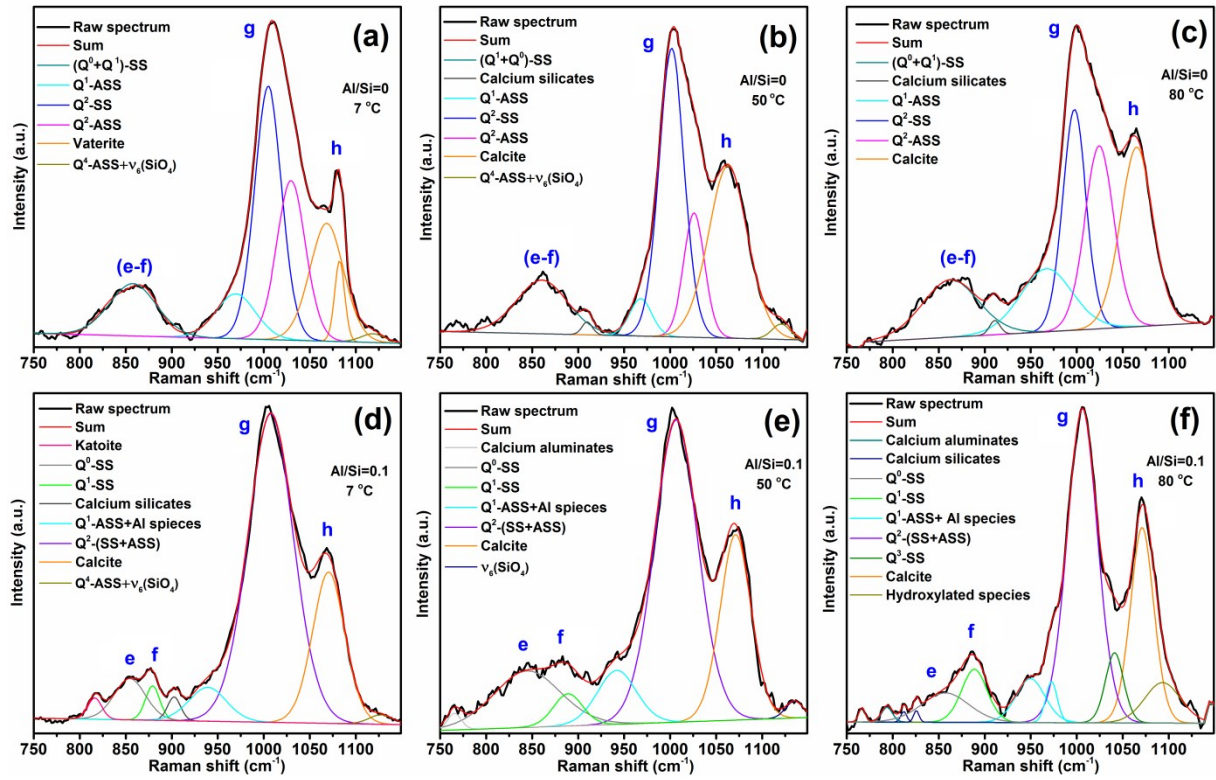


Figure S6. Deconvoluted spectra showing the effect of synthesis temperature on C-S-H at a) 7 °C , b) 50 °C , c) 80 °C and C-A-S-H samples with Al/Si=0.1 at d) 7 °C, e) 50 °C, f) 80 °C. The images have been extracted from Figure 5 at the range of 750 -1150  $\text{cm}^{-1}$ . For all samples, Ca/Si= 1.0.