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In Situ Observation of Wollastonite Carbonation in NaHCO_3 Aqueous Solution at Elevated Temperatures and Pressures via Raman Spectroscopy

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Mineral carbonation is a safe way for long term storage of CO_2 , since abundant natural silicate minerals such as wollastonite, serpentine and olivine, can trap CO_2 as stable carbonates, and have high capacity of storage. However, the natural carbonation process of silicate minerals is very slow, it is worthy to study how to accelerate the carbonation of silicate minerals. In this study, the experiments of wollastonite carbonation in NaHCO_3 aqueous solution were conducted in capillary high pressure optical cell (Chou et al., 2005) at elevated temperatures of 120, 150, 200 °C and pressures of 4, 7, and 20 MPa. The transition of wollastonite to calcium carbonates was monitored via in-situ Raman Spectroscopy.

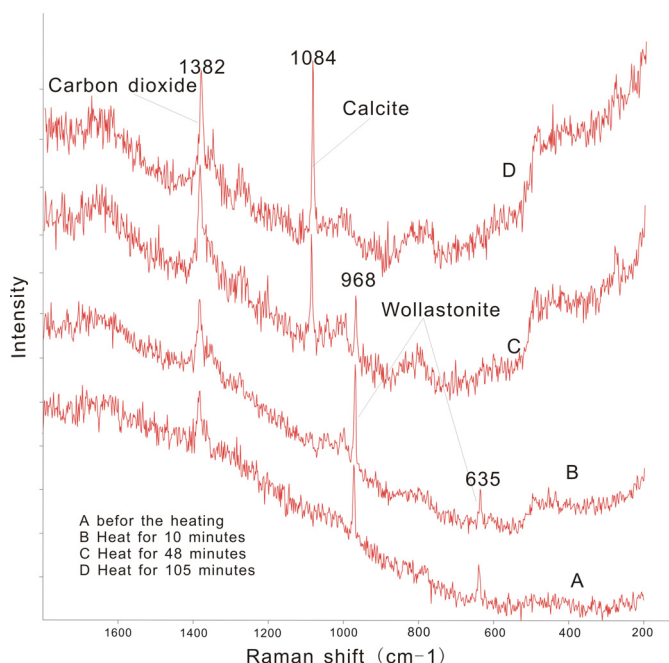


Fig. 1. Raman spectra collected on the surface of a wollastonite grain in 0.1 M NaHCO_3 solution at 120 °C, a peak at 1084 cm^{-1} occurred about two hours after heating indicates that calcite precipitated on the surface of the initial wollastonite grains

Calcium carbonates were detected on the surface the

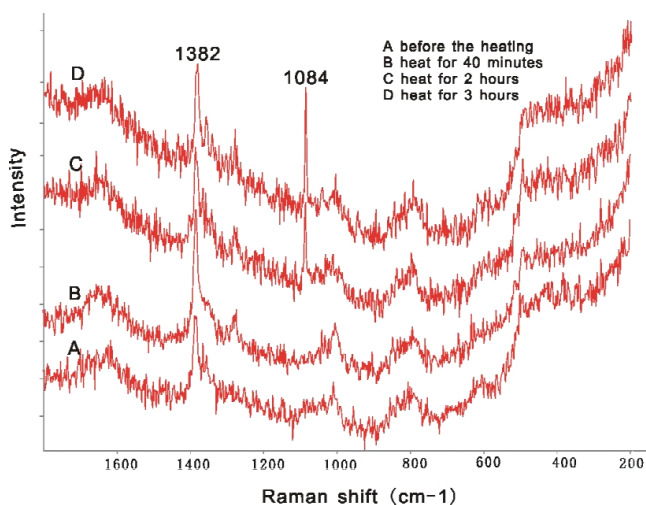


Fig. 2. Raman spectra collected for a section of 0.1 M NaHCO_3 solution away from wollastonite grains at 120 °C, a peak at 1084 cm^{-1} occurred about two hours after heating indicates that calcite deposited in the solution.

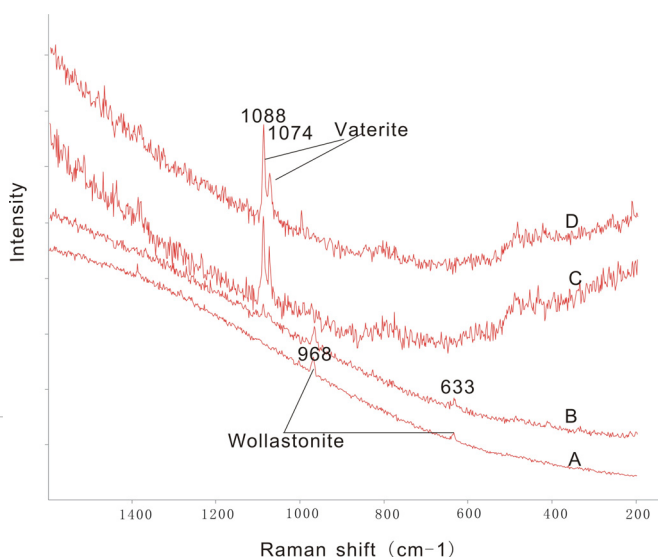


Fig. 3. Raman spectra of vaterite from wollastonite carbonation in 0.1 M NaHCO_3 at 120 °C

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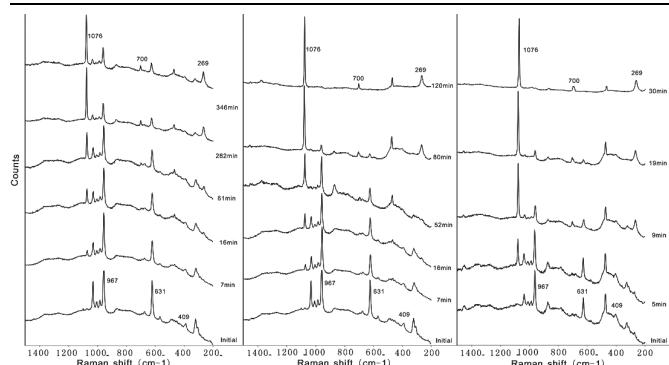


Fig. 4. Raman spectra collected at the surface of a wollastonite grain just located at the interface of the CO_2 -rich vapor and the 0.5 M NaHCO_3 solution at 120, 150 and 200°C, 7 MPa. A peak at 1076 cm^{-1} occurred earlier and the peak intensity was stronger with the increase of temperature that indicates the increased temperatures could accelerate the carbonation reaction.

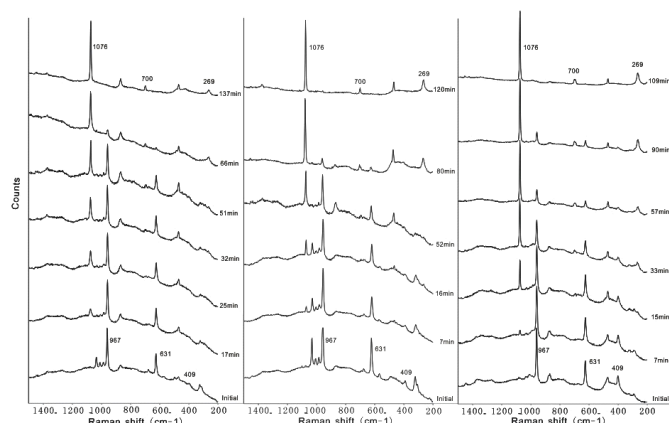


Fig. 5. Raman spectra collected at the surface of a wollastonite grain just located at the interface of the CO_2 -rich vapor and H_2O , 0.1 M and 0.5 M NaHCO_3 solution at 7 MPa, 150 °C. A peak at 1076 cm^{-1} occurred earlier and the peak intensity was stronger in the NaHCO_3 solution than that in the H_2O and higher NaHCO_3 solution concentration the peak at 1076 cm^{-1} occurred earlier and the peak intensity was stronger. The figure indicated the NaHCO_3 solution could accelerate the carbonation reaction.

wollastonite grains in both in the CO_2 rich vapor phase and aqueous solution (Fig. 1, 2), as well as in aqueous solution away from the wollastonite grains (Fig. 2), which indicates that Ca^{2+} can dissolve and diffuse into the solution and react with dissolved carbon dioxide. Calcium carbonates precipitated initially in the forms of vaterite, and transited to calcite eventually (Fig. 3). Experiments performed at different temperatures and pressures showed that increasing temperature could accelerate the carbonation reaction (Fig. 4), increasing pressure has smaller effect on acceleration of carbonation reaction. The observation of wollastonite carbonation in 0, 0.1, and 0.5 M NaHCO_3 solution at 7 MPa, 150 °C shows that, increasing concentration of NaHCO_3 can enhance the wollastonite carbonation in the solution (Fig. 5).

Key words: wollastonite carbonation; NaHCO_3 solution; Raman Spectroscopy

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