

Carbonate polymorphism controlled by iron redox dynamics at a natural CO₂ leakage site (Crystal Geyser, Utah)

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Crystal Geyser (Utah, USA) is a CO₂-rich low-temperature geyser and a natural analog for CO₂ leakage from geological storage sites [1]. We performed a detailed mineralogical characterization of fast-forming iron-rich calcium carbonate pisoids and terraced travertines precipitating from the fluids expelled by the geyser. The pisoids, located within a few meters from the geyser vent, are composed of concentric layers of aragonite and calcite. Calcite layers contain abundant ferrihydrite shrubs (frutexites) in which iron minerals are encasing bacterial forms (possibly iron-oxidizing bacteria which are abundant in the geyser waters [2]). The aragonite layers contain less abundant and finely dispersed iron, present either as iron-oxide microspherules or iron adsorbed to organic matter dispersed within the aragonite matrix. In the travertines, found more distally with respect to the geyser, aragonite and calcite do not form distinct layers but are finely intermixed. Iron is present as ferrihydrite dispersed in the carbonate matrix, and no spatial correlation between carbonate polymorphs and iron distribution is observed. We propose that carbonate polymorphism in the pisoids is mostly controlled by local fluctuations of the iron redox state of the fluids, caused by the geyser eruption cycle and episodic blooms of iron-oxidizing bacteria. Indeed, the waters expelled by Crystal Geyser contain >200 μM Fe²⁺, a known inhibitor of calcite growth. The calcite layers of the pisoids may record episodes of microbial iron oxidation, consistent with observations of iron-rich biofilms thriving in the stagnant pools around the geyser between eruptions. In turn, calcium carbonate polymorphism in the travertines is uncorrelated with iron speciation in the fluid, which in the travertine formation area contain relatively small amounts of dissolved Fe²⁺ (~1 μM). Calcium-carbonate polymorphism in the travertine may be controlled by other parameters such as CO₂ degassing and CaCO₃ precipitation rates. This study documents geochemical and microbial processes influencing mineral formation in CO₂-rich environments, and informs our understanding of the fate of CO₂ leaked from geological storage sites.

[1] Burnside, Shipton, Dockrill & Ellam (2013), *Geology* 41, 471–474.

[2] Emerson, Thomas, Alvarez & Banfield (2016), *Environmental Microbiology* 18:6, 1686–1703.