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Magnesite formation in playas: A natural analogue for carbon sequestration

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Non-marine carbonate deposits are of renewed interest as natural analogues for carbon sequestration and storage. Specifically, the sequestration of anthropogenic carbon dioxide (CO₂)in Mg-carbonate minerals is being actively investigated as a strategy for reducing greenhouse gas emissions¹. In northern British Columbia, hydromagnesitemagnesite playas (hectare-scale) have formed since the last deglaciation, suggesting that these minerals possess a level of stability required for long-term carbon storage². Quantitative mineralogical and hydrogeochemical data, as well as microscopy and field observations, were used to formulate a comprehensive facies model that describes the depositional environments for the formation of these playas. Over several millennia, there have been transitions from deposition of siliciclastic to subaqueous Ca-Mg-carbonate to subaerial Mg-carbonate sediments^{3,4}. Consequently, a complex assemblage of carbonate minerals is present within the playas including magnesite [MgCO₃], the most stable Mg-carbonate for storing CO₂. Magnesite precipitation at near-surface temperatures is kinetically inhibited due to the strong hydration of Mg²⁺ ions in solution⁵. Thus, understanding the rates of, and controls on, magnesite formation at low temperatures remains a challenge. Magnesite abundances at the surface (1 to 41 wt.%) and at depth (1 to 86 wt.%) within the playas are highly variable⁴. There is a propensity for hydrated Mg-carbonate minerals to undergo transformation to less hydrated, more stable forms (lansfordite > nesquehonite > dypingite > hydromagnesite)⁵; however, stable, radiogenic, and clumped isotope⁶ data as well as electron microscopy demonstrate that magnesite formation is likely dominated by direct precipitation from aqueous solution in the shallow subsurface (~3-10 °C). An observed variation in magnesite crystal morphology with depth is attributed to different crystal growth mechanisms induced by changes in magnesite saturation state. Particle size analyses show a positive correlation between magnesite abundance and mean particle size, indicating that magnesite formation is primarily limited by nucleation rather than crystal growth kinetics. We estimate that the rate of magnesite formation (nucleation + growth) is between 10^{-17} to 10^{-16} mol/cm²/s. Conversely, in the Ca-Mg-carbonate unit, magnesite may be forming via diagenesis of Ca-carbonate minerals. Our continued focus is to further constrain the rates and modes of magnesite formation in the context of long-term storage of CO₂.

[1] Power et al. (2013) *Rev. Mineral. Geochem.* **77**: 305-360. [2] Power et al. (2009) *Chem. Geol.* **206**: 302-316. [3] Power et al. (2007) *Geochem. Trans.* **8**: 13. [4] Power et al. (2014) *Sedimentology.* **61**:1701-1733. [5] Hänchen et al. (2008) *Chem. Eng. Sci.* **63**: 1012-1028. [6] Streit Falk and Kelemen, unpublished data.