Magnesite formation at Earth's surface

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Magnesite ($MgCO_3$) deposits and mineralization pathways are of renewed interest in the context of CO_2 sequestration strategies. The kinetic inhibition of magnesite precipitation at low temperature has hindered the understanding of natural magnesite formation and restricted technologies that capture and store CO_2 within magnesite to costly high-temperature systems. Thus, there is a need to better understand the fundamental geochemical processes of magnesite formation at Earth's surface over geologic timescales and to devise reaction pathways for accelerating magnesite formation at low temperature. Hydromagnesite-magnesite playas host a complex assemblage of carbonate minerals including abundant magnesite, up to ~90 wt.%. Stable, radiogenic, and clumped isotope data in concert with electron microscopy demonstrate that magnesite forms by direct precipitation from aqueous solution in the shallow subsurface at low temperature (~3-10 °C). Magnesite particle size distributions indicate that its formation is nucleation-limited with long induction times; while changing magnesite crystal morphology with depth suggests variable crystal growth mechanisms and formation rates (nucleation + crystal growth). We determine those rates to be in the range of 10^{-17} to 10^{-16} mol/cm^2/s based on field data. Furthermore, the abundance of magnesite with respect to hydromagnesite increases over time as its formation is *reaction-controlled* rather than *transport-controlled* for hydromagnesite. In microcosm experiments aimed at accelerating magnesite formation, carboxylated polystyrene microspheres caused desolvation of Mg^{2+} ions, minimizing the rate limitation and inducing magnesite precipitation at room temperature over 72 days (Power et al., 2017, *Cryst Growth Des* **17**:5652–5659). In comparison to natural magnesite formation, this is an acceleration of several orders of magnitude without requiring energy input. Incorporating carboxylated surfaces into mineral carbonation processes or the use of such ligands for deep geologic CO_2 storage may offer novel and economically viable strategies for permanent carbon storage.