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Chemical Reactivity in Thin Water Films on Minerals in Wet-scCO$_2$

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In geologic carbon sequestration (GCS), CO$_2$ captured from power plants or industrial sites is pumped into deep saline rock formations for long-term storage. Reservoir temperatures and pressures dictate that CO$_2$ is injected as a supercritical phase (scCO$_2$), and this fluid is usually anhydrous to prevent corrosion in transport pipe and well casings. However, with exposure to water in the reservoir, the CO$_2$ becomes spatially distributed as either variably wet scCO$_2$ or dissolved in an aqueous phase. Knowledge of the interactions between minerals and wet scCO$_2$ is of equal if not greater importance to the processes in aqueous dominated systems. Understanding mineral reactivity and transformations in wet scCO$_2$ is critical to reservoir site selection, operational efficiency and safety, and prediction of near- and long-term fate of injected CO$_2$.

In this presentation, we report laboratory experimental results, emphasizing *in situ* observations, on reactivity in wet scCO$_2$ of a range of natural and synthetic minerals that either are relevant to caprock and host rock in deep saline rock formations or are proxies to obtain insights into reaction processes unique to water bearing scCO$_2$ fluids. Minerals include montmorillonite, antigorite, enstatite, feldspar, brucite, fayalite, and forsterite. We show that in most cases, these minerals become coated in Ångstrom to nanometer thin, confined H$_2$O films when they are exposed to wet scCO$_2$. Transformations of minerals covered in thin films of adsorbed H$_2$O occur under unique chemical and physical constraints. For most minerals, there is evidence for a threshold adsorbed H$_2$O concentration, before which carbonation is limited but beyond which crystalline M^{2+}-carbonates continuously precipitate. Furthermore, surprising chemistry can occur just beyond the threshold, such as the emergence of relatively large μm-sized carbonate crystals, the growth of which is entirely mediated by nm-thick adsorbed H$_2$O films.

We detail our current understanding of the role of thin water films in mediating mineral carbonation reactivity in water-bearing scCO$_2$. We also state several hypotheses on water-film reactivity in low-water CO$_2$ fluids that are the subject of current and future research. This work provides important insights that will enhance modelling efforts designed to predict the fate and transport of subsurface stored CO$_2$. More broadly, our results fill a major scientific knowledge gap concerning interfacial chemistry in thin water films and mineral reactions in high-pressure fluids.