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# Low Temperature Magnesite Growth during Forsterite Carbonation in Thin $H_2O$ Films

**JOHN LORING**[1], SEBASTIAN MERGELSBERG[2], ODETA QAFOKU[2], CHRISTOPHER J THOMPSON[1], EUGENE S. ILTON[1] AND SEBASTIEN KERISIT[1]

[1]Pacific Northwest National Laboratory
[2]Pacific Northwest National Lab
Presenting Author: john.loring@pnnl.gov

Minerals in low-water environments are covered in Ångstrom-to-nanometer thin $H_2O$ films with unique properties that can lead to mineral transformation pathways that are very different from those in bulk water. For example, $H_2O$ films have extraordinarily high mineral-surface-to-water-volume ratios; thus, relatively minor amounts of mineral dissolution can rapidly yield high degrees of supersaturation with respect to secondary phases. Despite numerous examples of $H_2O$-limited domains in nature, we currently lack a fundamental understanding of coupled mineral dissolution, nucleation, and growth reactions in confined adsorbed $H_2O$ films.

Here, we present results of an investigation of the low temperature and low $H_2O$ activity carbonation pathway of forsterite ($Mg_2SiO_4$) to magnesite ($MgCO_3$) in thin $H_2O$ films in humidified high-pressure $CO_2$. Experiments were conducted on nanometer-sized forsterite at 25, 40, and 50 °C in 90 bar $CO_2$ at a relative humidity 62%, which corresponds to a $H_2O$ film thickness of 1.78 monolayers. *In situ* infrared (IR) spectroscopy was used to monitor forsterite carbonation as a function of time, and post reacted samples were characterized *ex situ* by scanning electron microscopy (SEM), high-energy X-ray diffraction (heXRD), and pair distribution function (PDF) analyses. These combined results evidenced for the first time a carbonation pathway of forsterite to magnesite through an amorphous magnesium carbonate (AMC) metastable phase [1].

Chemometric modeling of the IR spectroscopic data quantified the dissolution of forsterite and the formation of both AMC and magnesite as a function of time. Fitting of these data with a kinetic geochemical model in PHREEQC showed that magnesite grew in thin $H_2O$ films following a pseudo zero-order rate law, and magnesite remained supersaturated by two to three orders of magnitude. Hence, high reactant concentrations are the main drivers for why magnesite growth kinetics are enhanced during forsterite carbonation in thin $H_2O$ films [2].

This study furthers an understanding of divalent silicate carbonation under low-$H_2O$ $CO_2$-dominated fluids as part of a comprehensive effort to predict the fate of $CO_2$ stored in geologic reservoirs.

[1] Mergelsburg et al. (2020), Chem. Comm., 56, 12154-12157.

[2] Kerisit et al. (2021), ES&T Letters, Submitted.