

Nucleation of magnesium carbonate on quartz surfaces using *in situ* GISAXS

NAMHEY LEE^{1*}, BYEONGDU LEE²
AND GLENN A. WAYCHUNAS¹

¹Earth Science Division, Lawrence Berkeley National
Laboratory, Berkeley, CA, USA, nlee@lbl.gov

²Chemical and Material Science, X-ray Science Division,
Argonne National Laboratory, Argonne, IL, USA

Carbon trapping and sequestration involves the conversion of CO₂ into solid carbonate minerals, mostly to form calcium and magnesium carbonates due to these elements' natural abundance. The high concentration of divalent cations are provided by dissolution of mineral phases present in sites used for sequestration or by fluids in saline aquifers. Magnesium ion is found to be particularly rich in ultramafic rocks provided by minerals like olivine and brucite. However despite the abundance in many natural environments, magnesium does not readily form carbonate minerals at ambient conditions because of its strongly bound hydration sphere. Nevertheless, in reservoir conditions, the moderately elevated temperature (<100 °C) allows carbonate to directly bind with magnesium ion, resulting in significant precipitation of magnesium carbonate. Because of this requirement of higher temperatures, our observations and understanding on magnesium carbonate nucleation under sequestration conditions is rather limited.

Here we studied the nucleation of magnesium carbonate on quartz (100) surfaces under field relevant temperature conditions of 40 - 60°C at 1 bar. Grazing-Incidence Small-Angle X-ray Scattering (GISAXS) was used to measure the nucleation rate on quartz surfaces at varying saturation indices and temperatures. The nucleation rate was extracted using the invariant (Q), which is proportional to the total volume of the scattering particles. The data suggest that the precipitation is nucleation dominated. From these measurements, critical values directly applicable to field and modeling applications can be extracted, such as nucleation activation energy (E_a) and effective interfacial energies (α'). Further modeling analysis of the data will provide information on changes in the distribution of sizes and shape with particle evolution. The presence of mineral surfaces have been shown to lower the free energy barrier for nucleation. Hence quantification of the thermodynamic properties of heterogeneous nucleation will promote more accurate understanding and prediction of mineral trapping processes occurring in reservoir environments.