Nanoscale Chemical/Structure Analysis of Geological Minerals for Carbon Sequestration using Atom Probe Tomography

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Geological carbon sequestration is explored as one of the dominant means to mitigate the ever-growing anthropogenic CO_2 emission. Site-specific reactivity of minerals with supercritical CO_2 is of great interest in understanding the fundamental elementary reaction mechanisms, where the presence of impurities within the bulk mineral may affect reaction kinetics. A combination of atom probe tomography (APT) and scanning transmission electron microscopy (STEM) is being used to map the complex composition and nanometer-scale structure across various site-specific regions in order to better understand the mechanism upon reaction with sc-CO_2. The site-specific APT analysis of unreacted natural fayalite indicates the presence of nanometer scale hydrated iron oxide interfaces. In addition, Na impurities were found to concentrate within the hydrated interface while Mg and Mn were depleted from these regions. With the ability of APT to detect the chemical/structural heterogeneity by virtue of its high three-dimensional atomic-scale spatial resolution and ppm sensitivity, we find that APT will provide a viable means to correlate with ongoing experimental reaction studies and also provide guidance into theoretcial models of the heterogenous phase formation and reaction rates at precisely defined interfaces within minerals.