

Visualizing calcite dissolution in the presence of Pb^{2+} by 3D coherent X-ray diffraction imaging

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Crystallographic defects and strain can exert significant impact on the reactivity of minerals, influencing the reaction pathways and kinetics. A mechanistic understanding of geochemical reactions can be obtained by imaging the displacement fields in individual particles at the atomic scale. Bragg coherent diffraction imaging (BCDI) is a unique technique to obtain such information as it is capable of reconstructing the 3D electron density (i.e., shape) and displacement fields of individual (nano)particles simultaneously [1]. Here, we present BCDI results showing the strain and morphological evolution of individual calcite (calcium carbonate, CaCO_3) particles under cycles of dissolution in Pb^{2+} -containing aqueous solutions at acidic pH. Pb^{2+} is known to decrease calcite dissolution rates and alter the dissolution patterns with facets dissolving first instead of edges and corners [2]. These processes were interpreted to be controlled by the incorporation of Pb^{2+} in the calcite surface. Our BCDI results provide direct evidence that the dissolution reaction occurs preferentially on the most strained regions of the crystals, demonstrating a relationship between the strain and reactivity. They also anticipate how increased brightness of coherent x-rays through the application of the multi-bend achromat lattice technology, such as Advanced Photon Source Upgrade (APS-U), enables faster in situ measurements of relevant minerals with improved resolution, opening new opportunities for understanding the structural controls over the reactivities of minerals in various geochemical systems.

[1] Robinson, I.; Harder, R. Coherent X-Ray Diffraction Imaging of Strain at the Nanoscale, *Nat. Mater.* **2009**, 8, 291.

[2] Yuan, K. *et al.* Pb^{2+} -Calcite Interactions under Far-from-Equilibrium Conditions: Formation of Micropyramids and Pseudomorphic Growth of Cerussite. *J. Phys. Chem. C* **2018**, 122, 2238–2247.