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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 simultaneousl [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.

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