Abstract

The purpose of this work was to examine the effects of polished crystal-surface orientation and degree of solution undersaturation (\( \Delta \)) on the dissolution kinetics of calcite as a means of improving our understanding of fundamental reactions that may influence the efficacy of CO2 sequestration in geological formations. Crystallographic surface orientations utilized in this study included \( \sim 1 \text{ cm}^2 \) areas of natural calcite specimens polished approximately parallel to the plane, giving rise to surfaces with flat terraces with few steps, as well as fully kinked surfaces created by sectioning approximately parallel to the plane. Results from inductively coupled plasma (ICP-OES) and vertical scanning interferometry (VSI) investigations revealed how crystallographic orientations of calcite with higher initial surface morphologies were associated with greater Ca\(^{2+}\) release, greater surface retreat, and therefore, greater initial transient dissolution rates than those with lower initial surface morphologies. However, both the ICP-OES and atomic force microscopy (AFM) results confirm that the effects of crystal orientation become minimal under long-term conditions since (1.) varyingly oriented calcite surfaces exhibited similar —steady‖ rates and (2.) orientations with high initial reactive site densities developed lower energy morphologies. Results from this study
are significant for predicting long term calcite dissolution rates because they suggest the —steady‖ dissolution rate of any calcite surface with any degree of initial surface energy will be similar to that of a surface with natively low surface energy.