Dissolution kinetics of polycrystalline calcite

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Carbonate minerals are a significant component of sedimentary rocks and modern sediments. Their dissolution kinetics play an important role in many energy, economic and environmental problems: CO_2 sequestration, petroleum and geothermal reservoir characteristics, and ocean acidification provide diverse examples. Consequently, there is a need for a fundamental mechanistic understanding of dissolution kinetics, potentially yielding a realistic predictive capability. The large discrepancy of about 2-3 orders of magnitude of calcite dissolution rate data under identical chemical conditions hampers any authoritative predictions [1]. Instead of a single mean rate, the rate spectrum concept identifies critical components that combine to an overall reaction rate. The analysis of such concurrently active rate components provides quantitative insight into reaction rate evolution [2, 3].

In this study, we use this concept for the analysis of dissolution kinetics of polycrystalline calcite dissolution as a function of temporal and spatial scales. Two types of calcite marbles [4] are investigated, using a flow-through-cell and reaction periods of 2, 4, 6, 10 and 16 hours. Interferometric microscopy data are used to calculate rate maps that provide a graphic visualization of the spatial distribution of material flux during dissolution. A histogram analysis of these maps leads to multimodal rate spectra, and their deconvolution provides mechanistic information concerning the temporal evolution of critical rate components, as well as insight into the reaction kinetics of heterogeneous surfaces. We discuss the quantitative impact of crystal orientation, defect type and distribution, and grain boundary density in order to provide realistic constraints for predictive models.

[1] Arvidson et al. (2003), GCA **67**, 1623-1634. [2] Fischer et al. (2012), GCA **98**, 177-185. [3] Fischer & Luttge (2017), EPSL **457**, 100-105. [4] Zeisig et al. (2002), Geol Soc Spec Publ **205**, 65-80.