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Quantitative Analysis of Solid Solution Dissolution Kinetics

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During the last few decades, significant descrepancies between field and laboratory-derived dissolution rates have complicated the formulation of an empirical rate law of general application and significance, as it has been shown that measured rates are sensitive to a complex interaction of extrinsic and intrinsic parameters [1]. Whereas the influences of extrinsic parameters such as pH and temperature are quite well understood, variabilities in the surficial distribution of rates reflecting intrinsic mineral controls are as yet poorly understood, and require careful and systematic investigation.

In this study, we focus on the variability in surface reactivity as a function of compositional variability in single zoned crystals. The key motivation is to provide quantitative constraints to the range of reactivity in solid solutions. We utilize a combination of vertical scanning interferometry (VSI) and RAMAN spectroscopy. VSI data provide surface reaction rate maps and the identification of critical rate components that combine to produce an overall rate [2]. RAMAN microscopy data collected in parallel provide spatially-resolved chemical information. A time series of high-precision measurements of reacting surfaces that include their chemical signature provides details of reaction kinetics and controls via chemical feedback of reacting surfaces.

As a first example, we present dissolution data of zoned alkali feldspars. Flow-through reactor cell experiments were carried out far from equilibrium under alkaline conditions and elevated temperatures. We show how spatial patterns of reactivity identified in rate map data relate to compositional variation. These data are then used as the basis of a quantitative model of solid solution dissolution kinetics.

[1] White & Brantley (2003) *Chemical Geology* **202**, 479-506. [2] Fischer *et al.* (2014) *Applied Geochemistry* **43**, 132-157.