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Statistical scaling of nanoscale spatially heterogeneous dissolution rates at mineral-water interfaces

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Dissolution and precipitation processes taking place at the interface between water and the solid matrix of the host porous medium are key drivers for chemical weathering of minerals in subsurface environments. Direct high-resolution imaging of mineral substrates subject to dissolution/precipitation document that these reactions are driven by local mechanistic phenomena originating at natural defects embedded in the mineral lattice. These are typically viewed as randomly distributed across the mineral system and constitute a major element imprinting the pattern associated with the dynamic self-organization of the water-mineral interface. While their presence triggers the formation of characteristic interfacial patterns starting from the nanoscale, their action manifests across diverse spatial scales. It ultimately yields starkly heterogeneous distributions of reaction rates that can display several-fold variations across the same mineral surface. Sample probability density functions of mineral dissolution rates are documented to display complex non-Gaussian behavior. When homogenized across a given window of observation on a crystal surface, these distributions are characterized by multiple modes. These are, in turn, linked to mechanistic processes driving the reaction. Advancing our fundamental understanding and modeling capabilities of environmental scenarios underpinned by chemical weathering processes (including, e.g., carbon sequestration, pollutant migration/stabilization, or fracture morphology dynamics) requires incorporating spatial heterogeneity of reaction rates into reactive transport models. This spatial heterogeneity, which fundamentally originates at the nanoscale, plays a critical role in shaping system behavior. Accurately capturing and representing the statistical characteristics associated with these nanoscale variations within modeling frameworks is key for improving model results and fostering robust interpretations of these complex environmental phenomena.

In this broad context, understanding and providing a sound representation of the way in which key traits emerging from stochastic analysis of data transition with scale is a major research aspect. Here, we focus on calcite-water interfaces as a model geochemical system. We consider nanoscale spatial distributions of dissolution rates obtained from Atomic Force Microscopy measurements of the topography of a calcite sample subject to dissolution under continuous flow conditions. We investigate the scaling behavior of the ensuing random fields upon analyzing sample structure functions. The latter correspond to absolute q -th order statistical moments of spatial increments (i.e., differences between values of observed dissolution rates taken between two locations separated by a given distance or lag). Our analyses document that dissolution rates exhibit two distinct power-law scaling regimes. These are in turn associated with diverse degrees of persistence, as rendered through the classical Hurst exponent. We then provide a direct link between the occurrence of such regimes and the nanoscale mechanistic processes driving the evolution of the mineral-water interface.

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References

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