Pulsating dissolution of crystalline matter

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The reaction of crystalline material with fluids is of relevance for natural and technical processes. A basic assumption has been that the reaction products are continuously released from the crystal surface. New experimental and analytical results show something fundamentally different: Material is released in a series of reaction pulses [1]. We present reaction rate maps that are derived from sequences of topography maps and quantify the spatial distribution of reaction rates across the crystal surface. The first (rate acceleration) and second (rate jerk) temporal derivative of the rate quantify the dynamic formation and loss of reactive surface sites. The resulting variability in nanoscale roughness is a key factor that controls kink-site distribution and density and therefore may help explain why laboratory (bulk) dissolution rates are so variable.

Applied and theoretical implications impact both the upscaling of crystal dissolution kinetics, and more importantly, the problem of how dissolution and growth are connected via the equilibrium state. These results challenge the prevailing view that crystal dissolution is simply the inverse process of continuous crystal growth at crystal dislocations. Consequently, we need to examine how macroscopic crystal equilibrium reflects continuous or discontinuous processes in the microscopic state.

[1] Fischer, C., Luttge, A., 2018. Pulsating dissolution of crystalline matter. PNAS 115, 897-902.