

Is less more? Kinetic simulations of the dissolution of whole mineral grains

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It has been recognized since the work of Frank, Burton, and Cabrera [1,2] that mineral reactivity is fundamentally tied to the distribution of screw dislocations. These defects provide a continuous supply of sites during crystal growth at small departures from equilibrium, and more recently have also been recognized as a critical mechanism for dissolution as well [3]. However, during the dissolution of mineral grains, macroscopic discontinuities such as grain, twin, and related interfacial boundaries are also commonly recognized as preferred reactive sites. The importance of these sites should also increase with decreasing crystal diameter, implying complex changes in behaviour during the dissolution and growth of the entire crystal. We wish to understand these changes in a general way, and how they can be related to the integrated flux of material from the particle. This general kinetic problem can be best addressed by kinetic Monte Carlo modelling, beginning with simulations of dissolution and growth using a simple cubic Kossel crystal, and culminating in virtual experiments involving complex crystal structures. These simulations afford a practical approach to a variety of questions: (i) what is the contribution of these features to the integrated rate as a function of crystal diameter; (ii) how are the uptake and release at kink sites coordinated with diffusive transport; (iii) what is the role of reversible attachment and detachment operations during dissolution and growth; (iv) how does the area-specific reaction rate at edge sites compare to those on interior terraces as a function of defect density; and (v) what are the implications of this behaviour for poly-crystalline and nano-particulate materials.

1. Frank FC, Discuss. Faraday Soc. 5:48–54 (1949)
2. Burton WK, Cabrera N, & Frank FC, Phil. Trans. Roy. Soc. London Ser. A 243:299–358 (1951)
3. Lasaga AC & Luttge A, Science 291:2400–2404 (2001)