

DEDUCING GROWTH MECHANISMS FOR MINERALS FROM THE SHAPES OF CRYSTAL SIZE DISTRIBUTIONS

D. D. EBERL*, V. A. DRITS**, and J. ŚRODON***

ABSTRACT. Crystal size distributions (CSDs) of natural and synthetic samples are observed to have several distinct and different shapes. We have simulated these CSDs using three simple equations: the Law of Proportionate Effect (LPE), a mass balance equation, and equations for Ostwald ripening. The following crystal growth mechanisms are simulated using these equations and their modifications: (1) continuous nucleation and growth in an open system, during which crystals nucleate at either a constant, decaying, or accelerating nucleation rate, and then grow according to the LPE; (2) surface-controlled growth in an open system, during which crystals grow with an essentially unlimited supply of nutrients according to the LPE; (3) supply-controlled growth in an open system, during which crystals grow with a specified, limited supply of nutrients according to the LPE; (4) supply- or surface-controlled Ostwald ripening in a closed system, during which the relative rate of crystal dissolution and growth is controlled by differences in specific surface area and by diffusion rate; and (5) supply-controlled random ripening in a closed system, during which the rate of crystal dissolution and growth is random with respect to specific surface area. Each of these mechanisms affects the shapes of CSDs. For example, mechanism (1) above with a constant nucleation rate yields asymptotically-shaped CSDs for which the variance of the natural logarithms of the crystal sizes (β^2) increases exponentially with the mean of the natural logarithms of the sizes (α). Mechanism (2) yields lognormally-shaped CSDs, for which β^2 increases linearly with α , whereas mechanisms (3) and (5) do not change the shapes of CSDs, with β^2 remaining constant with increasing α . During supply-controlled Ostwald ripening (4), initial lognormally-shaped CSDs become more symmetric, with β^2 decreasing with increasing α . Thus, crystal growth mechanisms often can be deduced by noting trends in α versus β^2 of CSDs for a series of related samples.

INTRODUCTION

Crystal size distributions (size versus frequency plots, or CSDs) of minerals often have distinctive shapes which can convey information about crystal growth history (Wagner, 1961; Baronnet, 1982). A CSD first develops during nucleation, when crystals with sizes equal to or greater than that of the critical nucleus appear and grow in highly supersaturated solutions. If solution supersaturation falls to a level at which nucleation ceases, the CSD may continue to evolve by open system crystal growth without additional nucleation. The kinetics for this growth will be controlled either by how fast the crystal's surface grows given an essentially infinite supply of nutrients (surface-controlled growth kinetics) or may be limited by the rate at which nutrients reach the growing surface (supply-controlled kinetics). As supersaturation approaches equilibrium, less stable crystals may dissolve, yielding material for other crystals of the same phase to grow during a ripening process, thereby again modifying the shape of the CSD. Finally, crystals also may grow by agglomeration with other crystals, without significant dissolution.

Attempts to model crystal nucleation and crystal growth from classical kinetic theory have been disappointing (Ohara and Reid, 1973; Mullin, 1974; Dowty, 1980; Kirkpatrick, 1981; Lasaga, 1982). Some workers have abandoned such models altogether in favor of a chemical engineering approach that uses empirical methods, developed from the measurement of CSDs for crystals grown in industrial crystallizers, to describe nucleation and growth rates for minerals (Randolph and Larson, 1971;

* United States Geological Survey, 3215 Marine Street, Boulder, Colorado 80303-1066

** Institute of RAN, Pyzevskij per. D. 7, 109017 Moscow, Russia

*** Institute of Geological Sciences PAN, Senacka 1, 31002 Krakow, Poland

