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On the origin of size-dependent and size-independent crystal growth: Influence of advection and diffusion

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ABSTRACT

Crystal growth experiments were conducted using potassium alum and calcite crystals in aqueous solution under both non-stirred and stirred conditions to elucidate the mechanism for size-dependent (proportionate) and size-independent (constant) crystal growth. Growth by these two laws can be distinguished from each other because the *relative* size difference among crystals is maintained during proportionate growth, leading to a constant crystal size variance (β^2) for a crystal size distribution (CSD) as the mean size increases. The *absolute* size difference among crystals is maintained during constant growth, resulting in a decrease in size variance. Results of these experiments show that for centimeter-sized alum crystals, proportionate growth occurs in stirred systems, whereas constant growth occurs in non-stirred systems. Accordingly, the mechanism for proportionate growth is hypothesized to be related to the supply of reactants to the crystal surface by advection, whereas constant growth is related to supply by diffusion. Paradoxically, micrometer-sized calcite crystals showed proportionate growth both in stirred and in non-stirred systems. Such growth presumably results from the effects of convection and Brownian motion, which promote an advective environment and hence proportionate growth for minute crystals in non-stirred systems, thereby indicating the importance of solution velocity relative to crystal size. Calcite crystals grown in gels, where fluid motion was minimized, showed evidence for constant, diffusion-controlled growth. Additional investigations of CSDs of naturally occurring crystals indicate that proportionate growth is by far the most common growth law, thereby suggesting that advection, rather than diffusion, is the dominant process for supplying reactants to crystal surfaces.