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The carbonatation of gypsum: Pathways and pseudomorph formation

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Abstract

In this paper, we present an experimental study of the interaction between gypsum (010) surfaces and aqueous solutions of Na_2CO_3 with different concentrations. This interaction leads to the carbonatation (i.e., the transformation into carbonate minerals) of gypsum crystals, which under ambient conditions shows the characteristics of a mineral replacement and leads to the formation of pseudomorphs consisting of an aggregate of calcite crystals. Carbonatation progress was monitored by scanning electron microscopy (SEM) and glancing incidence X-ray diffraction (GIXRD). The carbonatation advances from outside to inside the gypsum crystal and occurs through a sequence of reactions, which involves the dissolution of gypsum and the simultaneous crystallization of different polymorphs of $CaCO_3$ [amorphous calcium carbonate (ACC), vaterite, aragonite, and calcite], as well as several solvent-mediated transformations between these polymorphs. The sequence in which $CaCO_3$ phases form is interpreted taking into consideration nucleation kinetics and the qualitative evolution of several chemical parameters in the aqueous solution. The textural characteristics of the transformed regions are described. The degree of faithfulness of the pseudomorphs obtained is related to the kinetics of the carbonatation process, which in turn depends on the initial concentration of carbonate in the aqueous solutions. Finally, changes in the rate at which the transformation front advances are discussed on the basis of both textural and physicochemical considerations.

Keywords: Carbonatation, dissolution-crystallization, mineral replacement, gypsum, calcium carbonate, pseudomorphism