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CARBONATION OF PORTLANDITE ($Ca(OH)_2$) - A PSEUDOMORPHIC, DISSOLUTION/PRECIPITATION REPLACEMENT REACTION

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The reaction between calcium hydroxide (portlandite) and carbon dioxide, the so-called carbonation, results in the formation of calcium carbonate ($Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$). This apparently straightforward reaction finds many scientific and industrial applications. For instance, homogeneous phase carbonation (i.e., in solution) results in precipitated calcium carbonate (PCC), which finds numerous applications as filler in paper, paint, food and pharmaceutical products. Heterogeneous phase carbonation (i.e., solid and fluid phases) is important in the field of geological storage of CO_2 (Regnault et al., 2009) and is crucial in the hardening of traditional lime mortars (Cowper, 1927). Research on the phase, morphology and texture evolution during portlandite carbonation is necessary to gain an insight on the behavior of lime mortars and plaster used in heritage conservation, as well as to understand the effects of cement carbonation in injection wells during underground CO_2 storage. Despite the numerous studies performed on the heterogeneous phase carbonation of portlandite, the mechanisms of this process are still poorly understood.

Here, the carbonation in air of two different types of portlandite crystals was studied: **A)** Hydrated lime powders (4 types, with different morphology, particle size, and surface area; all with a maximum particle size $< 10\ \mu$m) were mixed with de-ionized water to make a paste of appropriate consistency and placed in a plastic container with high RH (∼93%) at 20ºC. Samples were collected at different time intervals during a 4 months period and subsequently analyzed with XRD, TG, FESEM and TEM-SAED in order to calculate the degree of carbonation, texture and parent/product orientation relationship(s). The final morphology and the character of the crystal/aggregates depended on the type of hydrated lime, but the parent/product orientation relationship was always the same, with $(0001)_{portlandite}$//$(0001)_{calcite}$; **B)** Millimeter-sized portlandite single crystals, obtained by mixing NaOH with $CaCl_2$ (Johnston, 1914), were exposed to air at 20 ºC and ∼93% RH for over 2 months and analyzed with FESEM and 2D-XRD in order to determine the texture and parent/product crystallographic relationship(s) as carbonation progressed. Carbonation occurs via a pseudomorphic replacement reaction resulting in a highly oriented calcite. This process generates a significant amount of pores due to molar volume and solubility differences between $Ca(OH)_2$ and $CaCO_3$. The parent/product crystallographic relationship and the observed porosity generation are consistent with a coupled dissolution/precipitation replacement reaction involving a strong crystallographic control during the advancement of the reaction front.

References

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