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Solubility of Amorphous Magnesium Calcium Carbonate - Precipitation Experiments at 25°C

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Amorphous calcium carbonate (ACC) with variable Mg contents plays a significant role in calcium carbonate biomineralization processes of various organisms. In this context, it has been documented that ACC can accommodate up to 42 mol% Mg [1]. To date, however, the solubility of Mg-containing ACC (Mg-ACC) was not yet determined.

In an effort to shed light on the solubility product of Mg-ACC ($K_{Mg\text{-}ACC}$), precipitation experiments were carried out by the titration of a 0.6 M $(Ca,Mg)Cl_2$ solution at different Mg/Ca ratios (1/4 to 1/8) into a 1 M $NaHCO_3$ solution at 25°C and constant pH (8.3 or 8.8). The formation of Mg-ACC was monitored *in situ* by Raman spectroscopy, while homogeneous sub-samples of the solid and liquid phase were sampled over time in order to follow the chemical composition of the solution and precipitate. The activities of Ca and Mg ions in solution and the solid stochiometry were used to calculate ion activiy products ($IAP = K_{Mg\text{-}ACC} = (aCa^{2+})^{1-x} (Mg^{2+})^{x} (aCO_3^{2-})$) as a function of reaction time. Despite the fact that the Mg content of the forming ACCs ranges between 2 and 12 mol%, an almost constant $\log(K_{Mg\text{-}ACC})$ value of -6.07 ±0.07 were obtained for all conducted experiments. Previous work documented that the solubility of calcite increases with increasing uptake of Mg in its structure [2]. In contrast, our findings suggest that the fraction of $MgCO_3$ in ACC has no significant effect on the solubility of Mg-ACC. These differences may be best explained by the porous Mg-ACC structure and the high dynamic exchange between Mg-ACC and aquoeus solution. The obtained results are discussed together with literature data on ACC solubility and aim to shed light on the mechanisms of carbonate mineral formation via an ACC precursor in natural environments.

[1] Lin et al. (2015) *J. of Phys. Chem.* **C 119**, 7225-7233
[2] Busenberg and Plummer (1989) *Geochim. Cosm. Acta* **53**, 1189-12088