

The role of silica in the growth and stabilisation of Mg-carbonates

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Olivine addition to different natural media has been proposed as a potential method for the sequestration of CO₂ by facilitating its conversion into Mg-carbonate minerals. However, the breakdown of olivine that produces Mg²⁺ ions to react with bicarbonate or carbonate also releases silica. In nature, we observe that the direct products of olivine reactivity in aqueous conditions are typically dominated by Mg-silicates rather than Mg-carbonates [1], particularly in conditions with lower CO₂ partial pressures such as at the Earth's surface [2]. The formation of Mg-silicates can compete with the formation of Mg-carbonates, potentially decreasing the ability of the carbonate phases to form and thereby limiting CO₂ uptake. Furthermore, compared to Ca-carbonates, the formation of Mg-carbonates is a much more complicated process and at lower temperatures produces hydrated Mg-carbonates that may be metastable [3]. Here, we use crystal growth experiments and solution chemistry analysis coupled with phase identification to demonstrate that the presence of silica in solution can assist the formation of hydrated Mg-carbonate phases. In addition, in-situ observations of the transformation upon heating of precipitated Mg-carbonates phases also demonstrate that they become more stable if silica was present in the growth solution. This unexpected role indicates that silica in solution may enhance Mg-carbonate formation and that Mg-carbonate formation can out-compete Mg-silicate formation despite the much larger thermodynamic driving force for Mg-silicate precipitation as shown by PHREEQC calculations.

[1] Beinlich et al. *Terra Nova* 24.6 (2012): 446-455.

[2] Fuhr et al. *Frontiers in Climate* (2022): 39.

[3] Hänchen et al. *Chemical Engineering Science* 63.4 (2008): 1012-1028.