

## Magnesite formation during thermal decomposition of hydrated Mg-carbonates in a closed system: Winning back the CO<sub>2</sub>

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Carbon mineralisation is a promising technology for mitigating global warming by trapping atmospheric CO<sub>2</sub> within the structures of carbonate phases. Although highly-stable magnesite (MgCO<sub>3</sub>) is thermodynamically favoured to form at the Earth's surface, its precipitation from solution is inhibited by kinetics. Instead, magnesite forms via stepwise decomposition of hydrated Mg-carbonate minerals, such as nesquehonite (MgCO<sub>3</sub>·3H<sub>2</sub>O). It is therefore of fundamental importance to understand the decomposition pathways of these metastable hydrated phases, particularly given that they have recently been suggested as alternative long-term sinks for CO<sub>2</sub> [1, 2].

Here, we investigate thermal decomposition of dry nesquehonite within (a) an open vs. a closed system, and (b) with or without an external CO<sub>2</sub> source. Nesquehonite permanently releases structural CO<sub>2</sub> to form periclase (MgO) during decomposition in an open system at high temperatures (>300°C) when no external source of CO<sub>2</sub> is provided. Significantly, when confined to a closed system, CO<sub>2</sub> released from nesquehonite (and other metastable decomposition products) is reincorporated into the crystalline mass by magnesite formation.

This study improves the current understanding of nesquehonite decomposition and magnesite formation under varied environmental and engineered conditions, and explores a novel, stepwise route for CO<sub>2</sub> capture in MgCO<sub>3</sub>. Winning back the CO<sub>2</sub> generated during nesquehonite decomposition has the advantage of initial CO<sub>2</sub> capture in a low-temperature (and low-cost) mineral, followed by conversion of this metastable store into a permanent CO<sub>2</sub> sink.

[1] Ballirano *et al.* (2010) *J. Hazard. Mater.* **178**, 522–528. [2] Morgan *et al.* (2015) *Int. J. Greenh. Gas Control* **39**, 366–376.