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Complex reaction kinetics during direct air capture of CO_2 by MgO: what an improved fundamental understanding of mineral nucleation and growth can tell us.

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Limiting climate warming to less than 2 °C will likely require significant carbon removal from the atmosphere combined with geologic sequestration. MgO-based mineral looping direct air capture has been proposed as a potentially inexpensive and scaleable solution to capture atmospheric CO_2. However, the reaction kinetics and phase selection are mostly unknown. Here, we report first on our recent work to understand the reaction process of MgO with CO_2 and water vapor. To access decadal timescales, we conducted a "natural experiment" by characterizing the reaction layer of MgO crystals synthesized at ORNL in 1995 and compare that to short-term experiments on lab-reacted samples. Of particular interest are the reaction kinetics and phase selection. We find that the reacted layer acts to "armor" the material, slowing reaction rates over time, and that there are several different amorphous and crystalline phases forming. The net reaction rate on these crystals is more than two orders of magnitude slower than envisioned. However, parameters important to the rate, such as relative humidity and surface area, can be varied to enhance initial amounts of carbon capture. A yet-unsolved challenge in interpreting this data is determining whether the multiple reaction products represent a set of reactions in a linear series (e.g., Ostwald ripening), or whether reactions are occurring in parallel with multiple products forming simultaneously. This issue is an example of a deficiency in our ability to predict mineral reaction kinetics at a fundamental level: where the texture of the precipitate plays a role in determining the reaction rate. Time permitting, I will also connect this work to fundamental studies of the kinetics and mechanisms of heterogeneous nucleation from aqueous solutions, with accompanying modeling of precipitation textures and quantification. The results might have implications for not just direct air capture of CO2 using MgO, but other proposed strategies to remove CO2 from atmosphere, such as soil amendments with basalt.