

Effect of Impurities on Magnesium Oxide Hydroxylation and Carbonation

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Recently, the feasibility of MgO mineral looping for direct air capture of CO₂ has been analyzed using pure MgO in the absence of impurities. However, in nature, impurities in the source material and in the surrounding fluids may be present. The effect of neither solid nor solution phase impurities on hydroxylation and/or carbonation of MgO has been well studied, but the impurities may either cause passivation, thus decreasing CO₂ capture, or enhance reactivity, thus increasing uptake.

Here, we present a combined experimental and computational study on the effect of impurities, either incorporated or present in solution, on MgO reactivity. To test the effect of impurities in the solid, both batch experiments and in-situ X-ray reflectivity hydroxylation and carbonation experiments using Fe-doped MgO were performed. Samples were characterized ex-situ by a combination of scanning electron microscopy, transmission electron microscopy (TEM) and Raman spectroscopy. The effect of impurities on MgO hydroxylation and carbonation was simulated using density functional and density-functional tight-binding simulations.

Inhibition of both hydroxylation and carbonation was observed in in-situ X-ray reflectivity experiments for Fe-doped MgO. While formation of a several nm thick reaction layer was observed in TEM during pure MgO hydroxylation during 7 hours of reaction, no layer formation was observed on Fe-doped MgO. Computationally simulated interfacial electron density profiles of MgO and Fe-doped MgO in water are similar, but the dissociated OH⁻ groups from water were absorbed closer to the MgO surface than the Fe-doped MgO surface. We hypothesize that the presence of impurities changes the water structure at the mineral-water interface which could inhibit dissolution, and thus limit growth of the reaction layer.

In contrast, when impurities are present in solution, we observed secondary phase formation. In the presence iron chloride, an Fe-oxyhydroxide phase was formed. In the presence of NaCl, new particles of 100's of nm size were formed preferentially along pre-existing steps. Our results indicate that incorporated impurities potentially limit formation of hydrated and/or carbonated phases and that solution-based impurities may lead to surface passivation. Together, these results suggest that impurities may decrease the capacity of MgO to capture CO₂.