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The effect of sulphate on magnesite growth: in-situ and ex-situ nanoscale observations

Helen E. King (1), Hisao Satoh (2,3), Katsuo Tsukamoto (2), and Andrew Putnis (1)

(1) Institut für Mineralogie, University of Münster, Germany (hking_01@uni-muenster.de), (2) Department of Earth and Planetary Materials Science, Tohoku University, Japan, (3) Mitsubishi Materials Corporation, Japan

The composition of a solution has important implications for growth mechanisms and the incorporation of impurities during growth. For example, the presence of sulphate during CO_2 sequestration in Mg-silicate rocks such as ophiolites is expected to restrict magnesite ($MgCO_3$) growth due to the formation of Mg-sulphate ion pairs in solution (Pye et al. 1998), which lower the solution supersaturation with respect to magnesite. Conversely, direct interactions of sulphate with the magnesite surface, observed during dissolution (King and Putnis, 2013), could limit the negative effects of Mg-sulphate ion pair formation in solution by aiding growth through assisted desolvation of the Mg^{2+} ion (e.g., Piana et al. 2006). Furthermore, if the adsorbed sulphate is incorporated into the magnesite structure during growth it removes the need for expensive SO_2 flue gas scrubbers as both CO_2 and SO_2 can be sequestered simultaneously.

To explore the implications of sulphate in solution for CO_2 sequestration we have observed the growth of cleaved magnesite {104} surfaces in-situ using phase shift interferometry (PSI), a technique specially designed to monitor ultra-slow growing or dissolving mineral surfaces. In addition, we have conducted batch experiments in Teflon-lined steel autoclaves and examined the surfaces ex-situ using atomic force microscopy (AFM). All experiments were conducted at 90 ˚C in solutions of 0.2 M $NaHCO_3$ and 0.8 M NaCl or 0.4 M Na_2SO_4. Supersaturation of the solution was varied by changing the concentration of either $MgCl_2$ for Cl-rich or $MgSO_4$ in sulphate-rich solutions. For the PSI experiments a pressure of 1 MPa was used to prevent the formation of bubbles. In these experiments magnesite was grown in a flowing solution (100 μL/min) for 12 hours, whereas the batch experiments were conducted for 1-7 days under static conditions. The in-situ observations from the PSI experiments indicate that the presence of sulphate increased the rate of obtuse step propagation at etch pits present on the surface prior to the growth experiment, but that it limits 2D nucleation at the magnesite terraces. This is consistent with ex-situ observations from the AFM and the production of hydromagnesite ($Mg_2(CO_3)_4(OH)_24(H_2O)$) in Cl-bearing solutions during the batch reactions, identified using Raman spectroscopy.

Thus although sulphate can aid magnesite growth and possibly be incorporated into the obtuse step edge, it will have a negative effect during CO_2 sequestration into Mg-rich rock formations as nucleation in these scenarios is the critical step.

King, H.E. and Putnis C.V. (2013) *Geochim. Cosmochim. Acta.*, accepted for publication.

Piana S., Jones F. and Gale J.D. (2006) *J. Am. Chem. Soc.*, 128, 13568-13574.

Pye C.C. and Rudolph W.W. (1998) *J. Phys. Chem. A.*, 102, 9933-9943.