

Magnesium carbonate bearing minerals –Macroscopic to nano scale experimental data and observations

P. BENEZETH^{1*}, Q. GAUTIER^{1,2}, N. BERNINGER¹,
T. RONCAL-HERRERO³, R. KRÖGER³, D.
ALLOYEAU⁴,
V. MAVROMATIS¹, B. PURGSTALLER⁵ AND J.
SCHOTT¹

¹Géosciences Environnement Toulouse (GET)-
CNRS, France (pascale.benezeth@get.omp.eu)

²University Paris-est, Navier Laboratory, France

³Department of Physics, University of York, UK

⁴University Paris-Diderot-Paris 7, Paris, France

⁵ Graz University of Technology, Austria

The molecular mechanisms of reactions leading to the formation of Mg-carbonates from aqueous solutions are at the very beginning of their exploration despite their pivotal role in the global (bio)geochemical cycles of major elements, including carbon. We combined *ex-* and *in-situ* experimental tools at various length-scales (IR, AFM, SEM, STEM) to better understand and predict the formation of Mg-carbonates important for many environmental and industrial settings (e.g., deposits, weathering, climate reconstruction, CO₂ sequestration). Macroscopic techniques such as batch and flow-through experiments are suitable to quantify thermodynamics and kinetics of reacted minerals. We provide an overview of data on magnesite, hydromagnesite and dolomite, highlighting their puzzling formation and occurrence in nature [1]. Our studies focus on reacting surfaces, changes in morphology or growth [2-3]. In particular, real-time *in situ* imaging of dynamical, nanoscale processes in fluids were undertaken using liquid-cell scanning transmission electron microscopy (STEM), in combination with *in situ* Raman spectroscopy to follow nucleation and growth of Mg-carbonates. The preliminary results obtained identify i) the effect of radiation dose on the stability of the crystals formed, in particular rapid dissolution probably due to a local change in pH [4]; ii) a change of morphology/phase (amorphous to small crystallites) when increasing the temperature from 25 to 80°C by using a newly developed liquid cell TEM heating device (Protochips POSEIDON 210™). *In situ* Raman spectroscopy seems to indicate the formation of nesquehonite at T~60-70°C that we believe is corroborated by the STEM liquid cell observations.

[1] Bénézeth P. *et al.*, (2013) *Rev. Miner. Geochem.*, **76**, 81-133; [2] Gautier Q. *et al.*, (2015) *Geochim. Cosmochim. Acta*, **155**, 68-85; [3] Loring J. *et al.*, (2015) *Langmuir*, **31**, 7533-7543 ; [4] Schneider *et al.*, (2014) *J. Phys. Chem. C*, **118**, 22373-22382