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Study of dolomite dissolution at various temperatures –Evidence for the formation of nanocrystalline secondary phases at dolomite surface and influence on dolomite interactions with other minerals

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In most clay-rock geological formation studied for the storage of nuclear waste, pore water compositions are expected to be at equilibrium with carbonate minerals, which are always included in predictive models for pore water composition calculations [1]. Among the carbonates known to be present, dolomite may be problematic in the pore water composition calculation because its solubility spans a large range of values as a function of its crystallinity in thermodynamic databases. In addition, the composition of dolomite minerals observed in clay-rock formations such as Callovian-Oxfordian or Opalinus clay formation differs from this of a pure dolomite: the Ca/Mg stoichiometry is not ideal, and the minerals contain minor amounts of Fe and traces of many other elements [2].

To understand the influence of secondary phases precipitation during dolomite dissolution on pore water chemistry, the dissolution of monocrystals of dolomite were investigated at 25 °C and at 80 °C in a pH range 3 to 8 for various time periods (30 minutes to 21 days) in sealed PTFE reactors. Solution analyses evidenced a stoichiometric release of Ca and Mg in solution during dolomite dissolution. Scanning Electron Microscopy (SEM), Raman and X-Ray Diffraction (XRD) analyses did not evidence secondary Mg-bearing minerals precipitation, but revealed the formation of Fe-bearing particles on the dolomite surface. Morphological characterizations performed with Small-angle X-ray scattering (SAXS) evidenced that the precipitation occurs along a specific crystallographic plane of the dolomite monocrystal. Thus, the precipitated nanoparticles clustered on specific surface sites, and are made of Fe-rich phases poorly crystallized (carbonates, oxides and hydroxides).

[1] Tournassat et al. 2015. Ch. 3: Chemical Conditions in Clay-Rocks. *Natural and Engineered Clay Barriers*, Elsevier.

[2] Lerouge et al. 2011. *Geochim. et Cosmoch. Acta*, 2011, 75, 2633-2663.