

2.2.14

How do mineral coatings affect dissolution rates?

P. CUBILLAS¹, M. PRIETO¹, E. H. OELKERS²
AND S. KÖHLER³

¹Departamento de Geología. Universidad de Oviedo; Jesus Arias de Velasco s/n 33005, Oviedo, Asturias, Spain.
(pablo.cubillas@geol.uniovi.es;
manuel.prieto@geol.uniovi.es)

²Géochimie, Université Paul Sabatier, CNRS-UMR 5563, 38 rue des Trente-Six Ponts, 31400, Toulouse, France
(oelkers@lmtg.ups-tlse.fr)

³Institute for Engineering Geology and Applied Mineralogy, Rechbauerstraße 12, A-8010 Graz, Austria
(koehler@egam.tu-graz.ac.at)

One possible reason why dissolution rates in the field may be significantly lower than those measured in the laboratory are that natural mineral are often coated with surface precipitates [1]. This study attempts to quantify this effect through a series of experiments aimed at quantifying the rates of calcium carbonate dissolution in the presence of a precipitating surface phase.

The evolution of calcite, aragonite, and ground bivalve shells dissolution rates have been measured in open system reactors in solutions supersaturated with respect to otavite (CdCO₃). Otavite is found to precipitate on the surfaces of all initial CaCO₃ powders. The degree to which this precipitate affects CaCO₃ dissolution rates depends strongly on the similarity between the crystal structures of the precipitating phase and the dissolving solid, which itself affects strongly the precipitation mechanism [2].

Calcite possesses the same crystal structure as the precipitating otavite. Otavite precipitation proceeds via its epitaxial growth to a coating of only a few unit cells in thickness. This layer decreases calcite dissolution rates by more than one order of magnitude. In contrast, aragonite and aragonite composed shells possesses crystal structures different than that of the precipitating otavite. Otavite precipitation proceeds by three-dimensional nucleation of solid-solution crystals forming surface coatings of several microns thick on the original CaCO₃. Aragonite and aragonite composed shell dissolution rates decrease only slowing in response to precipitating otavite; nearly 6 microns of otavite coatings are required to arrest CaCO₃ dissolution. This observation suggests that three-dimensional nucleation is less effective at inhibiting dissolution than surface coatings formed by epitaxial growth.

References

- [1] Nugent M. A., Brantley S. L., Pantano C. G., and Maurice P. A. (1998) *Nature* **395**, 588-591.
- [2] Prieto M., Cubillas P., and Fernandez-González A., (2003) *GCA* **54**, 395-402.

2.2.15

Glauconite dissolution kinetics and application to CO₂ storage in the subsurface

P. AAGAARD¹, E.H. OELKERS² AND J. SCHOTT²

¹Dep of Geosciences, University of Oslo, Norway
(per.aagaard@geo.uio.no)

²Géochimie: Transferts et Mécanismes, CNRS/UMR 5563, Université Paul Sabatier (oelkers@lmtg.ups-tlse.fr; schott@lmtg.ups-tlse.fr)

Background

Accurate glauconite dissolution rates, are a requisite for assessing CO₂ sequestration potential in deep saline aquifers. The matrix sands of these aquifers are often marine sands with abundant glauconite, which are important reactants in mineral trapping. Previous geochemical simulations of CO₂ sequestering in such sands have been commonly based on the assumption that glauconite dissolution rates are equal to those of muscovite and biotite [1,2,3].

Experimental data

Dissolution experiments were performed in mixed flow reactors on aggregates of well crystalline glauconite (50-200 μm) at temperatures from 22-150 °C and pH conditions from 2-8. Mineral dissolution rates were calculated based on steady state solution compositions and either BET or estimated geometric surface area, and can be summarized as:

- Geometric surface area normalized dissolution rates are order of magnitude higher than that of other mica group minerals
- Apparent activation energy at pH 2 is about 35 kJ/mol and apparent reaction order with respect to protons -0.28.

Discussion

Both apparent activation energy and proton reaction order are in the range reported for dissolution of layer silicate minerals. The higher dissolution rate of glauconite favour the early mineral trapping phase of sequestration, and yet is low enough to ensure dispersed carbonate precipitation avoiding clogging of pores.

References

- [1] Perkins E.H. and Gunter W.D. (1995) WRI-8, 895-898.
- [2] Gunter W.D., Perkins E.H., and Hutcheon I. (2000) *Appl Geoch* **15**, 1085-1095.
- [3] Johnson J.W., Nitao J.J., Steefel C.I. and Knauss K.G (2001). *Proc. First Nat Conf Carbon Seq*, Washington DC 60 p.