Hydration of periclase at 350°C to 620°C and 200 MPa: An experimental study

Hakan Kuleci (1), Rainer Abart (1), and Christian Schmidt (2)

(1) University of Vienna, Department of Lithospheric Research, Vienna, Austria (hakan.kuleci@univie.ac.at), (2) German Research Centre for Geosciences, Potsdam, Germany

Hydration reactions play an important role in geodynamics. We chose the transformation periclase + H_2O = brucite as a model reaction to study the kinetics of hydration at crustal conditions.

To this end, dedicated hydration experiments were done using periclase single crystals, which were machined to cubes with 0.5 mm edge length and reacted with water at temperatures ranging from 350 to 620°C and at a pressure of 200 MPa using rapid-quench autoclaves. Run durations were between 5 and 40 min. Hydration produced reaction rims of fibrous brucite, which were separated from the reactant periclase by sharp reaction fronts. The reaction progress was determined using three different methods: Firstly the reacted samples were weighed before and after experiment, secondly the reacted samples were scanned with Micro-CT, and thirdly the remaining periclase and the newly formed brucite fractions were determined using a scanning electron microscope. For small to moderate reaction progress (less than about 30% of the original periclase converted) the brucite layers show relatively constant thickness. At higher reaction progress the geometry of the run product becomes rather complicated. The reaction progress was recalculated as the average thickness of the brucite layer.

Generally linear growth of the brucite layer is observed, and the growth rates increase with temperature from 350°C to about 530°C. At higher temperature, the growth rate decreases, and it vanishes, when the equilibrium temperature for the coexistence of per+H_2O+bru is approached at approximately 630°C.

We note that for an interface reaction the velocity of the reaction interface is equal to the thermodynamic driving force, the Gibbs free energy difference of the reaction ΔrG, multiplied by the "mobility" of the reaction interface, M.

$$v \ [mm/s] = M \ [m^4/s \cdot J] \bullet \Delta rG \ [J/m^3]$$

The Gibbs free energy difference was calculated from thermodynamic data and the interface mobility was extracted from the observed interface velocity. Interface motion is a thermally activated process and Arrhenius behavior can be assumed, so that

$$M(T) = M_0 \bullet e^{(-EA/(RT))},$$

where M_0 is a pre-exponential factor in m^4/s, EA the activation energy, T the absolute temperature, and R the gas constant. From our experiments, we estimated an activation energy of 50 kJ/mole and a pre-exponential factor of 0.0059 m^4/s. This yields a propagation rate of the reaction interface of 40 nm/s and 123 nm/s for 350°C and 530°C at 200 MPa, respectively.

The transformation of periclase to brucite is extremely quick. As the volume change of the solids associated with this reaction is about 100%, this should have interesting implications for the accommodation of the transformation strain in a solid phase aggregate.