



Mechanical effects of the hydration of periclase to brucite in calcite-periclase aggregates; an experimental study

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It has long been recognized that deformation and fluid flow are intimately coupled. For example hydration phenomena are often localized in zones of intense deformation. We investigated the opposite phenomenon looking at the deformation that is generated by hydration. This scenario may be relevant in the context of serpentinization, where large positive volume changes are associated with the hydration of olivine. This positive volume change may be accommodated by fracturing, which in turn provides fluid pathways leading to a positive feedback.

We select the model reaction:



which involves a positive volume change of the solids of about 100%, i.e. swelling by a factor of two with respect to the initial volume of periclase. Using the hot isostatic pressing technique we prepared dense (<5% porosity) calcite-periclase aggregates of cylindrical shape ($r=5\text{mm}$, $h=20\text{mm}$) with a calcite to periclase proportion of 90/10 by volume. The grain sizes of periclase and calcite were 100 to 200 and about $50\ \mu\text{m}$ respectively, in the aggregate. The samples were subjected to hydrothermal conditions of $P_{\text{H}_2\text{O}} = 200\ \text{MPa}$, and $T = 400^\circ\text{C}$ for 2 hours resulting in partial transformation of periclase to brucite. The associated volume increase led to fracturing in the calcite matrix essentially disintegrating the aggregate.

To understand the mutual feedback between hydration, swelling and fracturing it is crucial to know the kinetics of the hydration reaction. Periclase single crystals were machined to cubes with $3\times 3\times 3\ \text{mm}$ edge length and reacted with water at temperatures ranging from 400 to 530°C and pressures of 70 to $200\ \text{MPa}$ using cold-seal hydrothermal pressure vessels. Run durations were between 30 minutes and 2 hours. Hydration produced reaction rims of fibrous brucite, which were separated from the reactant periclase by sharp reaction fronts. The reaction progress was determined from measurement of the remaining periclase and the newly formed brucite volume fractions were recalculated as average thickness of the brucite layer. We assumed linear growth kinetics

$$d = k(T) * t,$$

where d is the thickness of the brucite layer in mm, $k(T)$ is the rate constant, and t is time in seconds, with

$$k(T) = k_0 * \exp(-E_A/R * T),$$

where T is absolute temperature, E_A is the activation energy and k_0 is the pre-exponential factor. From our experiments we estimated an activation energy of $66.35\ \text{kJ/mole}$ and a pre-exponential factor of $1.21\ \text{mm/s}$.

The hydration of periclase appears to be an extremely fast process with the reaction front proceeding at about $3\ \mu\text{m}$ per minute at 530°C and $200\ \text{MPa}$. We infer that hydration is so fast that it does not allow for the calcite matrix in the hot pressed calcite-periclase aggregates to accommodate the volume increase associated with hydration by creep thereby initiating fracturing.