



Experimental insight into the effect of water activity on mineral reaction rates at high pressure and temperature

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Using the MAX80 cubic multi-anvil press installed on the German synchrotron (HASYLAB-DESY, Hamburg), we have monitored from *in-situ* X-ray diffraction, the transformation rate of the exchange reaction, $\text{Ca}(\text{OH})_2 + \text{MgCO}_3 = \text{CaCO}_3 + \text{Mg}(\text{OH})_2$, at 1.7 GPa for temperatures below 600°C (*i.e.* in the field of aragonite). Although this reaction involves H_2O as chemical component, no water is release nor consumed in the course of the reaction. These experiments were performed either under controlled dry conditions (low H_2O activity) or under the humidity of the pressure assembly; the $\text{Ca}(\text{OH})_2 + \text{MgCO}_3$ starting material, composed of a fine powder, being initially submitted to the ambient air moisture. Under these pressure and temperature conditions, the free energy of this exchange reaction is of -30 to -35 kJ.mol.⁻¹. Additional experiments were performed under excess water conditions in a piston-cylinder apparatus (starting material sealed together with water in a gold capsule) at 1.7 GPa, 100, 120 and 150°C for different run durations.

Reaction progress plotted as a function of time shows kinetic curves with shapes which are consistent with a reaction dominated by nucleation and limited by the transport of the chemical species (*i.e.*, CO_2 and H_2O). SEM images show that Ca and Mg are immobile in first approximation ; CO_2 and H_2O are therefore the exchanged species. The major result of this experimental study is that for the same fine-grained starting

powder, nucleation occurs at the same kinetics under dry and excess-water conditions at two very different temperatures, 550 and 150°C, respectively. Extrapolation of the Arrhenius plot of half reaction-time shows that when half reaction is achieved within a few seconds at ca. 250°C under excess water, the same reaction progress will be never reached under dry conditions, even at the geological scale.

These experimental results are being interpreted using a “home made” numerical model which takes into account the respective kinetics of both nucleation and diffusion and grain-size effects. Then, these data will be extrapolated to naturally relevant conditions (i.e., dry and water saturated, large grain sizes etc . . .).