High-temperature kinetics of chlorite, biotite and illite

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Sheet silicates can comprise up to half of the mineral phase volume in some geothermal reservoirs targeted for stimulation, yet very few kinetic data are currently available for these minerals at geothermally relevant temperatures (≥ 100 °C). The reactivity of such phases is of special interest in geothermal fractures, where out-of-equilibrium fluid contacts minerals exposed at the fracture-wallrock interface. The lack of high-temperature kinetic data hampers our ability to confidently predict the extent and impact of geochemical alteration within fracture pathways and partially-filled veins, upon fracture-propping asperities, and on fluid exchange between fracture/matrix rock.

Suites of kinetic experiments have been conducted over pH ranges of 2.5 to 10, and temperatures from 100 to 280 °C for dissolution of three phyllosilicate minerals: chlorite (Mg-rich variety), biotite (Mg,Fe-containing variety), and illite. Individual rate expressions have been derived from these data and from combined datasets including available lower-temperature rate measurements. Among the three systems, we note that extrapolations of sub-100 °C rates often overpredict rates at higher geothermal temperatures. The conventional parabolic kinetic rate expression can be applied to these mineral reactions, although the magnitude of rate increase associated with increased levels of H+ and/or OH- is reduced depending on the likelihood of aluminum- or magnesiumhydroxide phase formation, respectively. Shifts in "neutral" fluid pH at elevated temperature also tend to dampen the dependence of rate with acid pH observed at lower temperatures. Solution chemistry modeling indicates that fluid saturation likely affects chlorite and biotite dissolution rates at $T \ge 100$ °C, although the data are not sufficient to fully describe the nature of the reaction affinity term. For the case of illite, fluid saturation may impact measured rates but it is of secondary importance to likely concurrent precipitation of other aluminosilicate phases. Variability in rates collected from differently substituted mineral varieties and among different laboratories must be considered when formulating kinetic rate expressions.