Experimental destabilization of reservoir rocks under low-grade metaphorphism : a potential source for CO_2 plumes in basins?

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Though the presence of CO_2 in gas fields is often produced through organic processes, low-grade metamorphic reactions (at $T<500°C$, $P<2500$ bar) can also generate CO_2 from the devolatilization of carbonates, when they are associated to alumino-silicate minerals. The ability to correctly predict the thermodynamic equilibrium of those reactions can be of great value to petroleum exploration, as it can help estimate the yield of CO_2 generated as a function of the mineral assemblage considered.

To verify the validity of our models, an experimental study was conducted on two different mineral assemblages with minerals common in sedimentary environments, containing (a) Ca-Mg-SiO_2-Al (dolomite, quartz, kaolinite) and (b) Ca-Mg-SiO_2-Al-K (dolomite, quartz, illite). Various experiments were conducted in batch reactors, at 300 bar and between 250°C and 300°C, to compare the experimental results with those predicted numerically for partial (250°C) and total (300°C) devolatilization of the initial carbonates.

The gas phase was sampled and quantified by gas-chromatography to follow the amount of CO_2 released through the experiments, and the liquid phase was analyzed by ICP-OES to determine the elements concentration released by the dissolution of the primary phase. The data was used for calculating the evolution of the fluid's affinity with respect to various secondary phases. Those thermodynamic calculations were made using the Arxim-GEM code and a modified database with numerous clay solid-solution phases. The analysis of the solid run products by x-ray diffraction and SEM shows a good fit with the models. In potassium-free systems, the initial dolomite $(Ca,Mg)(CO3)_2$ is progressively replaced by secondary calcite ($CaCO_3$), while the Mg released precipitates in a secondary phyllosilicate with a composition close to montmorillonite. On the other hand, in potassium-rich systems, an Mg-rich potassic mica, with a composition close to phlogopite, forms as illite dissolves. Those results allow us to discriminate between various thermodynamic data, while the good agreement with the models allow us to validate key reactions occurring at low temperature.