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Carbonate mineral dissolution kinetics in high pressure experiments

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The potential CO_2 reservoirs in the North German Basin are overlain by a series of Mesozoic barrier rocks and aquifers and finally mostly by Tertiary and Quaternary close-to-surface aquifers. The unexpected rise of stored CO_2 from its reservoir into close-to-surface aquifer systems, perhaps through a broken well casing, may pose a threat to groundwater quality because of the acidifying effect of CO_2 dissolution in water. The consequences may be further worsening of the groundwater quality due to the mobilization of heavy metals. Buffer mechanisms counteracting the acidification are for instance the dissolution of carbonates. Carbonate dissolution kinetics is comparably fast and carbonates can be abundant in close-to-surface aquifers.
The disadvantages of batch experiments compared to column experiments in order to determine rate constants are well known and have for instance been described by v. GRINSVEN and RIEMSDIJK (1992). Therefore, we have designed, developed, tested, and used a high-pressure laboratory column system to simulate aquifer conditions in a flow through setup within the CO_2-MoPa project. The calcite dissolution kinetics was determined for CO_2-pressures of 6, 10, and 50 bars. The results were evaluated by using the PHREEQC code with a 1-D reactive transport model, applying a LASAGA (1984) -type kinetic dissolution equation (PALANDRI and KHARAKA, 2004; eq. 7). While PALANDRI and KHARAKA (2004) gave calcite dissolution rate constants originating from batch experiments of log kacid = -0.3 and log kneutral = -5.81, the data of the column experiment were best fitted using log kacid = -2.3 and log kneutral = -7.81, so that the rate constants fitted using the lab experiment applying 50 bars pCO_2 were approximately 100 times lower than according to the literature data. Rate constants of experiments performed at less CO_2 pressure (pCO_2 = 6 bars: log kacid = -1.78; log kneutral = -7.29) were only 30 times lower than literature data.
These discrepancies in the reaction kinetics should be acknowledged when using reactive transport models, especially when modeling kinetically controlled pH-buffering processes between a CO_2 leakage an a receptor like a ground water well. Currently, further experiments for the determination of the dolomite dissolution kinetics are being performed. Here, the knowledge of the dissolution rate constants can be even more important compared to the (still) fast calcite dissolution.
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