DISSOLUTION OF SILICATES IN THE CONTEXT OF MINERAL CARBONATION FOR CARBON DIOXIDE SEQUESTRATION

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Mineral carbonation circumvents the long-term risks associated with other carbon dioxide disposal methods like geological or ocean sequestration by converting carbon dioxide into stable and benign carbonates. The currently most promising reaction process is aqueous carbonation, which mimics slow natural phenomena of mineral dissolution and precipitation. Its relatively high costs are caused by long reaction times, mostly measured in hours, costly mineral activation procedures, required to speed up the reaction, and expensive reaction vessels due to demanding reaction conditions. It is believed that the dissolution of the silicates is the rate-limiting step, although no thorough quantitative analysis of this issue has yet been carried out.

Dissolution kinetics for olivine and serpentine, the two main source minerals, has been studied by geochemists for quite some time; especially olivine has attracted noticeable interest, mainly due to its structural simplicity and high reactivity. However, almost all studies were conducted at temperatures between 25ºC and 65ºC and ambient pressure, whereas the optimal reaction temperatures for the carbonation process have been found to be between 150ºC and 200ºC and pressures up to 200 bar. Likewise, the effect of ligands or salinity, a major factor in the carbonation results reported so far, has only been looked at in a few cases. The available results for olivine mostly indicate agreement over the dissolution rate at low temperature and low pH, but are inconclusive for alkaline conditions and allow only for a range to be set for the activation energy. Under acidic to neutral conditions, a strong dependence on the solution pH is universally observed. We have analyzed the dissolution behaviour of olivine in a stirred and a non-stirred titanium autoclave. The first autoclave, stirred by a magnetically-coupled stirrer, can be operated at temperatures of up to 200ºC and 200 bar, both in batch and continuous mode. It is fed by a HPLC pump. The non-stirred autoclave can be operated at temperatures of up to 400ºC and 500 bar, in batch mode only. All parts in contact with the hot reactive fluid are made of titanium. Experiments are carried out over a range of temperatures and pH values, and various CO2 partial pressures. The dissolution rate is determined by measuring the concentration of the dissolved elements via Atomic Absorbtion Spectroscopy (AAS), Ion Chromatography (IC) and photospektrometric methods (Molybdate Blue). The rate is expressed relative to the BET surface area.