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Reservoir controls on the mineralization of carbon: implications for geological carbon storage

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Carbon mineralization is a natural weathering process that can be accelerated to store carbon dioxide in a solid state in the form of new minerals. In geologic formations, this chemical reaction involves the dissolution of mafic/ultramafic rocks that release cations (Ca^{2+}, Mg^{2+}, Fe^{2+}), which later bond with carbon in a CO_2-saturated brine to form carbonates (calcite, magnesite, siderite). However, reservoir conditions such as temperature, pressure and oxygen fugacity can have an important effect in the final solid products of this reaction.

Here we show the resulting solid products of four reactive flow experiments using ultramafic rocks and a CO_2-saturated brine. The experiments were performed in a servocontrolled pressure vessel under hydrostatic conditions and using isolated pore pressure fluid lines to simulate the pressure, temperature and fluid-rock ratios encountered in prospective geologic reservoirs for in-situ carbon mineralization.

In samples reacted at 142ºC we find abundant precipitation of magnesite, while at temperatures below 130ºC the precipitation of an amorphous form of organic carbon dominates the assemblage. Preliminary Raman spectroscopy analysis suggests that in experiments on dunite aggregates the organics are primarily composed of weakly crystalline graphite. This is indicated by two prominent peaks at 1580 cm^{-1} and 1340 cm^{-1}. Meanwhile, a peak around 1450 cm^{-1} appears in both dunite and serpentinite experiments, and is suggested to be related to organic carbon in the form of hydrocarbons.

These results suggest that the carbon species formed in each experiment are influenced by the oxidation of Fe in the sample to form Fe-oxides like magnetite and hematite. Assuming the oxygen fugacity (fO_2) in the sample is controlled by the magnetite-hematite buffer, at temperatures below 142ºC the corresponding fO_2 falls below the C^0-CO_2 equilibrium, favoring reduced carbon species such as graphite. In contrast, above 142ºC, fO_2 falls above the C^0-CO_2 equilibrium, favoring carbonate species, such as magnesite. This interpretation indicates that small changes in formation temperature can have strong controls on which species of carbon are mineralized in CO_2 reservoirs, and thus in how the physical and chemical properties of the storage site will change overtime.