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Laboratory investigations of stable carbon and oxygen isotope ratio data enhance monitoring of CO_2 underground

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Stable carbon and oxygen isotope data play an important role in monitoring CO_2 in the subsurface, for instance during carbon capture and storage (CCS). This includes monitoring of supercritical and gaseous CO_2 movement and reactions under reservoir conditions and detection of potential CO_2 leakage scenarios. However, in many cases isotope data from field campaigns are either limited due to complex sample retrieval or require verification under controlled boundary conditions. Moreover, experimentally verified isotope fractionation factors are also accurately known only for temperatures and pressures lower than commonly found in CO_2 reservoirs (Myrttinen et al., 2012). For this reason, several experimental series were conducted in order to investigate effects of elevated pressures, temperatures and salinities on stable carbon and oxygen isotope changes of CO_2 and water.

These tests were conducted with a heateable pressure device and with glass or metal gas containers in which CO_2 reacted with fluids for time periods of hours to several weeks. The obtained results revealed systematic differences in 13C/12C-distributions between CO_2 and the most important dissolved inorganic carbon (DIC) species under reservoir conditions (CO_2(aq), H2CO$_3$ and HCO$_3$-). Since direct measurements of the pH, even immediately after sampling, were unreliable due to rapid CO_2 de-gassing, one of the key results of this work is that carbon isotope fractionation data between DIC and CO_2 may serve to reconstruct in situ pH values. pH values reconstructed with this approach ranged between 5.5 and 7.4 for experiments with 60 bars and up to 120 °C and were on average 1.4 pH units lower than those measured with standard pH electrodes directly after sampling.

In addition, pressure and temperature experiments with H_2O and CO_2 revealed that differences between the oxygen isotope ratios of both phases depended on temperature, water-gas ratios as well as salt contents of the solutions involved. Such systematic knowledge of the extent of oxygen isotope fractionation between H_2O and CO_2 can help to reconstruct equilibration times, fluid-CO_2 ratios as well as temperature and salinity conditions.

Isotope results from systematic laboratory studies and the information they provide for assessing in situ reservoir conditions can be transferred to field applications concerning integrity of CO_2 reservoirs. They can also apply to natural systems and other industrial uses that involve monitoring of gases in the subsurface under similar pressure and temperature conditions.

Reference:

Myrttinen, A., Becker, V., Barth, J.A.C., 2012. A review of methods used for equilibrium isotope fractionation investigations between dissolved inorganic carbon and CO_2. Earth-Science Reviews, 115(3): 192-199.