Geophysical Research Abstracts
Vol. 12, EGU2010-12738, 2010
EGU General Assembly 2010

Isotope mass balances in deep formations: How to consider the influence of pressure, temperature and salinity

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Stable carbon isotopes are a sensitive tool to trace migration and to quantify mixing of CO_2 from different sources. This technique is well described for application at the surface and under close-to-surface conditions. However, in order to use isotope mass balances when monitoring the fate of CO_2 at carbon storage sites, some distinctive characteristics of deep formations have to be considered. High pressures (from 30 to 200 bar), temperatures (up to over 120 °C) and salinities (approx. 100 to 400 g/L) influence the carbonate equilibrium as activities, fugacities and the stoichiometric equilibrium constants (f_{CO2}, $K^*_{calcite}$, K^*_1, K^*_2) change with these parameters. Furthermore, isotopic fractionation is also affected by these parameters. Various relations are published, describing these dependencies with approaches of different complexity and exactness. In this field of application, available sampling data is usually limited, so that averaging and interpolation of input data may lead to noticeable error ranges. Under these conditions, the most elaborated algorithms do not necessarily perform better than more simple ones with respect to the overall error of the calculations. This work therefore compares the available approaches to describe temperature, pressure and salinity dependence in carbonate equilibrium calculations, as well as carbon isotope fractionation in this process with respect to the best ratio of accuracy in carbon storage site monitoring applications.

It stands out that the fugacity and the stoichiometric constants involved in DIC-speciation are heavily influenced by pressure, temperature and salinity in general, whereas the individual composition of the solution may be simplified, at least for NaCl-type brines. With respect to fractionation, temperature plays a key role; pressure and salinity variations contribute to the species distribution only to a small amount.

This study was conducted as a part of the R&D programme CLEAN, which is funded by the German Federal Ministry of Education in the framework of the programme GEOTECHNOLOGIEN.