

## Fundamental Geochemical Research on Long-Term Carbon Sequestration in Subsurface Environments

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To properly assess the viability of long-term CO<sub>2</sub> sequestration, accurate information is needed on the thermophysical properties, phase relations, stable-isotope systematics, and reaction kinetics of C-bearing fluids and minerals under subsurface conditions. Accordingly, we are performing various laboratory experiments to investigate: (i) the thermophysical properties and phase relations of CO<sub>2</sub>-CH<sub>4</sub>-H<sub>2</sub>O fluids; (ii) carbon and oxygen isotope partitioning during carbonate precipitation; and (iii) the utility of natural isotopic tracers in quantifying CO<sub>2</sub> residence times, storage capacity and reaction mechanisms in the subsurface.

The ultimate aim of the research on CO<sub>2</sub>-CH<sub>4</sub>-H<sub>2</sub>O fluids is to develop a comprehensive equation of state for binary and ternary mixtures of CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O at pressure-temperature (*P-T*) conditions representative of those in deep gas fields and saline aquifers. To acquire the data needed to create the model, two unique, custom-designed devices at the Oak Ridge National Laboratory—a high pressure vibrating-tube densimeter, and a hydrogen-service internally heated pressure vessel—are being used to measure the densities, excess molar volumes, miscibility limits and activity-composition relations of CO<sub>2</sub>-H<sub>2</sub>O, CH<sub>4</sub>-H<sub>2</sub>O and ternary CO<sub>2</sub>-CH<sub>4</sub>-H<sub>2</sub>O mixtures at *P-T* conditions near the vapor-saturation phase boundary in the H<sub>2</sub>O system.

To accurately determine the kinetics of carbonate precipitation from CO<sub>2</sub>-rich saline waters, and associated isotope partitioning, both inorganic and microbially mediated processes are being studied under environmental conditions encountered during CO<sub>2</sub> injection into the subsurface. Our results indicate that the behavior of isotopes is affected by the composition of water and the precipitation rate of carbonate minerals. Preliminary results on carbon isotope partitioning between CO<sub>2</sub> and hydrocarbon-saturated rock reacted statically at 25°C (an EOR injection scenario) suggest that a light isotopic component of CO<sub>2</sub> may be retained in the reservoir, leading to isotopically heavier CO<sub>2</sub> further down the flow path.

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