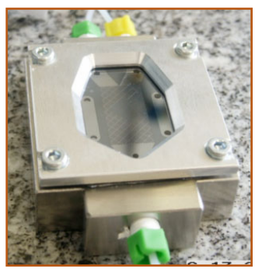
Molecular mineral transformation in real time

Lead investigator: Zheming Wang

PNNL researchers have developed an integrated, supercritical CO_2 optical spectroscopy platform for studying the molecular mineral transformation processes involved in geologic sequestration. In situ experimental studies will use a suite of optical spectroscopic techniques, including ultraviolet-visible spectroscopy, Fourier transform infrared resonance (FTIR), Raman, laser fluorescence spectroscopy and sum frequency generation. Initial experiments are underway to investigate the transformation kinetics of magnesium-chlorite, wallastonite, and forsterite into calcium and magnesium carbonate minerals. PNNL researchers also are studying the properties of dissolved water in supercritical CO_2.

Caprock-sealing efficiency and trapping mechanisms

Lead investigator: Mart Oostrom

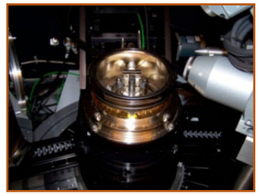


When the entry pressure at the caprock-reservoir interface is exceeded, supercritical CO_2 can move into the caprock, potentially compromising its sealing efficiency. Data related to these fluid-fluid and fluid-mineral interactions—such as capillarity, mass transfer, interfacial tension and wettability—are scarce. Researchers at PNNL are completing a series of micromodel experiments at supercritical conditions that address the following scientific challenges:

- Roles that porous medium properties, fluid properties, and wettability play during hydrodynamic trapping, when supercritical CO_2 displaces brine, and during capillary trapping, when brine displaces supercritical CO_2
- Effects of fluid-fluid interfacial tension, pore-size geometry, and wettability on caprock-sealing efficiency
- Relationships between capillary pressure, fluid saturation, and relative permeability during pore-scale displacement processes.

Caprock-mineral reactions using high-pressure X-ray diffraction

Lead investigator: H Todd Schaef



Hydrodynamic trapping by the caprock provides time for slow-trapping mechanisms to occur, such as CO_2 dissolution in pore water and precipitation of carbonate minerals. Scientists at PNNL are enhancing high-pressure X-ray diffraction (HXRD) capabilities to examine mineral reactions, including layer silicate minerals, and their kinetics when exposed to water solvated in supercritical CO_2 fluids under pressure, temperature, and fluid compositions typical for geologic sequestration.

Predicting the co-sequestration of CO_2, SO_x and NO_x

Lead investigator: Diana Bacon

Geologic co-sequestration of emission gases, such as CO_2, SO_x, and possibly NO_x, may be feasible in some geologic formations, greatly reducing the costs associated with post-emissions capture and potentially enhancing mineralization in calcium-rich igneous and weathered igneous mineral systems. The new IS^3 experimental capabilities will enable characterization of reactions when mixed gases are sequestered; however, numerical tools are lacking that combine the physics of multi-component/multi-phase mixtures with the chemical effects of mineral dissolution and precipitation. A new simulator that incorporates all of these complex processes is being developed and will be used to survey the effects of co-sequestration under a wide range of reservoir mineralogical and phase conditions.

FOR MORE INFORMATION, CONTACT:

Alain Bonneville
alain.bonneville@pnl.gov
(509) 371-7263

Ian Gorton
ian.gorton@pnl.gov
509-375-3850

Kevin Rosso
kevin.rosso@pnl.gov
(509) 371-6357

Tim Scheibe
tim.scheibe@pnl.gov
509-372-6065

PNNL-SA-70857

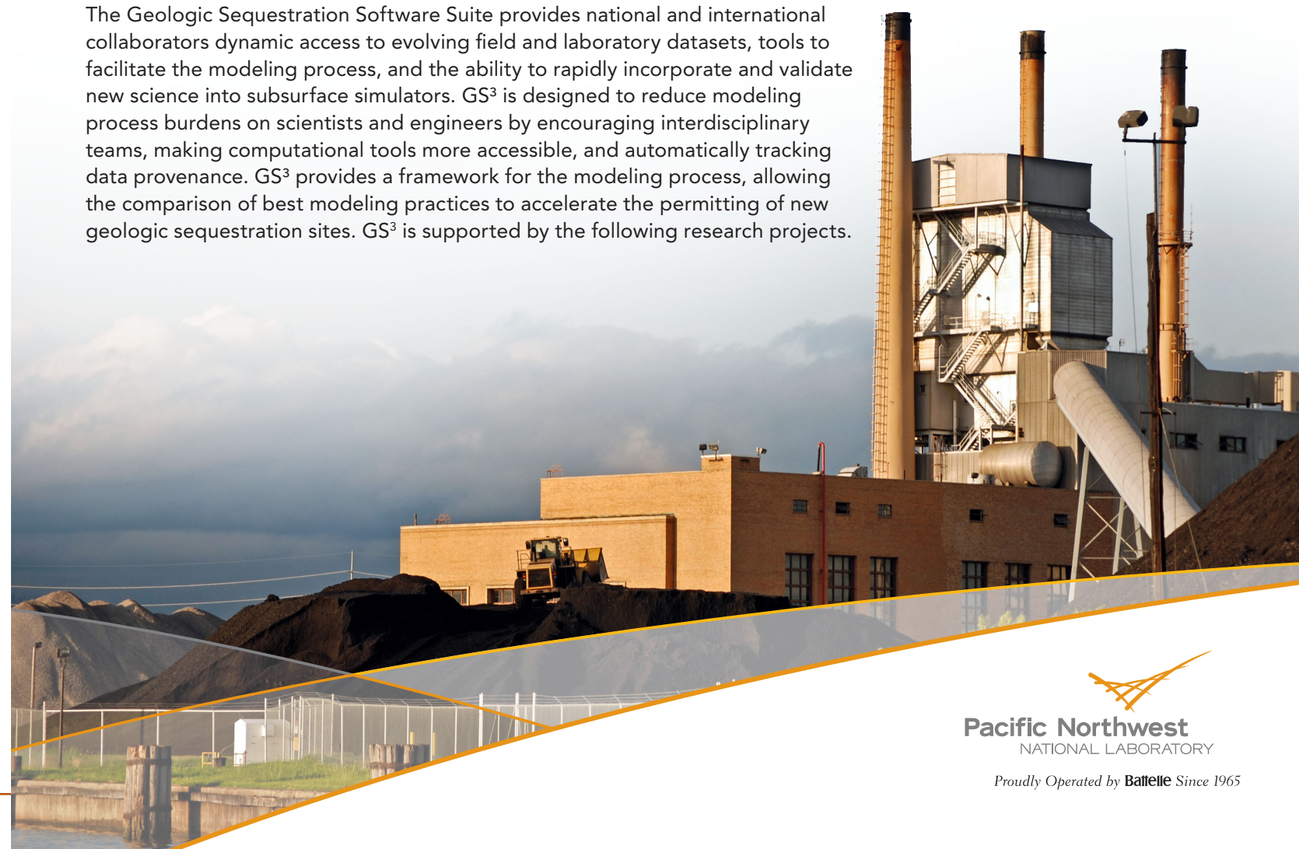
Science to Accelerate Sustainable Storage

The urgent need to reduce greenhouse gas emissions to the atmosphere will require global acceleration of commercial geologic sequestration. However, regulatory, liability and cost issues continue to slow progress and remain a barrier for industry in tackling first-of-a-kind demonstrations. Targeted science can provide the understanding needed to clarify many of the regulatory issues, reduce risk, provide the basis for improved monitoring criteria and tools, and ultimately reduce cost. But to make scientific discovery useful, it must be guided by field information and rapidly incorporated into modeling tools that are scientifically credible, transparent in their approach, continuously updated, and available for use at sequestration sites.

The science of geologic sequestration poses major experimental and theoretical challenges because the processes that impact long-term geologic sequestration occur under extreme pressure and temperature conditions over multiple scales of time and distance that require complex approaches. PNNL is addressing several of these key challenges by developing an experimental suite of tools, In Situ Supercritical Suite (IS^3), that is providing geochemical information to advance the predictive capability of simulators residing on a web-based modeling platform, Geologic Sequestration Software Suite (GS^3). These new tools, described below, are designed to accelerate translation of data into scientific understanding, simulation of site performance, and establishment of best standards.

Geologic Sequestration Software Suite (GS^3)

The Geologic Sequestration Software Suite provides national and international collaborators dynamic access to evolving field and laboratory datasets, tools to facilitate the modeling process, and the ability to rapidly incorporate and validate new science into subsurface simulators. GS^3 is designed to reduce modeling process burdens on scientists and engineers by encouraging interdisciplinary teams, making computational tools more accessible, and automatically tracking data provenance. GS^3 provides a framework for the modeling process, allowing the comparison of best modeling practices to accelerate the permitting of new geologic sequestration sites. GS^3 is supported by the following research projects.



Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle *Since 1965*

GS³ Projects

Developing a core architecture and simulation framework

Lead investigator: Ian Gorton

1 Advanced multi-phase flow and transport simulators play a crucial role in evaluating the long-term effects of geologic sequestration. Organizing, managing, and tracking the data used during the simulation process is critical to the integrity, verifiability, and repeatability of the analysis, yet the process is currently time consuming and prone to error. This effort is compounded by the need to provide history matching with monitoring data, which will occur over the life cycle of a sequestration project, and result in thousands of simulations over 50 or more years.

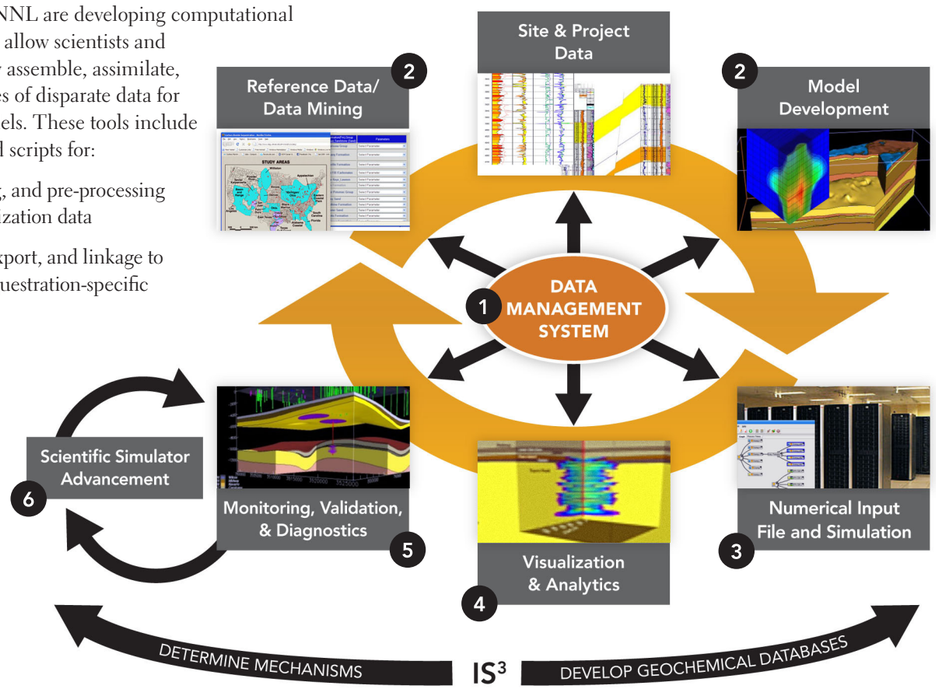
The GS³ framework is an open-source, web-based modeling platform that integrates seamlessly with data management capabilities. Ultimately, GS³ will vastly reduce the burden on modelers of manually organizing and tracking the volumes of data and simulations needed throughout the sequestration modeling life cycle.

Data assimilation tools for CO_2 reservoir model development

Lead investigator: Mark Rockhold

2 Researchers at PNNL are developing computational tools for GS³ that allow scientists and engineers to efficiently assemble, assimilate, and track large volumes of disparate data for building geologic models. These tools include programs, utilities, and scripts for:

- » evaluating, preparing, and pre-processing subsurface characterization data
- » facilitating import, export, and linkage to external geologic sequestration-specific modules
- » assimilating large, diverse data for building and parameterizing conceptual mathematical models, emphasizing data and processes that are specific to geologic sequestration
- » tracking authorship, data transformations, and analysis decisions to ensure efficient updating of auditable, fully traceable, and defensible models.



Numerical model development

Lead investigator: Mark Williams

3 This project provides tools and innovative methods to support the generation of simulator input files within GS³. New algorithms and tools will improve translation of conceptual models to the numerical framework required by simulators and provide the capability of generating multi-scale numerical models for long-term performance simulation.

The tools will support input file generation for multiple simulators, parallel computing platforms, stochastic simulations, and inverse modeling and parameter estimation. Researchers can rapidly develop and run simulations based on one or more conceptual models using various simulators, which will give a better understanding of the efficacy of a particular sequestration approach.

Visualizing Uncertainty in Conceptual and Numerical Models

Lead investigator: Luke Gosink

4 Scientific visualization and visual analytics provide insight into the complex events and processes modeled by numerical simulations and are integral to scientific discovery. This project will address the challenges associated with the increasing complexity and expanding scale of numerical simulations. Specifically, these tools will 1) support cross-model comparisons and analysis based on a collection of simulated data; 2) visually identify salient trends in numerical model uncertainty and sensitivity; and 3) provide distributed visualization support for large-scale data. These tools will visually create an information feedback loop to accelerate the process of developing, analyzing, and verifying the integrity of simulations.

A computational framework to validate numerical simulators

Lead investigator: Mark White

5 Simulator verification workshops and code comparison publications are static snapshots of the state of numerical simulation science for greenhouse gas sequestration. Researchers at PNNL are developing a dynamic validation and benchmarking capability, where numerical solutions evolve with modeling advances and benchmark problem complexity evolves with growth in available field and experimental data. GS3 will host 1) a computational framework for diagnosis and inter-comparison; 2) a suite of validation problems and solutions; 3) observational data sets for model validation; 4) standardized documentation of code features and characteristics; and 5) systematic accounting of disagreements for validation problems.

Multiscale investigations of CO_2 behavior

Lead investigator: Alexandre Tartakovsky

6 Pore-scale transport models have proved to be a valuable tool for estimating the constitutive relationships for non-reactive, two-phase flows and for studying the effect of complex pore-morphology on entrapment and dissolution of non-aqueous phases. PNNL researchers are developing multi-scale, multi-phase flow and transport models to simulate CO_2 flow and precipitation in the caprock at the pore and sub-pore scales. This hybrid model will bridge molecular dynamics surface chemistry models and pore-scale models, as well as describe the complex behavior of CO_2 in the caprock. The hybrid model serves as a basis for upscaling CO_2 behavior from the fundamental scales to the scale of practical importance.

In Situ Supercritical Suite (IS³)

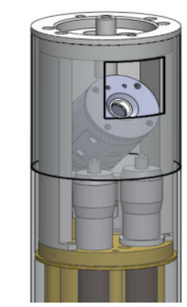
It has long been assumed that the supercritical CO_2 phase is chemically inert in deep saline systems. Research at PNNL has shown that water solvated in supercritical CO_2 is quite reactive towards both iron and silicate minerals under conditions expected for geologic sequestration. Moreover, simulators used in geologic sequestration assume water, not CO_2, as the solvating fluid, ignoring these important reactions near the borehole and caprock.

The In Situ Supercritical Suite is a group of one-of-a-kind instruments that will allow researchers to probe geochemical reactions under supercritical pressures and temperatures. These instruments will be used for mechanistic characterization of important mineral transformation and reaction kinetics, information that will be used to advance scientific simulators. Many of the projects below take advantage of unique instrumentation in the Environmental Molecular Sciences Laboratory, a U.S. Department of Energy user facility located at PNNL.

IS³ Projects

Trapping mechanisms using nuclear magnetic resonance

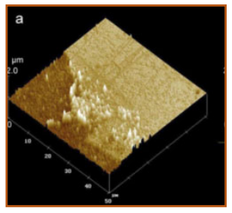
Lead investigators: David Hoyt and Jian Hu



A newly developed, low background ceramic cell is being used in nuclear magnetic resonance to observe geochemical reactions at pressures and temperatures found during geologic sequestration. PNNL scientists are studying the interfacial structure, kinetics and mechanisms of common primary and secondary mineral phase dissolution in CO_2-water mixtures, and carbonate nucleation and mineralization reactions.

Atomic force microscopy to image mineral-supercritical CO_2 reactions

Lead investigator: A. Scott Lea



The molecular-scale, site-specific reactions that take place at mineral-fluid interfaces for geologic sequestration are virtually unknown. Understanding and controlling the chemistry at these interfaces is necessary to assess the practicality of geologic sequestration and should be based on a quantitative understanding of these fundamental interactions. To address this, researchers at PNNL are building a hyperbaric atomic force microscope that is capable of imaging mineral-water-supercritical CO_2 systems in situ and in real time in order to obtain molecular-scale mineral transformation reaction kinetics. This instrument will be able to handle the pressures necessary to operate with supercritical CO_2.