

2026 Geosciences Principal Investigators' Meeting



Rockville, MD
February 9-10, 2026



U.S. DEPARTMENT OF
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Chemical Sciences, Geosciences, and Biosciences Division

Cover art: (Philip Wilk, 2025)

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Foreword

This abstract book provides a record of the 2026 U.S. Department of Energy Principal Investigators' meeting of the Geosciences program. Research described within was sponsored by the Chemical Sciences, Geosciences and Biosciences Division of the Office of Basic Energy Sciences.

The objective of this meeting is to provide a fruitful environment in which researchers with common interests will present and exchange information about their activities, will build collaborations among research groups with mutually reinforcing strengths, will identify needs of the research community, and will focus on opportunities for future research directions. With ample time for discussion and interactions, we emphasize that this is an informal meeting for exchange of information and building of collaborations; it is not a review of researchers' achievements or a forum to choose future directions.

It has been a privilege to serve as the manager of these research programs. In carrying out these tasks, I learn from the achievements and share the excitement of the research of the many sponsored scientists and students whose names appear on the papers in the following pages. I hope that this meeting enhanced research efforts and will nurture future collaborations. I would like to thank all the researchers whose dedication and innovation have advanced our research and made this meeting possible and productive.

Philip Wilk

Agenda

2026 Geosciences Principal Investigators' Meeting

Bethesda North Marriott Hotel & Conference Center, 5701 Marinelli Rd, Rockville, MD 20852

Monday, February 9 - Tuesday, February 10

Meetings Notes:

The meeting fee includes breakfast and lunch provided at the meeting.

People who did not pay the registration fee will need to pay for their meals.

Sunday, February 8

Arrival

7 -9 PM Informal No-Host Meetup at "The Corby Kitchen"

Bethesda North Marriott Hotel & Conference Center, 5701 Marinelli Rd, Rockville, MD 20852

Monday, February 9

8:00 - 9:00 Breakfast

9:00 Meeting Begins

Monday, February 9

8:00 - 9:00 Breakfast

9:00 Meeting Begins

Philip Wilk

Introduction, Agenda, Mini-Workshop Charge

9:15 Session 1

9:15 Benjamin Legg

Selective Nucleation of Rare Earth Element Phases on Mineral Surfaces

9:40 Alexander Gysi

Fractionation and Mobilization of Rare Earth Elements (REE) in Hydrothermal Ore Deposits: From Laboratory Experiments to Geochemical Modeling

10:05 Matthew Kohn

Uptake Mechanisms of REE in Sedimentary Phosphorite Mineral

10:30 **Break**

10:45 Jeffrey Catalano

Geochemical Mechanisms Controlling Rare Earth Element and Platinum Group Element Migration and Enrichment During Crustal Aqueous Alteration and Weathering

11:10 Young-Shin Jun

Geochemical Processes Controlling the Fate of Critical Elements During Carbonation of Mafic and Ultramafic Minerals

11:35 Craig Lundstrom

A Molecular Understanding of PGE Transport and Ore Formation Through an Integrated Experimental, Computational and Thermodynamic Modeling Approach/Elucidating REE Transport and Deposition through Molecular Understanding of Hydrous Carbonate Silicate Liquids

12:00 Working Lunch for Collaboration (poster room will be open)

1:00 Session 2

1:00 Sarah Slotznick

Magnetic Properties of Cobalt-bearing Minerals

1:25 Harry Lisabeth

Chemo-Rheological Controls on Subsurface Stress Heterogeneity in Geothermal Systems

1:50 Ahmadreza Hedayat

Geophysical Imaging of Creep Hydraulic Fracture Growth in Rocks

2:15 **Break**

2:30 Benjamin Holtzman

Project LISTEN-CLOSE

2:55 Jacques Rivière

Unraveling the Physics of Earthquake Precursors Using Ultrasonic Imaging and Physics-Informed Machine Learning

3:20 Yehuda Ben-Zion
Project: Properties and Dynamics of Fault Zones and the Shallow Crust

3:45 **Break**

4:00 **Poster Slam** Philip Wilk

(Each poster presenter gets one minute and one slide, no questions)

Poster Session 1: Even numbered posters (all posters are up)

6:00 Dinner on your own and Collaborate (poster room will be open)

Tuesday, February 10

8:00 - 9:00 Breakfast

9:00 Session 3

- 9:00 Sourabh Apte
Influence of Turbulence on Momentum Transfer at the River-Hyporheic Zone Interface: An Approach Combining Experiments, High-Performance Computing, and Upscaling Theory With Machine Learning
- 9:25 Daniel O'Malley
Quantum Computing and Machine Learning for Enhanced Understanding of Fracture Flow
- 9:50 Greg Beroza
Discerning Fault Geometry and Mechanics from Seismicity Point Clouds
- 10:15 **Break**
- 10:30 Weiqiang Zhu
Seismic Foundation Model for Integrated Subsurface Monitoring
- 10:55 Priya Vashishta
AI Assisted Neural Network Quantum Dynamics Simulations of Stress-Corrosion and Carbon Sequestration
- 11:20 Maarten de Hoop
Foundation models: Emerging Architectures and Fundamental Properties
- 11:45 Gail McLean
BES Update
- 12:30 Working Lunch for Collaboration (poster room will be open)
- 1:45 **Poster Session 2: Odd numbered posters (all posters are up)**
-

3:15 Program Update and Mini-Workshop Artificial Intelligence for Geosciences

Philip Wilk

5:45 Dinner on your own and Collaborate

Dismissal

Posters

- P1 Sang Soo Lee
pH-Driven Restructuring of Hydration Layers and Cation Adsorption at the Alumina–Water Interface
- P2 Ana Suzana
Imaging Defect-driven Dissolution of Calcite
- P3 Paul Fenter
Imaging Surface Topography with Coherent X-ray Reflectivity
- P4 Lauren Beckingham
Kinetics of Hydrogen-Mediated Pyrite Dissolution and Pyrrhotite Formation in Mixed Mineral Systems
- P5 Ian Bourg
Nanoscale Basis of the Permeability of Fine-Grained Sedimentary Rocks Eric Boyd
- P6 Reductive Biomineralization of Iron Sulfur Minerals
Jacquelyn Bracco
- P7 *Inhibition of Impurity Adsorption and Nucleation at Mineral-Water Interfaces via Chelator Adsorption*
- P8 Emily Brodsky
Measuring the Consequences of Stress Organization on a Laboratory Fault
- P9 Giuseppe Buscarnera
Fabric Effects in Granular Materials: A Geomechanics Perspective
- P10 Patricia Dove
Macromolecular Controls on Structure-Property Relationships during Crystal Nucleation
- P11 Thomas Goebel
Assessing the Relative Contributions of Fluid Pressure and Elastic Stress to Induced Seismic Energy Release
- P12 Ryan Hurley
X-rays Probes for Rock Texture, Stress, and Fracture in 3D
- P13 Shaina Kelly
Pore-Confinement Effects on Mineral Crystallization Behaviors in Geologic Multiphase Flow Systems
- P14 Todd Ladd
Platonic Forms of Ideal Stalagmites
- P15 Christopher Johnson
AI for Earthquake Physics: Learning Fault Slip and Earthquake Precursors to Failure
- P16 Hari Viswanathan
Elucidating Hydrothermal Fracture Flow, Rheology, and Transport Phenomena
- P17 Benjamin Gilbert
Chemical-Mechanical Controls on Subsurface Resources

- P18 Donald DePaolo
Isotopic Studies at the Interface of Geochemistry and Rock Deformation
- P19 Mengsu Hu
Serpentinization under Anisotropic Stress
- P20 Abdullah Cihan
A cDFT-Based Framework for Interfacial-Electrokinetic Control of Nanoscale Flow in Geologic Systems
- P21 Drew Syverson
Geochemical Controls on the Hydrogen Evolution Reaction
- P22 Piotr Zarzycki
Isotopic Signatures of Fluid-Rock Interactions Close to and Far from Equilibrium
- P23 Carl Steefel
Chemo-Mechanical Modeling with NMM-Crunch
- P24 Olufemi Olorode
Molecular Studies of CO₂ Storage in Gas Hydrates
- P25 Andrew Stack
Understanding How Interfacial and Reactive Processes Influence Mineral Nucleation
- P26 Ke Yuan
How Defects Impact Heterogeneous Nucleation and Mineral Growth
- P27 Lawrence Anovitz
Chemomechanics: Understanding the Roles of Annealing, Reaction, and Crystal Growth on Mechanics and Fracture
- P28 Sebastien Kerisit
Mechanisms of Ni and Co Hydroxide–Carbonate Competitive Heterogeneous Growth
- P29 Kevin Rosso
Understanding Multicomponent Fluid Effects on Permeability Evolution in Fractures
- P30 Duo Song
AI-Assisted, Web-Based Automation for Geochemical Modeling
- P31 Laura Pyrak-Nolte
Critical Links: Exploring the Central Elements of Fracture Networks
- P32 Cornelia Rasmussen
Position-Specific Isotope Analysis via Nuclear Magnetic Resonance Spectroscopy
- P33 Mark Rivers
Development of X-ray Tomography for Subsurface Rock and Fluid Studies
- P34 Seth Saltiel
Machine Learning Rheology of Brittle-ductile Experiments and Crust = MARBLE-C Project Pls
- P35 Heather Savage
Coseismic Chemistry
- P36 Parisa Shokouhi
Coupled in-situ X-ray imaging and Acoustic/Ultrasonic Monitoring of Fractured Rock and Fault Reactivation
- P37 Anastasia Ilgen
Equilibrium Reactions in Nanoconfined Aqueous Solutions
- P38 Tuan Ho
Energy-Structure Relationship During Particle Oriented Attachment

- P39 Kevin Leung
Molecular Simulation of Confinement Effects on Structural, Dynamic, and Electrostatic Properties of Fluids and Electrolytes
- P40 Daniel Stolper
Stable Hydrogen Isotopes as Tracers of H₂ Reactivity in Geological Systems
- P41 Joanne Stubbs
Next Generation Synchrotron X-ray Methods and Tools for Quantifying Chemical Complexity at Mineral-Fluid Interfaces
- P42 Laura Waters
Determinations of REEPO₄ Solubilities in Acidic, Saline Solutions at High Temperatures and Pressure Using Isotope Dilution Methods
- P43 Nicole Hurtig
Determinations of REEPO₄ Solubilities in Acidic, Saline Solutions at High Temperatures and Pressure Using Isotope Dilution Methods
- P44 Hans-Rudolf Wenk
Microstructures and Anisotropy of Shales, Slates and Organic Slates
- P45 Anna Herring
Fundamental Controls on Capillary Trapping of scCO₂ in Geologic Storage: Linking Pore and Reservoir Scales
- P46 Dorte Wildenschild
Optimizing CO₂ Storage via Capillary Trapping
- P47 Jie Xu
Transition Metal Sulfide Nanostructures in Nature: Cluster-Guided Assembly of Amorphous M-MO-S (M = Fe, Co, Ni, Cu) Under Low-Temperature Sulfide Conditions
- P48 Yida Zhang
Interpretation of Chemically Mediated Crack Growth and Healing Using Surface Force-Based Fracture Theory
- P49 Wenlu Zhu
Evolution of Pore Structure and Rock Strength in Subsurface Fluid-Rock Systems

Abstracts

Understanding Carbon Mineralization Processes in Adsorbed Water Nanofilms

Mohammad Javad Abdolhosseini Qomi, Principal Investigator

Civil and Environmental Engineering, Mechanical and Aerospace Engineering, and Materials Science and Engineering, University of California Irvine, Irvine, CA

Graduate Students: Ali Morshedi, Siavash Zare, Mehrdad Youzi, Julian Stapper, Pablo Martin

Collaborators: Kevin Rosso (PNNL), Quin Miller (PNNL), Todd Schaef(PNNL)

Overall research goals: The program at the University of California Irvine (UCI) aims to understand the fundamental mechanisms driving the mineralization processes in adsorbed water nanofilms. The program converges reactive and classical molecular simulations, theory, and spectroscopy to delineate the elementary interfacial processes under nanoconfinement, where the conventional dissolution-precipitation fluid-rock reactivity paradigm breaks down.

A high-level overview: The program was established in 2021 based on an Early Career Award. To draw the broader geoscience community's attention to mineral reactivity in adsorbed water nanofilms, the PI authored a review article, recently published in Nature Reviews Chemistry, that delineates why reactivity in these confined media fundamentally differs from bulk fluid-mediated rock reactivity [1]. In short, the properties of water as both a solvent and a reactant in adsorbed water nanofilms differ markedly from those of bulk water. These differences directly affect ion-pairing dynamics, solubility limits, and the transport properties of ions and ion pairs, thereby altering reaction pathways and associated reaction kinetics. Building on this conceptual framework, the PI and his team have extensively studied the formation of adsorbed water nanofilms on mineral surfaces exposed to humidified carbon dioxide. They have further probed the deprotonation of bicarbonate in adsorbed water nanofilms via an inverse proton-transfer mechanism, leading to the formation of interfacial magnesium carbonate ion pairs. In addition, the group has systematically investigated the mechanisms of magnesium ion dissolution in adsorbed water layers, highlighting the critical role of protons in interfacial metal-proton exchange reactions and the role of carbonate anions in ligand-assisted dissolution. Using advanced molecular simulations, the UCI team quantified the effective energy barriers associated with several elementary reaction steps occurring within adsorbed water layers. In collaboration with PNNL, the PI and his team also performed in situ X-ray diffraction measurements of carbon mineralization in pure forsterite nanoparticles, demonstrating that the overall effective energy barriers measured experimentally fall within the range predicted by molecular simulations. More specifically, by juxtaposing numerical and experimental results, the team showed that dissolution is the rate-limiting step of the carbon mineralization process under near-saturated humidity conditions. In contrast, at smaller film thicknesses corresponding to lower humidity levels, interfacial ion transport becomes increasingly dominant. The team further identified two distinct transport mechanisms—ion hopping and crawling. Although ion hopping has a lower energy barrier, this transport mode contributes more substantially to overall ion transport compared to the ion crawling mechanism.

Significant achievements during 2023- 2025: Within this program, the PI and his team have developed a reactive force field for aqueous and interfacial magnesium carbonate formation [2]. These reactive force fields are developed based on a range of *ab initio* calculations and are shown to capture the physico-chemical processes involving water, carbon dioxide, bicarbonate, carbonate, silicates and magnesium ions both in bulk and magnesium silicate mineral surfaces. This force field and the corresponding codes are available to the community to use when studying carbon mineralization in magnesium-bearing silicates. From the scientific perspective, the team made significant contributions in understanding interfacial reaction mechanisms in adsorbed water nanofilms. Using the Grand Canonical Monte Carlo (GCMC) Simulations, the team showed how the temperature, pressure, and relative humidity affect the thickness of adsorbed water layers on the surface of metal silicates [3]. These estimates were

compared against IR spectroscopy measurements performed at PNNL. These comparative studies show transferable interatomic potentials along with GCMC technique can quantitatively predict the water film thickness across a range of thermodynamic conditions. The research showed for the first time that bicarbonate reactivity in adsorbed water layers is mediated by a reverse proton transfer mechanism [4]. The process has a small energy barrier in the presence of surface OH⁻. The team also worked on understanding the fundamental mechanisms of dissolution and interfacial transport processes using enhanced sampling methods such as well-tempered MetaDynamics and kinetic Monte Carlo technique and XRD and TGA measurements [5-9]. The team showed that the dissolution of Me²⁺ from add-atom, kink, ledge, and terrace sites depend on the coordination number and local bonding environment with interfacial water molecules. The metal ion detachment was shown to have a complex dissolution path that involves breakage of Mg-O-Si bonds and formation of Mg-H₂O bonds. The group also studied the transport of Mg²⁺ on the surface and distinguished two fundamental mechanisms of ion hopping and crawling. The ion hopping is energetically a rare event with high energy barrier. The crawling was shown to be more prevalent with lower energy barrier.

Science objectives for 2026-2028: The team is currently focused on the last task of the proposal, developing a mechanistic theoretical model of interfacial reactivity. A transport-mediated dissolution-precipitation model is currently developed based on the level-set method and its parameters are determined from atomistic calculations of varying water film thickness. The group aims to compare the simulation results against the XRD reactivity of minerals across a wide range of pressure and temperature. The PI plans to submit a continuation proposal that centers on interfacial reactivity with applications in geothermal energy and critical mineral recovery from crystalline rocks.

Publications supported by this project since the last PI meeting:

1. M.J. Abdolhosseini Qomi, Q. Miller, S. Zare, H.T. Schaef, J. P Kaszuba, K. M Rosso, "Molecular-scale mechanisms of CO₂ mineralization in nanoscale interfacial water films." *Nature Reviews Chemistry*, **2022**, 6, 598-613, <https://doi.org/10.1038/s41570-022-00418-1>.
2. S. Zare, M.J. Abdolhosseini Qomi, "Reactive force fields for aqueous and interfacial magnesium carbonate formation." *Physical Chemistry Chemical Physics*, **2022**, 23, 23106-23123, <https://doi.org/10.1039/D1CP02627E>.
3. S. Zare, K.M.S Uddin, A. Funk, Q. Miller, M.J. Abdolhosseini Qomi, "Nanoconfinement matters in humidified CO₂ interaction with metal silicates." *Environmental Science: Nano*, **2022**, 9, 3766-3779, <https://doi.org/10.1039/D2EN00148A>.
4. S. Zare, A. Funk, M.J. Abdolhosseini Qomi, "Formation and dissolution of surface metal carbonate complexes: Implications for interfacial carbon mineralization in metal silicates" *Journal of Physical Chemistry C*, **2022**, 126, 11574-11584, <https://pubs.acs.org/doi/10.1021/acs.jpcc.2c02981>.
5. Y. Tao, S. Zare, F. Wang, M.J. Abdolhosseini Qomi, "Atomistic Thermodynamics and Kinetics of Dicalcium Silicate Dissolution" *Cement and Concrete Research*, **2022**, 157, 106833, <https://doi.org/10.1016/j.cemconres.2022.106833>.
6. B. Aguila, L. Hardee, H.T. Schaef, S. Zare, M.J. Abdolhosseini Qomi, J.V. Crum, J.E. Holliman Jr, E.T. Rodriguez, L.M. Anovitz, K.M. Rosso, Q. Miller, "Emerging investigator series: kinetics of diopside reactivity for carbon mineralization in mafic-ultramafic rocks" *Environmental Science: Nano*, **2023**, 10, 2672-2684, <https://doi.org/10.1039/D3EN00087G>.
7. M. Youzi, J.L. Stapper, S. Zare, H.T. Schaef, M.E. Bowden, K.M. Rosso, Q. Miller, M.J. Abdolhosseini Qomi, "Carbon mineralization pathways in interfacial adsorbed water nanofilms." *Journal Name or Abbreviation*, **2025**, 6, 478, <https://doi.org/10.1038/s43247-025-02436-5>.
8. Y. Tao, P. Martin, H. Manzano, M.J. Abdolhosseini Qomi, "Mesoscopic mechanisms of dicalcium silicate dissolution" *Cement and Concrete Research*, **2025**, 189, 107660, <https://doi.org/10.1016/j.cemconres.2024.107660>.
9. Y. Tao, Y. Gao, M.J. Abdolhosseini Qomi, R.J-M. Pellenq, C.S. Poon, "Exploring the aluminium paradox in calcium-aluminosilicate-hydrate decalcification" *Nature Communications*, **2025**, <https://doi.org/10.1038/s41467-025-67686-5>.

Influence of Turbulence on Momentum Transfer at the River-Hyporheic Zone Interface: An Approach Combining Experiments, High-Performance Computing, and Upscaling Theory With Machine Learning

Brian D. Wood (Principal Investigator)

Sourabh V. Apte, and Jim Liburdy (Co-Principal Investigators)

Oregon State University, Corvallis, OR

Graduate Students: Daniel Fust, Jordan Strahl, Zeeshan Memon

Overall research goals: The main research goal is to quantify momentum transfer at the interface between rivers and hyporheic zones as influenced by turbulent flow structures using (i) direct experimental observations using a refractive-index matched particle-image velocimetry (PIV) for detailed flow measurements, (ii) high-resolution numerical modeling of the Navier-Stokes equations using combination of large-eddy simulations (LES) and direct numerical simulations (DNS), (iii) development of upscaled theory and single closure model for unresolved stress based on machine learning and physics-informed modeling.

A high-level overview: The interface between rivers and groundwater has been identified as a critical component of watershed ecohydrology. The spatially and temporally fluctuating region where surface waters and groundwaters mix is termed as the hyporheic zone. The hyporheic zone has been identified as being one of the most important components controlling the exchange of nutrients (including carbon sources, electron donors, and electron acceptors such as oxygen) and other aqueous components (such as colloids and dissolved contaminants) between the surface and groundwater systems.

The overarching hypothesis is that turbulent flow structures interact with the riverbed to dramatically increase the net momentum flux (and, consequently, mass flux) through the hyporheic zone. Such increases in momentum transfer would not be captured by models using the Reynolds-averaged Navier-Stokes equations. This phenomenon has significant potential impacts on the modeling of nutrient and other dissolved chemical species at the interface between the river and groundwater. The main objective of this work is to improve understanding of this interfacial transport and develop new upscaled models that can be used in large-scale hydrologic models. Specifically, it is hypothesized that the standard permeability-porosity relations, applicable deep within the bed, are not valid in the sediment-water interface region of rapidly varying porosity and need modifications.

The chief novelty of this work involves (i) developing time-resolved, refractive-index matched PIV setup for turbulent flows over a bed of randomly packed particles and using it to quantify flow statistics in the interface region, (ii) applying volume-averaging techniques to existing and new pore-resolved, high-resolution numerical data of inertial and turbulent flows over a packed bed to quantify the unresolved subfilter stress and drag force, (iii) using the filtered data to develop improved porosity-permeability parameterizations in the interface region, (iv) developing physics-informed model corrections using machine learning techniques.

Significant achievements during 2023- 2025: Significant progress has been made over the past few years on all fronts.

- (i) A refractive index-matched flume for turbulent open channel flow over a randomly packed bed of monodispersed spherical particles was developed and used to collect flow data in the free flow and in the interface region for a range of permeability Reynolds numbers (0.01-2). This work resulted in a completed doctoral thesis (Jordan Strahl) and is being continued by Zeeshan Memon.
- (ii) Numerical simulations were performed to obtain detailed, three-dimensional flow data for a doubly periodic open channel flow over a randomly packed bed for a wide range of

permeability Reynolds numbers. This data was explicitly volume-averaged to obtain bed-normal variation of volume-averaged subfilter stress, drag force, porosity, and permeability. Different filter kernels; namely, box, Gaussian, quartic, and cellular, with different filter widths in the bed-normal direction to analyze the data. It was shown that the standard Carman-Kozeny permeability-porosity correlation significantly overpredicts the drag force and underpredicts the permeability in the interface region.

- (iii) To improve the closure models in the interface region, a kernel with varying filter width that is proportional to the pore size deep within the bed and smoothly transitions to grid resolution in the free flow is devised. A physics-informed, two-parameter correction based on Brinkman solution for the velocity within the bed is developed to correct the permeability model in the transition region with rapid porosity variation. This corrected model is shown to give very good results compared to the resolved data. A report has been published and a journal article (sent to the *Journal of Fluid Mechanics*) is in review.

Science objectives for 2026-2028:

- (i) Although the corrected model improves prediction, it does not capture the roughness effect of the top layers of particles. To capture these effects and hence the turbulence penetration across the sediment-water interface, a three-dimensional filter-kernel with local roughness-capturing porosity and permeability variation is necessary. A machine-learning-based nonlinear model is being explored and developed to relate permeability and mean drag force to pore-size, porosity, gradients in porosity, superficial velocity, and the gradient of the superficial velocity. A framework for such physics-informed machine learning-based closure has been developed and tested in the context of reactive transport by the PI. This framework is being explored for permeability modeling.
- (ii) A posteriori analysis of the new corrected model is being conducted, and a journal article is being prepared. Graduate student Dainel Fust will complete this article as part of his thesis.
- (iii) The above numerical framework will be extended to 'Physics-Informed Multiscale Modeling of Interfacial Thermal-Hydraulic-Chemical Coupling in Geothermal Systems' as a new research proposal with focus on enhanced geothermal systems.

Selected Publications supported by this project 2023-2025:

- (i) Daniel Fust, Sourabh V. Apte, S. Balachandar, "A Force Correction Scheme for A Volume-Filtered Formulation of Flow Over a Permeable Particle bed" *Proceedings of the 2024 Summer Program at Center For Turbulence Research*, **2024**
<https://ctr.stanford.edu/publications/summer-program-proceedings/proceedings-2024-summer-program>
- (ii) Daniel Fust, Sourabh V. Apte, Brian D. Wood, S. Balachandar, "Modeling Drag Force in Porous Media: A Physics-Informed Closure for Inhomogeneous Porosity" *Journal of Fluid Mechanics*, **2026**, under review.
- (iii) Ehsan Taghizadeh, Helen M. Byrne, and Brian D. Wood, "Explicit Physics-Informed Neural Networks for Nonlinear Closure: The Case for Transport in Tissues" *Journal of Computational Physics*, **2022**, 449, 110781, <https://doi.org/10.1016/j.jcp.2021.110781>
- (iv) Jordan Strahl, "Particle Image Velocimetry Study of Turbulent Flow Over Porous Media" PhD. Thesis, **2025**, Oregon state University,
https://ir.library.oregonstate.edu/concern/file_sets/tq57p1126?locale=es

Mechanisms of hydrogen interaction with earth materials in subsurface storage formations

Lauren E. Beckingham, Principal Investigator

Department of Civil & Environmental Engineering, Auburn University, Auburn, AL

Postdoctoral scholar: Dejene L. Driba

Graduate Student: Riana Rivera

Overall research goals:

Underground hydrogen storage (UHS) in subsurface porous formations is a promising means of energy storage. Salt caverns, depleted oil and gas reservoirs and saline aquifers are potential host formations where saline aquifers are particularly appealing due to their high potential storage capacities and geographic prevalence. While promising, interactions between the injected hydrogen, formation fluids, and formations minerals in saline aquifers may reduce the economic and technological viability of the storage system but are not well understood. Using advanced laboratory experiments, this work aims to enhance understanding of hydrogen-brine-mineral interactions under conditions pertinent for subsurface hydrogen storage in saline formations. Experiments will yield new understanding of mineral reactions, reaction rates, and impacts on formation properties. This fundamental knowledge is needed to design safe, effective, and efficient underground hydrogen storage systems.

Significant achievements during 2023-2025:

Following injection into a saline storage formation, hydrogen may be depleted by hydrogen-consuming redox reactions, namely pyrite reduction and subsequent pyrrhotite precipitation. Here, laboratory experiments mimicking hydrogen storage conditions were carried out to enhance understanding of hydrogen driven pyrite reduction mechanisms and kinetics. Significantly, we aimed to evaluate the hypothesis that the presence of coexisting minerals (siderite, hematite, barite) may modify reaction rates and mechanisms relative to pyrite-only systems. Batch experiments were conducted in a sealed high-pressure, high temperature batch reactor at 90–120 °C under 18 bar PH_2 (75% Ar/25% H_2). Crushed natural pyrite (63–90 μm) was reacted with non-buffered water (pH ~9, NaOH-adjusted) under anoxic conditions for 300 hours. Additional experiments included mixtures of pyrite with each coexisting mineral. $\text{H}_2\text{S}(\text{g})$ evolution was monitored by collecting headspace gas samples and analyzing them via gas chromatography (GC). At the end of the experiment, pyrrhotite (FeS) formation was analyzed using scanning electron microscopy (SEM) and X-ray diffraction (XRD). PHREEQC geochemical modeling was used to support interpretation. The result showed that Pyrite-only systems exhibit limited hydrogen consumption and low H_2S generation, indicating minimal hydrogen loss in the absence of other mineral phases. In contrast, systems containing co-existing iron-bearing and sulfate-associated minerals display enhanced hydrogen consumption, driven by sustained $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox cycling and sulfate reduction pathways that promote continued pyrite dissolution. These findings highlight the kinetics of H_2 -induced reductive dissolution of pyrite as well as the role of coexisting minerals in controlling pyrite reactivity, sulfide generation, and overall hydrogen storage efficiency during UHS.

Science objectives for 2026-2028:

In our ongoing work, we aim to understand the implications of our observed reaction rates and mechanisms in field systems, including in different regions of the storage system where cushion gas may be present. It has been well noted that reaction rates observed in batch, high reactive surface area systems do not reflect rates observed in field scenarios where porous media effects reduce reaction rates. Here, we seek to evaluate hydrogen driven pyrite reduction reaction rates in flow-through mixed mineral systems and impacts on host formation properties that control hydrogen

migration. Core-scale flow-through experiments will be performed and advanced analytical and imaging used to determine reaction rates, mechanisms, and changes in formation properties.

In field storage systems, injection of a cushion gas will proceed hydrogen injection, acting to create a gas saturated region and sustain desired operational conditions (e.g., pressure). CO₂ is a potential favorable choice of cushion gas but there is limited knowledge on its feasibility as a cushion gas from a geochemical perspective. Here, we aim to increase understanding of whether CO₂ used as a cushion gas can suppress hydrogen consumption by modifying fluid chemistry and mineral reactivity in the hydrogen–CO₂–brine mixing zone. This will be evaluated through advanced laboratory experiments mimicking potential variations in cushion gas to working gas ratios under hydrogen storage conditions.

Publications supported by this project 2023-2025:

1. Driba, Degene L., Beckingham, Lauren E. “Unraveling hydrogen induced geochemical reaction mechanisms through coupled geochemical modeling and machine learning” *Applied Geochemistry*, **2025**, 183, 106330, DOI: [10.1016/j.apgeochem.2025.106330](https://doi.org/10.1016/j.apgeochem.2025.106330).
2. Shi, Zhuofan, Driba, Degene L., Lopez Rivera, Nora, Kariminasab, Mohammad, Beckingham, Lauren E. “A review of coupled geochemical-geomechanical impacts in subsurface CO₂, H₂, and air storage systems” *Energies*, **2024**, DOI: [10.3390/en17122928](https://doi.org/10.3390/en17122928).

Impact of variations in molecular driving forces at substrate-mineral interfaces on mineral precipitation in porous media

Lauren E. Beckingham, Principal Investigator

Department of Civil & Environmental Engineering, Auburn University, Auburn, AL

Postdoctoral scholar: Md. Fahim Salek

Graduate Students: Mark Elungat, Samantha Mariano

Collaborators: Starchenko, Vitalii (ORNL), Lee, Sang Soo, Lee, Byeongdu (ANL)

Overall research goals:

Mineral precipitation reactions can occur in a range of natural and engineered environmental systems, resulting from natural weathering processes, contaminant transport, geologic carbon or hydrogen storage, and radioactive waste disposal, for example. Reactions can change critical formation properties including porosity and permeability, impacting flow, transport, and potentially the risk and integrity of the system. Accurate understanding and prediction of the rate and extent of precipitation, and impacts on formation properties, is critical to assess risks and engineer solutions to environmental problems. Predictive capabilities of mineral precipitation are largely limited by a lack of fundamental understanding of how variations in molecular driving forces at substrate-mineral interfaces control mineral nucleation and growth rates and mechanisms. In this work, we aim to increase understanding and predictive capabilities of mineral nucleation and growth in diverse environments in porous media using advanced laboratory and numerical simulation tools. Through controlled variation in experimental conditions, the molecular scale driving force for precipitation reactions will be altered and resulting reaction rates and mechanisms observed. New, pore scale numerical simulation tools will be leveraged to model reactions in diverse conditions.

Significant achievements during 2023- 2025:

Mineral precipitation is a complex process that involves nucleation and mineral growth, fundamentally distinct from mineral dissolution. Variations in reaction rates and mechanisms may occur with different solution composition, saturation index, surface composition, etc. Here, we are developing laboratory experiments and numerical simulations to understand how reaction rates and mechanisms vary in diverse environments. Towards this, a flow-through laboratory experimental system was designed and implemented to enable controlled precipitation experiments in single porous media. Trial tests refined experimental design, assessed reproducibility, and identified conditions suitable for investigating coupled transport and reaction processes in porous media. Trial experiments utilized 3D X-ray micro computed tomography to characterize pore structures, substrate surfaces, and early-stage precipitation. These analyses provided quantitative and qualitative insights into grain-size distributions, pore connectivity, surface alteration, and mineral nucleation. Partnerships with Argonne National Laboratory (Sang Soo Lee, Byeongdu Lee) have been initiated to leverage DOE user facilities for advanced X-ray scattering experiments, strengthening cross-institutional expertise and access to high-resolution, time-resolved characterization tools.

The objective of the numerical simulations is to model mineral precipitation and the associated evolution of pore permeability in a multi-pore nano and macro pore system. Simulations will leverage pore scale numerical simulation in OpenFOAM, integrating classical nucleation theory and a dynamic mineral-fluid interface mesh using a new generation customized hydro-geochemical solver within the OpenFOAM computational fluid dynamics framework. Uniquely, it considers mineral precipitation via heterogeneous nucleation using classical nucleation theory, where convection-diffusion accounts for ion transport and a dynamic mesh approach evolves the mesh surfaces as minerals precipitate. Preliminary simulations have successfully been conducted that provide the

steady state flow field and spatial distribution of the saturation index (SI) in an idealized micropore system. Initially, a simple geometry is considered, consisting of two connected pores. The flow simulation is carried out using the PISO algorithm implemented in OpenFOAM, and a customized solver is used to alter the solid interface using dynamic volume mesh and boundary mesh motion/relaxation. After certain time steps, the defined volume is re-meshed using snappyHexMesh to capture the changes. Varieties of sizes and shapes of pores are being tested during the initial stage, and the effect on the fluid flow distribution and transport of ions is observed. An experimental workflow is also being designed to conduct microfluidic experiments under conditions similar to those in the simulation to optimize and validate the simulation. Here, the micropore system is 3D-printed using stereolithography. While the solutions are being injected, the calcite precipitation is monitored using a high-speed camera. At the same time, a pressure transducer measures the pressure difference between the inlet and outlet to determine the evolution of permeability. This work is ongoing and results are yet to be analyzed.

Science objectives for 2026-2028:

Building on progress from trial experiments and advanced imaging, the next phase will focus on quantifying the mechanistic controls of pore- and substrate-scale heterogeneity on carbonate mineral nucleation and growth under flow conditions. Specific objectives include:

- **Implement in situ SAXS/WAXS measurements for dynamic characterization:** Conduct time-resolved, cross-scale measurements of particle size, shape, and phase evolution in flow-through columns to capture nucleation, growth, and aggregation processes under controlled chemical and hydrodynamic conditions.
- **Determine substrate-dependent behavior:** Evaluate how mineral composition and surface properties influence heterogeneous nucleation, growth rates, and precipitate aggregation in nano- and macro-pores.
- **Integrate experiments with modeling:** Pore-scale reactive transport simulations incorporating classical nucleation theory and evolving mineral-fluid interfaces will predict precipitation and permeability evolution across diverse porous media.
- **Publications:** Results from experiments and simulations will be published in peer-reviewed journals to advance understanding of molecular-scale controls on precipitation and permeability evolution in porous media, thereby advancing DOE priorities in energy, water, and environmental management.

Publications supported by this project 2023-2025:

Works are currently in preparation.

Project: Properties and dynamics of fault zones and the shallow crust

Yehuda Ben-Zion, Principal Investigator

Department of Earth Sciences, University of Southern California, Los Angeles, CA

Graduate Students and Postdocs: Shuye Huang, Guoliang Li, Zihua Niu, Sebastian Núñez-Jara, Nico Schliwa, Hao Zheng

Collaborators: Ahmed Elbanna (USC), Alice-Agnes Gabriel, Frank Vernon (UCSD), Florent Brenguier, Aurelien Mordret (Univ. Grenoble Alpes France), Grzegorz Kwiatek, Patricia Martínez-Garzón, Gerog Dresen (GFZ Germany), Francois Renard (Univ. of Oslo, Norway)

Overall research goals:

The project aims to advance the understanding of properties and dynamics of fault zones and the shallow crust by providing detailed *in-situ* information on spatio-temporal variations of P and S seismic velocities (V_p , V_s), attenuation coefficients (Q_p , Q_s), and microearthquakes around large faults and in the shallow crust. The research applies state-of-the-art seismic imaging, monitoring temporal changes of properties, and earthquake analysis techniques to continuous seismic waveforms recorded by dense arrays, borehole sensors, and regional seismic stations. Results of laboratory fracturing experiments and modeling are also used to clarify the processes associated with large failure events of crustal materials.

Significant achievements during 2023- 2025:

The imaging results reveal hierarchical rock damage structures around large faults with intense core damage zones that follow an overall flower-shape structure, with significant damage in the top few km that decreases in amplitude and width with depth. Shallow materials in the top 50-100 m have extreme properties (very low V_p , V_s , Q values; very high V_p/V_s ratios) that make it, together with the low normal stress, highly susceptible to failure and temporal changes. Large earthquakes produce widespread changes of seismic velocities that decay with distance but remain significant at shallow depth on a regional scale. The co-seismic velocity changes can be >30% in the top 20-30 m and are followed by $\log(t)$ recovery. Appreciable changes of shallow velocities generated by fluid and temperature effects are also observed on daily, seasonal, and longer timescales. Frequent upgoing high-frequency waves point to the occurrence of numerous small earthquakes in the top crust. Detecting pre-seismic temporal changes remains elusive but analysis of repeating body waves produced by heavy trains detected changes related to slow slip event on the San Jacinto fault in CA.

Science objectives for 2026-2028:

(1) Detecting and characterizing very small events (shear and tensile) in the top crust. (2) Clarifying the depth extent of property changes at different times. (3) Clarifying the rheology (brittle vs. viscous-like) of the top crust. (4) Merging the detailed imaging results for the top crust with regional velocity models to provide multi scale models that can be used to simulate ground motion from large scenario earthquakes. (5) Using the simulations to improve assessments of seismic hazard and risks to communities and critical infrastructure (6) Clarifying with *in-situ* and lab data the processes and signals (e.g., localization, “damage related radiation”) leading to large failure events.

Selective Publications supported by this project 2023-2025:

Huang, S., H. Meng and Y. Ben-Zion. A Comparative seismological study of the Banning and Mission Creek strands of the Southern San Andreas Fault, *J. Geophys. Res.*, **2025**, doi: 10.1029/2025JB031870.

Kwiatek, G., P. Martínez-Garzón, T. Goebel, M. Bohnhoff, Y. Ben-Zion and G. Dresen. Intermittent criticality multi-scale processes of stick-slip events on rough laboratory faults, *J. Geophys. Res.*, **2024**, doi: 10.1029/2023JB028411.

- Li, G. and Y. Ben-Zion. Daily and seasonal variations of shallow seismic velocities in southern California from analysis of H/V ratios and autocorrelations of seismic waveforms, *J. Geophys. Res.*, **2023**, doi: 10.1029/2022JB025682.
- Li, G. and Y. Ben-Zion. Multi-scale seismic imaging of the Ridgecrest, CA, region with full-waveform inversions of regional and dense array data, *J. Geophys. Res.*, **2024**, doi: 10.1029/2023JB028149.
- Mathiesen, J., McBeck, J., B. Cordonnier, Y. Ben-Zion and F. Renard. Deep-learning analysis of fracture networks leading to system-size failure in rocks, *J. Geophys. Res., Machine Learning and Computation*, **2024**, doi: 10.1029/2023JH000103.
- Meng, X., C. Pinilla-Ramos, A. Kottke, and Y. Ben-Zion. Investigating Near-fault Ground Motions Using Data Recorded by Dense Arrays around the 2019 M_w 7.1 Ridgecrest, CA, Earthquake Rupture, *Bull. Seism. Soc. Am.*, **2025**, doi: 10.1785/0120250092.
- Niu, Z., A.-A. Gabriel and Y. Ben-Zion. Delayed dynamic triggering and enhanced high-frequency seismic radiation due to brittle rock damage in 3D multi-fault rupture simulations, *J. Geophys. Res.*, **2025**, doi: 10.1029/2025JB031632.
- Núñez-Jara, S., P. Martínez-Garzón, G. Kwiatak, Y. Ben-Zion, G. Dresen, D. Becker, F. Cotton and M. Bohnhoff, 2025. Unraveling the spatiotemporal fault activation in a complex fault system: the run-up to the 2023 M_w 7.8 Kahramanmaraş earthquake, Türkiye, *Earth Planet. Sci. Lett.*, **2025**, doi: 10.1016/j.epsl.2025.119570.
- Qin, L., H. Qiu, N. Nakata, S. Deng, A. Levander and Y. Ben-Zion. Variable daily autocorrelation functions of high frequency seismic data on Mars, *Seis. Res. Lett.*, **2023**, doi: 10.1785/0220220196.
- Schliwa, N., A.-A. Gabriel and Y. Ben-Zion. Shallow fault zone structure affects rupture dynamics and ground motions of the 2019 Ridgecrest sequence to regional distances, *J. Geophys. Res.*, **2025**, doi: 10.1029/2025JB031194.
- Sheng, Y., A. Mordret, F. Brenguier, P. Boué, F. Vernon, T. Takeda, Y. Aoki T. Taira and Y. Ben-Zion. Seeking Repeating Anthropogenic Seismic Sources: Implications for Seismic Velocity Monitoring at Fault Zones, *J. Geophys. Res.*, **2023**, doi: 10.1029/2022JB024725.
- Sheng, Y., F. Brenguier, A. Mordret, Q. Higuieret, C. Aubert, L. Pinzon-Rincon, D. Hollis, F. Vernon, F. Wyatt and Y. Ben-Zion. In-situ velocity-strain sensitivity near the San Jacinto Fault Zone analyzed through train tremors, *Geophys. Res. Lett.*, **2024**, doi: 10.1029/2024GL110113.
- Yang, B., H. Meng, N. Gu, X. Liu, X. Chen, and Y. Ben-Zion. A Frequency Domain Methodology for Quantitative Evaluation of Diffuse Wavefield with Applications to Seismic Imaging, *J. Geophys. Res.*, **2024**, doi: 10.1029/2024JB028895.
- Zhang, S., B. Luo, Y. Ben-Zion, D. E. Lumley and H. Zhu. Monitoring terrestrial water storage, drought and seasonal changes in central Oklahoma with ambient seismic noise, *Geophys. Res. Lett.*, **2023**, doi: 10.1029/2023GL103419.
- Zhang, H. and Y. Ben-Zion. Enhancing regional seismic velocity models with higher-resolution local results using sparse dictionary learning, *J. Geophys. Res.*, **2024**, doi: 10.1029/2023JB027016.
- Zhao, C., M. S. Mia, A. Elbanna, and Y. Ben-Zion. Dynamic Rupture Modeling in a Complex Fault Zone with Distributed and Localized Damage, *Mechanics of Materials*, **2024**, doi: 10.1016/j.mechmat.2024.105139.

Discerning Fault Geometry and Mechanics from Seismicity Point Clouds

Gregory C. Beroza, Principal Investigator

Department of Geophysics, Stanford University, Stanford, CA, 94305-2215

Graduate Students: Albert Leonardo Aguilar-Suarez, Jaehong Chung, Yanlan Hu

Overall research goals:

Improvements enabled by deep learning have led to a rapidly proliferating new generation of earthquake catalogs. Many of these improvements are driven by deep learning-based phase picking models, which significantly improve the detection of S-waves and the picking of both *P*- and *S*-waves at low snr. The first component of this project focuses on the forensics of deep-learning-based catalogs, including: how well phase pickers work across different earthquake monitoring environments, how that manifests in terms of the magnitude of completeness, and based on this understanding, what can be done to improve the reliability of these catalogs. We are looking to improve all aspects of their construction. This will start with phase picking, but also extend to phase association, event location, and focal mechanism determination.

Significant achievements during 2023- 2025:

We started the project in 2025. We have submitted two articles for publication. Both are related to the first component of the project – assessing and improving machine learning catalogs. *Chung et al. (2026)* demonstrated that the probability of detecting seismic phases is qualitatively different for conventional vs. deep-learning-based phase pickers. Deep-learning-based pickers are more sensitive than conventional approaches, but the variability between stations and with distance/magnitude of deep-learning pickers is greater than for conventional methods. Understanding the source of this greater variability is the key to reducing it and making deep-learning results more consistent. *Aguilar-Suarez and Beroza (2026)* document the existence of pervasive errors in existing seismological benchmark datasets that are used for training and testing deep-learning-based pickers. The most common of these errors is the lack of labels for small earthquakes that are present in the waveforms but are unlabeled in the benchmark datasets. The prevalence of such errors could contribute to missing phase picks because deep-learning models will inadvertently be trained to ignore wave arrivals from small earthquake because they have been exposed to many examples of unlabeled picks. To improve our models, and to help the community improve their models, we have provided flags of these errors for existing benchmark data sets in the Seisbench working environment.

Science objectives for 2026-2028:

We don't have a publication on this year, but we have developed heuristic patterns to identify incorrect phase associations based on the pattern of associated vs. unassociated phases across a seismic network. If we identify sufficient numbers of bad associations, then it might be possible to train a machine learning model to identify them. We are working to optimize phase association such that we can reliably connect phases with the earthquakes that generated them. We plan to extend our focal mechanism determination method from assumed double-couple mechanisms to general moment-tensor sources. Work up to this point will be focused on a combination of catalog forensics followed by remediation of shortcomings we have exposed in the process.

For the second component of this project, we will focus on using the more comprehensive, reliable, and complete seismicity catalogs developed in the first part of the project as a foundation. For this we are working with deep learning segmentation models that have been created to discern surfaces (in our case piecewise planar fault surfaces) from distributed point clouds (in our case earthquake hypocenters). Individual earthquakes are treated as points in our proof-of-concept work to date, but higher-order information, such as focal mechanisms or waveform similarity, can also be used for this purpose.

The fault geometries determined from the second part of the project will serve as input into the third part of the project, which pursues the science goal of using the fault structures we discern, and the evolution of seismicity on them, to develop approaches to distinguish tectonic from human-induced earthquakes. We will include the analysis of tectonic earthquake swarms in this effort due to the

supposition that, like injection-induced earthquakes, they are triggered by changes in pore fluid pressure. We will combine the seismicity and fault structure information that emerge from these tasks to find relationships in the spatiotemporal evolution of seismicity that have the potential be used for probabilistic earthquake forecasting. We will develop our methods using earthquake catalogs from diverse settings, but we will apply what we have learned to study California as a natural laboratory due to the good instrumental coverage, the presence of significant numbers of tectonic earthquakes, and the presence of multiple sources of human-induced seismicity.

Publications supported by this project 2023-2025:

1. Aguilar-Suarez, Albert, Leonardo and Gregory C. Beroza, "Pervasive Errors in Seismological Machine Learning Datasets" *Seismica* **2026** (submitted).
2. Chung, Jaehong, Yifan Yu, Lauro Chiaraluce, Maddalena Michele, and Gregory C. Beroza, "Variability in performance of a machine-learning seismicity catalog: Central Italy, 2016-2017" *Seismological Research Letters*, **2026** (submitted).

Nanoscale Basis of the Permeability of Fine-Grained Sedimentary Rocks

Ian C. Bourg, Principal Investigator

Department of Civil and Environmental Engineering, Princeton University, Princeton, NJ

Graduate Students: Emily Sun, Xinyi Shen, Xiaohan Li, Avery Agles, Mitchell Jans, Caridad Estrada-Cardona, Jiaqi Wen, Bright Nyameasem

Collaborators: Thomas Underwood, Ayumi Koishi, Bastien Wild, Zhanar Zhakiyeva, Xiaojin Zheng, Sang Soo Lee (ANL), Paul Fenter (ANL), Kevin Rosso (PNNL), Jessica Rimsza (SNL), Alan Seltzer (WHOI), Alex Fernandez-Martinez (U of Grenoble), Carlos Vega (U Complutense de Madrid), Martina Havenith (U Ruhr-Bochum)

Overall research goals:

The overarching goals of this project are (i) to understand the fundamental, molecular-scale properties of water at interfaces and in nanopores and (ii) to develop new knowledge of how these properties influence geoscience phenomena observed at the human scale (e.g., at the scale of a soil, sediment, or rock sample) such as adsorption, solute transport, isotope fractionation, fluid flow, capillary phenomena, colloid transport, and poromechanics. Key molecular-scale systems of interest include water-clay interfaces (the predominant mineral-water interface in most soils, sediments, and sedimentary rocks) and water-air interfaces (which play important roles in the geochemistry and transport properties of unsaturated porous media).

Significant achievements during 2021- 2025:

Since the last PI meeting in August 2021, this project has made significant achievements on several important questions related to the overall research goals. A selection of these achievements is listed here. First, the project discovered an important transition in the properties of confined liquid water, whereby water becomes a distinctly less cohesive fluid when confined to dimensions below ~4 nm [2,4,7]. This transition appears to be associated with the highly collective nature of water's hydrogen bond network and has important implications for colloidal interactions in water [4] and for water-organic phase separation in small clusters [7].

Second, the project developed molecular [6], coarse-grained [12], and continuum-scale [11] representations of coupled transport, mechanics, and thermal properties of compacted clay systems at temperatures up to 373 K. This unlocks new abilities to predict the performance of enhanced geothermal systems and nuclear waste repositories through multi-scale simulations of shales and other clay-rich media at elevated temperatures.

Third, the project developed several new molecular simulation approaches to characterize the properties of adsorbed water films on mineral surfaces, including an order-of-magnitude improvement in the ability to resolve the energetics of these films (i.e., their disjoining pressure) [8]. This has led to important new understanding of the relationships between capillary and adsorbed water on mineral surfaces [5,8] and the impact of ions on mineral hygroscopicity [3,8], two phenomena that drive fluid flow in unsaturated porous media.

Fourth, the project generated new insight on the energetics of aggregation of organic molecules at water-mineral [9] and water-air interfaces [10]. Results demonstrate that clustering of organic molecules is driven by both polar and hydrophobic interactions in ways that can be either additive or opposite. These results have important implications for our understanding of the migration of organic compounds in geologic media.

Science objectives for 2026-2028:

Key objectives for 2026-2028 include three major thrusts. First, the project has been developing a new framework to understand how molecular fluctuations in fluid-filled nanopores give rise to coupled

transport properties (e.g., coupling between fluxes of solution, salt, heat, and ionic current). A major objective will be to complete and publish this new framework. This will lay the groundwork for its application in key systems (compacted clay, nanoporous brucite) and for future theoretical developments, including efforts to develop new understanding of clay mechanics and new rock characterization approaches using impedance spectroscopy.

Second, the project has recently developed a multi-scale simulation framework that can predict coupled flow, mechanics, and thermal effects in clay-rich media at elevated temperatures. This framework has been developed and validated in the case of pure Na-smectite clay. A major objective will be to extend this new framework to other aqueous chemistries and more complex mineralogies relevant to clay sediments and clay-rich rock formations.

Third, the project will maintain a significant focus on generating new fundamental understanding of the properties of water at interfaces and in nanopores. An important emphasis will be on completing ongoing work on the high-frequency vibrational dynamics of water and solutes, including comparisons with THz spectroscopy and isotope fractionation results.

Publications supported by this project 2021-2025:

Since the last PI meeting in August 2021, this project has supported 20 publications, including one currently in review. A selection of these publications is listed below.

1. Sang Soo Lee, Ayumi Koishi, Ian C. Bourg, and Paul Fenter, "Ion correlations drive charge overscreening and heterogeneous nucleation at solid-aqueous electrolyte interfaces" *Proc. Natl. Acad. Sci. U.S.A.*, **2021**, <https://doi.org/10.1073/pnas.2105154118>.
2. Thomas R. Underwood and Ian C. Bourg, "Dielectric properties of water in charged nanopores" *J. Phys. Chem. B*, **2022**, <https://doi.org/10.1021/acs.jpcc.1c09688>.
3. Ayumi Koishi, Sang Soo Lee, Paul Fenter, Alejandro Fernandez-Martinez, and Ian C. Bourg, "Water adsorption on mica surfaces with hydrophilicity tuned by counterion type (Na, K, Cs) and structural fluorination" *J. Phys. Chem. C*, **2022**, <https://doi.org/10.1021/acs.jpcc.2c04751>.
4. Xinyi Shen and Ian C. Bourg, "Interaction between hydrated smectite clay particles as a function of salinity (0 to 1 M) and counterion type (Na, K, Ca)" *J. Phys. Chem. C*, **2022**, <https://doi.org/10.1021/acs.jpcc.2c04636>.
5. Emily W.-H Sun and Ian C. Bourg, "Impact of organic solutes on capillary phenomena in water-CO₂-quartz systems" *J. Colloid Interface Sci.*, **2023**, <https://doi.org/10.1016/j.jcis.2022.08.124>.
6. Xiaojin Zheng, Thomas R. Underwood, and Ian C. Bourg, "Molecular dynamics simulation of thermal, hydraulic, and mechanical properties of bentonite clay at 298 to 373 K" *Appl. Clay Sci.*, **2023**, <https://doi.org/10.1016/j.clay.2023.106964>.
7. Xiaohan Li and Ian C. Bourg, "Phase state, surface tension, water activity, and accommodation coefficient of water-organic clusters near the critical size for atmospheric new particle formation" *Environ. Sci. Technol.*, **2023**, <https://doi.org/10.1021/acs.est.2c09627>.
8. Xiaohan Li and Ian C. Bourg, "Hygroscopic growth of adsorbed water films on smectite clay particles" *Environ. Sci. Technol.*, **2024**, <https://doi.org/10.1021/acs.est.3c08253>.
9. Thomas R. Underwood, Ian C. Bourg, and Kevin M. Rosso, "Mineral-associated organic matter is heterogeneous and structured by hydrophobic, charged, and polar interactions" *Proc. Natl. Acad. Sci. U.S.A.*, **2024**, <https://doi.org/10.1073/pnas.2413216121>.
10. Amélie C. Lemay and Ian C. Bourg, "Interactions between per- and polyfluoroalkyl substances (PFAS) at the water-air interface" *Environ. Sci. Technol.*, **2025**, <https://doi.org/10.1021/acs.est.4c08285>.
11. Xiaojin Zheng and Ian C. Bourg, "A multiscale approach to simulate non-isothermal multiphase flow in deformable porous materials" *Water Resour. Res.*, **2025**, <https://doi.org/10.1029/2025WR041300>.
12. Xiaojin Zheng and Ian C. Bourg, "Microstructure, transport, and mechanics of compacted clay simulated at the 0.1 μm scale (1400 smectite clay particles) using a coarse-grained model with explicit counterions" *J. Phys. Chem. C*, **2025**, *in review*.

Reductive Biomining of Iron Sulfur Minerals

Eric Boyd, Principal Investigator

Department of Microbiology and Cell Biology, Montana State University (MSU), Bozeman, MT,
Graduate Students and Postdocs: Hunter Fausett (MSU), James Larson (MSU), Rachel Spietz (MSU),

Collaborators: Brian Bothner (MSU), Joan Broderick (MSU), Eric Shepard (MSU)

Overall research goals:

Pyrite (FeS_2), the most abundant iron sulfide in Earth's crust, is a reservoir of trace elements of national strategic importance including nickel (Ni), cobalt (Co), and molybdenum (Mo). Traditional methods to extract such elements from pyritic ores employ microbial oxidative dissolution strategies that generate sulfuric acid and can lead to acid mine drainage that can have negative consequences for aquatic ecosystems. Recently, we discovered that a variety of anaerobic microorganisms, including methanogens, sulfate reducers, and fermenters, can reductively dissolve FeS_2 through a process that does not generate acid. During reduction of FeS_2 , cells assimilate dissolution products to meet their iron (Fe) and sulfur (S) demands, including for biosynthesis of simple and complex metallocusters. This includes metallocusters in enzymes that reduce carbon dioxide to methane, reduce dinitrogen to ammonia, and reduce protons to hydrogen, all of which are value-added products. Further, cells bioconcentrate Fe during growth with FeS_2 suggesting a potential application of reductive dissolution in biomining of metals from low grade FeS_2 ore. The goals of our research are to forge new understanding of the cellular mechanisms that enable the reductive dissolution of FeS_2 and the assimilation, trafficking, and bioconcentration of metals. A suite of physiological, biochemical, -omic, and spectroscopic approaches are being combined to address these research goals. Further, state of the art imaging and computational approaches are being used to assess the reaction mechanisms, kinetics, and chemical transformations that take place at the mineral-water-cell interface during reduction. Through this integrated approach, our goal is to identify the enzymes and pathways that allow anaerobes to bio-mine Ni, Co, and Ni from FeS_2 ore to enable the cost-effective recovery and/or conversion of raw substrate into catalysts for electrochemical energy generation.

Significant achievements during 2023- 2025:

Physiology. Sulfide is commonly used as the sulfur source for cultivation of anaerobes. Yet, sulfide markedly decreases the solubility/availability of trace metals for these anaerobes, a phenomenon that was shown to drastically alter the metalloproteome of cells. Further, data indicate that cells can synthesize all nitrogenase metallocofactors from FeS_2 and molybdate ion. Growth was enhanced in this condition relative to cells grown with soluble Fe(II) , sulfide, and molybdate which was attributed to the higher bioavailability of molybdate when grown with FeS_2 . This was due to thiolation of molybdate ion to form tetrathiomolybdate, which we also showed can be used as a Mo source, and ultimately molybdenum sulfide, the latter of which is far less bioavailable. **Computational Chemistry and Geochemistry.** Atomic scale models of FeS_2 nanoparticles have been built, validated, and used to model surface transformations and reactivity during reduction. The model for FeS_2 reduction suggests initial release of sulfide into solution is concomitant with the precipitation of pyrrhotite (Fe_{1-x}S) on the mineral surface. Dissolved Fe, but not S, from Fe_{1-x}S then combines with sulfide (HS^-) in solution to form aqueous iron sulfur clusters (FeS_{aq}) that are neutrally charged and likely passively diffuse into the cell. To examine the kinetics of FeS_{aq} cluster growth, dynamic light scattering was used to reveal the influence of temperature, $\text{Fe(II)}/\text{HS}^-$ concentration, pH, and temperature on FeS_{aq} growth kinetics; such information provides constraints on FeS_{aq} utilization by cells. FeS_2 was synthesized that contains Co-, Ni-, and Co/Ni impurities for additional growth studies. **Biochemistry and Spectroscopy.** Experiments (EPR, Mössbauer, UV-Vis, XAS) on purified protein homologs or on whole cells heterologously expressing these proteins highlight their role in metal bioaccumulation.

Comparison of spectroscopic data from laboratory-synthesized minerals to that coordinated by purified proteins is narrowing the candidates for identification of the biomineralization product. Expression of protein homologs in *E. coli* affects growth kinetics and metal accumulation in those cells, which can be optimized to balance stable biomass accumulation and metal accumulation.

Science objectives for 2026-2028:

Ongoing research is aimed at further defining the cellular mechanisms of selective metal recovery from sulfide minerals. Our objectives moving forward are to use whole cell and purified proteins to identify the mechanisms of mineral reduction and molecular determinants of metal binding, trafficking, and accumulation in cells. Target Ni, Co, and Fe binding proteins have been identified and cell genetic backgrounds and growth conditions are being optimized to achieve this objective. Further, we aim to move from selective recovery of metals from laboratory-synthesized minerals to naturally occurring minerals. This includes recovery of Co from cobaltite (CoAsS), while minimizing release of free arsenic (As) from the mineral structure.

Publications supported by this project 2023-2025 (Representative):

1. Manjinder Kour, Heather M. Callaway, and Eric S. Boyd. Environmental controls on the kinetics of iron-sulfur cluster nucleation and nanoparticle formation. *J. Nanopart. Res.*, **2025**, 27, 256, 10.1007/s11051-025-06445-5.
 2. Eric S. Boyd and Devon Payne. Expanded diversity of microbial groups capable of anaerobic pyrite reduction and assimilation of dissolution products. *Environ. Microbiol.*, **2025**, 27, e70125, 10.1111/1462-2920.70125.
 3. Devon Payne, Lisa Keller, James Larson, Brian Bothner, Daniel R. Colman, and Eric S. Boyd. Alternative sources of molybdenum for *Methanococcus maripaludis* and their implications for the evolution of molybdoenzymes. *Comms. Biol.*, **2024**, 7, 1337, 10.1038/s42003-024-07049-w.
 4. Hunter Fausset, Rachel L. Spietz, Savannah Cox, Gwendolyn Cooper, Scott Spurzem, Monika Tokmina-Lukaszewska, Jennifer DuBois, Joan Broderick, Eric Shepard, Eric Boyd, and Brian Bothner. 2024. A shift between mineral and nonmineral sources of iron and sulfur causes proteome-wide changes in *Methanosarcina barkeri*. *Microbiol. Spectrum*, **2024**, 12, e00418-23, 10.1128/spectrum.00418-23.
 5. James Larson, Monika Tokmina-Lukaszewska, Rachel L. Spietz, Devon Payne, Hunter Fausset, Md Gahinger Alam, Brooklyn K. Brekke, Jordan Pauley, Ethan J. Hasenöhr, Eric M. Shepard, Eric S. Boyd, and Brian Bothner. Impact of mineral and non-mineral sources of iron and sulfur on the metalloproteome of *Methanosarcina barkeri*. *Appl. Environ. Microbiol.*, **2024**, 90, e00516-24, 10.1128/aem.00516-24.
 6. Manjinder Kour, Attila Táborosi, Eric S. Boyd, and Robert K. Szilagyi. Development of molecular cluster models to probe pyrite surface reactivity. *J. Comp. Chem.*, **2023**, 44, 2486-2500, 10.1002/jcc.27213.
 7. Rachel L. Spietz, Devon Payne, and Eric S. Boyd. Methanogens acquire and bioaccumulate Ni during reductive dissolution of nickelian pyrite. *Appl. Environ. Microbiol.* **2023**, 89, e00991-23, 10.1128/aem.00991-23.
 8. Devon Payne, Rachel L. Spietz, Dennis L. Newell, Paul J. Dykstra, and Eric S. Boyd. Influence of sulfide on diazotrophic growth of a methanogen and its implications for the origin of nitrogenase. *Comm. Biol.*, **2023**, 6, 799, 10.1038/s42003-023-05163-9.
 9. Eric S. Boyd, Rachel L. Spietz, Manjinder Kour, and Daniel R. Colman. Crystal ball: A naturalist perspective of microbiology: Examples from methanogenic archaea. *Environ. Microbiol.*, **2023**, 25, 184-198, 10.1111/1462-2920.16285.
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Inhibition of Impurity Adsorption and Nucleation at Mineral-Water Interfaces via Chelator Adsorption

Jacquelyn Bracco, Principal Investigator

School of Earth and Environmental Sciences, Queens College, City University of New York, Flushing, NY

Graduate Students: Evan Ostroff, George Charalambous

Collaborators: Alice Paskin (Argonne National Laboratory – postdoctoral researcher), Sang Soo Lee (Argonne National Laboratory)

Overall research goals:

Organic chelators are commonly used in energy/water production and industrial processes to inhibit nucleation and growth of scaling minerals that cause detrimental consequences such as clogging and failure of oil and gas pipelines or reduction of reverse osmosis membrane capacities. The overarching goal of this project is to develop a fundamental understanding of the molecular-scale mechanisms by which organic chelating agents modify surface structure, ion adsorption, and nucleation at geologically and environmentally relevant conditions. Our research objectives are: 1) resolve the mechanisms by which chelator molecules interact with reactive sites on mineral surfaces; 2) determine how chelating agents affect the kinetics of impurity ion incorporation and adsorption at mineral surfaces; and 3) quantify the impact of chelating agents on nucleation and epitaxial growth of secondary phases. To address these goals, we use a combination of batch geochemical techniques to measure dissolution rates, microscopy imaging to track changes in surface morphology induced by the chelator molecules, and X-ray scattering characterizations to determine how the chelator affects surface structure and the sorption behavior of impurity ions at the molecular level.

Significant achievements during 2023- 2025:

We determined the effect of the number of acidic functional groups of organic chelators on the dissolution rates and surface structure of common scale-forming minerals. Experiments were conducted using two chelating agents, EDTA and DTPA on three minerals, barite (BaSO_4), celestite (SrSO_4), and calcite (CaCO_3). These minerals exhibit orders-of-magnitude differences in dissolution rates under similar aqueous conditions, enabling us to explore different timescales and reaction conditions.

As part of research objective one, we investigated how increasing chelator concentration and pH control mineral dissolution mechanisms and rates using atomic force microscopy (AFM), batch dissolution experiments, and crystal truncation rod (CTR) measurements. Dissolution primarily occurred by step-edge retreat at acidic and near-neutral pH (~ 2 and ~ 6), whereas etch pit nucleation and expansion dominated at basic pH (~ 11.5). Dissolution rates were largely constant over short time periods but decreased at longer reaction times, likely due to adsorption of the chelator molecules to the surfaces that physically hindered the interaction of reactive surface sites with aqueous species. For calcite, dissolution was faster with DTPA than with EDTA, presumably due to the larger number of functional groups in DTPA, which can bind dissolved ions more strongly. In contrast, this chemical dependence was less pronounced for barite. Instead, both chelating agents almost equally affected the chemical composition and roughness of the barite (001) surface by displacing surface barium and sulfate ions, as confirmed by time-resolved CTR measurements.

For research objective 2, we measured the surface structure of barite and celestite in the presence of lead (Pb) while varying EDTA concentration. When $[\text{Pb}] \leq [\text{EDTA}]$, we observed strong inhibition of Pb sorption at the barite (001) surface. However, when $[\text{Pb}] > [\text{EDTA}]$, we observed systematic increases in Pb uptake via three mechanisms: surface incorporation, inner-sphere surface complexation, and growth of a lead-rich $(\text{Pb,Ba})\text{SO}_4$ phase. Pb sorbed at the surface could not be

fully removed even after exposure to deionized water, indicating a high energy barrier against desorption of structurally incorporated Pb and/or the presence of chemically bonded EDTA films that physically hindered desorption of Pb underneath the films.

To address research objective 3, we used a combination of CTR and AFM to measure the formation and removal of gypsum, anglesite, and (Pb,Ba)SO₄ overgrowths on barite in the presence of EDTA. Dissolution of the overgrowths in the presence of EDTA primarily occurred through etch pit formation and step retreat. During nucleation of gypsum and anglesite on barite and celestite, we observed changes in CTR shape consistent with formation of a secondary phase whose out-of-plane lattice spacing is smaller than that of the barite or celestite substrate. At higher EDTA concentrations, the overgrowths were preferentially removed, indicating that these secondary phases were thermodynamically less stable compared to the substrates.

Overall, our results suggest that chelating agents can enhance dissolution of scale-forming minerals by promoting removal of metal cations from the surface, but that adsorption of the chelator to the surface can reduce dissolution rates over longer time periods. This adsorption can also inhibit removal of secondary phases that form on the mineral surfaces.

In addition to our research objectives, this grant supported training of three undergraduate students and two graduate students in geochemical techniques, including operation of an optical microscope purchased as part of this grant. Two of the undergraduate students (Bill Lu and Julia Sandke) and a graduate student (George Charalambous) travelled to Argonne National Laboratory for training in atomic force microscopy. An undergraduate student (Bill Lu) and a master's student (George Charalambous) also received training in X-ray scattering data collection and analysis at the Advanced Photon Source.

Science objectives for 2026-2028:

Until this grant expires, our primary experiments will focus on determining how chelators affect the mechanisms and rates of (1) dissolution of scaling mineral calcite and (2) desorption of impurity metal ions (i.e., Pb, Sr) incorporated in the calcite structure. Calcite dissolution rates will be measured using AFM in comparison with our previously collected batch dissolution measurements and digital holographic microscopy observations. The experiments will be conducted as a function of temperature to determine the impact of organic agents on intrinsic reaction kinetics, such as activation energy for dissolution. Metal desorption rates will be measured using X-ray scattering (CTR and resonant anomalous X-ray reflectivity); the required beamtime is scheduled for April 2026. In parallel, we will finalize our data analysis for previously collected X-ray scattering data, prepare the results for publication, and support Mr. Charalambous in completing and defending his thesis.

Publications supported by this project 2023-2025:

1. Amanda Dorfman, Anna K Wanhala, Sang Soo Lee, Peter J Eng, Joanne E Stubbs, Lexi Kenis, Jacquelyn N Bracco, "Lead Adsorption and Desorption at the Barite (001) Surface in the Presence of EDTA" *ACS ES&T Water*, **2025**, 1, 264-273, <https://doi.org/10.1021/acsestwater.4c00836>.

Determining scale-dependent shear strength in the crust

Emily E. Brosky, Principal Investigator

Dept of Earth and Planetary Sciences, University of California, Santa Cruz, CA

Graduate Students: Minghan Yang, Huiyun Guo

Postdoctoral Scholars: Will Steinhardt, Valere Lambert, Gaspard Farge

Overall research goals:

Under the natural loading of the overlying crust, rocks fail in shear. Characterizing and predicting the conditions of failure is key to the sustainability of any subterranean human activity including carbon sequestration, geothermal energy, groundwater management, and oil and gas reservoir management. Much of what we know about the shear failure of rocks has its origins in earthquake physics and geotechnical engineering, where laboratory experiments are commonly performed on centimeter-scale samples. These results are then extrapolated to field settings where failure can occur on kilometer-scale structures. Such extrapolation is potentially problematic given the known scale-dependence of tensile strength, and the presence of geological structures at the relevant scales. Thus, scaling shear stress is a grand challenge in the geosciences. Here we:

1. Capitalize on exhumed fault surfaces as multi-scale archives of the shear strength of rocks. We provide a firm theoretical foundation for interpreting fault roughness as a measure of scale-dependent rock strength.
2. Use a new laboratory apparatus to perform laboratory experiments on analog materials to capture the dynamics of fully confined ruptures that naturally organize the stress of the fault across scales. The resulting distribution controls the stress drop of earthquakes, precursory locking events and the tendency for events to link together to make larger events. These seismological observables are commonly interpreted as measurements of fault stress or strength at scale, but in fact reflect the self-organized distribution of stress due to the repeated earthquake cycles.

Significant achievements during 2023- 2025:

Fault surface topography is nearly unique as an observable that can be measured from micron to kilometer scale. Prior work by our group had suggested that the scale-dependent roughness reflected scale-dependent strength based on an order of magnitude analysis. Now, we have developed a rigorous theory for the elastoplastic deformation of the surface that allows interpretation of the preserved roughness in terms of the scale-dependent strength (Lambert and Brodsky, 2025). We show that the relationship between surface roughness and scale-dependent strength is governed by two limits. One is a plastic limit where the scale-dependent yield proportional to the average aspect ratio of fault bumps. If faults are rougher than what the yield stress prescribes, non-recoverable deformation dominates and smooths them. A distinct elastic limit defines the condition where all deformation can be accommodated elastically and all roughness will survive. The elastic limit occurs when strength scales as the square of aspect ratio. Numerical solutions show that the inelastic deformation of surfaces between the elastic and plastic limits is small and thus real surfaces likely have roughness that sits between the elastic and plastic limits. As a result of this analysis, scale-dependent strength of rocks can be determined in situ directly from the rock record.

We have also studied scale-dependent stress field using an analog laboratory model. Steinhardt et al. (2023) presented a scale model built of transparent rubber (PDMS) that allowed sequences of slip events to be simulated. Importantly, because the material is soft, most slip events are entirely confined within the laboratory fault. Slip events redistribute strain

energy to the edges of the slip patches and, after a few earthquake cycles, a heterogeneous stress distribution emerges that influences the qualitative and quantitative behavior of the earthquake parameters commonly observed by seismologists. For instance, the stress drop of the individual slip events in the experiment is independent of the normal stress on the fault (Steinhardt et al., 2023). Locking events emerge in addition to slip events and the locking events are predictive of future locations and time of slip. (Steinhardt and Brodsky, 2025). These locking events would not occur without a particular distribution of stress. As before, recognizing that stress distributions can self-organize is the key to understanding why the fault slips as it does.

The experiment also provides insight into how an earthquake grows across scales (Steinhardt and Brodsky, under review). Sometimes there is enough strain energy around a single, strongly stressed patch to become a large earthquake. Other times, a moderately stressed patch triggers another moderately stressed area and creates a large earthquake by linking the patches together. These two paths to growth have different implications for size predictability. The growth of a single rupture implies that only information about that region is necessary in order to anticipate the size of the ensuing earthquake. Linked growth implies that information is required about the energy of the whole population of patches before size can be anticipated. We find that any process that results in high stress concentrations results in a greater percentage of linked events.

These results are synthesized with related observations in a major conceptual article in press at PNAS titled "How Earthquakes Organize Stress in the Crust." This article described how earthquakes organize the stress in the crust by redistributing it through slip events. As a result, fault systems evolve to preferred, reproducible states as evidenced by natural experiments that measure statistical distributions of stress from failure, strain energy and scale-dependent rock strength. Knowing the mathematical form, and even establishing existence of the distributions has proven to be a powerful tool for tracking changes, characterizing systems and explaining rupture behavior. If the primary role of earthquakes in the system is to redistribute stress, then self-organization is a central feature of the Earth's crust.

Science objectives for 2026-2028:

- 1) Directly measure the scale-dependent stress distribution in the laboratory fault
- 2) Utilize machine-learning to forecast seismicity on the laboratory fault and compare performance to standard parameterized statistical seismicity forecast methods.

Publications supported by this project 2023-2025:

1. Lambert, Valère, and Emily E. Brodsky. "Competition between roughness and strength for scale-dependent surfaces." *Physical Review E*, **2025**, 10.1103/PhysRevE.111.065502.
2. Steinhardt, Will, and Emily E. Brodsky. "Precursory locking precedes slip events on laboratory fault." *Geophysical Research Letters*, **2025**, 10.1029/2024GL112882.
3. Steinhardt, Will, Sam Dillavou, Mary Agajanian, Shmuel M. Rubinstein, and Emily E. Brodsky. "Seismological stress drops for confined ruptures are invariant to normal stress." *Geophysical Research Letters* **2023**, 10.1029/2022GL101366
4. Yang, Minghan, Valere Lambert, and Emily E. Brodsky. "The Role of Normal Stress and Shear Stress Heterogeneity in the Inferred Depth-Independence of Stress Drop." *Geophys. Res. Lett.*, **2026**.
5. Brodsky, E. E. and G. Farge, "How Earthquakes Organize Stress", *PNAS*, **in press**.
6. Steinhardt, W. and E. E. Brodsky, "Distinguishing Single and Linked Ruptures in the Laboratory and Nature", *Geophys. Res. Lett.*, **in review**.

Explaining the Nonlinear Mechanics of Geophysical Granular Solids in terms of Anisotropic Force Chain Transmission and Contact Area Adaptivity

Giuseppe Buscarnera, Principal Investigator

Northwestern University, Department of Civil and Environmental Engineering, Evanston, IL

Graduate Students: Divyanshu Lal, Shubjot Singh.

Postdoctoral Scholars: Dawei Xue, Yize Pan, Pengjia Song.

Collaborators: Mark Rivers (University of Chicago; GESCARS), Gioacchino Viggiani (Université Grenoble Alpes).

Overall research goals:

Granular geomaterials are involved in virtually every physical process shaping the Earth's surface and its upper lithosphere. Their deformation emerges from collective interactions among grains, as well as from the propagation of these interactions across scales. This project aims to explain one of the unique features of granular matter, i.e., its ability to adapt to the physical conditions prevailing in the subsurface. In this project we aimed to explain the adaptivity of granular geomaterials with an integrated description of how the contact dynamics contributes to their mechanical anisotropy and, in turn, to the propagation of elastic waves and the onset of strain discontinuities in granular rocks.

More specifically, during this project we formulated a continuum framework bridging the adaptivity of interparticle contacts with the elastodynamic signature of geomaterials. This goal was pursued by using micromechanics to inspire new definitions of granular fabric that (i) reflected the adaptivity of stressed contacts during both reversible and irreversible deformation; (ii) could be verified with multi-scale experiments resolving the microstructural arrangement and the bulk stress-strain response of grain packings (e.g., mechanical tests assisted by X-ray microtomography); (iii) led to averaged descriptors that could be readily used as state variables in a continuum thermodynamics models.

Significant achievements during 2023- 2025:

Some key achievements of our prior project are outlined as follows:

1) New computational methods to regularize the analysis of deformation discontinuities were proposed, from viscoplastic models to nonlocal plasticity. It was shown that shear bands can form only if the material fulfils specific criteria, reflected by its constitutive properties. In addition, we defined a closed-form relation between the thickness of a shear band and a combination of local and nonlocal rheological parameters. This finding warrants further research to uncover the links between faulting and geomaterial composition (e.g., gradation, shape, mineralogy, fabric). Specifically, our work indicates that a way to discover such links is to reinterpret geomaterial failure through the lenses of micromechanics-based rheological models.

2) Particle-scale experiments were integrated into a new model of grain crushability for particles of arbitrary morphology. This effort was guided by concepts of nonlinear contact mechanics, which we linked to both deterministic and statistical fracture criteria. The resulting expressions embedded the role of the morphology of grain contacts (e.g., their curvature) on the strain energy storage and its dissipation via tensile fracture. These results were validated with strength measurements collected for sand grains of different shapes, whose morphology was constrained with X-ray imaging. By shedding light on one of the controls of yielding in granular packings (i.e., breakage), these results are a springboard to explain how particle characteristics influence nonlinear mechanical interactions.

3) Discrete and continuum models were formulated to study the elastic anisotropy of granular solids, such as sand. Specifically, we proposed a new interpretation of the granular fabric that isolates the mechanical sources of anisotropy (i.e., the ability of stress to change stiffness directionality) from its inherent microstructural causes (e.g., grain orientation due to their deposition history). Closed-form

expressions of this multi-modal fabric were also obtained, which led to the definition of an augmented tensor capturing the nonlinear elasticity of granular solids, which we called contact area fabric. This tensor is sensitive to stress (i.e., it emerges from contact mechanics), while also retaining the known effects of contact density and orientation. Not only does this allow us to rationalize the origin of mechanical anisotropy in the subsurface, but it is also a steppingstone to infer non-destructively the granular fabric (and its evolution) from elastodynamic measurements.

4) Through the Discrete Element Method, we examined the link between elasticity and fabric prior to frictional failure. These numerical experiments allowed us to freeze the microstructure at selected states to inspect the patterns of force chain alteration and consequent grain slip. Our results pinpoint a strong link between contact slip and the decay of the shear modulus prior to failure. In addition, they show that particle slips occur along preferential directions and alter irreversibly the mechanical anisotropy. Most notably, since these processes take place before shear failure, our findings are instrumental to: (i) infer continuum variables that couple the frictional response with the nonlinear elasticity of a granular packing; (ii) define fabric-based precursors of impending slip that to be used within augmented rate-and-state friction laws.

Science objectives for 2026-2028:

For the next project period we aim to use insights from our research on the mechanical adaptivity of granular packings to inform the frictional rheology of particle systems during dynamic slip. For this purpose, we envision an integration of the concepts of fabric-dependent elasticity and plasticity for granular solids with the established formalism of rate-and-state friction laws. In particular, we plan to build models based on the hypothesis that the quasi-static and the dynamic (i.e., rate-dependent) frictional regimes cannot be compartmentalized, in that they are entangled with one another. Hence, we will postulate a coupling between behavioral regimes enacted by the joint dependence of all the constitutive functions on the same state variables (e.g., the contact fabric).

To pursue these objectives, we will continue to use a combination of experimental, theoretical, and numerical strategies. From an experimental standpoint, we envision using X-ray microtomography in conjunction with non-destructive measurements to track the coupled energetics of local particle slip and collective frictional failure. From a theoretical standpoint, we propose a synergistic use of discrete and continuum mechanics to define state variables deployable into both quasi-static and dynamic friction laws. Finally, from a computational standpoint we will study simulated faults with embedded frictional interfaces, where friction is obtained as an emergent property stemming from grain-scale models that are explicitly resolved to account for features such as grain size and shape, contact morphology, and the directionality of such properties.

Selected publications supported by this project 2023-2025:

1. Xue, D., Lü, X., Lim, K. W., Buscarnera, G., "Nonlocal implicit gradient enhancements for strain localization informed by controllability criteria for plastic solids", *Computer Methods in Applied Mechanics and Engineering*, **2023**, <https://doi.org/10.1016/j.cma.2023.116275>.
2. Seo, D., Tengattini, A., Viggiani, G., Buscarnera, G., "Experimental and analytical assessment of fracture criteria for non-spherical sand grains", *Granular Matter*, **2024**, <https://doi.org/10.1007/s10035-023-01372-5>.
3. Xue, D., Shahin, G., Lü, X., Buscarnera, G., "Simulation and interpretation of compaction patterns around boreholes excavated in high-porosity rocks", *Acta Geotechnica*, **2024**, <https://doi.org/10.1007/s11440-023-01978-2>.
4. Lal, D., Xue, D., Buscarnera, G., "Exploring the impact of hydro-mechanical coupling on compaction localization", *Computers and Geotechnics*, **2025**, <https://doi.org/10.1016/j.compgeo.2025.107393>.
5. Singh, S., Buscarnera, G., "Examining the adaptive elastic anisotropy of granular materials", *Géotechnique*, **2025**, <https://doi.org/10.1680/jgeot.23.00190>.

Geochemical Mechanisms Controlling Rare Earth Element and Platinum Group Element Migration and Enrichment during Crustal Aqueous Alteration and Weathering

Jeffrey G. Catalano, Principal Investigator

Eugene S. Ilton, Co-Principal Investigator

Department of Earth, Environmental, and Planetary Sciences, Washington University in St. Louis, Saint Louis, MO 63130

Pacific Northwest National Laboratory, Richland, WA 99352

Graduate Students: Xicheng He, Greg J. Ledingham, Bavan Rajan, Elmira Ramazanov, Neha Sharma, Emily G. Wright

Collaborators: Daniel E. Giammar, Elaine D. Flynn, Yihang Fang, Yanting Qian, Olwen Stagg (Washington University in St. Louis), Eric J. Bylaska, Sebastian T. Mergelsberg, Duo Song (PNNL), Sang Soo Lee (ANL), Joanne E. Stubbs, Peter J. Eng (University of Chicago)

Overall research goals:

The overall research goal of this project is to elucidate the roles of mineral surfaces and structures in controlling the migration and enrichment of rare earth elements (REEs) and platinum group elements (PGEs) in weathering environments. REEs and PGEs in regolith, laterites, and other residual deposits formed by weathering represent new potential resources for future exploitation but the fundamental geochemical processes that dictate their migration and enrichment are poorly constrained. We seek to identify the dominant molecular-scale mechanisms of REE and PGE adsorption at mineral-water interfaces and to establish the substitution mechanisms of REEs and PGEs in iron oxides and phyllosilicate clays and how these affect retention in mineral structures.

Significant achievements during 2023- 2025:

1. Interfacial Geochemistry of REEs: Goethite, the dominant iron oxide in regolith deposits, produces a weak fractionation towards heavy REEs via adsorption. Citrate ligands invert this adsorption-induced fractionation to favor the binding of light REEs to goethite. Kaolinite adsorbs REEs through multiple mechanisms, with cation exchange dominant below pH 6 and surface complexation above pH 6. Competition between surface binding and aqueous complexation has offsetting effects that yields negligible REE fractionation, including upon addition of citrate. Comparison of Nd(III) and Yb(III) binding to two individual surfaces of alumina using element-specific surface X-ray scattering methods reveal two to three separate species form for each REE on each surface. In addition to established inner- and outer-sphere binding mechanisms, we identified a new type of disordered REE surface species associated with Al vacancy surface defects. Each binding mechanism showed distinct affinities for Nd and Yb on each surface. This indicates that surface- and mechanism-specific REE fractionation occurs, with the net fractionation on mineral particles comprising the mass balance sum of fractionations produced at individual surfaces and binding sites.

2. Structural Geochemistry of REEs: The substitution of Nd(III), Dy(III), Yb(III) and Lu(III) into goethite generate Fe vacancy defects. While Yb and Lu fully substitute into the goethite structure, substantial fractions of Nd and Dy occur as separate amorphous phases. AIMD-informed EXAFS analysis and AC-HAADF-STEM demonstrate that Yb and Lu enter structural Fe sites in goethite associated with protonated Fe vacancies. These substitution mechanisms create substantial fractionation in favor of heavy REEs. Aging of REE-ferrihydrite coprecipitates into goethite caused complete incorporation of Yb but only partially entrapped Lu, fractionating these heavy REEs. Ligand-promoted dissolution of goethite induces the stoichiometric release of structural Yb, demonstrating that ligand-promoted REE mobilization must be coupled to substantial iron leaching. Clay synthesis experiments reveal that negligible structural substitution of REEs occurs for kaolinite, but REEs readily enter the octahedral sheet of montmorillonite, with incorporation strongly favoring the smaller heavy REEs.

3. Interfacial Geochemistry of PGEs: Complexation by chloride consistently suppresses the adsorption of Pd(II) and Pt(II) to goethite and hematite, the dominant iron oxides in laterites and

regolith formed from the weathering of PGE-enriched ultramafic rocks. Net Pd and Pt binding reflect offsetting inhibition by aqueous complexation and enhancement by ternary surface complexation with chloride. Systematic comparison reveals that the canonical explanation of fractionation of Pd from Pt during weathering, distinct strengths of aqueous complexation of chloride, is invalid. Instead, mineral-specific interfacial reactions cause this fractionation. Hematite-dominated regolith in semi-arid systems preferentially retains Pt but goethite-dominated regolith in humid regions will maintain sub-equal proportions of Pd and Pt. This shows that the PGE resource potential of ultramafic-derived regolith will vary with mineralogy that is ultimately controlled by local (paleo)climate.

4. Structural Geochemistry of PGEs: Iron oxides enriched in Pt and Pd occur in weathering zones and imply that these PGEs are structurally incorporated, but our work reveals substantial barriers to such entrapment. All pathways explored for Pd(II) result in the formation of discrete PdO, and most routes for Pt(II) similarly produce a disordered PtO-like precipitate. However, introduction of Pt(IV) into a solution containing Fe(II) generated Pt(II)-bearing goethite. AIMD-informed EXAFS shows that Pt(II) retains a square planar geometry when substituting into a structural Fe site. This work demonstrates that redox cycling is a viable pathway to Pt entrapment in goethite.

Science objectives for 2026-2028:

As this project transitions from a joint university-laboratory collaborative activity to a university-led endeavor, the core focus will shift to study cooperative and competitive interfacial controls on the accumulation, mobilization, and fractionation of critical minerals. Three linked research directions are being pursued that investigate the interfacial reactions of REEs and PGEs with clay and oxide minerals which dominate the reactivity of weathering zones. First, the impacts of inorganic and organic ligands on critical mineral adsorption behavior and mechanisms will be studied, with a focus on fluoride, phosphate, and citrate for REEs and ammonia, chloride, and oxalate for PGEs. Ligand competition will be explored for both critical mineral groups. Second, fractionations of critical minerals via competitive interfacial reactions will be investigated. For REEs, the manifestation of binding affinity versus surface site competition modulated by ligand complexation on fractionations will be evaluated. For PGEs, fractionation of Pd(II) from Pt(II) via competitive adsorption by trace metals abundant in ultramafic source rocks will be studied. Third, the effects of aging on the reversibility of critical mineral adsorption and associated fractionations will be examined. Studies with REEs will focus on potential irreversible binding following adsorption while investigations of PGEs will explore how slow ligand exchange rates create distinct adsorption behavior for Pd(II) and Pt(II).

Select publications supported by this project 2023-2025:

1. Sebastian T. Mergelsberg, Alex J. Kugler, Elaine D. Flynn, Duo Song, Eric J. Bylaska, Jeffrey G. Catalano, Eugene S. Ilton, "Structural incorporation into goethite fractionates rare earth elements" *RSC Adv.*, **2025**, 15, 28815-28826, DOI: [10.1039/D5RA04022A](https://doi.org/10.1039/D5RA04022A).
2. Elmira Ramazanov, Neha Sharma, Elaine D. Flynn, Olwen Stagg, Jeffrey G. Catalano, Daniel E. Giammar, "Adsorption of REEs to kaolinite via ion exchange and surface complexation as a function of water chemistry" *ACS Earth Space Chem.*, **2025**, 9, 1430-1442, DOI: [10.1021/acsearthspacechem.4c00389](https://doi.org/10.1021/acsearthspacechem.4c00389).
3. Yihang Fang, Sang Soo Lee, Greg J. Ledingham, Joanne E. Stubbs, Peter J. Eng, Jeffrey G. Catalano, "Complex adsorption behavior of neodymium and ytterbium on structurally-distinct alumina surfaces" *Environ. Sci. Technol.*, **2025**, 59, 3972-3981, DOI: [10.1016/j.gca.2024.12.029](https://doi.org/10.1016/j.gca.2024.12.029).
4. Emily G. Wright, Xicheng He, Elaine D. Flynn, Daniel E. Giammar, Jeffrey G. Catalano, "Competitive and cooperative effects of chloride on palladium(II) adsorption to iron (oxyhydr)oxides: Implications for mobility during weathering" *Geochim. Cosmochim. Acta*, **2025**, 391, 203-217, DOI: [10.1016/j.gca.2024.12.029](https://doi.org/10.1016/j.gca.2024.12.029).
5. Xicheng He, Neha Sharma, Olwen Stagg, Elaine D. Flynn, Jeffrey G. Catalano, Daniel E. Giammar, "Adsorption of neodymium, dysprosium, and ytterbium to goethite under varying aqueous chemistry conditions" *ACS Earth Space Chem.*, **2024**, 8, 1224-1235, DOI: [10.1021/acsearthspacechem.4c00032](https://doi.org/10.1021/acsearthspacechem.4c00032).

A cDFT-Based Framework for Interfacial-Electrokinetic Control of Nanoscale Flow in Geologic Systems

Abdullah Cihan, Task Lead; Ben Gilbert, Principal Investigator

Energy Geosciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA

Collaborators: Ben Gilbert (LBNL), Mengsu Hu (LBNL), Carl Steefel (LBNL), Zhao Hao (LBNL), Piotr Zarzycki (LBNL)

Overall research goals:

The goal of this research is to develop a predictive theoretical framework for multiphysics transport that resolves molecular-scale interfacial forces and couples them to continuum-scale transport, enabling quantitative prediction of nanoscale flow in geologic systems. By integrating classical density functional theory (cDFT) with hydrodynamics, electrokinetics, capillarity, and interfacial reactions, the work provides a consistent theoretical and computational framework linking molecular interfacial physics to macroscopic transport processes under geologically relevant conditions.

Significant achievements during 2023- 2025:

The significant achievements are summarized as follows:

- Established a cDFT-based description of mineral-water interfaces that account for intermolecular interactions, surface tension, and interfacial forces including electrostatic interactions and short-range hydrogen bonding.
- Predicted and explained experimentally observed anomalous water film thicknesses on hydroxylated quartz surfaces. For the first time, the model provided a mechanistic explanation for long-standing experimental observations by showing that attractive hydrogen-bonding and van der Waals interactions initiate water layering, while the electrostatic forces arising from the charged hydroxylated surface drive film thickening (Cihan et al., 2024).
- Developed the first fully coupled nanocapillary filling model that integrates cDFT, hydrodynamics, electrokinetics, ion transport, and capillarity (Cihan et al., 2025).
- Resolved long-standing questions in nanofluidic experiments by demonstrating that classical $\sqrt{t}$ capillary filling behavior can emerge from molecularly informed interfacial forces, while quantitatively distinguishing the roles of electrokinetic resistance and interfacial forces.
- Identified a previously unexplored transport regime of nanocapillary flow at elevated temperature and pressure in which classical capillary scaling breaks down and interface motion halts due to an evaporation-driven pinning mechanism (Figure 1).

Science objectives for 2026-2028:

- Extend the cDFT-based framework to reactive and evolving interfaces, including multiphase flow (H_2 - H_2O), reactive gas generation during serpentinization, mineral dissolution and precipitation, and feedback between surface chemistry, interfacial forces, and transport.
- Integrate cDFT-based models with optical imaging, nanotomography, spectroscopic observations to determine the conditions under which serpentinization rates are limited by water transport through nanopore and microfracture networks.
- Establish AI/ML accelerated modeling strategies informed by cDFT results to enable large scale predictions while retaining molecular-level fidelity.

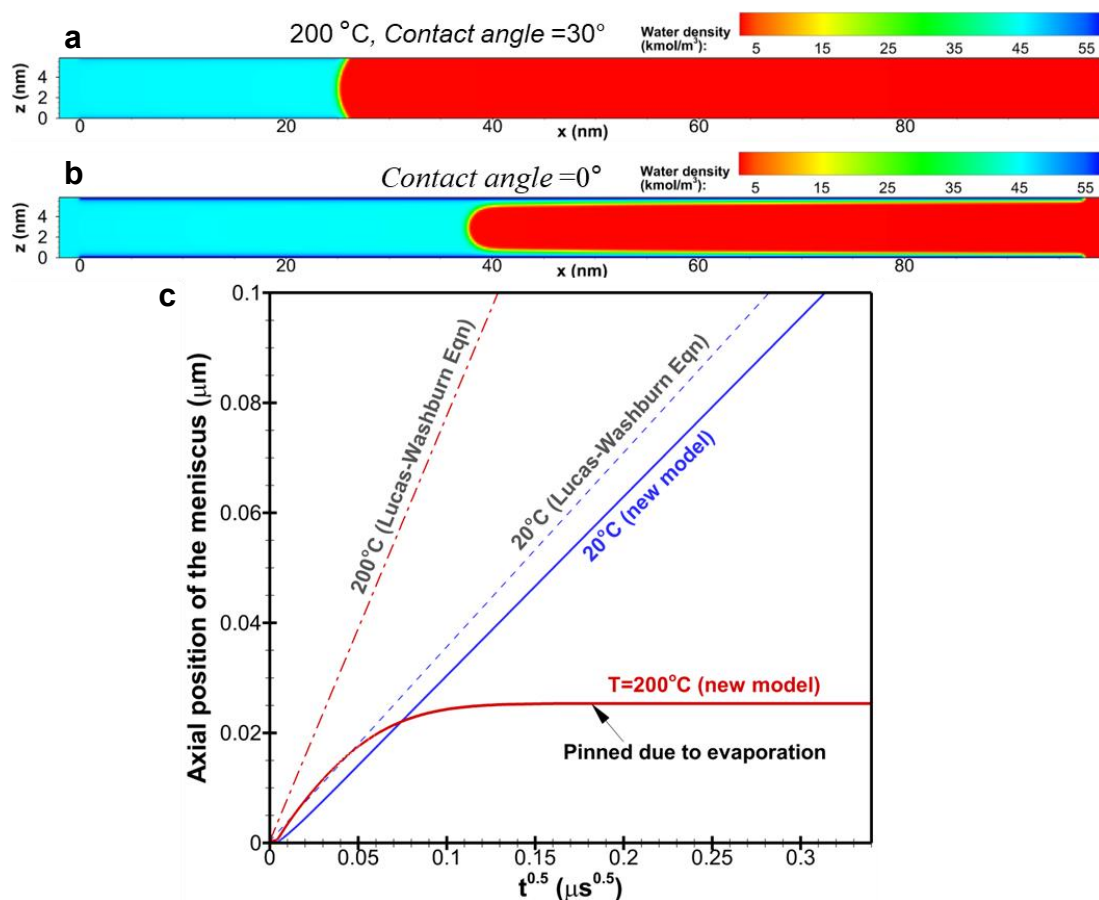


Figure 1. Predicted effect of elevated temperature on nanoscale capillary filling. (a) Pinned liquid–vapor interfaces for two wettability conditions at relative humidity = 0.3: Contact angle = 30° and (b) Contact angle = 0° . (c) Comparison of predicted axial meniscus position versus $\sqrt{t}$ at 20°C and 200°C (Contact angle = 30°), showing breakdown of the classical Lucas–Washburn capillary scaling at elevated temperature.

Selected Publications supported by this project 2023-2025:

1. Abdullah Cihan, Piotr Zarzycki, Zhao Hao, “Surface Hydroxylation-Induced Electrostatic Forces Thicken Water Films on Quartz” *Langmuir*, 40(29), 15099-15106, DOI: <https://doi.org/10.1021/acs.langmuir.4c01461>
2. Abdullah Cihan, Ben Gilbert, Pramod Bhuvankar, Mengsu Hu, Carl Steefel, “Interfacial–electrokinetic control and evaporation–driven pinning of nanocapillary filling from ambient to serpentinization temperatures” *Communications Physics*, Under review.

Learning the stochastic dynamics of complex geophysical systems

Maarten V. de Hoop, Principal Investigator

Simons Chair in Computational and Applied Mathematics and Earth Science, Rice University

Graduate students and PostDocs: A. Lara Benitez (Rice University), A. Siahkoobi (now at University of Central Florida), T. M. Roddenberry (Rice University), T. Teolis (Rice University)

Collaborators: E. Chaves (OVSICORI, Universidad Nacional, Costa Rica), I. Dokmanić, (University of Basel, Switzerland), T. Furuya (Doshisha University, Japan), J. Garnier (École Polytechnique), A. Kratsios (McMaster University), M. Lassas (University of Helsinki, Finland), M. Mahoney (University of California, Berkeley), G. Peyré (École Normale Supérieure, France)

Overall research goals, 2025:

Scientific ML promises deeper insight into complex phenomena, yet extracting interpretable scientific understanding from complex systems, specifically in Earth's subsurface, remains challenging. The physics of granular materials, geomechanics in the presence of fracture networks, friction, thermodynamics, multiphase fluid content and flow, and tidal forces are all expected to play a role in Earth's subsurface at discrepant spatial and temporal scales. Even the basic constitutive laws governing these processes have not been fully characterized.

The multi-physics and stochastic nature of subsurface phenomena renders monolithic models ill-suited for forecasting and modeling tasks. New data and recently developed learning algorithms present an opportunity to build solid foundations for data-driven scientific discovery focused on subsurface dynamics. This project develops new ML tools and theory that fuse and extend recent developments in foundation models, deep sequence models, jump diffusion models, and deep learning for inverse problems to learn the dynamics of complex systems in the Earth's subsurface. Also necessary for this endeavor is foundational research on the interplay between training dynamics, "grokking" phenomena, and uncertainty quantification.

Significant achievements during 2023- 2025:

Under nonlinear operator learning, we developed universal approximators of continuous maps between arbitrary Polish metric spaces, together with quantitative guarantees for Hölder-like maps arising from operators in nonlinear inverse problems. We then resolved a key open problem concerning learning inverse operators by providing a framework for extending the range of the forward operators. We obtained the first generalization and out-of-distribution estimates for neural operators upon introducing stochastic depth, and developed a mixture of experts softening the curse of dimensionality. We also established optimal rates and designed an architecture for learning geometry-aware operators with applications in wave imaging.

While score-based diffusion models have demonstrated success in finite-dimensional settings, we extend the models to infinite-dimensional function spaces in preconditioned Langevin dynamics with score-based generative models for infinite-dimensional linear Bayesian inverse problems, and dimension-free multimodal sampling via preconditioned annealed Langevin dynamics. Furthermore, we proposed autophagy penalized likelihood estimation, an unbiased alternative to maximum likelihood estimation inspired by model autophagy disorder.

We developed multi-scale clustering and source separation techniques for seismic data using novel factorial Gaussian Mixture Models. In the multi-sensor setting, we developed a procedure for "association without association" named HARPA. SeisLM, our basic large language or foundation model for continuous seismic data, showed, via self-supervised learning, remarkable performance across a range of tasks, including identification, forecasting, and prediction. Without task-specific supervision, SeisLM uncovered physically meaningful structure from raw data, including the autonomous clustering of tremors important to learning fault dynamics.

While foundation models have recently emerged as a powerful paradigm in scientific machine learning, their theoretical underpinnings and physical scope remain largely unresolved. We addressed this gap by developing a principled framework for foundation models in infinite dimensions, revealing natural connections to physics. Central to this framework are measure-theoretic transformers, which we showed are intimately linked to interacting particle systems and kinetic theory, in particular Vlasov flows. At the kinetic level, transformers can be adapted to collisional dynamics. Via graph measures, these transformers can be used for operator learning, such as in wave scattering and inverse problems, while flows motivated an alternative architecture based on self warping, named FLOWERS. Making the connection to physics stronger, we then introduced NeurDE, a novel neural architecture rooted in Lattice Boltzmann Models. Discretization is addressed with entropically stabilized "grids."

Science objectives for 2026-2028:

To further develop our foundation models for learning (stochastic) dynamics, autoregressive rollout, and forecasting; to develop a deep learning, generative model of jump diffusion type on graphs, which is motivated by the fact that the linked stress release model as well as its extensions lead to a backward Kolmogorov equation; to study the state space models (SSMs) on graphs associated with these, and new hybrid architectures; to develop a provable method of learning these; to develop power diagram dynamics as a physics-based model for training dynamics; to develop sampling-based neural operators for discovering functional clusters; to develop dimension-free multimodal sampling via preconditioned annealed Langevin dynamics for nonlinear inverse problems; to integrate SSMs and diffusion models for sequential UQ.

Selected publications supported by this project 2023-2025:

1. Thomas Muser, Alexander Spitzer, Matti Lassas, Maarten V. de Hoop, Ivan Dokmanic, "Flowers: A Warp Drive for Neural PDE Solvers," submitted to the International Conference on Machine Learning (ICML), **2026**.
2. Lorenzo Baldassari, Josselin Garnier, Knut Solna, Maarten V. de Hoop, "Preconditioned Langevin Dynamics with Score-Based Generative Models for Infinite-Dimensional Linear Bayesian Inverse Problems," *Advances in Neural Information Processing Systems*, **2025**, to appear, <https://doi.org/10.48550/arXiv.2505.18276>.
3. Maarten V. de Hoop, Nikola B. Kovachki, Matti Lassas, Nicholas H. Nelsen, "Extension and neural operator approximation of the electrical impedance tomography inverse map," arXiv preprint, **2025**, <https://doi.org/10.48550/arXiv.2511.20361>.
4. Takashi Furuya, Maarten V. de Hoop, Gabriel Peyré, "Transformers are universal in-context learners," *International Conference on Learning Representations (ICML)*, **2025**, pp. 80820–80845, <https://doi.org/10.48550/arXiv.2408.01367>.
5. Jose Antonio Lara Benitez, Kareem Hegazy, Junyi Guo, Ivan Dokmanić, Michael W. Mahoney, Maarten V. de Hoop, "Neural equilibria for long-term prediction of nonlinear conservation laws," arXiv preprint, **2025**, <https://doi.org/10.48550/arXiv.2501.06933>.
6. Jose Antonio Lara Benitez, Takashi Furuya, Florian Faucher, Anastasis Kratsios, Xavier Tricoche, Maarten V. de Hoop, "Out-of-distributional risk bounds for neural operators with applications to the Helmholtz equation," *Journal of Computational Physics*, **2024**, Vol. 513, Art. 113168, <https://doi.org/10.1016/j.jcp.2024.113168>.
7. Tianlin Liu, Jannes Münchmeyer, Laura Laurenti, Chris Marone, Maarten V. de Hoop, Ivan Dokmanić, "SeisLM: A foundation model for seismic waveforms," arXiv preprint, **2024**, <https://doi.org/10.48550/arXiv.2410.15765>.

Isotopic studies at the interface of geochemistry and rock deformation

Donald DePaolo and Harrison Lisabeth, Principal Investigators

Energy Geoscience Division, Lawrence Berkeley National Laboratory, Berkeley, CA

Graduate Students: Ziman Wu (UC Berkeley)

Collaborators: John N. Christensen (LBNL), Shuo Zhang (Tsinghua University), Chuan Liu (UC Berkeley postdoc), Andrew Turner (UC Berkeley/LBNL postdoc), Daniel Stolper (UC Berkeley/LBNL), Wenming Dong (LBNL staff)

Overall research goals:

The broad objectives of this part of Thrust 1 and 3 of the LBNL BES program are to understand the role of geochemistry in rock deformation including feedbacks between deformation and chemistry. Our contribution to this problem is to use trace element and isotope geochemical tracers to measure chemical fluxes between solids and fluid while simultaneously measuring strain and strain rate. This “application” of geochemistry raises questions about the degree to which we understand the controls on trace element and isotopic effects during dissolution, transport and precipitation, which in turn are determined by molecular to micro-scale processes at grain surfaces and transport along grain boundaries and through confined fluid at grain-grain contact surfaces.

In the process of using trace element and isotope geochemistry to further understanding of the role of chemistry in rock deformation we have recognized that rock deformation, specifically pressure solution, can be used with a flow-through column to access near-equilibrium conditions for mineral precipitation that are difficult to achieve otherwise. We are using this approach to measure near-equilibrium isotopic fractionation in the calcite-water system, and to better characterize near-equilibrium precipitation rates for calcite. The driver for these studies is the fact that many or even most natural geochemical processes take place over extended time periods at reaction rates that are far slower than can be achieved in laboratory experiments. To further constrain these ultra-slow processes we carry out parallel studies of deep sediment pore fluids, which remain in contact with the enclosing solids for millions of years and interact with the solid at near-equilibrium conditions.

In addition to conducting laboratory experiments, we use natural experiments taking place in the environment on long time scales to provide important constraints on whether measured rates in the laboratory can be scaled to processes in geological systems.

Significant achievements during 2023- 2025:

Our major achievement is represented by a paper in GCA (Lisabeth et al., 2024) that presents results from our first sets of calcite compaction experiments where we measured strain and dissolution/precipitation rates using a specially designed flow-through column. The paper includes protocols for using the geochemical data to infer reaction rates, and geometrical models to relate reaction rates to strain. Our conclusions are that chemical reaction rates are likely to be the most important limitation on deformation rates by pressure solution, rather than diffusion control along grain surfaces as typically inferred. But also, the evolution of chemical reaction rates with time can be different, and usually much slower than, what is calculated solely on the basis of normal stresses at grain contacts. Reaction rates also slow with time, and may be responsible for most of the observed time-dependence of strain rates.

In our first experiments using the flow through column to measure near-equilibrium Ca isotope fractionation between calcite and fluid we have a number of self-consistent experiments and a few that did not behave well due to experimental problems. The conclusions are not finalized but suggest that the equilibrium isotopic fractionation is close to 1.000 for $^{44}\text{Ca}/^{40}\text{Ca}$ which agrees with most data from deep sea carbonate sediments, but implies that reaction rates during diagenesis are extremely

slow; less than 10^{-10} to 10^{-12} mol/m²/s and perhaps much lower. This observation underscores the difficulty in applying results from laboratory studies to natural systems.

We conducted isotopic measurements of multiple substituted carbonate isotopologues from deep sea carbonate sediments extending to over 1 km burial depth and ranging in age up to ~75 million years. The sediments from the bottom portion of the core have fabric indicative of extensive pressure solution. We model the isotopic data accounting for slow dissolution and precipitation that occurs in the upper portion of the core absent pressure solution and reactivation due to pressure solution deeper in the core. This study provides deformation and reaction rates occurring on geologic timescales that relate to the transitions from ooze to chalk to limestone.

Other notable papers are two with collaborator Zhang on calcite-pore fluid reaction rates in deep sea sediments. In one paper we combined pore fluid Sr/Ca ratios with measurements of pore fluid Ca concentrations to show that pore fluids remain slightly undersaturated even as the sediment (which has 50 to 70% porosity) is buried to 100's of meters depth. So these sediments are not "closed chemical systems" as most investigators typically assume, but remain in diffusive communication with seawater for millions of years as they are buried by younger sediment. In the other paper we developed a model that can account for unexpectedly high rates of isotopic exchange between calcite and fluids in laboratory experiments, but it requires "solid state" diffusion in calcite with Ca, C, and O diffusivity values of order 10^{-24} m²/s.

Science objectives for 2026-2028:

We are planning new flow-through compaction column experiments using a newly designed and fabricated column and making other adjustments to improve reaction rate estimates. This work will be partly focused on using the compaction column to measure near-equilibrium isotopic and trace element partitioning between calcite and fluid, and extending this system to other minerals. The other focus will be on extending and improving our data on the relationship between strain rates and reaction rates, and exploring the effects of temperature and fluid composition.

In parallel we have started chemostat experiments designed to test a new model for Mg isotopes in calcite that we have developed. There is currently a large discrepancy between near-equilibrium isotopic fractionation measured in the laboratory and in deep sea pore fluids. Another objective is to test a model we developed for aqueous diffusion effects on isotopic fractionation during mineral precipitation; this requires fast growth rates and variable stirring rates.

The above mentioned studies, plus a planned field study in serpentinite using Mg isotopes to estimate serpentinization reaction rates are integrated with other ongoing and planned work in Thrust 2 and other parts of Thrust 1 and 3.

Publications supported by this project 2023-2025:

1. Lisabeth, Harrison, DePaolo, Donald J., Pester, Nicholas J., Christensen, John N., 2024, Concurrent measurement of strain and chemical reaction rates in a calcite grain pack undergoing pressure solution: Evidence for surface-reaction controlled dissolution. *Geochimica et Cosmochimica Acta* 385, 184-198 <https://doi.org/10.1016/j.gca.2024.09.018>
2. DePaolo, Donald J. and Zhang, Shuo, 2024, Calcite-fluid near-equilibrium dissolution and exchange rates: Uncertainties and ambiguities of isotopic tracer methods. *Geochimica et Cosmochimica Acta* 379, 233-243. <https://doi.org/10.1016/j.gca.2024.06.004>
3. Zhang, Shuo, DePaolo, Donald J., Zhou, Renjie, Huang, Yuefei, Wang, Guangqian, 2023, Origin and significance of ultra-slow calcite dissolution rates in deep sea sediments. *Geochimica et Cosmochimica Acta* 351, 181-191. <https://doi.org/10.1016/j.gca.2023.04.026>

Investigating the Physical Basis of Biomineralization

Patricia Dove, Principal Investigator

Department of Geosciences, Virginia Tech, Blacksburg, VA

Department of Chemistry, Virginia Tech, Blacksburg, VA

Department of Materials Sciences and Engineering, Virginia Tech, Blacksburg, VA

Graduate Students: Brenna Knight, Christina McCutchin, Ronnie Mondal, Connor Gallagher.

Collaborators: James De Yoreo, Chun-Long Chen, Biao Jin, Yuna Bae, Ying Chen (PNNL), Kevin Edgar, Louis Madsen, Michael Schulz, Valerie Vassier-Welborn (Virginia Tech).

Overall research goals:

Establish a quantitative and molecular-level understanding of how macromolecule chemistry controls the kinetics and energetics of crystal nucleation with a focus on timing and placement in biological mineralization. Our foundational studies using polysaccharides and peptoids seek to build structure-property relationships that elucidate how bioactive functional groups and their regiospecificities influence the onset of crystallization. Connect findings with efforts to develop new materials and novel separations processes for applications in subsurface and engineered systems.

Significant achievements during 2023-2025:

In 2023, our research shifted from using the quantitative tools developed over the course of our foundational research of biomineralization processes to focus on macromolecule chemistry and controls on biocrystallization of CaCO_3 and SiO_2 systems. This new collaboration with synthetic chemists who specialize in structure-function relationships of biopolymers has proven to be a valuable investment. By derivatizing natural polysaccharides into tailored compositions, we obtain unique palettes of hydrogel compositions that can pose specific and testable hypotheses.

Calcification. Initial studies focused on synthesizing and characterizing a library of chitosan (chitin)-based biopolymers with varied degrees of sulfation and regiospecificities. Sulfate groups are highly bioactive and abundant in biomineralization systems, but the community has traditionally focused on carboxylates. Measurements of calcite nucleation rate onto sulfated materials show solvent-functional group interactions dominate the thermodynamic barrier (excess free energy) to nucleation. MD simulations indicate the highest probability of Ca^{2+} - HCO_3^- interactions occurs at some distance apart from the sulfate groups. This finding supports recent evidence from nucleation studies of other systems, indicating nucleation occurs adjacent to, but not directly on, a macromolecule or solid substrate-water interface. We probe this prediction using liquid-cell TEM for real-time observations of calcite nucleation onto polysaccharides. By coating NH_4^+ -Si nanoparticles (np) with heparin (negatively charged by sulfation) and chitosan (small positive charge), we found nuclei form in the polysaccharide-water interphase region at a constant distance from the np. Our observations, however, could not resolve a distinct interface, thus preventing direct comparisons to model predictions. Ca^{2+} binding, $K_{b,\text{Ca}^{2+}}$ (25°C) to sulfated heparins was quantified by isothermal titration calorimetry to resolve the influence of SO_3^- and COO^- content and SO_3^- position. We find ΔG_{rxn} is dominated by entropic contributions due to favorable water release from polar, hydrophilic groups. Returning to published studies, we found that Ca^{2+} -polysaccharide interactions contain a shared thermodynamic signature that suggests these interactions are generally dominated by water restructuring with little conformational change. We further show that this relationship is distinct from Ca^{2+} -protein binding, with biological implications. The findings reiterate the role of macromolecular solvation during CaCO_3 formation and suggest broader applications to other sparingly soluble salts.

Macromolecule controls on silica nucleation (new). Polyamines and polysaccharides are known to control silica biomineral formation. However, a molecular-level understanding of how their composition and conformation influence Si condensation remains unclear. The macromolecular structure-property relationships that control silica condensation may also have multiple applications

to critical minerals recovery, including: delaying silica precipitation while keeping target elements mobile; redirecting silica nucleation away from critical ion-rich domains; promoting (or inhibiting) selective precipitation. In new work, we investigate a library of differently charged (and structured) macromolecules to quantify rates of silica condensation and find a previously unrecognized relationship with insights for molecular-level energetics.

Science objectives for 2026-2028:

1. Establish and validate a new approach to measure kinetics of silica nucleation using NMR to obtain a quantitative and molecular-level understanding of pathways to Si condensation.
2. Use the newly developed NMR approach and insights to design an experiment to investigate the mechanistic basis for how macromolecules promote (and inhibit) silicification.
3. Conduct complementary studies to determine the mechanistic basis for our measurements that find systematics between macromolecule composition and silica condensation kinetics.
4. Peptoids. Use the cationic and anionic peptoids that we synthesized at PNNL last summer to determine the energetic basis for why specific composition motifs and configurations catalyze the kinetics of silicification. Use these insights to design peptoids with enhanced abilities to promote (or inhibit) the kinetics of silica condensation.

Publications supported by this project 2023-2025:

1. Knight, Brenna M., Edgar, Kevin J., De Yoreo, James J., and Dove, Patricia M. Chitosan as a canvas for studies of macromolecular controls on CaCO_3 biological crystallization. *Biomacromolecules*, **2023**, 24, 1078 -1102. <https://doi.org/10.1021/acs.biomac.2c01394>.
2. Bañuelos, José Leobardo, Borguet, Eric, Brown, Gordon E. Jr., Cygan, Randall T., DeYoreo, James J., Dove, Patricia M., Gageot, Marie-Pierre, Geiger, Franz M., Gibbs, Julianne M., Grassian, Vicki H., Ilgen, Anastasia G., Jun, Young-Shin, Kabengi, Nadine, Katz, Lynn, Kubicki, James D., Lützenkirchen, Johannes, Putnis, Christine V., Remsing, Richard C., Rosso, Kevin M., Rother, Gernot, Sulpizi, Marialore, Villalobos, Mario, Zhang, Huichun. Oxide–and silicate–water interfaces and their roles in technology and the environment. *Chemical Reviews*, **2023**, 123, 6413 - 6544. <https://doi.org/10.1021/acs.chemrev.2c00130>.
3. Knight, Brenna M., Mondal, Ronnie, Han, Nizhou., Pietra, Nicholas F., Edgar, Kevin J., Welborn, Vaissier V., Madsen, Louis A., De Yoreo, Jame J., and Dove, Patricia M. Kinetics of Calcite Nucleation onto Sulfated Chitosan Derivatives and Implications for Water–Polysaccharide Interactions during Crystallization of Sparingly Soluble Salts. *Crystal Growth & Design*, **2024**, 24, 6338 – 6353. <https://doi.org/10.1021/acs.cgd.4c00602>.
4. McCutchin, Christina A., Edgar, Kevin J., Chen, Chun-Long, and Dove, Patricia M. Silica–biomacromolecule interactions: toward a mechanistic understanding of silicification. *Biomacromolecules*, **2024**, 26, 43 - 84. *Featured as journal cover illustration*, <https://doi.org/10.1021/acs.biomac.4c00674>.
5. Knight, Brenna M., Gallagher, Connor M., Schulz, Michael D., Edgar, Kevin J., McNaul, Caylyn D., McCutchin, Christina A., and Dove, Patricia M. Thermodynamics of calcium binding to heparin: Implications of solvation and water structuring for polysaccharide biofunctions. *Proceedings of the National Academy of Sciences*, **2025**, 122, e2504348122. <https://doi.org/10.1073/pnas.2504348122>.
6. Knight, Brenna M., De Yoreo, James J., and Dove, Patricia M. *In situ* transmission electron microscopy observations of CaCO_3 crystallization onto polysaccharide-coated nanoparticles **2026**, *Crystal Engineering Communications*. 2026, Submitted.
7. Knight, Brenna M., Giuffre, Anthony J., De Yoreo, James J., Dove, Patricia M. Effect of ionic strength and polysaccharide chemistry in regulating kinetics of calcite nucleation through competition of interfacial forces. *Royal Society of Chem. Advances*. **2026**, near-submission.
8. Gerlach, Kyle, Ambrosi, Brittani, Tschetter, Rachel, Chen, Michael, McCutchin, Christina, Dove, Patricia. Alginate- CaCO_3 hydrogel composites for controlled drug delivery in transdermal applications. *Carbohydrate Polymer Tech. and Applications*. **2026**, near-submission.

Mineral-Fluid Interactions: Synchrotron X-ray Studies

Paul Fenter, Principal Investigator

Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL

Senior Investigators: Sang Soo Lee, Ana Suzana

Post-Doctoral Fellows: Anusheela Das, Yihang Yuan

Collaborators (unfunded): Irene Calvo-Almazán (University of Zaragoza, Spain), Neil Sturchio (University of Delaware, retired)

Overall research goals:

This project seeks to obtain a mechanistic understanding of the processes that govern mineral-water interface reactivity and elemental transport in low-temperature geochemical systems in order to establish the scientific foundations needed for the efficient extraction of resources (e.g., critical materials) and the disposition of energy byproducts in Earth's sub-surface. Our approach is to use direct, in situ, and real-time observations of well-defined systems at the molecular scale, by focusing on three inter-related themes. Studies of "intrinsic" mineral reactivity on planar mineral surfaces examine interfacial hydration and ion adsorption controlled by surface charge, ion hydration strength, and mineral chemistry at well-defined mineral-water interfaces (e.g., muscovite mica and alumina). This is complemented with studies of the "extrinsic" controls that influence interfacial reactivity, including crystal shape, size, reactant transport, solution flow, and local disequilibria, with a focus on calcium carbonate minerals. Synergies between these activities are enabled by the use and development of novel "coherent X-ray imaging" capabilities that leverage the unique characteristics of 4th generation hard X-ray sources (e.g., the Advanced Photon Source Upgrade, APS-U). These capabilities allow direct observation of structural complexity of geochemical systems (i.e., the spatial distribution of defects at planar surfaces and three-dimensional nano- to micron-sized crystals), providing a pathway for establishing robust structure-function relationships that govern geochemical interfacial reactivity.

Significant achievements during 2023- 2025:

The "intrinsic" structures and chemical reactivities of planar mineral-water interfaces were studied using X-ray reflectivity (XR) and resonant anomalous X-ray reflectivity (RAXR). The salinity-dependent evolution of the electric double layer (EDL) structure was observed via direct visualization of correlated cation-anion sorption structures at the muscovite (001)-water interface during the transition from classical (i.e., dilute) to non-classical (i.e., saline) regimes [1]. Rb⁺ and Br⁻ distributions were probed as a function of ion concentration. Overcharging was manifested as the formation of alternating cation and anion layers, and the onset of overcharging was found to be sensitive to the choice of anion (Cl⁻, Br⁻, I⁻). Additional studies of the adsorption of uranyl [U(VI)O₂²⁺] on the alumina Al₂O₃(012)-water interface revealed multiple coexisting inner- and outer-sphere species whose relative amounts were controlled by pH [2]. Notably, the outer-sphere uranyl species became more dominant at higher pH, indicating an enhanced role for hydrogen bonding in stabilizing adsorbed uranyl at the interface.

The role of "extrinsic" factors (e.g., strain, morphology, and ion transport) was investigated by observing the reactivity of calcite in undersaturated (acidic) solutions containing dissolved metal impurities (e.g., Pb²⁺). Operando observations of calcite (104) surfaces during dissolution under transport-controlled conditions (using digital holographic microscopy, DHM) revealed that the average calcite dissolution rate was systematically inhibited with increasing [Pb²⁺], and that the spatial variability of reactivity produced complex interfacial morphologies [3]. This inhibition of dissolution at elevated [Pb²⁺] was shown (by XR and RAXR) to be controlled by a novel ion sorption mode in which Pb²⁺ substituted for Ca²⁺ within the top four molecular layers at a dynamically dissolving surface [4]. The role of lattice strain in altering the reactivity of calcite

nanoparticles was demonstrated by Bragg coherent diffraction imaging (BCDI) and scanning electron microscopy (SEM), revealing that dissolution rates were enhanced at highly strained surface regions and this surface reactivity subsequently altered the lattice strain distribution within the reacted crystal [5].

X-ray coherence provides fundamentally new opportunities to observe the structural heterogeneity of geochemical interfaces. BCDI revealed the sensitivity of the internal nano-crystallinity in rhombohedral calcite particles ($\sim 1\ \mu\text{m}$ in size) to the synthesis conditions (i.e., equilibrium vs. non-equilibrium conditions) [6]. We also used computational simulations to establish the theoretical basis for observing nm-scale topography of mineral surfaces using coherent X-ray reflectivity [7].

Science objectives for 2026-2028:

We will systematically explore how the EDL structure at the mineral-water interface evolves as a function of ion concentration for different cation/anion pairs, with an emphasis on understanding the changes in EDL structure under static and dynamically flowing solutions. We will expand our understanding of how dissolved metal cations influence the reactivity of calcium carbonate minerals in acidic solutions. This study will compare the effects of dissolved metal ions (i.e., Cd^{2+} , Zn^{2+} , Sr^{2+} , Co^{2+} and Cu^{2+}) that were selected to distinguish different impurity-induced inhibition mechanisms (e.g., due to ion size, carbonate phase stability, and cation water-exchange rate). DHM, SEM, and BCDI will be used to track the topographic evolution of mineral surfaces. We will also demonstrate the application of new coherent X-ray imaging capabilities to geochemical systems. BCDI will be used to probe the coupling changes in morphology and internal strain in mineral particles during reactions with these dissolved metal ions. We also will continue to expand the use and application of new coherent X-ray capabilities. Three-dimensional ptychographic imaging will be used to image $>1\ \mu\text{m}$ sized mineral grains, complementing the BCDI-based capabilities. Coherent X-ray reflectivity will be used to probe interfacial topography of buried interfaces and surfaces of extended samples using through experimental observations and calculations.

Selected publications supported by this project 2023-2025:

1. Julia Neumann, Sang Soo Lee, Eric J. Zhao, Paul Fenter, "Direct Experimental Observations of Ion Distributions during Overcharging at the Muscovite-Water Interface by Adsorption of Rb^+ and Halides (Cl^- , Br^- , I^-) at High Salinity", *ChemPhysChem*, **2023**, 24, e202300545(1-9), [DOI](#).
2. Julia Neumann, Amanda J. Carr, Jessica Lessing, Lynda Soderholm; Jeffrey G. Catalano, Paul Fenter, Sang Soo Lee, "Unravelling pH-Dependent Changes in Adsorption Structure of Uranyl on Alumina (012)", *Journal of Physical Chemistry Letter*, **2024** 15, 3493–3501, [DOI](#).
3. Bektur Abdilla, Sang Soo Lee, Paul Fenter, Neil C. Sturchio, "Dynamic Surface Incorporation of Pb^{2+} Ions at the Actively Dissolving Calcite (104) Surface", *Environmental Science & Technology*, **2024** 58, 16525-16534, [DOI](#).
4. Bektur Abdilla, Sang Soo Lee, Paul Fenter, Neil C. Sturchio, "Dynamic Inhibition of Calcite Dissolution in Flowing Acidic Pb^{2+} Solutions", *Environmental Science & Technology*, **2024**, 58, 7133–7143, [DOI](#).
5. Ana F. Suzana, Sang Soo Lee, Irene Calvo Almazán, Wonsuk Cha, Ross Harder, Paul Fenter, "Insights into Strain Change of Calcite by Imaging Defect-driven Dissolution in the Presence of Pb^{2+} ", *Communications Materials*, **2025** 6, 285(1-10), [DOI](#).
6. Ana F. Suzana, Sang Soo Lee, Irene Calvo Almazán, Wonsuk Cha, Ross Harder, Paul Fenter, "Visualizing the Internal Nano-crystallinity of Calcite during Non-classical Crystallization by 3D Coherent X-ray Diffraction Imaging", *Advanced Materials*, **2024**, 2310672(1-11), [DOI](#).
7. Paul Fenter, Irene Calvo Almazán, "Imaging Interfacial Topography with Coherent X-ray Reflectivity", *Physical Review B*, **2024**, 109, 094112(1-13), [DOI](#).

Chemical-Mechanical Controls on Subsurface Resources

Benjamin Gilbert, Principal Investigator

The BES CSGB Core Program at Lawrence Berkeley National Laboratory, Berkeley, CA

Team Members: Mengsu Hu (LBNL; Thrust 1 Lead), Harry Lisabeth (LBNL; Thrust 2 Lead), Daniel Stolper (UC Berkeley; Thrust 3 Lead), Jill Banfield (UC Berkeley), John Christensen (LBNL), Abdullah Cihan (LBNL), Don DePaolo (LBNL), Wenming Dong (LBNL), Ben Gilbert (LBNL), Zhao Hao (LBNL), Michael Manga (UC Berkeley), Seiji Nakagawa (LBNL), Carl Steefel (LBNL), Drew Syverson (University of North Carolina at Charlotte), Christophe Tournassat (LBNL and University of Orléans), Yuxin Wu (LBNL), and Piotr Zarzycki (LBNL)

Graduate Students: Ziman Wu, Tyler Cadena, Yarong Qi (PhD candidates, UC Berkeley), Rundong Huang (PhD candidate, UC Riverside), Hooman Dadras (PhD candidate, UCo Boulder), Siqin Yu (PhD intern, Stanford University), Serhat Sevgen (PhD intern, University of Calgary)

Postdocs: Arun Bhattecharjee, Anne Voigtlaender, Melese Dessie

Overall research goals:

The goals of the Geoscience core program at Berkeley Lab are:

1. Thrust 1: Establish the relative roles of stress and fluid chemistry on the three dominant pathways through which porous carbonate reservoirs evolve permeability and strength
2. Thrust 2: Test the hypothesis that mineral serpentinization rates, the yield of hydrogen, and the risk of seismicity, are governed by anisotropic stresses originating from tectonic forces or through the mineral transformations themselves.
3. Thrust 3: Advance the foundations of equilibrium and kinetic isotopic distributions between aqueous fluids and mineral phases, and use that knowledge in Thrusts 1 and 2

A high-level overview

Geologic formations in the Earth's crust offer essential resources for current and future societal energy demands. Carbonate reservoirs, sources of fossil fuels for almost two centuries, may be repurposed for long-term storage of carbon dioxide or short-term storage of hydrogen. Iron-bearing silicate formations are source rocks for geologic hydrogen generated via serpentinization reactions occurring in the geologic past or through future subsurface engineering. Exploiting these subsurface systems and resources requires the injection or withdrawal of fluids that alter the stress state and shift the chemical equilibrium of the formation. Establishing the mechanisms and rates of processes in which flow, chemistry and mechanics are combined, however, is a basic science grand challenge. This project addresses that challenge through three aligned and collaborative research Thrusts (Fig. 1).

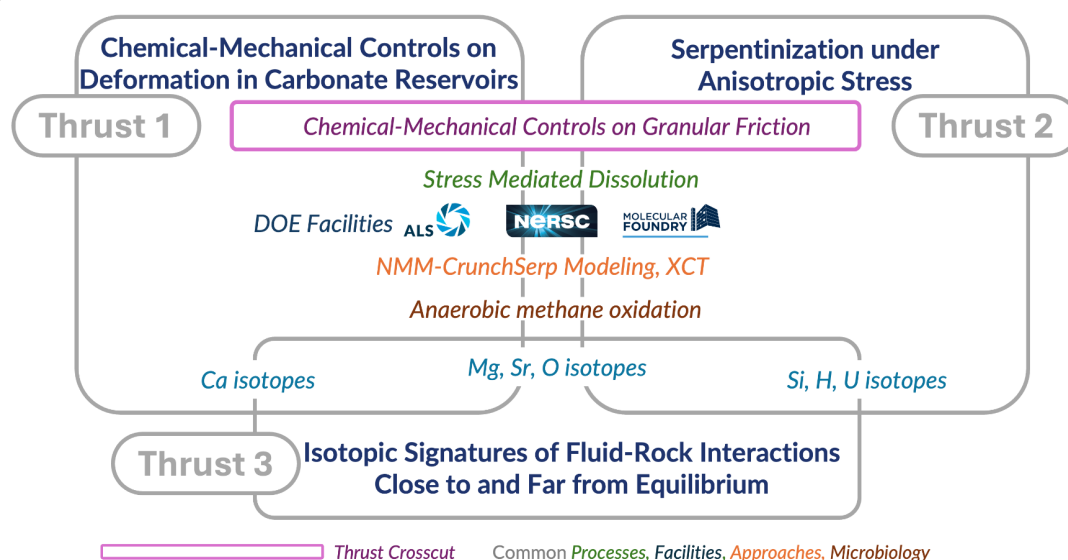


Figure 1 Organization of Thrusts and major crosscutting approaches and activities

Significant achievements during 2023-2025:

In the first project period, we created a collaborative multidisciplinary team to discover and model the fundamental processes of subsurface system evolution in regimes of stress and chemical reactivity both close to and far away from equilibrium.

The team developed new experimental and modeling tools:

- The NMM-Crunch code for predictive pore-scale chemical-mechanical simulation
- Theory and numerical implementation of electrochemical effects on nanopore flow
- Free-energy based continuum density functional theory (cDFT) of nanoscale flow, reaction and stress
- Double torsion instrument and methods for chemical effects on crack opening and healing
- Surface forces instrumentation and methods for single-interface pressure solution
- X-ray nanotomography instrument for *in situ* fluid-rock deformation and alteration
- Quantum sensing instrument for nanoscale fluid chemistry
- Triple oxygen isotope instrumentation and methods for fluid-rock interaction mechanisms
- Advances in cryogenic electron microscopy for atomistic fluid-nanoparticle imaging
- Development of non-linear complex conductivity models for geophysics monitoring
- Accurate prediction of mineral surface speciation using deep neural networks

The team applied these tools to achieve new insights into fluid-mineral interactions, including:

- Integration of molecular surface forces into predictive model of crack processes (Dadras et al., 2025)
- Use of fluid isotopic data and modeling to constrain granular calcite compaction pathways (Lisabeth et al. 2024)
- Coupling of electrostatics and mechanics for new model of clay swelling (Whittaker et al., (2023))

Science objectives for 2026-2028:

Thrust 1: Establish predictive understanding of pressure solution and crack propagation and healing in calcite, identify the mechanisms of calcite interface friction and shear, and develop an integrated model for carbonate rock deformation and failure.

Thrust 2: Establish the mechanistic controls on hydrogen generation, transport and loss during serpentinization at all spatial scales, including molecular scale iron and hydrogen redox chemistry, microbial hydrogen, iron and carbon cycling, nanoscale fluid flow, and grain-to-formation scale shear stress, fracturing, flow and reaction.

Thrust 3: Achieve an order of magnitude increase in the ability to predict equilibrium isotopic fractionation and develop new isotopic signatures of mineral growth rates and pathways.

Publications supported by this project 2023-2025:

46 peer reviewed papers were fully or partially supported by this project 2023 - 2025. Selected papers illustrating multidisciplinary advances on chemical-mechanical processes are:

1. H Lisabeth, DJ DePaolo, NJ Pester, JN Christensen, "Concurrent measurement of strain and chemical reaction rates in a calcite grain pack undergoing pressure solution: Evidence for surface-reaction controlled dissolution." *GCA* **2024** 385, 184-198, 10.1016/j.gca.2024.09.018.
2. ML Whittaker, M Shoaib, LN Lammers, Y Zhang, C Tournassat, B Gilbert, "Smectite phase separation is driven by hydration-mediated interfacial charge." *J Coll Interf Sci* **2023**, 647, 406-420, 10.1016/j.jcis.2023.05.085.
3. H Dadras, M Eskandari-Ghadi, S Nakagawa, B Gilbert, Y Zhang. "Complex dependence of calcite crack kinetics on salinity: The role of DLVO and hydration forces." **2025**, *J Geophys Res* 130 (9), e2025JB031332.
4. M Hu, CI Steefel, J Rutqvist, B Gilbert, "Microscale THMC Modeling of Pressure Solution in Salt Rock: Impacts of Geometry and Temperature". *Rock Mech Rock Eng* **2023** 10.1007/s00603-022-03162-6.

Chemistry of CaCO₃ phase transitions

Pupa Gilbert, Principal Investigator

Department of Physics, University of Wisconsin-Madison, and Chemical Sciences Division, Lawrence Berkeley National Laboratory, 94720 Berkeley CA

Co-PI: Daniel Stolper, Department of Earth and Planetary Science, University of California Berkeley, and Energy Geosciences Division, Lawrence Berkeley National Laboratory

Graduate students: Ziman Wu (UC Berkeley) and Aiden Gustafson (UW-Madison)

Postdoctoral students: Cristian Castillo Alvarez and Laurent S. Devriendt, LBNL

Collaborators: Sylvie Tambutté, Centre Scientifique de Monaco, Principality of Monaco, Giovanni De Giudici, University of Cagliari, Sardinia, Italy.

Overall research goals:

Understanding how calcium carbonates are formed biogenically, synthetically, and geologically

Significant achievements during 2023-2025:

We made several significant discoveries, including: (1) precursors to CaCO₃ formation are not only 2 amorphous phases, as previously known, but also 2 metastable crystalline phases, precursors to biogenic aragonite (CaCO₃) in corals and nacre¹. (2) Surprisingly, the 4 precursors all transform to aragonite following an exponential decay law in space and time, and do so with the same time constants for all phases². (3) We observed for the first time multiple twinning in geologic and biogenic aragonite, up to quadruple twinning³. (4) We experimentally tested whether there are isotopic signatures of phase transitions in synthetic CaCO₃⁴. (5) We found a clear correlation of B concentration and isotopes with growth rate in corals⁵. (6) Most recently, and excitingly, initial data indicate that the same 4 precursor phases observed in coral skeletons and nacre are also precursors to aragonite formation in geologic stalactites from Sardinia⁶. Other papers on CaCO₃ systems were published, under review, or in preparation⁷⁻¹⁵.

Science objectives for 2026-2028:

We plan to confirm the observation of the 4 precursor phases in fresh, forming aragonite stalactites from Sardinia, Italy. If confirmed, this observation will provide a transformative new understanding of geologic mineral formation, for aragonite and potentially for other minerals and rocks. The impact of such a discovery could be transformative in space and time, because it will show that formation via metastable precursors is a common, general mechanism for the formation of carbonate minerals on Earth today and likely in the geologic past. These include, but are not limited to, ooids, carbonate platforms, and stromatolites, all of which will be subject of the upcoming renewal proposal. For now, it is critical that we confirm this initial observation. Following this, we will expand to calcite (CaCO₃) stalactites, and to other important carbonates.

Publications supported by this project 2023-2025:

1. CA Schmidt, E Tambutté, AA Venn, Z Zou, C Castillo Alvarez, LS Devriendt, H Bechtel, CA Stifler, S Anglemeyer, CP Breit, CL Foust, A Hopanchuk, CN Klaus, IJ Koehler, IM LeCloux, J Mezera, MR Patton, A Purisch, V Quach, JS Sengkhamee, T Sristy, S Vatter, EJ Walch, M Albéric, Y Politi, P Fratzl, S Tambutté, PUPA Gilbert, "Myriad Mapping of nanoscale minerals reveals calcium carbonate hemihydrate in forming nacre and coral biominerals" *Nature Communications*, **2024**, 15, 1812, <https://doi.org/10.1038/s41467-024-46117-x>.

2. Z Rechav, E Tambutté, IM LeCloux, S Anglemeyer, NE Beltz, NA Chou, BE Dixon-Kruijff, J Domagk, AM Larson, SW Lewis, R Rich, LO Saheed, JL Schwenk, JS Sengkhamee, CA Waltenberg, J Ye, BQ Achinuk, AA Venn, S Tambutté, PUPA Gilbert, "Exponential crystallization in corals" *Nature Communications*, **2026**, accepted for publication on Jan 5, 2026.

3. C Schmidt, W Nachlas, B Schneider, X Mao, E Tambutté, AA Venn, S Tambutté, PUPA Gilbert, "Multiple twinning in nacre and aragonite" *Advanced Functional Materials*, **2025**, e18248, 1-12, <https://doi.org/10.1002/adfm.202518248>.
4. LS Devriendt, DA Stolper, PUPA Gilbert, "Carbon, oxygen, and clumped isotopes in calcite are not altered by formation via amorphous precursors" *Geochimica et Cosmochimica Acta*, **2026**, in preparation.
5. C Castillo Alvarez, DA Stolper, E Tambutté, H Jurikova, JWB Rae, AA Venn, S Tambutté, PUPA Gilbert, "Boron, growth rates, and more accurate pH reconstructions from coral skeletons" *Nature Geosciences*, **2026**, in preparation.
6. A Gustafson, I Carlomagno, L Sanna, CA Schmidt, BQ Achinuq, S Anglemeyer, NA Chou, J Domagk, SW Lewis, LO Saheed, JL Schwenk, CA Waltenberg, G De Giudici, PUPA Gilbert, "Amorphous and crystalline precursors to speleothem formation" *Nature*, **2026**, in preparation.
7. PUPA Gilbert, DR Green, P Mahoney, D Guatelli-Steinberg, WS McGraw, E Lagan, F Kyalo Manthi, S Muteti, E Ndiema, F Ramirez Rozzi, CA Stifler, CA Schmidt, BQ Achinuq, A Scholl, B Gilbert, MC O'Hara, "Human and primate enamel nanocrystal c-axis misorientation varied with diet" *Nature*, **2026**, under review.
8. J Wilcots, KD Bergmann, PUPA Gilbert, AJ Cross "Nanoscale crystal fabric preserved in dolomite ooids at the onset of the ediacaran Shuram excursion" *The Sedimentary Record*, **2025**, 23, 1-17, [10.2110/001c.145144](https://doi.org/10.2110/001c.145144).
9. Z Wu, E Tambutté, C Castillo Alvarez, S Tambutté, PUPA Gilbert, DA Stolper, "Clumped isotope thermometry away from equilibrium in synthetic and biogenic CaCO_3 systems" *Geochimica et Cosmochimica Acta*, **2026**, in preparation.
10. C Castillo Alvarez, E Tambutté, V Mavromatis, S Tambutté, DA Stolper, PUPA Gilbert, "Magnesium concentration and isotopic composition correlates with crystal growth rate in synthetic and biogenic CaCO_3 systems" *Geochimica et Cosmochimica Acta*, **2026**, in preparation.
11. EW Lees, C Tournassat, AZ Weber, PUPA Gilbert, "eCoral: How electrolysis could restore seawater conditions ideal for coral reefs" *J Phys Chem Letters*, **2024**, 15, 12206-12211, <https://doi.org/10.1021/acs.jpclett.4c02715>.
12. AJ Lew, CA Stifler, A Tits, CA Schmidt, A Scholl, A Cantamessa, L Müller, Y Delaunois, P Compère, D Ruffoni, MJ Buehler, PUPA Gilbert, "A Molecular Scale Understanding of Misorientation Toughening in Corals and Seashells" *Advanced Materials*, **2023**, 35, 2300373, <https://doi.org/10.1002/adma.202300373>.
13. AJ Lew, CA Stifler, A Cantamessa, A Tits, D Ruffoni, PUPA Gilbert, MJ Buehler, "Deep learning virtual indenter maps nanoscale hardness rapidly, non-destructively, revealing mechanism, and enhancing bioinspired design" *Matter*, **2023**, 6, 1975-91, <https://doi.org/10.1016/j.matt.2023.03.031>.
14. PUPA Gilbert, "Biomaterial mesostructure" *MRS Bulletin*, **2023**, 48, 413-420, <https://doi.org/10.1557/s43577-023-00479-7>.
15. C Castillo Alvarez, JL Grimsich, CA Schmidt, H Lisabeth, A Voigtländer, PUPA Gilbert, "Calcite twinning in mollusk shells and Carrara marble" *Advanced Functional Materials*, **2023**, 34, 2304288, <https://doi.org/10.1002/adfm.202304288>.

Assessing the Relative Contributions of Fluid Pressure and Elastic Stress to Induced Seismic Energy Release

Thomas H. Goebel, *Principal Investigator, Center for Earthquake Research and Information, University of Memphis*

Graduate Students: Roshan Koirala and Navin Thapa, *Postdocs:* Valerian Schuster, German Research Centre for Geosciences, *Collaborators:* Georg Dresen, German Research Centre for Geosciences, Emily Brodsky, University of California, Santa Cruz, Trenton Cladouhos, Cyrq Energy (now Quaise Energy), Daniel Minquez, Cyrq Energy

Overall research goals:

The project focuses on a multi-scale analysis of hydrothermal alterations, slip-induced permeability changes and poroelastic effects in geothermal reservoirs. The work addresses three primary questions: (1) What is the contribution of poroelastically induced events to total moment release? (2) What conditions promote large-magnitude induced earthquakes? (3) What are realistic permeability poroelastic coupling coefficients for fault damage zones? The work integrates coupled poroelastic models with reservoir and laboratory measurements and tests the effects of seismic and aseismic slip. Reservoir-scale injection tests and permeability measurements are conducted at an active geothermal reservoir in Nevada. Using both reservoir-scale measurements and controlled laboratory injection experiments, we characterized surface deformation, hydrothermal alterations, and induced seismicity in Blue Mountain. We calculated the relative differences in seismic moment release from long-term operations which are pressure and temperature dominated vs. poroelastically induced events after rapid operational changes. Results from this research have immediate implications on induced seismicity assessment and improved geothermal reservoir characterization.

Significant achievements during 2023- 2025:

We completed three distinct studies that explored 1) long and short-term deformation of geothermal reservoirs, 2) induced earthquake magnitudes in enhanced geothermal systems, and 3) poroelastic deformation and permeability changes in geothermal rock samples. These three studies are summarized in the following.

1) Geothermal reservoir operations can lead to seismic activity, for instance, due to increased pore pressure, but some observations are difficult to explain by pressure changes alone. Such observations include unexpected, induced events after injection well shut-in when pore pressures are thought to decrease. We analyzed induced seismicity and surface deformation in Blue Mountain, Nevada using seismic, geodetic, and hydraulic data. Long-term surface subsidence reached up to 1 cm/yr above the reservoir and was associated with modest seismic activity concentrated within the geothermal reservoir at 0.5 - 2.5 km depth.

We numerically modelled abrupt seismicity rate changes during two maintenance shutdowns. The observed seismic activity can be explained by short-term poroelastic effects and increased Coulomb stressing rates. Our findings demonstrate that poroelastic coupling can result in abrupt increases in seismic activity following well shutdowns, while aseismic fault slip and thermal contraction dictate the long-term static deformation in the Blue Mountain reservoir.

2) We investigated the effect of fault roughness and pore fluid pressure on induced earthquake magnitudes in the laboratory and compared them with hydro-shearing in Enhanced Geothermal Systems. Spatial-temporal variations in b-values (exponent of the frequency-magnitude distribution) are a result of increased differential stress which is further amplified by higher pore pressure close to failure. Higher pore pressures generally lead to lower b-values.

Spatial variations during EGS stimulation are thought to be structurally controlled so that high damage zones promote more small magnitude earthquakes and higher b-values. We conclude that b-value variations outside the lab require cautious interpretation because of these multiple underlying causes.

3) We experimentally analyzed the effect of hydrothermal alteration on the petrophysical properties, mineral composition, microstructural characteristics, volumetric deformation and permeability of various altered dikes and phyllites obtained from the Blue Mountain, Nevada. Hydrothermal alterations increase phyllosilicate mineral content, porosity, and hydraulic storativity of dikes from Blue Mountain, while reducing their density, stiffness, and thermal conductivity. Due to elevated porosity, altered dikes exhibit increased poroelastic coupling, which is expressed by high effective pressure coefficients of volumetric deformation. Propylitic alterations tend to be stiff and large, which reduces the stress sensitivity of permeability and porosity of altered dikes. These rocks are more susceptible to pore pressure induced deformation and subsidence during fluid extraction, but long-term production decline is limited. Although fractures can increase permeability, an increase in effective pressure due to the closure of compliant and eccentric pores is likely to result in a significant decrease in flow rate, as observed in microcracked granite and silicified phyllite. Our results highlight the effects of argillic–propylitic and silicification on porosity and pore geometry of geothermal reservoir rocks and how these affect poroelastic and hydraulic behavior. Accounting for these effects is crucial to assess reservoir performance and plan well hydraulics.

Science objectives for 2026-2028:

There are many observations of distributed and localized strain accumulation and release around geothermal fields, however, seismic hazard implications remain insufficiently understood. To address this knowledge gap, we will tackle three fundamental questions: 1) How can we measure and quantify aseismic deformation across different geothermal reservoirs? 2) What controls the relative amount of aseismic deformation in a geothermal reservoir? 3) Do reservoirs with higher levels of aseismic deformation produce lower magnitude induced events? This work builds on our recent publications of results from Blue Mountain, Nevada where reservoir-scale induced strain is predominantly released aseismically and the largest magnitude induced event was only ML1.5. The proposed work will concentrate on an integration of InSAR surface displacements, numerical models and seismicity analysis to quantify the relative amount of aseismic deformation across different geothermal reservoirs. We will compare relative aseismic deformation with the magnitude of the largest, induced event. Results from this work are expected to improve the understanding of aseismic deformation processes during long-term geothermal operations and implications for seismic hazards.

Publications supported by this project 2023-2025:

1. Thapa, Navin, Georg Dresen, and Thomas H. Goebel. "Does b-value increase with pore-pressure?: Insights from laboratory experiments and induced seismicity." *Geophysical Research Letters* 52.11 (2025), <https://doi.org/10.1029/2025GL115740>.
2. Schuster, Valerian, et al. "The effect of hydrothermal alteration and microcracks on hydraulic properties and poroelastic deformation: A case study of the Blue Mountain geothermal field." *Journal of Geophysical Research: Solid Earth* 130.4 (2025). <https://doi.org/10.1029/2024JB030541>
3. Koirala, Roshan, et al. "Induced seismicity and surface deformation associated with long-term and abrupt geothermal operations in Blue Mountain, Nevada." *Earth and Planetary Science Letters* 643 (2024). <https://doi.org/10.1016/j.epsl.2024.118883>
4. Pandey, Thapa, Dresen, Goebel, "Strain Localization Increases Repeating Earthquakes: Perspective from Laboratory Seismicity" (in prep. for *Pure and Applied Geophysics*)

Fractionation and mobilization of rare earth elements (REE) in hydrothermal ore deposits: from laboratory experiments to geochemical modeling

Alexander Gysi, Principal Investigator; Nicole Hurtig, co-Investigator; Laura Waters, co-Investigator

NM Bureau of Geology and Mineral Resources, New Mexico Institute of Mining and Technology, Socorro, NM

Dept. Earth & Environmental Science, New Mexico Institute of Mining and Technology, Socorro, NM

Graduate Students: Debarati Banerjee, Charles Kershaw, Kevin Padilla, Yerko Figueroa Penarrieta

Collaborators: Hannah Han (NMT), Sarah Smith-Schmitz (NMT), Artas Migdisov (LANL), Chen Zhu (IUB)

Overall research goals:

The rare earth elements (REE) are critical due to the potential disruption in their market supply and because they are essential materials for the high-tech and green energy industries. Expertise on the fundamental behavior of REE in geologic systems is required to more efficiently explore for potential critical mineral deposits and improving targeted exploration discoveries. Major REE deposits include carbonatites and (per)alkaline systems, which form through a complex interplay of magmatic-hydrothermal processes during their emplacement in the Earth's crust. Magmatic-hydrothermal fluids are capable of mobilizing, transporting, and depositing the REE. Slightly different chemical behavior of the REE can lead to fractionation of the larger light (L)REE from the smaller heavy (H)REE. However, understanding the underlying processes that affect the REE in these geologic fluids requires fundamental knowledge of their thermodynamic properties in minerals and aqueous species, their coordination chemistry and their molecular-level behavior. Surprisingly little is known about the mechanisms that lead to fractionation, transport, and enrichment of the REE in critical mineral deposits, particularly in high-temperature (400–600 °C) supercritical fluids, which we hypothesize, represent the optimal transport conditions for REE in geologic systems. The overall research goals of this project are to: 1) experimentally determine the solubility of REE minerals as a function of temperature, pressure, pH, salinity, and ligand concentrations (Cl^- , OH^- , SO_4^{2-} , CO_3^{2-}); 2) determine *in situ* speciation of REE in hydrothermal fluids using spectroscopic techniques and identify REE aqueous complexes controlling REE mineral solubilities and L/HREE fractionation; 3) derive thermodynamic constants and build a thermodynamic database that permits modeling the behavior of REE in hydrothermal ore-forming systems.

Significant achievements during 2023- 2025:

In the last few years, we have developed new experimental techniques and thermodynamic optimization methods at New Mexico Tech to determine the properties of REE aqueous species and REE minerals. We have developed calorimetric, UV-Vis spectrophotometric, potentiometric, and REE hydroxide solubility experiments to determine the properties of the REE^{3+} aqua ion and the hydrolysis constants for REE including for La, Nd, Dy, and Er from 25 to 250 °C. These data permit predicting accurately the speciation of REE hydroxyl complexes as a function of pH. We have developed new experimental methods to determine the solubility of REE phosphates (monazite, xenotime) at sub- to supercritical conditions from 350 to 700 °C and saturated water vapor pressure (P_{sat}) to 2 kbar in chloride-bearing solutions as a function of pH. These experiments permit deriving thermodynamic constants for REE chloride and hydroxyl complexes indicating significant L/HREE fractionation mechanisms being controlled by ligand type, pH and temperature, and orders of magnitude increase in REE mineral solubilities at supercritical conditions. We have developed new *in situ* confocal Raman characterization techniques for the speciation of aqueous Nd, Dy, and Yb chloride and sulfate complexes using quartz capillaries up to 300 °C. We also developed a new

multivariate curve resolution method for the interpretation of Raman spectra based on the perturbation of the water band by REE-ligand complexes. This method is promising for quantification of Nd and Yb aqua ions and chloride species at room temperature. Finally, we built a hydrothermal diamond anvil cell to measure the solubility of REEPO₄ (monazite, xenotime) and the speciation of REE sulfate complexes at high pressure (0.2-2 GPa) from 25 to 600 °C using Raman and synchrotron XRF. Experimental results are summarized in the ThermoExp REE database and results of thermodynamic optimization for REE species have been implemented in the MINES thermodynamic database which permit modeling accurately the solubility of REE phosphates from 25 to 300 °C, and future updates will include results at supercritical conditions.

Science objectives for 2026-2028:

The new DOE EPSCoR grant supports the NMT CritMin experimental research hub and aims at investigating the solubility of REE mineral solid solutions at sub- to supercritical conditions, measuring thermodynamic constants for predicting the speciation of REE chloride, sulfate, carbonate complexes as a function of pressure-temperature, and develop new methods to determine calorimetry of REE adsorption on clay minerals. Furthermore, we are working on developing equation of states for modeling the speciation of REE across the entire range of conditions studied, and we plan on further investigating the importance of pressure on REE complexation and resulting L/HREE fractionation. A key objective will be to integrate all experimental data into the MINES database, make it available for a range of geochemical modeling programs, and build new predictive tools for fitting thermodynamic properties of REE in hydrothermal fluids and minerals.

Selected publications supported by this project 2023-2025:

1. Banerjee, D., Waters, L., Hurtig, N.C., Gysi, A.P., Harlov, D., Zhu, C., Migdisov, A., "NdPO₄ solubility and aqueous neodymium speciation in supercritical fluids: An experimental study at 500–700°C and 1.7 kbar" *Geochim. Cosmochim. Acta*, **2025**, 406, 211–226, <https://doi.org/10.1016/j.gca.2025.01.004>.
2. Figueroa Penarrieta, Y., Gysi, A. P., "Hydrothermal solution calorimetry in acidic aqueous solutions and revisiting the standard partial molal thermodynamic properties of Nd³⁺ from 25 to 300 °C" *Journal Name or Abbreviation*, **2025**, 406, 227–242, <https://doi.org/10.1016/j.gca.2024.07.014>.
3. Gysi, A. P., Hurtig, N. C., Pan, R., Miron, G. D., Kulik, D. A., "MINES thermodynamic database, New Mexico Bureau of Geology and Mineral Resources, version 23" New Mexico Bureau of Geology and Mineral Resources, **2023**, DOI. <https://doi.org/10.58799/mines-tdb>
4. Han, H. J., Gysi, A. P., "UV-Vis spectrophotometric determination of rare earth elements (REE) speciation at near-neutral to alkaline pH. Part II: hydrolysis of Er from 25 to 75 °C" *Dalton Trans.*, **2025**, 54, 5153–5163, <https://doi.org/10.1039/D4DT03037K>.
5. Hurtig, N.C., Gysi, A.P., Smith-Schmitz, S., Harlov, D., "Raman spectroscopic study of anhydrous and hydrous REE phosphates, oxides, and hydroxides" *Dalton Trans.*, **2024**, 53, 9964–9978, <https://doi.org/10.1039/D4DT01086H>.
6. Maciag, B. J., Gysi, A. P., Hurtig, N. C., "Quantification of the REE³⁺ aqua ions and chloride species in aqueous fluids by in situ Raman spectroscopy using perturbations of the water band" *Chem. Geol.*, **2025**, 122684, <https://doi.org/10.1016/j.chemgeo.2025.122684>.
7. Padilla, K., Gysi, A.P. "The solubility of La hydroxide and stability of La³⁺ and La hydroxyl complexes at acidic to mildly acidic pH from 25 to 250 °C" *Geochim. Cosmochim. Acta*, **2025**, 406, 175–188. <https://doi.org/10.1016/j.gca.2024.12.006>
8. Pan, R., Gysi, A.P., Miron, G.D., Zhu, C. "Optimized thermodynamic properties of REE aqueous species (REE³⁺ and REEOH²⁺) and experimental database for modeling the solubility of REE phosphate minerals (monazite, xenotime, and rhabdophane) from 25 to 300 °C" *Chem. Geol.*, **2024**, 643, 121817. <https://doi.org/10.1016/j.chemgeo.2023.121817>
9. Smith-Schmitz, S. E., Hurtig, N. C., Gysi, A. P. "In situ Raman investigation of Dy complexation in Cl-bearing aqueous solutions at 20–300 °C", *Dalton Trans.*, **2025**, 54, 4286–4302. <https://doi.org/10.1039/D4DT02170C>

Uncovering Physical Drivers Behind Graphic Structure within Nano-Seismic Catalogs

Jesse Hampton, Principal Investigator

Geological Engineering, Civil Engineering, University of Wisconsin-Madison, Madison, WI

Graduate Students: Ellie Johnson, Ana Paula Villaquiran Vargas

Overall research goals:

The long-term goal of this Early Career Research Program (ECRP) project is to establish a physics-based framework for interpreting nano-seismicity catalogs as evolving space-time networks that encode subsurface stress, fracture geometry, and failure state. Rather than treating acoustic-emission and microseismic events as independent points in time and space, this project seeks to understand how their topological organization—specifically triggering-triggered network structure and its evolution—reveals the physical drivers of fracture nucleation, propagation, and coalescence in geologic materials.

By combining large-scale polyaxial laboratory experiments, distributed fiber-optic strain sensing, and advanced graph-theoretic and machine-learning methods, this research aims to link seismic network topology to stress, pore pressure, and fracture state. This capability is foundational for predicting and mitigating induced seismicity and for monitoring fracture development in Enhanced Geothermal Systems (EGS) and geologic carbon storage.

Significant achievements during 2023- 2025:

This ECRP was awarded in 2025 and initiated in Fall 2025. Major accomplishments to date include establishment of the experimental and computational infrastructure required for the program, including preparation of the polyaxial testing system for coupled acoustic-emission and distributed fiber-optic strain measurements, development of triggering-triggered network construction pipelines, and implementation of baseline graph-theoretic and graph neural network (GNN) analysis workflows. Preliminary nano-seismicity datasets from prior laboratory experiments have been organized to support method development and benchmarking.

Science objectives for 2026-2028:

From 2026–2028, the project will execute a coordinated experimental and analytical campaign to quantify how nano-seismicity network topology reflects stress state, fracture geometry, and failure.

A first objective is to generate three classes of scale-invariant laboratory nano-seismicity catalogs under controlled conditions: (1) dry shear on a critically stressed fault, (2) fluid-driven shear on a critically stressed fault, and (3) hydraulic fracture in intact granite. These experiments will be performed in a true-triaxial apparatus using large prismatic specimens instrumented with dense acoustic-emission arrays and embedded distributed fiber-optic strain sensors, enabling direct measurement of evolving stress-strain fields alongside microseismic activity.

A second objective is to convert these catalogs into directed space-time networks using physically based triggering-triggered correlations and to identify topological signatures associated with stress orientation, fracture localization, and the transition from distributed microcracking to macroscopic rupture. Graph-theoretic metrics, persistent path homology, and assortativity will be used to quantify network structure and evolution.

A third objective is to apply graph neural networks and dynamic network change-point detection to determine whether transitions in fracture state and driving mechanism (poroelastic versus non-poroelastic) are detectable directly from seismic network topology. By integrating distributed strain measurements with nano-seismicity networks, the project will produce experimentally validated, physics-based tools for interpreting seismic catalogs as evolving graphs that encode subsurface mechanical state.

Publications supported by this project 2023-2025:

None.

In Situ Sylvite K-Ca Dating to Establish Long-term Geologic Stability of Nuclear Waste Repositories

Mark Harrison, Principal Investigator

Department of Earth, Planetary and Space Sciences, University of California, Los Angeles

Graduate Students: Linxi Lui

Collaborators: None

Overall research goals: We are developing a K-Ca thermochronometer capable of assessing the thermal and deformation histories of nuclear waste repositories are designed to isolate artefactual radioactivities from the human environment until they have largely decayed to stable or exceedingly long-lived isotopes. Depending on the nature of the waste, this could require isolation in rock for many tens of thousands of years. Evaporite deposits have physical properties that make them appealing for a secure nuclear waste repository, including a high thermal conductivity, very low permeability, and a tendency to respond to deformation by sealing cracks. The most direct way to demonstrate geologic stability is to apply geochronometric methods to minerals that comprise the repository lithologies to assess if they have remained closed systems over their lifetime. Geologic disturbances, such as recrystallization, dissolution-reprecipitation, and deformation, are expected to either partially or completely reset a geochronometer.

In situ K-Ca dating of sylvite (KCl), a relatively common mineral in evaporite deposits, permits direct imaging of the distribution of daughter $^{40}\text{Ca}^*$ relative to parent ^{40}K permitting its use to assess whether the system has been 'open' since primary mineral precipitation and, if so, when that happened and how much isotopic equilibration occurred. Since recrystallization of salt due to fluid action or deformation (e.g., due to diapirism) resets the ^{40}K - ^{40}Ca system, μ -scale, in situ, $^{40}\text{Ca}^*/^{40}\text{K}$ variations should permit assessment of the mechanism of disturbance. Furthermore, if the crystal has remained intact and lost $^{40}\text{Ca}^*$ only by diffusion, that in principle could be used to assess the thermal history of the host rock if we know the transport kinetics of Ca in KCl. In situ ion microprobe K-Ca dating was limited by the ultrahigh mass resolving power required to separate $^{40}\text{K}^+$ from $^{40}\text{Ca}^+$, which even today remains beyond available capabilities. However, development of the 'double-plus' method (i.e., measurement of Ca^{++} and K^{++}) transcends that limitation permitting virtually full transmission of those secondary ions while leaving $^{40}\text{Ca}^{++}$ free from any significant interference from $^{40}\text{K}^{++}$. The remaining requirement is fabrication of a sylvite K-Ca relative sensitivity factor (RSF) standard using a ^{44}Ca ion implanted lithium borosilicate glass spiked with ^{42}Ca for concentration calibration. The ion implanted sylvite will also be used as starting material in 1-atm thermal annealing experiments to calibrate the Ca diffusion law for KCl needed for the thermochronometric calculations.

Significant achievements during 2023-2025:

Efforts to calibrate a natural sylvite K-Ca age showed this to be unfeasible requiring synthesis of an RSF standard. We initially chose to fabricate a mixed lithium metaborate-lithium tetraborate glass (as both pure endmember compounds crystallize on quenching rather than make homogeneous glasses) with a known trace concentration of spiked Ca but found that reagent grade oxides did not meet their claimed low [Ca] specifications. We did acquire lithium metaborate which proved, via SIMS analysis to have an exceedingly low [Ca] but 'high-purity' lithium tetraborate powders acquired from three different manufacturers were all found (via SIMS analyses) to have [Ca] about two orders of magnitude higher than the lithium metaborate. We thus pursued an alternate approach of making a lithium borosilicate glass using Puratronic silica and lithium carbonate reagents together with our high purity lithium metaborate powder. Specifically, lithium silicate-lithium metaborate mixtures with 40 and 80 wt% SiO_2 , melted at 1070°C , avoid a two liquid field and can be quenched to a glass (Sastry & Hummel, 1960, J. Am. Ceram. Soc. 43, 23). A further advantage is that SiO_2 -rich glasses

are more durable than lithium borate glasses and polish well. Melting experiments using these mixtures are promising in terms of the quality of the glass produced and await analytical confirmation of their purity (i.e., ultra-low [Ca]). We have obtained ~50 mg enriched spikes of both ^{42}Ca (92.9%) and ^{44}Ca (96.2%), the former to use as the gravimetric standard in the implantation glass fluence monitor (as we will be implanting near pure ^{44}Ca), and the latter to gauge [Ca] levels in SIMS surveys of the lithium-borosilicate glasses. Fabrication of the lithium borosilicate glass to be used as our ion implantation target awaits confirmation of overall sub-ppm level [Ca] in the starting mixture.

Science objectives for 2026-2028:

Our priority in the coming year is to fabricate the ^{42}Ca -spiked lithium borosilicate glass and then ^{44}Ca ion implant it along with a 1 cm² synthetic KCl crystal to create our SIMS K-Ca relative sensitivity monitor. We can then begin in situ SIMS K-Ca dating of sylvite samples obtained from existing German (Gorleben, Morsleben) and proposed (Teutschenthal) nuclear waste repositories to assess their geologic histories and whether any meet the federal legal requirement of having remained undisturbed for the past 34 Ma. Having now found analytical conditions appropriate to obtaining meaningful diffusion profiles from our original experimental CaCl_2 -KCl couple, we will undertake long term 1 atm annealing's (50° to 300°C) to establish a diffusion law for Ca in sylvite to be used for thermochronologic applications.

Publications supported by this project 2023-2025:

Nothing to report

Geophysical Imaging of Creep Hydraulic Fracture Growth in Rocks

Reza Hedayat, Principal Investigator

Department of Civil and Environmental Engineering, Colorado School of Mines, Golden, CO

Graduate Students and Postdocs: Awais Butt, Sana Zafar, Mehrdad Imani Tilenoiei, Jun Young Song

Collaborators: Gabriel Walton (Mines); Dimitrios Papadomarkakis (National Technical University of Athens)

Overall research goals:

This project aims to improve the understanding of the seismic and aseismic microcrack processes governing delayed yet stable fracture growth under time-dependent (creep) hydraulic fracturing conditions. Fracture growth in rocks is often time-dependent and can occur even when stresses remain below the material's instantaneous strength via subcritical crack growth. In hydraulic fracturing applications, continuous pressurization under constant borehole pressure below the monotonic breakdown pressure, commonly referred to as creep injection, can induce delayed yet stable fracture propagation. The central goal is to elucidate the physics of hydraulic fracture initiation and propagation in the creep and subcritical crack growth regimes by quantifying the spatio-temporal evolution of fractures, their source mechanisms, energy budget, and resulting fracture patterns. These processes are particularly relevant to hydrocarbon and geothermal reservoirs, where fracture growth often occurs under constant or slowly varying injection pressures and may involve substantial aseismic deformation.

Significant achievements during 2023- 2025:

The motivation for this project, which started in Sept. 2025, builds on outcomes from our completed DOE Early Career project (2018–2024), which focused on identifying geophysical signatures of crack initiation and growth in natural rocks under different loading regimes. Two complementary experimental research thrusts were pursued: (1) uniaxial compression testing of rock specimens with and without pre-existing flaws, and (2) triaxial compression testing of natural rock blocks.

This first research thrust focused on investigating the micromechanics of fractures in rocks through uniaxial compression testing combined with advanced geophysical and imaging techniques, specifically acoustic emission (AE) monitoring, ultrasonic imaging, and 2-dimensional digital image correlation (2D-DIC). By examining damage processes, insights into damage localization, crack initiation, and fracturing mechanisms were gained (Imani et al., 2024, 2025). Application of the absolute sensor calibration technique to the AE response enabled us to estimate key parameters, including magnitude, source dimension, stress drop, and radiated seismic energy, associated with the fractures (Zafar et al., 2023, 2024). The second research thrust focused on enhancing the understanding of the hydraulic fracturing processes in natural rocks through triaxial compression testing, real-time AE monitoring, and ultrasonic monitoring. We investigated the impact of various factors, such as fracture propagation regimes, injection parameters, rock types, and pre-existing conditions, on the hydraulic fracture behavior (Butt et al., 2023a). Coupled analysis of passive microseismicity and active ultrasonic signal attributes enabled systematic identification of distinct hydraulic fracturing stages, including aseismic deformation, fracture initiation and fluid permeation, pressure buildup, fracture propagation, and pressure release and fluid leak-off (Butt et al., 2023b). Collectively, these results demonstrated the effectiveness of integrated active–passive monitoring for resolving both seismic and aseismic fracture processes.

We have extended this framework to laboratory hydraulic fracturing experiments that explicitly compare monotonic and creep (constant-pressure) injection schemes. Using true-triaxial tests

on Barre granite with high-viscosity fracturing fluid, we conducted coupled AE and ultrasonic monitoring to characterize fracture evolution under time-dependent loading. Constant pressure experiments exhibited longer fracture propagation times, reduced microseismic activity, and a higher proportion of stable fracture growth relative to the monotonic injection. Microseismic event distributions indicate that monotonic injection promotes localized planar fracture growth, whereas creep injection produces more tortuous, branched fracture geometries. Active ultrasonic measurements revealed pronounced velocity and energy changes preceding detectable AE activity, particularly under constant-pressure conditions, highlighting the importance of aseismic deformation, micro-cracking, and fluid infiltration during fracture initiation. The combined analysis of active and passive seismic responses provides a more complete description of hydraulic fracture evolution and confirms that creep hydraulic fracturing is characterized by more tortuous fracture geometries and reduced seismic radiation.

Science objectives for 2026-2028:

These results confirm that a significant portion of fracture growth under time-dependent loading is aseismic and cannot be fully captured using passive microseismic monitoring alone. The next phase of this project will focus on: (a) characterizing the spatio-temporal evolution, source mechanisms, and fracture patterns associated with monotonic and creep hydraulic fracturing in sedimentary and granitic rocks; (b) quantifying the energy budget, including seismic source parameters and seismic efficiency, for different hydraulic fracturing schemes; and (c) assessing the role of fluid infiltration and subcritical crack growth in controlling fracture behavior in both granitic and sedimentary rocks. The overarching hypothesis is that fracture processes under time-dependent loading are dominated by aseismic mechanisms, and that integrating active ultrasonic imaging with calibrated passive AE monitoring provides a robust framework for identifying, quantifying, and modeling the full spectrum of deformation processes governing creep hydraulic fracture growth.

Selected publications supported by this project 2023-2025:

1. Imani, M., Hedayat, A., Walton, G. "Critical State of Damage at the Onset of Acceleration Creep in Stanstead Granite Evaluated Using 2D-DIC and Percolation Theory Approach" *Rock Mechanics and Rock Engineering*, 2025, <https://doi.org/10.1007/s00603-025-05114-2>
2. Imani, M., Walton, G., Moradian, O., and Hedayat, A. "Temporal and Spatial Evolution of Non-Elastic Strain Accumulation in Stanstead Granite During Brittle Creep." *Rock Mechanics and Rock Engineering*, 2024, 58, 275-299. <https://doi.org/10.1007/s00603-024-04170-4>.
3. Butt, A., Hedayat, A., and Moradian, O. "Microseismic Monitoring of Laboratory Hydraulic Fracturing Experiments in Granitic Rocks for Different Fracture Propagation Regimes" *Rock Mechanics and Rock Engineering*, 2023a, 57, 2035-2059. <https://doi.org/10.1007/s00603-023-03669-6>.
4. Butt, A., Hedayat, A., and Moradian, O. "Laboratory investigation of hydraulic fracturing in granitic rocks using active and passive seismic monitoring." *Geophysical Journal International*, 2023b, 234 (3), 1752-1770. <https://doi.org/10.1093/gji/ggad162>.
5. Zafar, S., Hedayat, A., and Moradian, O. "Stress Dependency of Brittle Creep in Granite: Insights into Source Mechanisms and Parameters." *Rock Mechanics and Rock Engineering*, 2024, 57, 10349-10366. <https://doi.org/10.1007/s00603-024-04097-w>
6. Zafar, S., Hedayat, A., and Moradian, O. "Energy Budget of Brittle Fracturing in Granite Under Stress Relaxation and Creep." *Rock Mechanics and Rock Engineering*, 2023, 57, 921-943. <https://doi.org/10.1007/s00603-023-03593-9>.

Fundamental Controls on Capillary Trapping of scCO₂ in Geologic Storage

Anna Herring, Principal Investigator

Civil and Environmental Engineering, University of Tennessee Knoxville, TN

Graduate Students: Shukrah Adesola-Olabiyi, Christopher Allison

Collaborators: Dorte Wildenschild (OSU)

Overall research goals:

Recent research has shown that for when multiple fluids are present within a porous medium, capillary trapping of the nonwetting fluid (e.g. carbon dioxide, CO₂) can be significantly enhanced via cyclic injection schemes; i.e. multiple repeated consecutive injections of water and nonwetting phase. We aim explore the potential mechanisms of enhanced trapping during cycling -- including fluid property variation, surface chemistry alterations, and clay fines migration -- and determine how these mechanisms manifest across length scales, from the pore-scale to the core-scale. The use of CO₂ as a nonwetting fluid for these experiments provides a unique opportunity to control and manipulate fluid properties (viscosity, interfacial tension, density), due to the proximity of the CO₂ critical point to common subsurface storage reservoir temperature and pressure conditions. Exploration across fluid properties and sample types will enable delineation of potential trapping enhancement processes.

This project is a collaborative project: a broad range of “screening” core-flooding experiments are being conducted at the University of Tennessee, Knoxville (UTK); these results are analyzed to identify select conditions to undertake imaging experiments conducted at Oregon State University (OSU). Our goals are: (1) establish a robust OSU-UTK collaboration with complementary project foci; (2) conduct cyclic supercritical CO₂-brine injection experiments under a range of pressure and temperature conditions to determine saturation and pressure trends during cycling in three sample types (quarry sandstone, synthetic, reservoir sandstone); and (3) quantify changes in surface roughness and water quality effluent to observe potential surface alterations and fines migration. The results from UTK-based experiments will inform conditions to target to high-resolution imaging studies to be undertaken at OSU. Ultimately, our collaborative experimental results will inform subsurface fluid storage applications and benefit design of optimal fluid injection/withdrawal strategies for subsurface fluid and energy storage schemes.

Significant achievements during 2023- 2025:

The project was initiated in Fall 2024. Two UTK-based Graduate Research Assistants (Adesola-Olabiyi and Allison) have worked together at UTK to conduct multiple sets of supercritical experiments at pressure-temperature conditions ranging from 20-40°C and 1250-1800 PSI for quarry sandstone. Saturation trends and pressure measurements were recorded and presented via poster presentation by GRA Adesola at the International Society for Porous Media (Interpore) Annual Conference in Albuquerque, NM, May 19-22, 2025, with additional presentations anticipated at the Gordon Research Conference on Flow and Transport in Permeable Media in July 2026 in Switzerland. Samples have been acquired for Diffuse Reflectance Infrared Fourier Transform (DRIFTS) spectral analysis, water quality, and roughness quantification; these analyses are ongoing. The OSU and UTK teams have developed a strong collaborative group, with multiple visits (UTK to OSU, and vice-versa) for experimental runs during 2025.

In completed core-flooding experiments, we observed a significant deviation in cyclic trapping behavior at injection conditions nearest to the supercritical point of CO₂, which was coupled with a repeatable, unstable pressure differential across the core at these conditions. Injection pressure

differential increases for subsequent drainages after the primary drainage, indicating that trapping during cyclic injections is not well-characterized via the conventional Land trapping curve.

DRIFTS measurements show distinct spectral features when comparing pre- and post-core flooding. Post-flood measurements suggest the presence of adsorbed CO₂ or carbonate species, along with minor shifts and intensity reductions in kaolinite-associated O-H and Al-OH bonds, indicative of interactions with clay minerals.

Science objectives for 2026-2028:

Key future objectives include expanding the range of pressure-temperature conditions and samples types for analysis. Comparison of quantitative data during cyclic injections for samples with varying levels of clays and initial surface characteristics, and across the six pressure-temperature conditions, will enable determination of the mechanisms underlying enhanced trapping. Specifically, we will compare:

- Differential pressure and saturation response during cyclic injections of CO₂ and water
- Pre- and post-flooding DRIFTS spectral maps of core inlets and outlets to observe mineralogical modifications
- Microscopy and BET isotherm quantification of granular surface areas to identify minute solid dissolution resulting in alterations in surface roughness
- Water quality analysis to identify mobilization elements indicative of clay fines migration

Experiments with exceptional behavior (i.e. significant increases in capillary trapping throughout cycling) will be identified for high-resolution X-ray micro-Computed Tomography (μCT) imaging experiments at OSU to enable direct observation of pore-to-core scale expressions of these mechanisms, including via non-wetting phase topology and distribution.

Publications supported by this project 2023-2025:

Since project initiation in Fall 2024, we have not yet published results. We do however have multiple publications in preparation for submission:

1. Christopher Allison, Jaydeep Kolape, Anna Herring, "Bentheimer Mineral Surface Alteration by CO₂-Brine Solutions Under Storage Conditions." *In preparation for submission to Chemical Geology.*
2. Christopher Allison, Shukrah A-Olabiyi, Anna Herring, "The Effects of Supercritical Phase Compressibility on Geologic Storage Technology", *In preparation for submission to Energy and Fuels.*
3. Shukrah A-Olabiyi, Christopher Allison, Daniel Enebe, Dorthe Wildenschild, Anna Herring, "Evolution of CO₂ Saturation under Multicycle Drainage-Imbibition on Bentheimer Sandstone." *In preparation for submission to International Journal of Greenhouse Gas Control.*
4. Shukrah A-Olabiyi, Christopher Allison, Anna Herring, "Impact of Dynamic Flow on Fines Mobilization and Mineral Alteration in CO₂-Flooded Core-Scale Bentheimer" *In preparation for submission to Chemical Geology.*

Project LISTEN-CLOSE
Ben Holtzman, Principal Investigator

Earth Resources Lab, Massachusetts Institute of Technology, Cambridge, MA
Lamont Doherty Earth Observatory, Columbia University, New York, NY

Postdocs: Eric Beaucé (LDEO)

Collaborators: Hoagy O’Ghaffari (MIT), Matej Pec (MIT), Ulrich Mok (MIT) Tushar Mittal (Penn State), Kangan Li (Penn State University), Brian Bonner (LBNL), André Chrysochoos, Loic Daridon (Univ. Montpellier, France)

Overall research goals:

This project, entitled “LISTEN-CLOSE” was funded initially by BES grant “Towards machine-learning a fully-coupled constitutive model for thermal- hydraulic fracture in geothermal systems: phase I [DE-SC-0024678],” and now by “Laboratory-driven, machine- learned constitutive models for fracture network evolution in the deep crust, with applications to thermal energy and critical metals extraction [DE-SC-0026145]“. The project addresses challenges in understanding and controlling subsurface fracture networks, which are crucial for applications like deep geothermal heat mining and deep-crustal minerals/metals/hydrogen extraction. The research focuses on advancing the understanding of coupled thermal-hydro-mechanical-chemical (THMC) processes in geologic materials, particularly under the high temperature and pressure conditions found in the deep crust. This seed grant focused specifically on thermal cracking and the development of new constitutive models for THMC using continuum thermodynamics and machine learning.

Significant achievements during 2023- 2025:

To study micro-scale fracture formation, the seed project demonstrated the ability to create thermal cracking under stress in granite samples using a Paterson Gas-medium Deformation Apparatus. A primary accomplishment was the application of novel high-temperature, near-field “needle” sensors made to record acoustic emissions (AEs) during these experiments. A high-resolution event detection methodology based on matched filtering was applied to continuous acoustic recordings, over 270,000 acoustic events in a single experiment, providing new resolution into the internal deformation processes. Thermal cracking tends to produce random, unclustered acoustic emissions, that, as stress increases, become more clustered as the sample approaches rupture. The results suggest a competition between increasing temperature and stress influences the patterns of energy dissipation as the samples enter the brittle-ductile transition. Statistical approaches from earthquake seismology applied to AEs indicate increased temporal clustering leading up to failure. Unsupervised machine learning methods were also successfully applied to extract diagnostic spectral features from these acoustic signals, which aims to characterize different cracking regimes and material states.

The project also made significant progress in developing novel constitutive models that satisfy the laws of thermodynamics, in the context of reversible (elastic) and irreversible (viscous/ plastic) processes. The core concept for a novel neural network architecture to learn these constitutive models from data was validated on multiple non-linear viscoelastic models. This physics-informed machine learning approach is designed to discover the underlying thermodynamic potentials that describe reversible and irreversible processes in deforming solids.

Overall, the seed grant successfully validated the proposed integration of novel laboratory experiments, advanced acoustic analysis, and thermodynamics-informed neural networks (TIP-INN) for discovery of constitutive models to characterize fracture network evolution.

Science objectives for 2026-2028:

We will be continuing the three main aspects of this work: 1) thermal cracking experiments with near-field acoustic sensing, for example, with exploration of effects of confining pressure on cracking rate; 2) extensive machine learning analysis of the acoustic data from the high-temperature sensors (AEs and continuous recordings) and micro-structural characterization of fracture networks; 3) constitutive model development through the application of forward models based in thermodynamics of irreversible processes, and inverse modeling with the TIP-INN (which at present, does not incorporate the thermal aspects of the free energy, but will soon). The key outcome of this research will be the development of strongly coupled mechanical laws describing the effective properties of evolving micro-fracture networks, that can be directly incorporated into large-scale reservoir models used for deep heat mining and resource extraction.

Publications (partly in prep.) supported by this project 2023-2025:

1. Benjamin Holtzman, Hoagy O’Ghaffari, Eric Beaucé, Tushar Mittal, Anna Barth, Ulrich Mok, and Matěj Peč, “Towards multi-scale coupled thermo-hydro-mechanical constitutive models: Experiments on thermal cracking under stress with near-field acoustic data.” *Stanford Geothermal Workshop Proceedings, 2025*, <https://pangea.stanford.edu/ERE/pdf/IGAstandard/SGW/2025/Holtzman.pdf>
2. Hoagy O’Ghaffari, Benjamin Holtzman, Ulrich Mok, Matěj Peč, “High-temperature near-field acoustic sensing of thermal cracking in granite.” *to be submitted: Journal of Geophysical Research: Solid Earth*.
3. Benjamin Holtzman, Eric Beaucé, Tushar Mittal, Anna Barth, Hoagy O’Ghaffari, Matěj Peč, Brian Bonner, “Experiment study of thermal cracking under stress with near-field acoustic data” *to be submitted: Journal of Geophysical Research: Solid Earth*.
4. Kangan Li, Tushar Mittal, Benjamin Holtzman, Seth Saltiel, Loic Daridon, and André Chrysochoos, “Thermodynamics-informed neural networks for the rheology of rocks: A data-driven approach to learning complex constitutive models.” *to be submitted: Journal of Geophysical Research: Machine Learning and Computation*.

Serpentinization under Anisotropic Stress

Mengsu Hu, Thrust Lead; Ben Gilbert, Principal Investigator

Energy Geosciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA

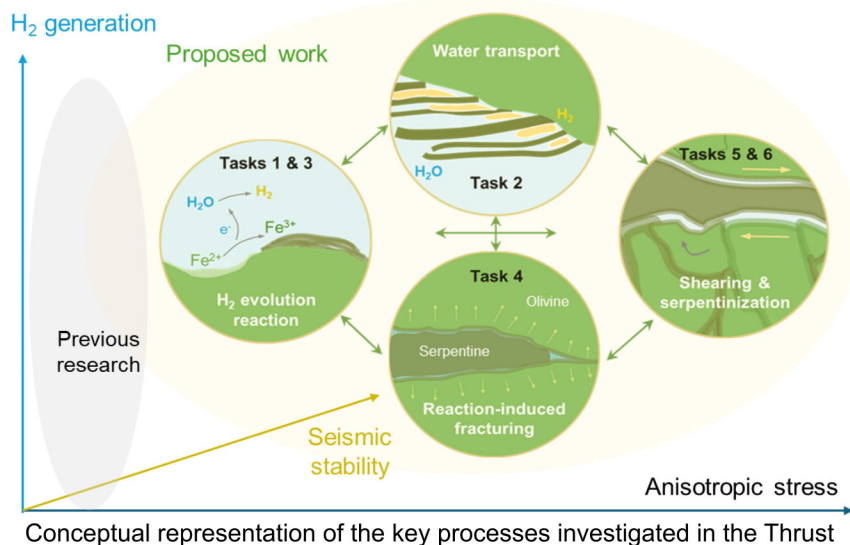
Co-PIs: Jill Banfield (UC Berkeley), John Christensen (LBNL), Abdullah Cihan (LBNL), Don DePaolo (LBNL), Wenming Dong (LBNL), Ben Gilbert (LBNL), Zhao Hao (LBNL), Harry Lisabeth (LBNL), Michael Manga (UC Berkeley), Seiji Nakagawa (LBNL), Carl Steefel (LBNL), Daniel Stolper (UC Berkeley), Drew Syverson (University of North Carolina at Charlotte), Christophe Tournassat (LBNL and University of Orléans), Yuxin Wu (LBNL), and Piotr Zarzycki (LBNL)

Graduate Students: Yarong Qi, (PhD candidate, UC Berkeley), Siqin Yu (PhD intern, Stanford University), Serhat Sevgen (PhD intern, University of Calgary)

Overall research goals: Serpentinization of Mg-Fe(II)-bearing ultramafic rocks is an important mechanism by which large quantities of geologic hydrogen can be produced. This process involves a highly complex and only partly understood set of coupled chemical-mechanical (CM) processes. **Serpentinization under Anisotropic Stress** is a new thrust within the LBNL BES program **Chemical-Mechanical Controls on Subsurface Resources**, funded for FY26–FY28. The overall research goal is to determine the chemical-mechanical controls on serpentinization and associated seismic stability across a range of stress and temperature regimes. *We hypothesize that anisotropic stress promotes the fastest serpentinization rates, resulting in the highest hydrogen generation rates and yields while simultaneously increasing the likelihood of reaction-induced fracturing and modifying fault frictional behavior.*

A high-level overview

We conduct a suite of tightly integrated Tasks informed by shared laboratory, field, and modeling crosscuts. These Tasks aim to elucidate the serpentinization reaction pathway, water transport, isotopic signatures, reaction-induced fracturing, and the coupled interplay between shearing and serpentinization.



The research integrates molecular and nanoscale experimental and simulation studies of mineral and redox transformations with mesoscale investigations on the coupled processes affecting fracturing, flow, shearing, and overall hydrogen production. Laboratory experiments are conducted at high temperatures using batch and flow-through reactors including gold-bag systems. Several of the tasks use natural samples from California in experiments or characterize natural samples

over a range of scales.

Significant achievements during 2023- 2025:

The Thrust was initiated in FY25. The following are the highlighted achievements:

- Optimization of solution chemistry and catalysis on serpentinization and H₂ generation rates (see poster by Gilbert, Syverson, et al.)

- A new chemo-mechanical rate law applicable to incongruent reactions that unifies the description of pressure solution and stress-mediated crystallization to elucidate stress controls on serpentinization (see poster by Steefel and Hu)
- Unraveling the mechanism of residual trapping governed by the interplay between the pore geometry and injection flow rate (Yu, Hu*, Steefel, Battiato, 2024, ADWR)
- The first fully coupled nanocapillary filling model, resolving long-standing debates on interfacial and electrokinetic effects and uncovering a new high-temperature, high-pressure regime where classical capillary scaling fails (see poster by Cihan, et al.)
- Measured isotope effects associated with H₂ production from iron-bearing minerals.
- Documentation of natural evidence of serpentinization, fracture growth and stress history via micro-CT imaging at LBL Advanced Light Source.

Science objectives for 2026-2028:

- To establish the role and mechanisms of iron redox reactions associated with silicate minerals in generating hydrogen and the potential for redox active iron in silicate minerals supporting subsurface microbial metabolism that cycles hydrogen and iron through pathways including anaerobic methane oxidation
- To determine the regimes under which the rate of serpentinization is limited by water transport through nano-capillary pumping and/or microscale channelized flow during serpentinization
- To develop isotopic and geochemical methods to constrain the stress-mediated reaction rates at selected field sites.
- To determine the effects of anisotropic stress on the crystallization rate of serpentine and reaction-induced fracturing
- To understand how serpentinization affects seismic stability and how shearing affects the serpentinization rate in actively serpentinizing fault interfaces and granular structures
- To identify the field sites that can provide suitable rock, fluid and microbiological samples, and the field data that records natural spatiotemporal chemical-mechanical alteration.

Publications supported by this project 2023-2025:

1. Siqin Yu, Mengsu Hu, Carl I. Steefel, and Ilenia Battiato, "Unraveling residual trapping for geologic hydrogen storage and production using pore-scale modeling." *Advances in Water Resources* 2024, 185, 104659.
2. Mengsu Hu, Carl I. Steefel, Jonny Rutqvist, and Benjamin Gilbert. "Microscale THMC modeling of pressure solution in salt rock: impacts of geometry and temperature." *Rock Mechanics and Rock Engineering* 2023, 56, 7071-7089.
3. Abdullah Cihan, Ben Gilbert, Pramod Bhuvankar, Mengsu Hu, Carl Steefel. "Interfacial–electrokinetic control and evaporation–driven pinning of nanocapillary filling from ambient to serpentinization temperatures." Submitted to Communications Physics 2026.
4. Abdullah Cihan, Piotr Zarzycki, Zhao Hao. "Surface Hydroxylation-Induced Electrostatic Forces Thicken Water Films on Quartz." *Langmuir* 2024, 40(29), 15099-15106.
5. Youzheng Qi and Yuxin Wu. "Impedance Spectroscopy of the Mineral–Electrolyte Interfaces." *Reviews in Mineralogy and Geochemistry* 2025, 91(1), 85–104.
6. Youzheng Qi and Yuxin Wu. "Revisiting the relationship between induced polarization and surface conductivity: Ratios from laboratory to field." *Journal of Geophysical Research: Solid Earth* 2025, 130(4), e2024JB030406.
7. Youzheng Qi and Yuxin Wu. "Induced polarization of clayey rocks and soils: Non-linear complex conductivity models." *Journal of Geophysical Research: Solid Earth* 2024, 129(3), e2023JB028405.

Direct 3D X-ray Probes of Rock Texture, Structure, and Stress Heterogeneity, and Digital Twins to Study Their Effects

Ryan Hurley, Principal Investigator

Department of Mechanical Engineering, Johns Hopkins University, Baltimore, MD

Graduate Students: Subham Bose (Civil and Systems Engineering, Johns Hopkins University)

Collaborators: Jun-Sang Park (APS), Peter Kenesei (APS)

Overall research goals:

The goal of this research project is to determine the competing roles that crystallographic texture, microstructure, and initial grain stresses and their evolving heterogeneity play in controlling rock fracture in sandstone. To achieve this goal, we perform synchrotron experiments pushing the frontiers of miniature rock deformation testing with in-situ X-ray tomography (XRCT) and diffraction microscopy (near-field or nf-HEDM, far-field or ff-HEDM, and point-focused or pf-ff-HEDM). Our objective (beginning in 2025) is also to build, validate, and use digital twins of rock specimens to parametrically study the competing roles of texture, structure, and initial stresses on fracture.

Significant achievements during 2023- 2025:

Achievement 1: We demonstrated the first successful application of both XRCT and ff-HEDM to Berea and Nugget sandstones¹ (see Fig. 1). The samples, measuring around 1 mm, were stressed to failure in unconfined uniaxial compression at the CHESS FAST 3a beamline. Our study revealed sample failure through progressive inter-granular cement bridge destruction, i.e., opening or shear failure of pre-existing defects at the micron scale (Fig. 1(c)). Through statistical analysis of ff-HEDM measurements, our study revealed increasing vertical alignment of principal compressive stresses throughout both samples during the uniaxial compression experiments (Fig. 1(d)). Heterogeneity in principal stresses remained significant and increased upon application of vertical load (Fig. 1(d)).

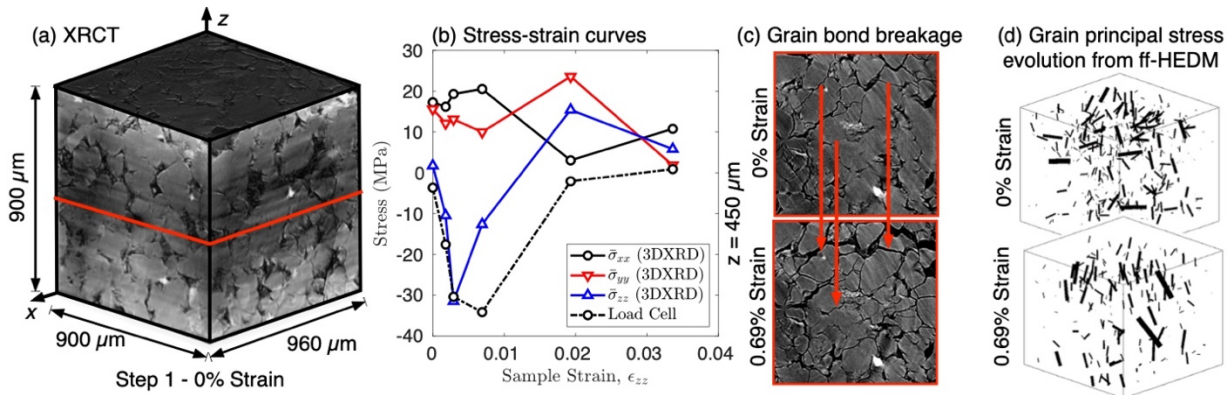


Figure 1 - Select results from first application of XRCT and nf-HEDM to sandstone.¹ (a) An XRCT image of Nugget sandstone imaged during a beamtime at CHESS. (b) Stress-strain curves for the sample, one curve from the load cell and one from ff-HEDM (also called 3DXRD). (c) A horizontal cross-section of the sample highlighted in red in (a) illustrating sample failure through grain bond breakage. Arrows aid the eye in seeing bonds which open dramatically during sample failure. (d) Principal compressive stresses, scaled linearly in length and thickness by magnitude, for each grain analysed with ff-HEDM. The box represents the sample extents shown in (a).

Achievement 2: We demonstrated the first known application of nf-HEDM, XRCT, and ff-HEDM to a sandstone^{2,3} (see Fig. 2 for select results). The sample, measuring around 1 mm³, was stressed to failure in unconfined uniaxial compression at the APS 1-ID beamline. nf-HEDM, processed with a novel combination of ML-based image segmentation and forward diffraction modeling, revealed 3D crystal texture of the sandstone for the first time, demonstrating that grain overgrowths and

misorientations vary widely even over very short distances (Fig. 2(b)). XRCT, processed with a combination of ML-based image segmentation and DVC, revealed fracture networks suggesting the emergence of vertically-aligned load bearing structures. ff-HEDM revealed the emergence of force-chain-like loading bearing structures hypothesized to exist in rocks but never, to the best of our knowledge, demonstrated to exist as in our study (Fig. 2(d)).

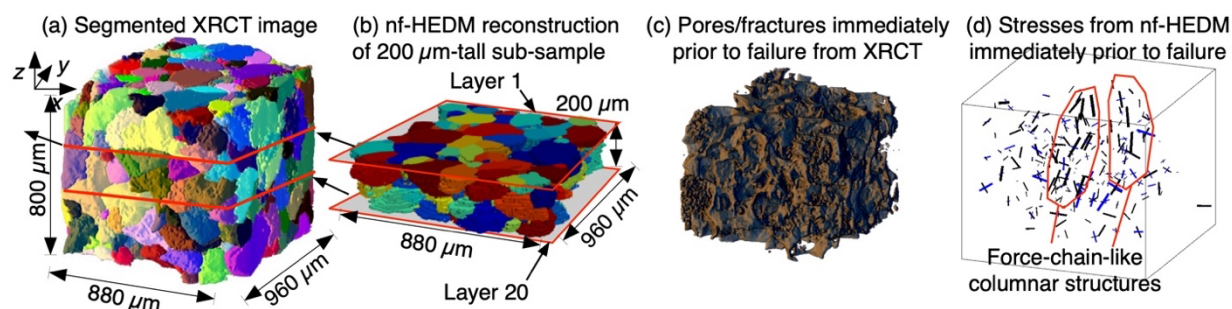


Figure 2 - Select results from combined application of XRCT, nf-HEDM, and ff-HEDM.² (a) Segmented XRCT image of sample of Nugget sandstone. (b) nf-HEDM reconstruction of a 200 μm tall section of the sample shown in (a). (c) Stress-strain curve for the sample. (d) Principal compressive (black) and tensile (blue) stresses, scaled linearly in length and thickness by magnitude, for each grain analysed with ff-HEDM. The box is 1.2 x 1.2 mm.

Science objectives for 2026-2028:

In the next several years, our overarching scientific objective is to build a deeper understanding of the competing effects of crystal texture, microstructure, and initial stresses on fracture in sandstones. Crystal texture, microstructure, and initial stresses are known to influence wave propagation, stress concentrations, and fracture nucleation, but their competing roles are not fully understood. We will perform targeted rock deformation experiments at high pressures using a custom rig at APS 1-ID and 20-ID while using simultaneous in-situ XRCT, nf-, ff-, and pf-ff-HEDM to reveal texture, structure, and initial and evolving stresses during loading. The triaxial rig permits 200 MPa confining pressures and has been used extensively in laboratory XRCT experiments. We will also develop, validate (using X-ray measurements), and use digital twins of rock specimens in the finite element method to parametrically study the influence of length scale, loading conditions, and initial texture, structure, and grain stress on failure susceptibility. We plan to perform experiments on rocks with variable porosity and grain sizes, including but not limited to Berea Gray (21% porosity, 175-micron grain size), Bandera Brown (20% porosity, 130-micron grain size, and Nugget (11% porosity, 150-micron grain size). Digital twin modeling will extend preliminary work described in recent work.²

Publications supported by this project 2023-2025:

1. Ryan C. Hurley, Ghassan Shahin, "In-Situ X-ray Measurements of Grain Stress and Strain During Uniaxial Compaction of Miniature Sandstone Specimens" 57th U.S. Rock Mechanics/Geomechanics Symposium, Atlanta, Georgia, USA, June 2023, ARMA-2023-0958. <https://doi.org/10.56952/ARMA-2023-0958>
2. Ryan C. Hurley, Ye Tian, Jun-Sang Park, Peter Kenesei, Hemant Sharma, Adyota Gupta, Kwangmin Lee, "Crystallographic texture, structure, and stress transmission in Nugget sandstone examined with X-ray tomography and diffraction microscopy" *Journal of Geophysical Research: Solid Earth*, **2025**, 130(7), e2025JB031690. <https://doi.org/10.1029/2025JB031690>
3. Subham Bose, Mohmad Thakur, Ghassan Shahin, Katherine Shanks, Amlan Das, Jun-Sang Park, Peter Kenesei, Hemant Sharma, Ryan C. Hurley, "Stress and strain heterogeneity and persistence in uniaxially and triaxially-loaded sandstone" *ESS Open Archive (and in review)*. <https://doi.org/10.22541/essoar.176280047.73950008/v1>

Molecular Interactions in Geologic Nanopores

Anastasia G. Ilgen, Lead Principal Investigator

Geothermal Research Department, Sandia National Laboratories, Albuquerque, NM

Co-Principal Investigators: Kevin Leung (SNL), Jeffery Greathouse (SNL), Tuan Ho (SNL)

Postdoctoral Fellows: Hasini Senanayake (current, SNL), Anthony Baldo (former, SNL), Bidemi Fashina (former, SNL)

External Collaborator: Ward Thompson (Kansas University)

Nanoconfined chemical environments, also referred to as nanopores, are prevalent in geologic systems, including soils and sedimentary rocks. The large surface-to-volume ratios in these environments can influence the behavior of chemical species. Specifically, the presence of solid surfaces affects the properties of nanoconfined water and aqueous solutions, including ion solvation and the thermodynamics of chemical reactions within nanopores. To advance this important research field, our work combines theory, modeling, and experiments to identify molecular-scale processes in geologic nanopores filled with aqueous solutions. We focus on the effects of nanoconfinement on solvent structure and dynamics, solvation properties, and the state of chemical equilibria in nanopores. Additionally, we research how dielectric response governs ion solvation in relation to ion concentrations. We aim to introduce conceptual advances such as activity coefficients under confinement, descriptors of electrostatic environments that explain dielectric properties, and short-range hydration forces and ion-ion correlations that control oriented attachment of nanoparticles. A distinguishing characteristic of this research program is its emphasis on both short- and long-range interactions and solvent dynamics that impact chemistry at the nanoscale, which requires that we consider both pore size and interfacial effects. This knowledge will enable predictions of chemical transformations relevant to evolving geologic and engineered systems and extending to applications in chemical separations and water purification.

During the 2023-2025 cycle, our team addressed three major research tasks: the structure and dynamics of water and solutes in nanopores, force field (FF) development for mineral-water interactions, and molecular-scale controls on particle-particle interactions.

In the first task we investigated the dielectric response of electrolytes in confined nano-slits, revealing a strong dependence on electrolyte properties and boundary conditions. We also quantified hydrogen bonding interactions in nanopores and found evidence for two co-existing water populations (near- and far- from the silica surface). Our classical molecular dynamics (CMD) simulations indicated that the equilibrium dielectric response of confined electrolytes increases towards infinity as salt concentration rises, aligning with broad-band dielectric measurements. This research has implications for reinterpreting atomic force microscopy measurements in electrolytes and free energy calculations performed using *ab initio* codes.

To understand how nanoconfinement may impact chemical speciation inside nanopores and the state of chemical equilibria, we studied (de)protonation reactions of carboxylic acids and the formation of ion pairs in cylindrical silica nanopores using Raman spectroscopy and MD simulations. We found that the acidity of carboxylic acids and bicarbonate ions decreases in silica nanopores with negative surface charge, while ion pairing of di- and tri-valent cations with acetate anion becomes more favorable. These findings indicate that equilibrium shifts towards complexes in nanopores. In a separate study investigating the influence of ion pairing on ion and counterion retention in neutral nanopores of gibbsite and kaolinite, we found that the Na–Cl pair behaves as an effective dipole. In gibbsite nanopores, this dipole aligns with the local water orientation, resulting in a more favorable free energy than that of the ion pair in bulk solution. In contrast, the more hydrophobic kaolinite nanopore disfavors the partitioning of all species, including Na⁺, Cl[–], and the Na–Cl ion pair. This raises questions about whether the observed changes in equilibrium state are due to variations in

the free energy of reactions or changes in the activities of chemical species due to nanoconfinement. In the upcoming FY26-28 cycle, we aim to resolve this question and develop new thermodynamic knowledge about chemical factors governing reactions in nanopores.

In the second task, a significant effort was dedicated to the development of new FF parameters for accurate simulations of interfacial systems. We developed a flexible silica FF (Silica-DDEC) for CMD simulations of amorphous and crystalline silica, utilizing density functional theory (DFT)-based atomic charges derived from periodic DFT calculations. This new FF accurately predicts the experimental bulk density of silica as well as the DFT-predicted electrostatic environment at the silica surface. Follow-on work resulted in improved Lennard-Jones parameters for nonbonded for silica-water and silica-CO₂ interactions. These methods will be used to investigate nanoconfinement effects on solvent properties in the FY26-28 cycle.

In the third task, we explored the energetics of particle-particle interactions during approaching, rotating, and sliding motions of gibbsite. We detailed the roles of atomistic mismatch, hydrogen bonding, and properties of confined water on the oriented attachment (OA) of particles with no surface charge. Our findings indicate that the highest energy barrier for OA is associated with removing the final water layer to enable a jump to contact, with mutual rotation being more probable than sliding motion. Additionally, we examined the role of hydrogen bonds in gibbsite particle attachment, revealing that changes in the surface-water and water-water hydrogen bonding networks are strongly coupled with the free energy profile during translation. These results highlight the importance of particle-slab misalignment and hydrogen bonding networks in the OA of mineral particles in aqueous solutions, informing our planned experimental measurements of hydrogen bond strength in nanoconfined systems in the FY26-28 cycle.

Selected publications supported by this project 2023-2025:

1. Sahan Godahewa, Thanuja Jayawardena, Ward Thompson, and Jeffery A. Greathouse, Accurate Force Field for Carbon Dioxide-Silica Interactions Based on Density Functional Theory. *J. Phys. Chem. B* **2025**, DOI 10.1021/acs.jpcc.4c07413.
2. Thanuja Jayawardena, Sahan Godahewa, Ward Thompson, and Jeffery A. Greathouse, Beyond Force Field Mixing Rules to Model Silica-Water Interfaces. *J. Phys. Chem. C* **2025**, DOI 10.1021/acs.jpcc.5c02442
3. Tuan A. Ho, Influence of Ion Pairing on Ion and Counterion Retention in Neutral Nanopores of *Gibbsite and Kaolinite*, *Nanoscale*, **2025**, *17*, 19666-19674. DOI 10.1039/D5NR01746G
4. Bidemi Fashina, Anthony Baldo, Heath Watts, Kevin Leung, James Kubicki, and Anastasia G. Ilgen, Enhanced ion pairing of calcium, zinc, and lanthanum with acetate in silica nanopores. *Micro. Mesoporous Mater.* **2025**, DOI 10.1016/j.micromeso.2025.113538
5. Anthony P. Baldo, Anastasia G. Ilgen, and Kevin Leung, Deprotonation of formic, acetic acids and bicarbonate ion in slit silica nanopores at infinite dilution and in the presence of electrolytes. *J. Coll. Interface Sci.* **2024**, DOI 10.1016/j.jcis.2024.05.160
6. Kevin Leung, Finding Infinities in Nanoconfined Geothermal Electrolyte Static Dielectric Properties and Implications on Ion Adsorption/Pairing. *Nano Lett.* **2023**, DOI 10.1021/acs.nanolett.3c01865
7. Anastasia G. Ilgen, Hasini Senanayake, Ward Thompson, and Jeffery A. Greathouse, Structure and energetics of hydrogen bonding networks in dilute HOD/H₂O solutions confined in silica nanopores *Environ. Sci. Nano*, **2023**, DOI 10.1039/D3EN00291H
8. Hasini Senanayake, Pubudu Wimalasiri, Sahan Godahewa, Ward Thompson, and Jeffery A. Greathouse, Ab Initio-Derived Force Field for Amorphous Silica Interfaces for Use in Molecular Dynamics Simulations. *J. Phys. Chem. C* **2023**, DOI 10.1021/acs.jpcc.3c02270
9. Tuan A. Ho, Kevin M. Rosso, and Louise Criscenti, Atomistic Mismatch Defines Energy-Structure Relationships during Oriented Attachment of Nanoparticles, *J. Phys. Chem. Letter*, **2022** DOI 10.1021/acs.jpclett.2c02511

Learning Fault Slip Physics and Failure

Christopher W Johnson, Principal Investigator

Earth and Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM

Graduate Students: Keegan Davis, Laura Laurenti, Kenzi Meni, Lucas Tiede; Postdoctoral

Researchers: Ryley Hill, Pablo Laura, Michal Chamarcruk, Kun Wang

Collaborators: Paul Johnson (LANL), Andrew Deloey (LANL), Robert Guyer (LANL), Richard Alfaro-Diaz (LANL); leveraging, not funded Esteban Rougier (LANL), Chris Marone (Sapienza), Harsha Bhat (ENS)

Overall research goals:

The key problem we address is identifying and characterizing undiscovered or poorly constrained aspects of fault physics and the earthquake cycle, including when a fault is critically stressed. Our hypothesis is that the continuous seismic signal is a high-fidelity proxy for fault physical and material property changes. There are still many unknowns in this frontier of science. The central focus is to establish the presence, types, and significance of signals discernable with deep learning techniques, and their physical meaning regarding faulting and stress accumulation in the crust. The enormous challenge is applications to seismogenic faults—the ones that host large magnitude earthquakes and produce damaging ground shaking. The laboratory-to-earth application has produced enormous success with models inspired by laboratory results successfully applied to different tectonic regions.

Significant achievements during 2023- 2025:

- Applied laboratory inspired AI models to predict fault slip on moderate sized earthquakes (~M5.5) associated with the 2018 caldera collapse at Kīlauea volcano, Hawaii. This is a major success as it shows for the first time that the general approach developed works for seismogenic faults.
- Completed study demonstrating the ability of audio foundation models for their application to geophysical time-series data that is analogous to the complex patterns in recorded speech. This is a critical step in developing more advanced waveform foundation models.
- Completed study investigating the characteristics of the data used with the machine learning models to understand the dynamics of the mechanical system. This understanding allows more informed decision making when developing models to translate continuous seismic waveforms to physics informed phenomenon.
- Completed study in the laboratory that reproduces numerical simulation results studying the ability of determining the permeability from the seismic noise. The machine learning modeling shows that information about permeability can be extracted directly from continuous recordings.
- Completed numerical study on nonlinear grain interactions and developed software package to facilitate rapid generation of numerical mesh for grain-to-grain interaction using laboratory derived images of sample composition.
- Co-organized the Kaggle Competition with the director of the Center for Information & Systems Engineering that focuses on AI at Boston University. The competition brought together leading scientists in geophysics and computer engineering for machine learning applications applied to advancing earthquake physics. The event was a forum for 8 leading geoscientist to present cutting edge research to a diverse group of computer engineers to develop interdisciplinary collaborations that will advance DOE OBES initiatives.
- Co-organized the 2024 and 2025 conference “Machine Learning in Solid Earth Geophysics” in Santa Fe, NM sponsored by LANL Center for Nonlinear Studies and SC-BES. The conference has >80 participants with 40 speakers total ranging from PhD candidates to senior scientists with AI experts from Caltech, Uni. of Perugia (Italy), SNL, Stanford, Sapienza Uni (Italy), Uni of Edinburgh (UK), Uni of Texas, Kyoto Uni (Japan), and 2 DOE-BES SBIR funded startups.

Science objectives for 2026-2028:

- Obj. 1: Develop a seismic foundation model applicable to analyzing continuous waveform data. This effort will assess different foundation model designs to test key metrics when developing a functioning model specific to continuous seismic waveform analysis.
- Obj. 2: Investigate mechanistic processes that control faulting throughout the continuous seismic cycle. The primary focus here is analyzing the continuous wavefield with the aim of developing a complete understanding of fault zone deformation during the full seismic cycle and determine if a preparation phase is detectable in the seismic waveforms
- Obj. 3: Measure time dependent nonlinear elastic material behavior in a fault zone. Producing nonlinear elastic measurements from continuous waveforms in fault zones will allow us to investigate direct links of the evolution of fractured material in a fault zone.
- Obj. 4: Quantify permeable fracture networks that control time dependent rheological changes in the crust. Separating nonlinear elastic processes (pressure, fracture density, and fluids) that are producing a material velocity change will be investigated through numerical simulations for different grain configurations, fracture networks, pore space, and fluid pressure in the void space.

Publications supported by this project 2023-2025:

1. Hill, R.G., R.A. Guyer, P.A. Johnson, C.W. Johnson, "Material-defined two-dimensional numerical model for grain-scale nonlinear elasticity", *The Journal of the Acoustical Society of America*, **2025**, 158, 4985-4993. <https://doi.org/10.1121/10.0034637>
2. Hill, R.G., K.S. Davis, C.W. Johnson, "Grain2mesh: A Python and cubit mesh generator from unprocessed mesoscale images", *SoftwareX*, **2025**, <https://doi.org/10.1016/j.softx.2025.102427>
3. Johnson, C. W. and ., Donahue, C. M, "Distributed acoustic sensing to estimate the permeability" *Geophysical Research Letters*, **2025**, 52(18). <https://doi.org/10.1029/2025GL116944>
4. Johnson, C. W., Wang, K., & Johnson, P. A., "Automatic speech recognition predicts contemporaneous earthquake fault displacement", *Nature Communications*, **2025**, 16(1), 1069. [doi:10.1038/s41467-025-55994-9](https://doi.org/10.1038/s41467-025-55994-9)
5. Hsu, Y.-J., Bürgmann, R., Jiang, Z., Tang, C.-H., Johnson, C. W., Chen, D.-Y., . . . Yang, X., "Hydrologically-induced crustal stress changes and their association with seismicity rates in Taiwan", *Earth and Planetary Science Letters*, **2025**, 651. [doi:10.1016/j.epsl.2024.119181](https://doi.org/10.1016/j.epsl.2024.119181)
6. Johnson, C. W., & Johnson, P. A., "Seismic Features Predict Ground Motions During Repeating Caldera Collapse Sequence", *Geophysical Research Letters*, **2024**, 51(11). [doi:10.1029/2024GL108288](https://doi.org/10.1029/2024GL108288)
7. Guyer, R. A., Marty, S., Marone, C., Johnson, P. A., & Johnson, C. W., "On the anatomy of acoustic emission", *The Journal of the Acoustical Society of America*, **2024**, 156(6), 4116-4122. [doi:10.1121/10.0034637](https://doi.org/10.1121/10.0034637)
8. Ueda, T., Kato, A., Johnson, C. W., & Terakawa, T., "Seasonal Modulation of Crustal Seismicity in Northeastern Japan Driven by Snow Load", *Journal of Geophysical Research: Solid Earth*, **2024**, 129(3). [doi:10.1029/2023JB028217](https://doi.org/10.1029/2023JB028217)
9. Umlauf, J.*, Johnson, C. W.*, Roux, P., Trugman, D. T., Lecointre, A., Walpersdorf, A., . . . Johnson, P. A., "Mapping Glacier Basal Sliding Applying Machine Learning", *Journal of Geophysical Research: Earth Surface*, **2023**, [doi:10.1029/2023JF007280](https://doi.org/10.1029/2023JF007280) (* Contributed equally)
10. Euser, B., Johnson, C. W., Guyer, R. A., Rougier, E., Donahue, C. M., Guthrie, G. D., Johnson, P. A., "Straining to find the permeability". *Earth and Planetary Science Letters*, **2023**, [doi:10.1016/j.epsl.2023.118132](https://doi.org/10.1016/j.epsl.2023.118132)
11. Paul Johnson and Christopher W Johnson, "Probing Seismogenic Faults with Machine Learning" *IEEE International Conference on Image Processing (ICIP)*, **2022**, <https://doi.org/10.1109/ICIP46576.2022.9897239>

Geochemical Processes Controlling the Fate of Critical Elements During Carbonation of Mafic and Ultramafic Minerals

Young-Shin Jun, Principal Investigator

Washington University in St. Louis (WashU), St. Louis, MO

Graduate Students: (WashU) Ying Wang, Xueyi Liu, Taylor Barnett, Zixuan Xie, Tori Cordista, Andrea Stoica, and Mingyang Song; Undergraduate Students: (WashU) Jacqueline Silva, Andrea Carbonell, Tobias Zaidman, Young-Ha Lee, Cindy Borrayo-Carrera, Christina Alexakos, and Marlo Underhill; (Pacific Northwest National Laboratory, PNNL) Bavan Rajan, Tenley Webb, and Bridgette Carven; Postdoctoral Scholars/Staff Scientists: (WashU) Jennifer Houghton, Jonghan Moon, Zoltan Vaci, John Haywood, Clive Jones, and Karthikeyan Rengasamy; (PNNL) Kelly Peterson, Xinyi Shen, Michel Sassi, and Chris Thompson

Collaborators (Co-PIs): Sebastien N. Kerisit (PNNL), David A. Fike (WashU), John S. Loring (PNNL), and J. Mark Meacham (WashU)

Overall research goals: Grounded in the needs for innovative critical element (CE) recovery such as nickel (Ni) and cobalt (Co), our proposed project develops a fundamental, molecular-level understanding of the geochemical processes that control surface passivation and the partitioning of Ni and Co during carbonation and sulfidization of silicate minerals from low-grade mafic and ultramafic ores. In pursuit of this goal, we have defined four project goals (Gs): G1 is to quantify the fundamental processes of CE release, silicate passivation, and carbonate nucleation. G2 is to develop a predictive understanding of CE partitioning into solid products of coupled carbonation and sulfidization processes. G3 is to understand the effects of water activity on silicate surface passivation and the fate of CEs. G4 is to identify the role of reactive fluid flow in determining the outcomes of carbonation and sulfidization in porous media.

Significant achievements during 2022- 2025: In Task 1, we determined the rates of CE release during silicate carbonation and the mechanisms of carbonate nucleation on reacted silicate surfaces (G1). We examined Ni dissolution from San Carlos olivine at high temperatures and high CO₂ pressure. Iron in olivine formed iron(III) (hydr)oxide layers, impeding olivine dissolution. The addition of sodium dithionite (Na₂S₂O₄), a reducing agent, significantly improved olivine dissolution by preventing iron(III) (hydr)oxide formation. To connect this Ni and Co dissolution, we conducted *Ab initio* molecular dynamics (MD) simulations. These simulations revealed that the atomic-level distribution of Co and Ni in abundant mineral deposits depends on their concentrations. To systematically understand the impacts of silicate structure, experimentally we synthesized CE-bearing forsterite (with isolated tetrahedral SiO₄) and CE-bearing enstatite (containing chains of corner-sharing tetrahedral SiO₄). We varied CE contents to examine their carbonation and sulfidation. At equal molecular CE levels, CE-bearing enstatite was less reactive for carbonation than Ni-bearing forsterite, because the chain-structured silicates are harder to break. To investigate carbonate nucleation, classical MD simulations demonstrated that heterogeneous amorphous carbonate nucleation is independent of quartz facet. Quartz surfaces draw water out of hydrated amorphous carbonate nuclei to form a partial hydration layer, leading to interfacial energies independent of crystal facet, and higher than for homogeneous nucleation. When classical MD simulations modeled the nucleation of AMC (MgCO₃·H₂O) during forsterite carbonation, the epitaxial match between forsterite and magnesite (MgCO₃) catalyzed the heterogeneous transformation of AMC into magnesite. Comparing these computational findings, we conducted synchrotron-based grazing incidence small angle X-ray scattering to observe real-time *in situ* Ni carbonate and Co carbonate nucleation on quartz and amorphous silica. We found that Co carbonate nucleation is faster compared to Ni carbonate. Co carbonate nucleation was significantly promoted on amorphous silica compared to quartz, while Ni carbonate has a faster nucleation rate on quartz over amorphous silica.

Task 2 introduced different sulfur sources and examined the preferential transformation of CEs from

primary silicate minerals or secondary carbonate phases to sulfides. We specifically focused on efficiently segregate Ni and Co due to their differential reactivities to each other and other abundant cations (e.g., Mg, Fe) in natural mafic and ultramafic minerals (G2). When sequential carbonation and sulfidation of Co were conducted, we observed the formation of MgCO_3 shells with nanochannel-like pore structures, and Co sulfide nanoparticles nucleated inside these nanochannels. This core-shell structure occurs due to differences in solubilities between Co carbonate and MgCO_3 and the rapid conversion of Co carbonate to Co sulfide during sulfidation. The lattice mismatch between Co phase and MgCO_3 results in the nanochannels, which provides transport pathways for reactants.

Task 3 investigated the effects of water activity on CO_2 mineralization and the fate of CEs during carbonation and sulfidization in thin water films (G3). We examined the reactivity of both the Ni- and Co-bearing forsterites at 5% and 25% Co concentration levels under low-water CO_2 at 85 and 96% relative humidity. Under these low-water conditions, water dissolved in the CO_2 adsorbs onto the forsterite in the form of angstrom- to nanoscale thin water films. CE-bearing samples predominantly form magnesite, whereas samples without CE yields magnesite, hydromagnesite, and dypingite. Ni-bearing forsterite is less reactive than forsterite without Ni. Co-bearing forsterite shows nonmonotonic reactivity: Faster at 5% Co and slower at 25% Co concentration, due to thermodynamic drivers that lower carbonate supersaturation.

Task 4 determined the roles of reactive fluid flow and the impacts of pore/fracture clogging during reactions in porous media/beds (G4). We conducted high-temperature and low-pressure fluid cell experiments filled with olivine mineral particles and studied MgCO_3 formation with and without CE precursors. In the presence of Ni, MgCO_3 nucleation occurred faster and filled the fluid cell more significantly compared to the system without Ni. This result suggests that the CEs significantly influence MgCO_3 formation, which in turn affects reactive flow and impact their transport and recovery. To enhance our fluid cell experiments, we developed micromodels that can be used under high pressure (< 80 bar) and high temperature ($< 100^\circ\text{C}$) for *in situ* monitoring (e.g., Raman spectroscopy) of carbonation and sulfidization processes under reactive fluid flow.

This project is a collaboration between WashU and PNNL, leveraging state-of-the-art experimental and computational capabilities funded by DOE-BES, as well as DOE Office of Science user facilities. Our experimental approaches are closely integrated with advanced computational molecular modeling to provide molecular-scale insights into CE distribution, recovery, carbonation, and concurrent sulfidation. This project spurs the development of new technologies that can enhance the recovery of CEs, lower the processing costs of low-grade ores, save water, and reduce waste.

Science objectives for 2026-2028: This project under DE-FOA-0002676 concludes in 2026, yielding valuable new information. We plan to further advance the science of CE recovery in 2026-2028.

Publications supported by this project 2023-2025: A total of 14 publications, 4 in revision, 1 doctoral dissertation, 7 in preparation, 32 talks, 11 conference/session organized. Selected publications are

1. Ying Wang and Young-Shin Jun*, Dithionite Inhibits Iron(III) (Hydr)oxide Formation during Olivine Dissolution Advancing Simultaneous CO_2 Mineralization and Nickel Recovery. *Environmental Science & Technology*, **2025**, 59, 49, 26886–26896, DOI:10.1021/acs.est.5c13307
2. Xinyi Shen, Micah P Prange, Sebastien N Kerisit*, Impact of Interfacial Structure on Heterogeneous Nucleation of Amorphous Carbonates. *The Journal of Physical Chemistry Letters*. **2025**, 16, 4440-4448, DOI:10.1021/acs.jpclett.5c00629
3. Loring, J. S.; Webb, T. E.; Bowden, M. E.; Engelhard, M. H.; Kerisit, S. N. Cobalt substitution slows forsterite carbonation in low-water supercritical carbon dioxide. *Physical Chemistry Chemical Physics*, **2024**, 26 (41), 26465-26471, DOI: 10.1039/D4CP02092H.
4. Jennifer Houghton*, John M. Haywood, Ying Wang, Young-Shin Jun, and David A. Fike, Selectivity of Tris complexation for Ni(II), Co(II), and Fe(II) and its effect on carbonate precipitation under alkaline conditions, *Applied Geochemistry*, **2025**, 191, 106512, DOI:10.1016/j.apgeochem.2025.106512

Probing CO₂ dissolution and Mineralization in Deep Saline Aquifers with Experiment and Simulation

Aaditya Khanal, Principal Investigator

Russell School of Chemical Engineering and McDougall School of Petroleum Engineering,
University of Tulsa, Tulsa, OK

Graduate Students: Md Fahim Shahriar, Prince Otabir, Ebenezer Ofosu, Maliha Anzuman

Collaborators: Mojdeh Delshad (UT Austin), Ryosuke Okuno (UT Austin)

Overall research goals:

The goal of this project is to advance fundamental understanding of subsurface gas storage in deep geological formations. The research focuses on assessing convective dissolution, mineral trapping, and wettability effects in heterogeneous reservoirs, with emphasis on how geological variability, fluid composition, and fluid–rock interactions influence injectivity, trapping behavior, and long-term storage security. While the primary application is carbon dioxide (CO₂) storage in saline aquifers, the underlying processes and methodologies are broadly relevant to other subsurface gas storage systems.

Significant achievements during 2023- 2025:

Reactive transport simulations were conducted to quantify convective dissolution under heterogeneous reservoir conditions representative of deep saline aquifers. A range of permeability contrasts, layering configurations, and spatial correlation lengths were examined to assess their influence on dissolution and trapping behavior. Results show that fine-scale heterogeneity significantly alters the onset, morphology, and persistence of convective fingers, leading to non-monotonic effects on dissolution trapping compared to homogeneous systems. Certain heterogeneity structures enhance lateral mixing while suppressing vertical plume migration, highlighting the importance of resolving sub-grid geological variability when assessing storage performance.

Additional modeling examined the impact of injected gas composition, with particular focus on impurities relevant to industrial gas streams. These simulations incorporated coupled multiphase flow and geochemical reactions to evaluate how impurities influence near-well and reservoir-scale behavior. Results indicate that the presence of H₂S can accelerate halite precipitation near the injection region, reducing local permeability and injectivity while also redistributing gas among mobile, dissolved, and trapped phases. Sensitivity studies further showed that the extent and spatial distribution of salt precipitation depend strongly on well geometry, injection rate, and reservoir heterogeneity. This work provides new insight into how fluid composition and operational parameters jointly control injectivity and trapping outcomes. A manuscript describing these results has been submitted for peer review.

Laboratory sand-pack experiments were conducted to examine the influence of wettability on subsurface flow behavior and gas-brine interactions. Controlled flooding tests demonstrated that shifts toward more water-wet conditions lead to measurable increases in injectivity and changes in gas saturation distribution. These experimental observations were complemented by field-scale simulations based on benchmark storage scenarios, which showed consistent trends in injectivity and dissolution behavior under altered wettability conditions. Together, the experiments and simulations establish a mechanistic link between wettability, flow pathways, and dissolution dynamics.

To synthesize results across modeling and experimental studies, a comprehensive simulation database was developed and analyzed using global sensitivity analysis techniques. These analyses confirm that wettability and reactive surface areas are dominant controls on mineral trapping

efficiency, exceeding the influence of several kinetic and thermodynamic parameters within the ranges examined. The results provide a quantitative basis for prioritizing key parameters in future modeling and experimental efforts and support the development of reduced-order and machine-learning-based predictive frameworks.

Science objectives for 2026-2028:

Future work will continue to assess dissolution, mineralization, and wettability effects under increasingly realistic geological conditions. Planned efforts include expanded laboratory experiments and additional reactive transport simulations to evaluate the robustness of observed behaviors across a broader range of reservoir properties, including heterogeneity structure, fluid composition, and injection conditions.

A key objective for this period is to integrate data-driven and machine-learning approaches with physics-based modeling to improve assessment of subsurface gas storage behavior. Simulation and experimental datasets generated in earlier phases of the project will be used to train statistical and machine-learning models capable of identifying dominant controls, quantifying uncertainty, and capturing nonlinear interactions among governing parameters. These models will be developed to complement, rather than replace, high-fidelity simulations, enabling efficient exploration of parameter space and improved interpretability of storage performance trends.

The established modeling and experimental frameworks will also be used to examine the transferability of identified mechanisms to other subsurface gas storage systems, such as hydrogen and natural gas, while remaining aligned with the project's original scientific scope. Collectively, these efforts will advance predictive understanding of subsurface gas storage processes and provide a foundation for integrated physics-informed and data-driven assessment tools.

Publications supported by this project 2023-2025 (Selected):

1. Prince N. Y. M. Otabir, Aaditya Khanal, Marcos Vitor Barbosa Machado, Mojdeh Delshad, "Effects of CO₂-H₂S Mixtures, Well Orientation, and Heterogeneity on Halite Precipitation in Geological Storage," *Gas Science and Engineering*, 2025, submitted. DOI: pending.
2. Ebenezer A. Ofosu, Prince N. Y. M. Otabir, Aaditya Khanal, "Quantifying Local Capillary Trapping of CO₂ in Heterogeneous Shale-Sand Systems," *ACS Energy & Fuels*, 2025, in preparation.
3. Aaditya Khanal, Md Irfan Khan, Md Fahim Shahriar, "Comprehensive parametric study of CO₂ sequestration in deep saline aquifers ", *Chemical Engineering Science*, DOI: <https://doi.org/10.1016/j.ces.2024.119734>

Uptake mechanisms of REE in sedimentary phosphorite mineral

Matthew J. Kohn, Principal Investigator

Department of Geosciences, Boise State University, Boise, ID

Graduate Students: John Fink (Boise State University), Prachi Garg (University at Buffalo)

Collaborators: Baishakhi Mazumder (University at Buffalo), Richard Hervig (Arizona State University), Darin Schwartz (Boise State University)

Overall research goals:

Rare-earth elements (REE) are crucial to numerous technologies, including battery-, computer screen-, glass-, green-, defense-, magnet-, laser-, and medical-technologies. China seemingly controls the global supply of all REE, although REE deposits are widespread and voluminous in the US. In fact, US phosphorite deposits, dominated by the mineral apatite, are already being mined commercially for their phosphate (for fertilizer), and contain large reserves of REE. This source of REE could, in principle, provide sufficient REE for the foreseeable future. However, little is known about the distribution of REE in phosphorites, or the mechanisms by which REE are transported. For example, we do not know (a) whether REE reside inside apatite nanocrystals or on nanocrystal surfaces, (b) whether LREE vs. HREE occupy different crystallographic locations within or around nanocrystals, and (c) whether REE move via lattice diffusion, grain boundary diffusion, dissolution-reprecipitation, recrystallization, or some combination. The purpose of our project is to better understand REE siting and movement to help inform future extraction techniques.

Significant achievements during 2023- 2025:

Our research focuses on tooth enamel as an ideal, initially REE-free substrate for characterizing uptake rates and mechanisms. Activities with students have focused on three main objectives: (1) Identify fossil materials with uptake profiles, (2) Experimentally determine REE diffusion rates in tooth enamel, and (3) Use atom probe tomography (APT) to characterize distributions of REE on a nm scale to identify uptake mechanisms. Other activities include fundamental research on the ionic radii of REE in common minerals (Kohn and Schwartz, 2025a, 2025b), paleoecological research involving the geochemistry of fossils (Stiles et al., 2025; Corecco et al., 2024, 2025), and undergraduate research developing a new technique for quantifying OH content in apatite.

For objective (1), we have identified numerous fossils with relatively high concentrations of REE (100s to 1000s of ppm), suitable for APT analysis. Concentration profiles were measured using laser-ablation, inductively-coupled plasma, mass spectrometry at Boise State University, and generally conform to prior research that shows high concentrations near tooth surfaces, with quasi-exponential or error function decreases in concentrations into tooth interiors. APT tips have been milled from near-surface regions of these teeth using focused ion beam milling, and a subset of tips has been analyzed using APT.

For objective (2), we have conducted hydrothermal experiments at Boise State over temperature ranges of 23 °C to 90 °C for periods ranging from 1 week to 7 months. Composition profiles were measured using Secondary Ion Mass Spectrometry at Arizona State University, and show a near-surface mixing profile, succeeded at depth by a quasi-exponential or error-function decrease in concentration into the tooth interior. Near-surface data are biased by ion mixing, but compositions farther from the surface yield diffusivities for Sc, lanthanides, and U that are broadly similar to each other, and decrease from $\sim 10^{-20}$ m²/s at 90 °C to $\sim 10^{-21}$ m²/s at 23 °C. Diffusion of Sr, Ba, and Pb is 1 to 3 orders of magnitude faster: $\sim 10^{-17}$ m²/s at 90 °C and $\sim 10^{-20}$ m²/s at 23 °C.

For objective (3), sub-nm-scale distributions of atoms were measured using APT at University of Buffalo. In general, these data confirm that uptake is not merely surface adsorption but involves complex charge compensation substitutions (e.g., $\text{Na}^+ + \text{REE}^{3+} \rightarrow 2\text{Ca}^{2+}$) within crystallites. Atomic scale differences between LREE and HREE uptake appear governed by competition between lattice

strain and electronic stability. High- and low-concentration domains also mimic original crystallite geometries, suggesting primary biogenic crystal control on REE uptake and potentially providing guidance for chemical extraction of REE. Original biogenic crystallites have low concentrations of REE, while their surfaces are enriched in REE.

Additional research shows that updates to ionic radii relative to Shannon (1976; *Acta Crystallographica*) violate crystal-chemical principles (e.g., the lanthanide contraction). A new approach that relies on mineral partitioning experiments refines the ionic radii of trivalent REE to within $\pm 0.002\text{\AA}$. Ionic radii are fundamental to research in (geo)chemistry, physical chemistry, and materials science, and the new estimates provide a stronger foundation for future research on REE speciation, mineralization, materials properties etc. Raman spectra on apatite with variable OH content shows complications related to crystallographic orientation, but promise future *in situ* determination with high precision (a few percent in the mole fraction of OH; Negrete et al., 2025). A novel comparison of tooth enamel apatite crystallography across the tree of life is currently in final stages of writing for submission to a high-profile journal (Fink et al., 2024, in prep.)

Science objectives for 2026-2028:

Activities in pursuit of objective 1 (identifying suitable materials for APT analysis) are completed. For objective 2, we are currently completing time-series analyses of diffusivities to ensure our data are not biased by diffusion transients. We are also conducting experiments at intermediate temperatures (40-60 °C). We are currently in the midst of objective 3, and plan APT analysis of several more tips to verify or refine our interpretations of trace element distributions in our first samples. Raman analysis of apatite for OH content will require extensive new data with careful control of crystal orientation. All work requires compilation and writing for publication.

Publications supported by this project 2023-2025: peer-reviewed articles

1. Kohn, M.J., and Schwartz, D.M. "Revision of Y^{3+} ionic radii in common minerals based on trace element partitioning" *American Mineralogist*, **2025**, 110, 2029–2033, [DOI:10.2138/am-2025-9771](https://doi.org/10.2138/am-2025-9771)
2. Kohn, M.J., and Schwartz, D.M. "Rebooting REE³⁺ radii in common minerals – a practical approach" *Contributions to Mineralogy and Petrology*, **2025**, [DOI:10.1007/s00410-025-02248-y](https://doi.org/10.1007/s00410-025-02248-y)
3. Stiles, E., Gelfo, J.N., Raigemborn, M.S., Kohn, M.J., Canares, B.A.D., Trayler, R.B., Ibañez-Mejía, M., Erra, G., Goin, F., and Strömberg, C.A.E. "Phytoliths reveal Paleocene–Eocene forest expansions preceded dryland vegetation in southern South America" *Geological Society of America Bulletin*, **2025**, 138(1-2), 823–838, [DOI:10.1130/b38275.1](https://doi.org/10.1130/b38275.1)
4. Corecco, L., Kohn, M.J., Pereira, V.P., Reynard, L.M., and Schultz, C.L. "Paleoenvironmental and paleoecological reconstruction of Santa Maria Supersequence units (Middle to Late Triassic, Brazil) based on stable isotope data: Influence of the Carnian Pluvial Episode" *Chemical Geology*, **2025**, 674, [DOI:10.1016/j.chemgeo.2024.122589](https://doi.org/10.1016/j.chemgeo.2024.122589)
5. Corecco, L., Kohn, M.J., and Schultz, C.L. "Triassic climate and the rise of the dinosaur empire in South America" *Journal of South American Earth Sciences*, **2024**, 142, [DOI:10.1016/j.jsames.2024.104977](https://doi.org/10.1016/j.jsames.2024.104977)

Publications supported by this project 2023-2025: selected abstracts

6. Negrete, M, Kohn, MJ, and Fink, J "Towards a quantitative calibration of hydroxyl content in apatite by Raman spectroscopy", **2025**, GSA annual meeting, San Antonio.
7. Fink, JT, Kohn, MJ, and Hervig, RL (2025) "New constraints on trace element diffusion rates in tooth enamel bioapatite" **2025**, GSA annual meeting, San Antonio.
8. Garg, P, Fink, JT, Kohn, MJ, and Mazumder, B "Atomic scale origins of rare earth element enrichment in phosphorites for critical mineral recovery" **2025**, Materials Research Society Annual Meeting, Honolulu.
9. Fink, JT, Farfan, GA, Rakovan, JF, Kohn, MJ, Bemis, KE, Hsu, TY, Hawkins, MT "Unit cell refinements of enamel and enameloid bioapatite from across the tree of life" **2024**, AGU fall meeting, Washington, DC

Selective nucleation of rare earth element phases on mineral surfaces

Benjamin A. Legg, Principal Investigator

Physical Sciences Division, Pacific Northwest National Laboratory, Richland, WA

Post Doctoral Researchers: Zsombor Molnar, Shuhong Song, Maxime Pouvreau

CCI Intern: Daniela Sepulveda Gonzalez

Collaborators: Christopher Mundy, Mohammadhasan Dinpajooh, Sebastian Mergelsberg, Venkateshkumar Prabhakaran, Hyou Park, and Evan Mondarte (PNNL).

Overall research goals:

Molecular-scale processes of crystal nucleation, interfacial precipitation, and ion adsorption play a fundamental role in controlling the distribution, speciation, and fractionation of elements in geochemical settings. These processes are strongly influenced by the presence of mineral substrates that can catalyze nucleation and host varied modes of interfacial precipitation/adsorption. Moreover, as unconventional mineral resources present novel combinations of critical elements, mineral substrates, and geochemical conditions, we anticipate that new modes of interfacial behaviour may also occur. Examples include recently discovered phenomena, such as the electrostatically stabilized interfacial nanostructured phases. Understanding such phenomena is of outstanding importance, to enable the identification of new sources of critical minerals and support the development of efficient recovery strategies.

This project focuses on understanding how mineral surfaces drive the selective nucleation of rare earth element (REE) phases. Specific objectives include (1) *elucidating the key molecular-scale interfacial forces that drive REE surface precipitation*, (2) *testing how specific combinations of geochemical conditions and mineral surfaces can drive divergent precipitation outcomes*, and (3) *determining whether mineral surfaces can be leveraged to drive separation and/or enrichment of specific REEs from solutions containing mixtures of diverse cationic species*.

Significant achievements during 2023- 2025:

Since funding in late 2023, this early career research project has assembled a research team of three postdoctoral researchers with expertise in geochemistry, crystal growth, molecular modeling, and atomic scale imaging. We have also completed a comprehensive set of simulations on the pH-dependent behavior of heavy and light rare earth elements and a detailed suite of experiments characterizing the pH-dependent interactions of heavy and light REEs with gibbsite surfaces.

Modeling the formation of complex, oligomeric REE species at interfaces is a key goal of this project. To support this, we have completed a series of ab initio molecular dynamics (AIMD) simulations for hydrolysis for REE ions across the lanthanide series. This involved detailed benchmarking of quantum basis sets and functionals, and comparison of two distinct simulation approaches: a widely-used alchemical proton insertion/deletion method (for La, Nd, Sm, Gd, Dy, Er and Yb), and a newer umbrella sampling method to generate potentials of mean force (PMFs) with respect to a complex reaction coordinate (for La, Gd, and Yb). Using both methods, we have predicted pK_a values across the lanthanide series, reproducing the expected trend of lower pK_a with increasing atomic number. This allowed us to validate the PMF approach, while simultaneously generating a key dataset of molecular trajectories that will enable the refinement of ML potentials for future simulation of larger-scale collective behavior, such as oligomerization. This work will be submitted in an upcoming publication:

Maxime Pouvreau, Christopher J. Mundy, Mohammadhasan Dinpajooh, Benjamin A. Legg
“Simulating the hydrolysis of rare earth elements” **2026**, *in prep.*

In parallel, we have recently completed a complete experimental campaign on the pH-dependence of heavy and light REE adsorption on gibbsite nanoparticles, that included surface potentiometry, mass spectrometric, atomic resolution scanning transmission electron microscopy (STEM), and in situ atomic force microscopy (AFM). Our work showed that adsorption increased significantly at higher pH and was dominated by the formation of nanoscale oligomeric species (i.e. hydroxide ions) that are neglected in existing surface complexation models. Importantly, we also found major differences in the sorption of heavy and light REEs in the environmentally-relevant pH range of 7-9, which we hypothesize is key to driving elemental fractionation. A manuscript describing these results has been completed:

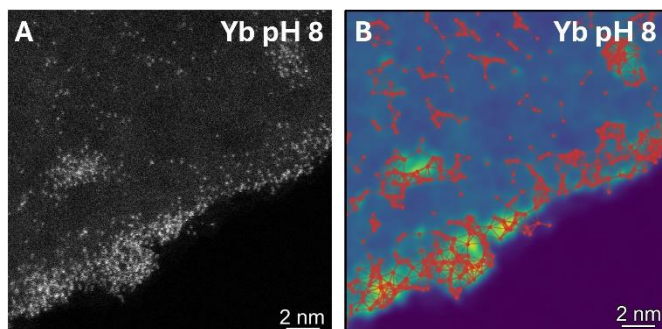


Figure. 1: Atomic resolution STEM imaging with custom-written automated image-analysis routines demonstrated the emergence of nanoscale Yb clusters under slightly basic conditions (pH 8), that are essential to regulating Yb adsorption on gibbsite. The experiments also revealed systematic differences with ion type (i.e. increased oligomerization of heavier REEs) and facet-specific interactions (i.e. preferential adsorption at gibbsite edges) that will enable improved modeling.

Zsombor Molnar, Shuhong Song, Hyoju Park, Daniela Gonzalez-Speulveda, Benjamin Adam Legg
“Selective adsorption of rare earth ions and oligomers on gibbsite surfaces.” **2026**, *In submission.*

We have also conducted a complementary series of atomic force microscopy experiments utilizing advanced in situ 3D force mapping to investigate the physical drivers of facet-specific adsorption. Data analysis and complementary finite-volume modeling efforts are currently underway to explore the couplings between nanometer-scale electric fields generated at molecular-scale features (i.e. gibbsite step edges) and ion adsorption.

Science objectives for 2026-2028:

Major priorities for 2026-2028 include understanding (1) differences in behavior between aluminum hydroxides (e.g. gibbsite) and common phyllosilicates (e.g. swelling clays and micas), (2) characterizing the influence of carbonate ions on adsorption and precipitation, with a focus on the competition between hydroxide and carbonate phases, (3) building molecular and continuum scale models of surface adsorption and precipitation, and (4) testing whether the distinct modes of precipitation observed can be harnessed to drive selective enrichment and separation of REE elements. Significant progress is already underway with respect to goals 1 and 2, as we have already collected a detailed dataset of surface potential and STEM data for pH-dependent adsorption on the clay minerals, and the influence of carbonate on REE interactions with gibbsite. For goal 3, we have built collaborations with PNNL simulation experts C. Mundy and M. Dinpajooh at PNNL and are currently generating an efficient simulation framework to model complex collective behavior of charge-frustrated REE systems. Finally, we are currently working to meet goal 4 by preparing proposals to DOE synchrotron radiation sources that will enable element-specific, molecular-scale characterization of adsorbates on aluminum (oxy)hydroxide surfaces.

Publications supported by this project 2023-2025:

1. Benjamin A. Legg, Yandi Hu, Ayumi Koishi, M. Whittaker, “Pathways for Nucleation and Growth in Confined Spaces and at Interfaces” *Elements*, **2025**, 21, 40-45, [DOI:10.2138/gselements.21.1.40](https://doi.org/10.2138/gselements.21.1.40).

Chemo-Rheological Controls on Subsurface Stress Heterogeneity in Geothermal Systems

Harry Lisabeth, Principal Investigator

Chemical Sciences Division, Hawkins National Laboratory, Hawkins, IN
Physical Sciences Division, Hawkins National Laboratory, Hawkins, IN

Graduate Students: None

Collaborators: John Christensen, Nobumichi Tamura, Donald DePaolo (LBNL), Clay Jones (University of Utah), Greg Boitnott (New England Research)

Overall research goals:

This project seeks to quantify the coupled mechanical and chemical controls on subsurface stress evolution in geothermal reservoirs in order to improve predictability of fracture behavior, reservoir stimulation, and long-term system performance. Subsurface stress governs fracture initiation, propagation, and permeability development, yet its evolution in high-temperature crystalline geothermal environments remains poorly constrained. The project addresses this gap by integrating laboratory rock mechanics, isotope geochemistry, and machine-learning approaches to develop a physically grounded predictive framework for stress heterogeneity, with direct application to enhanced geothermal systems (EGS).

Significant achievements during 2023- 2025:

Since project initiation in October 2025, the first quarter of work has focused on establishing the experimental and material foundation required for the research program. Key accomplishments include identification of target lithologic horizons within the FORGE geothermal system suitable for rheological and geochemical investigation, and close coordination with the FORGE core curation team to secure representative granitoid core material for laboratory testing. In parallel, design of the custom high-temperature, high-pressure deformation apparatus has begun, including development of engineering specifications for the integrated mechanical loading and in-line fluid sampling system. The project team has also initiated procurement activities and is actively working toward execution of a purchase contract for the experimental system. Preliminary characterization of recovered core materials has commenced, including sample preparation and baseline assessment of physical properties in support of upcoming forced-oscillation and creep experiments. Together, these efforts establish the infrastructure, sample suite, and experimental readiness required for the project's primary scientific objectives.

Science objectives for 2026-2028:

The primary scientific objective of the next project phase is to develop a physically constrained, quantitatively validated framework for predicting subsurface stress evolution in crystalline geothermal reservoirs, using material and data from FORGE as a baseline. This will be accomplished through coordinated advances in experimental rock mechanics, isotope geochemistry, and data-driven modeling.

A central goal is to determine the appropriate viscoelastic rheological description for FORGE granitoid under in-situ geothermal conditions. Low-frequency forced-oscillation experiments will be conducted on core samples over a wide range of temperatures (to ~350 °C), confining pressures (to ~100 MPa), differential stresses, and fluid saturation states. These experiments will yield frequency-dependent storage and loss moduli, attenuation, and dispersion curves that directly constrain time-dependent deformation behavior. The experimental results will be used to evaluate competing rheological models, including linear

and nonlinear viscoelastic formulations (e.g., Maxwell, Kelvin-Voigt, Burgers, power-law Burgers, and Andrade models), and to identify the constitutive parameters governing stress relaxation across relevant timescales. These measurements will provide the first comprehensive rheological characterization of geothermal reservoir granite under true reservoir conditions and establish the mechanical basis for modeling long-term stress redistribution in EGS.

A second major objective is to quantify the role of chemically mediated deformation mechanisms in controlling creep and stress relaxation. Long-duration hydrothermal creep experiments will be conducted using a newly developed high-temperature, high-pressure deformation system with integrated in-line pore-fluid sampling and stabilization. By combining real-time mechanical measurements with high-precision isotopic and elemental analyses of experimental effluents (including $^{29}\text{Si}/^{28}\text{Si}$, $^{87}\text{Sr}/^{86}\text{Sr}$, $^{44}\text{Ca}/^{40}\text{Ca}$, Ge/Si, and Sr/Ca), the project will directly resolve concurrent mineral dissolution and precipitation rates during deformation. This approach will allow the relative contributions of crystal plasticity, pressure solution, subcritical cracking, and fluid-assisted mass transfer to be quantitatively deconvolved as a function of temperature, stress, mineralogy, and fluid chemistry. Establishing the mechanistic controls on creep at geothermal conditions is essential for parameterizing the viscoelastic stress-relaxation models developed in the first research thrust.

The third objective is to integrate laboratory-derived rheological and geochemical constraints with field data to produce predictive models of subsurface stress heterogeneity. A comprehensive dataset will be assembled from FORGE well logs, core measurements, stress calibration data, and experimental results. Machine-learning models, including random forests, adaptive neuro-fuzzy inference systems, and functional networks, will be trained to predict minimum horizontal stress as a function of depth using physically meaningful input features. The models will be grounded in the viscoelastic stress-relaxation framework, enabling laboratory-scale constitutive behavior to inform field-scale stress predictions. Model performance will be rigorously evaluated through cross-validation, uncertainty quantification, and comparison with independent field stress measurements.

Collectively, these objectives will deliver a unified experimental–computational framework for predicting subsurface stress evolution in geothermal systems. The resulting capability will directly inform geothermal reservoir stimulation design, mitigate operational risks such as induced seismicity and fluid loss, and improve long-term EGS performance, while also advancing fundamental understanding of chemo-mechanical deformation processes in the Earth's crust.

Publications supported by this project 2023-2025:

This is a newly initiated project.

A molecular understanding of PGE transport and ore formation through an integrated experimental, computational and thermodynamic modeling approach/Elucidating REE transport and deposition through molecular understanding of hydrous carbonate silicate liquids

Craig Lundstrom, Principal Investigator

Professor, Dept of Earth Science and Environmental Change, University of Illinois at Urbana Champaign, Urbana IL 61801

Graduate Students: Jacob Huffaker, Ines Gautier

Post-doctoral researchers: Qi Chen, Patrick Manselle

Collaborators: Demitry Sverjensky (Johns Hopkins University), Giulia Galli (University of Chicago)

Overall research goals:

Most of the constituents of DOE's Critical minerals inventory reflect deposition in igneous systems with magmas being the ultimate source of a critical element—however, in most cases, the ore formation reflects deposition from fluids associated with magma and not the magma itself. Thus, understanding the magmatic-hydrothermal transition is critical to understanding deposit formation. These projects aim to understand the molecular controls on PGE and REE transport and deposition from hydrous alkali silicate (+/-) carbonate liquids through solubility determinations combined with bonding and valence characterization using XAS.

Significant achievements during 2023-2025:

The dominant form of PGE deposit globally are “reefs” in layered mafic intrusions. The origin of these thin, highly concentrated (1000x enrichment) layers is debated: the orthomagmatic model posits gravitational settling of PGE sulfides from silicate melt while the hydrothermal model posits upflow of H₂O fluids. The system H₂O-Na₂Si₂O₅ shows full liquid miscibility at <400°C; a liquid in equilibrium with quartz and albite shows continuous change from silicate-rich “melt” at 800°C to liquids with 50 wt% H₂O at 330°C providing unique insight into the magmatic-hydrothermal transition. We performed high pressure (1 kbar) experiments at variable controlled *f*O₂ and temperature (500-750°C) to determine the solubility of Pd and Pt into hydrous alkali silicate liquids saturated in quartz and albite. The silicate melts quenched to vapor saturated glass with ~8-9 wt% H₂O. ~2 wt% Cl was added to some experiments to examine the role of Cl complexing PGE in these liquids. Oxygen fugacity was buffered using double capsule techniques and ranged from Ni-NiO to Mn₃O₄-Mn₂O₃. All glasses contain PGE metal nuggets, even under extreme *f*O₂ of Mn₃O₄-Mn₂O₃ buffer. Measurements of nugget free glass by LA-ICPMS find Pt solubility varies from <10 ppm at NNO to 220 ppm at Mn₃O₄-Mn₂O₃. The presence of 2 wt% Cl in the glass appears to make no difference to solubility. These findings are corroborated by XAS. Using Pt L3-edge or Pd K-edge spectra with reference standards, we find that the Pt and Pd oxidation states increase systematically with increasing *f*O₂. Under the most oxidizing condition (Mn₃O₄-Mn₂O₃), Pt is bonded to O with an effective valence near 3+, i.e., a mixed Pt⁴⁺-and Pt²⁺ environment, with a first-shell Fourier Transform peak occurring at ~ 1.70 Å (phase-shift uncorrected). Pd at the same buffer is also O bonded reflecting Pd²⁺ and a Fourier Transform peak at ~ 1.44 Å. A decrease in both the absorption edge and white line was observed under more reduced conditions, indicating reduction to lower valence states for both elements. At NNO +0.5, both Pt and Pd spectra closely approach their respective metal signals. Notably, no Pt-Cl or Pd-Cl scattering is detected in any of the measured samples, despite up to 2.0 wt.% Cl dissolved in the melt, demonstrating that chloride does not enter the first coordination sphere of Pt or Pd in these hydrous peralkaline melts. These results indicate that under *f*O₂ relevant to layered intrusions, Pt and Pd are metals with limited transport possible. To further explore PGE behavior in a simulated magma chamber mush, we performed temperature gradient experiments where 2% PtS

or PdS were homogeneously added to a 93% Olivine-Orthopyroxene aggregate and 5% (30% H₂O:60% Na₂Si₂O₅:10% Na₂CO₃) powder and placed into cm long Ag or Quartz tubes. The sealed tubes were placed into imposed temperature gradients (offsets of either 1100-1000°C or 900-400°C) for week-long durations. Line scans using LA-ICPMS were set up to examine resulting transport and deposition of PGEs. In both types of experiments, we observed isotherm-parallel PGE blebs with prominent Pt and Pd peaks appearing in the line scans indicating diffusive transport and deposition of PGE sulfides at specific temperatures in each capsule. In the high temperature, low water content silica tubes, the peak occurs at ~1066°C. In the water-rich Ag capsule experiments (at 1 kbar), deposition occurs at ~750°C. These results are consistent with Lundstrom and Gajos (Econ. Geo. 2014) who showed base metal sulfides diffused and deposited at specific temperatures in similar experiments. PGE transport behavior was also constrained by molecular dynamics calculations. MD results show that Pt complexes with water and CO₂ under two pressure conditions (1GPa and 11GPa; Le and Galli, 2025). Notably, they found that Pt (IV) formed bidentate complexes with carbonate ions at the lower pressure conditions. MD predicts a prominent Raman band signature of this complex which will be examined in coming hydrothermal diamond anvil cell work. Lastly, falling sphere synchrotron measurements of viscosity indicate 1×10^{-3} Pa·s for 50:50 H₂O: Na₂Si₂O₅ liquids at 250°C.

Science objectives for 2026-2028:

The biggest global REE deposits are carbonatite associated but again reflect a hydrous fluid derived from carbonatite and not deposition from carbonatite liquid directly. Prior work shows that a hydrous peralkaline silicate liquid coexists with feldspar and nepheline at 330°C (1 kb) but only if wt% levels of carbonate are present. We will use this liquid in a series of experiments to determine solubility, transport and deposition of REE to understand carbonatite related REE deposits. Specifically, we will experimentally determine liquid compositions transitioning from hydrous silicate melts to alkali carbonate silicate liquids with 20-40% H₂O at high temperatures and pressures and determine the solubility dependence of DyPO₄ (synthetic xenotime) over the range in liquid composition. We will measure synchrotron EXAFS spectra of Al and Dy in the continuum of liquid compositions (quenched to glass) ranging from hydrous silica undersaturated melts at 800° to aqueous, alkali silicate-carbonate rich liquids at 300°C. We will also build a high P&T flow reactor and use it to examine reactive transport of hydrous alkali silicate-carbonate liquids through mineral aggregate rocks at 1 kb and 300-550°C. This will allow us to test hypotheses for the origin and evolution of REE deposits in rocks by natural fluids using chemical mass transfer models in which comparison between natural systems and flow reacted experimental material are made.

Publications supported by this project 2023-2025:

1. Le, L., & Galli, G. (2025). Platinum Carbonates in Aqueous Fluids under Extreme Conditions. *The Journal of Physical Chemistry Letters*, 16(8), 2048–2056. <https://doi.org/10.1021/acs.jpclett.4c03653>.
2. Chen Q., Huffaker J., Newville M., Lanzirotti A., Lundstrom C.C. (in press), Transport of Pt and Pd in hydrous sodium silicate melt: a study from X-ray absorption spectroscopy. *Geochim. Cosmochim. Acta*. <https://doi.org/10.1016/j.gca.2025.12.061>.
3. Huffaker, J. (2024). Concentrations of Pt and Pd in a hydrous silicate glass (Master's thesis, University of Illinois at Urbana-Champaign). UIUC IDEALS. <https://hdl.handle.net/2142/125643>
4. Huffaker J. and Lundstrom, C. C. (2025). Sulfide migration in a temperature gradient as a means for PGE Concentration in Layered Mafic Intrusions. *Goldschmidt 2025 Conference Abstracts*.

Coarse-Grained Molecular Studies of CO₂ Storage in Gas Hydrates

OluFemi Olorode, Principal Investigator

Petroleum Engineering Department, Louisiana State University, Baton Rouge, LA

Graduate Students: Temitayo Adeyemi, Meisam Adibifard

Postdoctoral Researcher: Clement Afagwu

Undergraduate Student Worker: Gideon Benson

Overall research goals:

The overarching goal of this project is to obtain molecular-level insights into “large-scale” mechanisms that control the trapping of CO₂ nanobubbles during gas hydrate formation. This project was born out of our observation that gas nanobubbles with diameters of ~9 nm were getting trapped within growing solid hydrates when we simulated domains as large as 96 nm x 12 nm x 12 nm. Considering the potential to enhance gas storage and curtail subsurface gas leakage through hydrate formation, this project seeks to understand the role of the MD simulation domain size in observing larger-scale physical phenomena that were impossible in the smaller domains studied in the literature. The specific goals of this project are summarized as follows:

- Unravel large-scale phenomena that cannot be observed in the small simulation domains typically studied by calibrating the monoatomic water model for CO₂ hydrate studies.
- Conduct a rigorous study that quantifies the potential of increasing hydrate-based CO₂ storage via the trapping of CO₂ gas nanobubbles in a growing solid hydrate.
- Confirm the stability of the trapped nanobubbles using large-scale atomistic MD simulations that leverage physics-informed graph neural networks.

Significant achievements during 2023- 2025:

Our significant achievements since 2023 are summarized as follows:

- Successfully calibrated the mW model for CO₂ hydrate studies and observed that the trapping of these nanobubbles can enhance hydrate-based CO₂ storage by over 37%. Published in the Materials Today Sustainability Journal in 2025.
- Conducted atomistic studies of CO₂ hydrate formation (to generate data for the deep learning accelerated studies of large-scale systems at the atomic scale) and observed that simulation domains smaller than a threshold were too small to observe the formation and trapping of gas nanobubbles. To be presented at the ACS Spring 2026 Meeting.
- Extended this work to conduct atomistic and experimental studies of the enhancement of hydrate-based methane and hydrogen storage, and secured an NSF CAREER award for the enhanced hydrogen storage studies.

Science objectives for 2026-2028:

Our objectives moving forward are summarized as follows:

- Obtain new insights on the observed structural changes in the hydrates formed using coarse-grained force fields by completing ongoing studies that employ a combination of physics-constrained graph neural networks and atomic-scale simulations.
- Conduct atomistic MD and experimental studies to confirm and quantify this idea of enhancing CO₂ storage via the trapping of gas nanobubbles.
- Understand the changes in the mechanical properties of gas hydrates because of the trapped gas nanobubbles, in the context of the mechanical stability of subsurface gas hydrates.

Publications supported by this project 2023-2025:

1. Meisam Adibifard and Olufemi Olorode. "Coarse-Grained Molecular Studies Reveal Potential for Increased CO₂ Storage in Hydrates". *Materials Today Sustainability Journal*, **2025**, Vol 30, 101106, <https://doi.org/10.1016/j.mtsust.2025.101106>.
2. Meisam Adibifard, 2025. "A Coarse-Grained Molecular Dynamics Study of Gas Hydrate Formation and Dissociation". *LSU Doctoral Dissertations*, **2024**, https://repository.lsu.edu/gradschool_dissertations/6578.
3. Olufemi Olorode, Oladoyin Kolawole, Chibuzor Igweonu, and Harun Rashid. "Taking bio-induced precipitation to the field for sustainable geo-energy storage: Experimental and numerical studies of leakage mitigation" *Journal of Energy Storage*, **2025**, Vol 112, 115474, <https://doi.org/10.1016/j.est.2025.115474>.

Quantum Computing and Machine Learning for Enhanced Understanding of Fracture Flow

Daniel O'Malley, Principal Investigator

Earth & Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM

Graduate Students: Bharat Srikishan (Stevens Institute of Technology), Austin Pechan (UC, Berkeley)

Collaborators: John Kath (LANL postdoc), Agnese Marcato (Politecnico di Torino), Arnab Mazumder (LANL postdoc), Chris Johnson (LANL), Allon Percus (Claremont Graduate University), Hari Viswanathan (LANL)

Overall research goals:

This Field Work Proposal tackles the long-standing challenge of modeling fluid flow and seismic waves in densely fractured rock, a problem central to hydraulic fracturing, geothermal development, carbon storage, and hydrogen systems. One thrust uses machine learning to characterize and image the subsurface with a focus on understanding fluid flows and fractured systems. A second, parallel thrust designs fault-tolerant quantum algorithms to solve the sparse linear systems that govern fracture flow and to extend similar quantum techniques to elastic wave equations. We aim to provide resource estimates for upcoming hardware and chart a path toward practical quantum algorithms for subsurface energy science.

Significant achievements during 2023- 2025:

We have made progress on multiple fronts to achieve our goals. Our machine learning work has begun to develop a foundation model of the Earth's subsurface. This model, which we call Transparent Earth, can currently predict 8 properties of the Earth's subsurface globally with strong performance on held-out validation data. In modeling reactive flow through fracture networks with machine learning, we have developed graph-transformer models that match the accuracy of baselines without depending on tedious feature engineering that is key to the baseline's accuracy. Additionally, we developed a foundation model that can simulate fracture mechanics. This model showed excellent generalization across a wide range of fracture conditions and materials, including few-shot learning from as few as 1 training example.

On the quantum side, block-encoding formulations for large fracture-flow operators have been derived, giving a path toward for fault-tolerant implementations. This work provides the first scaling advantage for quantum algorithms over classical algorithms for simulating fluid flow through fracture networks. Additionally, it determined the first resource estimates for such a quantum algorithm, showing an exponential reduction in memory requirements compared to classical computers. We also developed algorithms for fracture flow that utilize quantum-accelerated uncertainty analysis. This is critical for fracture flow because so much uncertainty surrounds the fracture properties (e.g., aperture, length, width) and fracture network properties (e.g., inter-fracture connectivity) that impact flow. Additionally, this increases the amount of quantum computational work performed relative to the amount of information extracted – a central bottleneck that limits many quantum algorithms.

On top of research, we organized the Machine Learning in Solid Earth Geoscience conference, which was held in January 2024 and 2025 in Santa Fe.

Science objectives for 2026-2028:

Going forward, we hope to improve the scaling advantages of our quantum algorithms compared to classical algorithms for fracture flow. This work follows two tracks. First, we aim to extend the modest polynomial speed-up that we have achieved into a more substantial polynomial speed-up or, possibly, an exponential speed-up. Second, we will combine our quantum algorithms for solving fracture flow equations and uncertainty quantification. This has the effect of stacking the speed-ups to further improve performance.

We have also begun work on quantum algorithms for simulating seismic wave propagation through subsurface fracture networks. The goal with this work is to obtain speed-ups and resource estimates similar to the ones we have obtained for flow through subsurface fracture networks. In particular, we aim to leverage recently developed quantum algorithms for simulating large systems of coupled harmonic oscillators to model elastic wave propagation at resolutions beyond what is tractable classically. By doing so, we seek to quantify how fracture network topology and multiscale heterogeneity influence scattering, attenuation, and wavefield complexity, and to establish concrete resource estimates that clarify when fault-tolerant quantum hardware could enable fundamentally new seismic modeling capabilities for fractured subsurface systems.

A complementary machine learning objective is the development of Transparent Earth, a multimodal foundation model for the Earth's subsurface that integrates heterogeneous geophysical, geological, and geochemical observations to infer subsurface properties at scale. The goal of this effort is to learn generalizable representations of the subsurface that can support imaging, characterization, and uncertainty quantification across a wide range of geoscience applications, including but not limited to fractured systems. In parallel, we are advancing graph-based learning methods for modeling flow and reactive transport in discrete fracture networks, using graph transformers that operate on compact network representations to faithfully capture multiscale topology and long-range dependencies. These machine learning efforts provide fast, physics-aware surrogates for complex subsurface processes and allow observational data and high-fidelity simulations to be used in a consistent way. This leads to more reliable predictions and clearer insight into subsurface systems relevant to energy applications.

Publications supported by this project 2023-2025:

1. Ryley G. Hill, Kai Gao, Aleksandra Pachalieva, Agnese Marcato, Xiaoyu Wang, Pascal Grosset, Esteban Rougier, Zhou Lei, Javier E. Santos, Vinamra Agrawal, Qinqun Kang, Jeffrey D. Hyman, Abigail Hunter, Christine M. Sweeney, Nathan DeBardeleben, Earl Lawrence, Hari Viswanathan, Daniel O'Malley, "Material Fracturing and Failure Simulation Datasets" *Scientific Data (Sci Data)*, **2025**, In Press, DOI: [10.1038/s41597-025-06412-8](https://doi.org/10.1038/s41597-025-06412-8).
2. John Kath, John K. Golden, Allon G. Percus, Daniel O'Malley, "Predicting Flow in Fracture Networks With Quantum Algorithms" *Water Resour. Res.*, **2025**, Vol. 61, e2024WR039784, DOI: [10.1029/2024WR039784](https://doi.org/10.1029/2024WR039784).
3. Arnab Mazumder, Javier E. Santos, Noah Hobbs, Mohamed Mehana, Daniel O'Malley, "The Transparent Earth: A Multimodal Foundation Model for the Earth's Subsurface" *arXiv e-prints*, **2025**, In Press (NeurIPS 2025 AI for Science Workshop), DOI: [10.48550/arXiv.2509.02783](https://doi.org/10.48550/arXiv.2509.02783).
4. Harun Ur Rashid, Aleksandra Pachalieva, Daniel O'Malley, "Differentiable multiphase flow model for physics-informed machine learning in reservoir pressure management" *Scientific Reports*, **2026**, In Press, DOI: [10.48550/arXiv.2508.19419](https://doi.org/10.48550/arXiv.2508.19419).
5. Austin Pechan, John K. Golden, Daniel O'Malley, "Block encoding the 3D heterogeneous Poisson equation with application to fracture flow" *arXiv e-prints*, **2025**, Under Review, DOI: [10.48550/arXiv.2508.07125](https://doi.org/10.48550/arXiv.2508.07125).
6. Agnese Marcato, Aleksandra Pachalieva, Ryley G. Hill, Kai Gao, Xiaoyu Wang, Esteban Rougier, Zhou Lei, Vinamra Agrawal, Janel Chua, Qinqun Kang, Jeffrey D. Hyman, Abigail Hunter, Nathan DeBardeleben, Earl Lawrence, Hari S. Viswanathan, Daniel O'Malley, Javier E. Santos, "A Foundation Model for Material Fracture Prediction" *arXiv e-prints*, **2025**, Under Review, DOI: [10.48550/arXiv.2507.23077](https://doi.org/10.48550/arXiv.2507.23077).
7. Aleksandra A. Pachalieva, Jeffrey D. Hyman, Daniel O'Malley, Gowri Srinivasan, Hari Viswanathan, "Learning the factors controlling mineral dissolution in three-dimensional fracture networks: applications in geologic carbon sequestration" *Frontiers in Environmental Science*, **2025**, Vol. 12, Article 1454295, DOI: [10.3389/fenvs.2024.1454295](https://doi.org/10.3389/fenvs.2024.1454295).

Fracture Intersections: Exploring the Central Element of Fracture Networks

Laura J. Pyrak-Nolte, Principal Investigator

Department of Physics & Astronomy, Purdue University, West Lafayette, IN

Graduate Students: Alexandra Clark

Collaborators: Hari Viswanathan & Erin Heilman (Los Alamos National Laboratory), Peter Kang (University of Minnesota), Antonio Bobet & David Nolte (Purdue University), Hongkyu Yoon (Sandia National Laboratory)

Overall research goals:

Geothermal and other subsurface reservoirs benefit from the creation and control of subsurface fracture networks. The simplest topological element of a fracture network is composed of two fractures coupled by one intersection. The mechanical and hydraulic behavior of fracture intersections is central to how a fracture network evolves in response to changing physical and geochemical processes that can either enhance or reduce network connectivity. The importance of intersections is well recognized, but little is understood about their physical properties, raising key questions. Our fundamental research goals are: (1) to investigate how fracture intersections deform under stress and affect the flow paths through a network; (2) to determine how complex network geometry affects fluid mixing and distributions; (3) to determine the mineralogical, structural, and mechanical factors that control the formation of intersection void geometry; (4) to determine the mechanical interactions among multiple topological elements and how they affect network connectivity; and (5) to determine how fracture networks and their components contribute to energy partitioning of elastic waves. Our research lays the groundwork for advancing our understanding of fracture network connectivity and how networks change in response to physical processes. Successful completion of the work will advance the goal of characterizing and adaptively controlling subsurface fracture networks.

Significant achievements during 2023- 2025:

- Imaged, for the 1st time, the deformed geometry of fracture intersections between 2 fractures using 3D X-ray tomography. 3D reconstructions of the fracture network show that the orientation of a fracture network relative to an applied load affects the amount of deformation and the void geometry of fracture intersections. For a "+" orientation relative to a vertical load, network connectivity is maintained by the vertical fracture that does not deform significantly under load. However, for an "x" orientation, the void volume and connectivity between the two fractures decrease significantly and nearly vanish at the highest load as the topology transitioned from "X" to "Z" to "V". These findings suggest that hydro-mechanical modeling of fracture networks that are used to estimate or predict reservoir performance should carefully consider how fracture network topology might evolve or be perturbed from changes in stress caused by subsurface engineering activities.
- Determined, experimentally, that spatial correlations in the aperture distributions and roughness along the individual fracture planes are a dominant factor in determining the void geometry of fracture intersections. For the same applied loads, a fracture network with a random distribution of apertures exhibits more connections between the two fractures than for the spatially correlated case. Just as for single fractures, networks with spatially correlated aperture distributions are dominated by only 1 or 2 flow paths that close more rapidly under stress than networks with random aperture distributions that can support multiple flow paths as stress is increased.
- Demonstrated that fluid mixing is strongly affected by partial closure of an intersection under stress by combining the fracture network geometries imaged in the laboratory with pore-scale numerical modeling. As an intersection closes, the void area for fluid flow and diffusion decreases, leading to significant deviations between conventional mixing models and full pore-scale modeling. A new mixing model was proposed that accounts for intersection deformation, which is essential for

accurate modeling of solute transport and mixing through fracture networks. The conventional mixing rules currently used in discrete fracture network modeling of subsurface reservoirs may not correctly estimate fluid mixing in subsurface reservoirs.

- Demonstrated through a percolation and finite-size scaling analysis that: (1) the contribution of intersections to fracture network connectivity depends on the stress-dependent spatial correlations in fracture aperture distributions; (2) intersections enhance connectivity in flow directions parallel to intersections; (3) correlated clusters on one fracture may be disconnected from correlated clusters on the other fracture, reducing the connectivity of a network; and (4) percolation probabilities are found to be scale dependent which has important consequences for the connectivity of larger fracture networks composed of the fundamental element studied here.

Science objectives for 2026-2028:

We made important progress on understanding how the orientation of the simplest topological elements relative to an applied stress field affect intersection geometry as well as fracture connectivity and fluid mixing. However, fundamental questions about the formation and interaction of topological elements must be answered to meet the challenge of creating, controlling, and maintaining fracture networks in the subsurface. In the next period, our research objectives are: (1) to perform experiments to provide insight on the formation of fracture intersection geometry during crack propagation and interaction; and (2) to determine the effect of global versus local shearing on the interactions among multiple topological elements that affect fracture connectivity. Achievement of these objectives will provide insight into the roles that interacting topological elements, and the geometry of these elements, play in determining fracture network connectivity.

Selected Publications supported by this project 2023-2025:

1. Wang, Chaoyi, Jiang, Liang, Bobet, Antonio, Yoon, Hongkyu, and Laura J. Pyrak-Nolte, "Fabric controls on fracture surface roughness of an architected rock material", *Mechanics Research Communications*, **2024**, 135, 104223, <https://doi.org/10.1016/j.mechrescom.2023.104223>.
2. Sumner, Madelyn, and Laura J. Pyrak-Nolte. "Visualization of Fracture Intersections from 3D X-Ray Imaging", *Proceedings of the 15th ISRM Congress*, **2023**, Salzburg, Austria, October 2023, Paper Number: ISRM-15CONGRESS-2023-150.
3. Nolte, David D., Pyrak-Nolte, Laura J. and Chet Hopp, "Monitoring Fracture Hydromechanical Evolution in the Lab and Field using Unsupervised Metric Learning", *Journal of Geophysical Research – Machine Learning and Computation*, **2025**, vol 2, no. 3, e2025JH000657, <https://doi.org/10.1029/2025JH000657>.
4. Pyrak-Nolte, Laura J., and David D. Nolte. "Role of intersections in fracture connectivity", *Journal of Geophysical Research: Solid Earth*, **2025** 130.12: e2025JB033007.

Position-specific carbon isotope analysis via novel NMR tools

Cornelia Rasmussen, Principal Investigator

University of Texas Institute for Geophysics, University of Texas at Austin, Austin, TX

Collaborators: David Hoffman (University of Texas at Austin), Arndt Schimmelmann (Indiana University)

Overall research goals:

For decades, carbon isotope ratios ($^{13}\text{C}/^{12}\text{C}$) have been utilized to trace the source and transformation history of Earth's organic carbon, with applications ranging across many scientific disciplines. Most isotope measurements have relied on isotope ratio mass spectrometry (IRMS), and report isotope ratios as averages over bulk organic matter or individual compounds. While effective for many applications, the traditional IRMS approach obscures the isotopic variability within molecules, where information related to synthesis, metabolic transformation, and degradation pathways is encoded at specific atomic positions.

Position-specific isotope analysis seeks to uncover this information by determining isotope ratios at individual carbon atom positions within organic molecules. Access to intramolecular isotope distributions has the potential to substantially increase the diagnostic power of carbon isotope analysis, enabling differentiation of chemically identical compounds and providing direct insight into the physiological, kinetic, and equilibrium isotope effects that govern molecular formation and transformation. Potential applications range from distinguishing biotic from abiotic systems, establishing environmental conditions (CO_2 , O_2) at time of formation, or distinguishing sources, for example, of petroleum.

Nuclear magnetic resonance (NMR) spectroscopy has the potential to play a key role in advancing position-specific isotope analysis. Unlike mass-spectrometry-based approaches, NMR enables isotope data to be obtained without molecular fragmentation and provides an independent determination of absolute isotope ratios. Moreover, NMR can access intramolecular isotope distributions within complex organic molecules and mixtures that are currently not amenable to IRMS-based methods. NMR thus provides unique information, and is inherently complementary to mass spectrometry and can help identify future analytical targets for other techniques.

The goal of this work has been to develop NMR tools who take advantage of these prerequisites, and we are pursuing three specific aims: (1) develop NMR acquisition and data-processing strategies capable of disentangling overlapping NMR signals originating from complex samples; (2) apply the resulting tools to complex molecules and mixtures derived from natural samples, focusing on complex organics derived from autotrophs and heterotrophs. These molecular classes provide a tractable yet challenging test case for resolving overlapping signals and enables direct comparison of position-specific isotope distributions in molecules shared across all domains of life, yielding novel insight into the physiological, kinetic, and equilibrium isotope effects; and (3) develop, in collaboration with the US Geological Survey (USGS), a suite of reference materials optimized for determining $^{13}\text{C}/^{12}\text{C}$ ratios by NMR and for intercalibrating NMR- and IRMS-based measurements. These materials will be made available through the USGS to support broader implementation of the developed methods.

Significant achievements during 2023- 2025:

We developed and validated new best-practice strategies for NMR data acquisition and processing that enable reliable determination of position-specific $^{13}\text{C}/^{12}\text{C}$ ratios in the presence of overlapping signals and complex impurity/mixture backgrounds. By adapting a stacked-acquisition approach analogous to methods used in astronomical imaging, we demonstrated that sets of short-duration NMR spectra can be jointly analyzed to improve sensitivity, mitigate instrumental drift and artifacts, and enhance analytical robustness. In parallel, we advanced the use of proton (^1H), fluorine (^{19}F)

and phosphorus (^{31}P) NMR to isolate $^{13}\text{C}/^{12}\text{C}$ signals of interest from complex organic mixtures, enabling position-specific isotope analysis of anthropogenically relevant organic compounds and their degradation products. All approaches were verified using official reference materials supplied by the USGS.

After establishing these foundations, we began applying those tools to biologically derived, natural, samples. We purified RNA from multiple autotrophic and heterotrophic organisms and obtained position-specific carbon isotope data for nucleobases, alongside bulk isotope measurements of whole cells and RNA. These results provide initial evidence that physiological traits, carbon concentrating mechanisms, and metabolic pathways can imprint distinct and recognizable intramolecular isotope patterns, particularly at the C8 position of adenine and guanine.

Finally, we developed three new carbon isotope reference materials (USGS 106-108) optimized for both NMR and IRMS analysis. These materials exhibit excellent homogeneity, precision, and long-term stability, enable direct intercalibration between different analytical techniques, and support refinement of the absolute $^{13}\text{C}/^{12}\text{C}$ ratio of the VPDB primary standard. These materials are available via USGS to all researchers.

Science objectives for 2026-2028:

The primary focus of the coming years will be to apply the developed tools to complex molecules and mixtures derived from natural samples. Ongoing efforts focus on position-specific isotope analysis of organics sourced from autotroph and heterotroph organisms, to understand how aspects such as environmental conditions and metabolism contribute to intramolecular carbon isotope fractionation patterns. Moreover, these studies contribute to ongoing methods development for analyzing complex mixtures at low concentrations for their position-specific isotope information across multiple compounds.

Position-specific isotope analysis is an emerging field of research, with methods still being developed and refined, and complex intramolecular isotope data now becoming available for the first time. Hence, robust interpretations of these novel isotope fingerprints are still in early stages. Our next science objectives are focused on providing the necessary knowledge base for robustly interpreting those data and applying this knowledge to natural systems such petroleum sources, hydrothermal systems, and other systems containing organic-mineral-fluid interactions.

By developing a framework that connects laboratory experiments to natural systems, our goal is to understand how conditions governing molecule formation, reactivity, and long-term system evolution can produce the isotope signals found within organic carbon. Results will provide new molecular-scale constraints to enhance our understanding of reaction mechanisms, rates, and pathways that control carbon and associated element cycling in geochemical systems.

Publications supported by this project 2023-2025:

Selected items:

1. Cornelia Rasmussen & David Hoffman, "Fingerprinting Organofluorine Molecules via Position-Specific Isotope Analysis" *Environmental Science & Technology*, **2024**, 58, 13426-13433, <https://doi.org/10.1021/acs.est.4c02250>.
2. David Hoffman & Cornelia Rasmussen, "Position-specific carbon stable isotope analysis of glyphosate: isotope fingerprinting of molecules within a mixture" *Analytical and Bioanalytical Chemistry*, **2024**, 416, 3847-3856, <https://doi.org/10.1007/s00216-024-05326-5>.
3. Cornelia Rasmussen & David Hoffman, "Absolute carbon stable isotope ratio in the Vienna Pee Dee Belemnite isotope reference determined by ^1H NMR spectroscopy" *Analytical Chemistry*, **2022**, 94, 5240-5247, <https://doi.org/10.1021/acs.analchem.1c04565>.

Chemical and Rate Effects on Fracture

Alex Rinehart, Principal Investigator

Earth and Environmental Science Department, New Mexico Tech, Socorro, NM

Graduate Students: Bradley Ackroyd

Collaborators: Deep Choudhuri (NMT), Jamie Kimberley (Merrimack College), Dustin Crandall (NETL)

Overall research goals:

The fundamental goal of this research program is to place rate dependent fracture propagation into a general framework based on rates of energy dissipation from chemical work, chemomechanical coupling, mechanical work, and inertial and fluid feedbacks. More specifically we will understand how changing chemical and loading rate will impact the geometry and damage zone of mode-1 fracture as well as the atomistic mechanisms of this energy dissipation. We will use a combination of experimental measurements and molecular dynamics simulations of SiO₂ materials (quartz, silicate glass and quartzite rock) to place the chemical and rate effects into a kinetic and thermodynamic framework across 12 orders of magnitudes of rates. Water is the most common reactant leading to weakening (i.e., stress corrosion) at low to quasistatic rates. At high rates, fluid viscosity and surface tension provide additional feedbacks to inertial stress waves dissipating energy and slowing fracture propagation.

Experimentally, we will be using a combination of traditional laboratory fracture testing geometries (double torsion, drilled compression double cantilever, notched Brazilian) that will allow us to systematically vary fracture propagation rates from 10^{-9} m/s to 10^3 m/s. During these tests, the change in mechanical energy dissipation and damage zone geometries will be monitored. These tests will be conducted on a range of load frames, but two custom load frames will allow the integration of the different rates and geometries for verification. During tests, we will be holding temperature constant and will vary the activity of water from near non-existent to close to saturation levels. The water will be dissolved in air and in glycerin, allowing end members of effective diffusion coefficients, surface tension and viscosity to be examined – these are additional controls on fluid-feedbacks on fracture propagation. Monotonic and cyclic fatigue will be examined, allowing for differing flaw-lengths and off-crack damage to accrue prior to unstable rupture. Lastly, we will be testing a local fine-grained metamorphic quartzite and then also the silica-glass formed by melting the quartzite with <10% flux to (1) accelerate chemical kinetics, and (2) decrease the material flaw size.

For most samples, distributed surface strain fields will be monitored visually with low-rate and high-rate cameras appropriately, and will be inverted using commercial digital image correlation software. Comparison to analytical LEFM solutions and identification of visible fractures will allow for the quantification of process or damage zone extent. A subset of experiments will be conducted using X-ray computed tomography monitoring for creep, fatigue and quasi-static monotonic loading for both quartzite and quartzite-derived silicate glass samples. The inclusion of these 4D imaging techniques will allow a better quantification of process zone and other non-LEFM fracture styles as a function of the same variables as for the standard laboratory experiments.

Simulations will be performed in LAMMPS and VASP frameworks for force-field molecular dynamics simulations (FFMD) and *ab initio* simulations (AIMD). FFMD simulations will focus on the problem of fracture across rates and with varying pseudo-activities of water at the molecular scale. This work will be informed by recent work by J. Rudnicki at UTEP improving of force-fields for water-SiO₂ FFMD simulations, as well as applying machine learning to relevant AIMD

simulations. AIMD simulations will focus on the details of water-SiO₂ surface interactions and the change of properties of water in confinement and when SiO₂ lattices are stressed.

To handle the vast sea of spatiotemporal data that we are generating we are considering several options. Some of these are standard DIC and digital volume correlation methods. However, to interrogate the fracture process it appears that we will need to develop AI-accelerated methods to estimate potential fields for microstructural or mesh-free methods – methods that can handle fragmentation of samples naturally but struggle with calibration and computational efficiency.

Significant achievements during 2023- 2025:

- Successful development of staged XRCT imaging of fracture propagation under controlled relative humidity and temperature using modified DCDC fracture testing geometry at NETL.
- Acquisition of >500 GB of XRCT fracture imaging from 10 experiments.
- Recruitment of two graduate
- Acquisition of custom load frames and ‘shakedown’ experiments.
- Acquisition of high-temperature furnace.
- Recruitment of two graduate students (one with visa pending).
- Finding a source for fine-grained quartzite, which are not commonly commercially available, and chemical and mineralogical characterization of quartzite.
- Development of streamlined sample preparation methods.
- Development of temperature and humidity controls on creep to low-rate dynamic load frames.

Science objectives for 2026-2028:

Each of the following will lead to at least on manuscript that will be submitted prior to the end of the project.

- Develop sample preparation methods for glass samples.
- Finish DCDC, DT and notched Brazilian experiments for all activities of water in air and rates in laboratories at NMT, Merrimack College, in glass and
- Finish DCDC, DT and notched Brazilian experiments for all activities of water in glycerin and rates.
- Image 4D fracture processes across activities of water in air and glycerin, and a subset of rates at NETL and APS.
- Construct semi-automated analysis and simulation methods to quantify fracture networks from 2D and 3D datasets.
- Conduct FFMD simulations of quartz and silicate glass fracture using ML-derived potentials.
- Conduct AIMD simulations of confined water, water-SiO₂ interfacial interactions and similar modeling pathways.
- Begin synthesizing the results into a ‘phase-field’-like framework.

Development of X-ray Tomography for Subsurface Rock and Fluid Studies

Mark Rivers, Principal Investigator

Center for Advanced Radiation Sources, University of Chicago, Chicago, IL
X-ray Sciences Division, Argonne National Laboratory, Lemont, IL

Postdoctoral Scholars: Maksim Yakovlev

Collaborators: Yanbin Wang (University of Chicago), Wenlu Zhu (University of Maryland), Andrew Stack, Lawrence M. Anovitz and Julianne Weber (Oak Ridge National Laboratory), Parisa Shokouhi, Jacques Riviere (Pennsylvania State University), Giuseppe Buscarnera, Alessandro Rotta Loria (Northwestern University), Khalid Alshibli (University of Tennessee), Lazaro Perez Fonseca (Oregon State University).

Overall research goals:

This project seeks to make significant developments in the x-ray microtomography capabilities at GSECARS for studying rock/fluid interactions under subsurface conditions. The conditions we will be able to simulate while doing high-resolution imaging include those important for oil and gas extraction, for CO₂ sequestration, and for understanding shallow earthquakes. We will enable a wide community of scientists to rapidly obtain high-resolution images in complex sample environments vital to understanding earth's subsurface environment.

Expanding the range of pressure, temperature and stress conditions that can be probed, and maximizing the speed at which fluid rock interactions can be studied, will lead to new, high-level views of mineral-fluid chemical systematics, enabling broad predictive capabilities for understanding grain fracturing, porosity and fluid flow, as well as mineral dissolution and growth mechanisms. These new initiatives at elevated pressures and confined fluids will allow us to obtain important insight on macroscale subsurface phenomena critical to energy security, including fracture propagation in rock, energy resource management, and contaminant sequestration at the sub-micron scale.

Significant achievements during 2023- 2025:

One of the most significant achievements during this period was the construction, commissioning, and use of a new cell for in-situ tomographic imaging at shallow crustal conditions. The cell is based on a design called the Heitt Mjolnir from Ian Butler's group at the University of Edinburgh. It is designed for cylindrical samples 10 mm diameter and 20 mm long. It can operate at confining pressures up to 30 MPa, axial loads to 600 MPa, temperatures to 300°C, and with pore-fluid flow. We have extended the design to have both the original high-strength aluminum sample chamber and new PEEK sample chambers for much better X-ray imaging at lower confining and axial pressures. This system is now in use by several external user groups during each APS run cycle, and the first papers using the system have now been published. In addition, the pumping system we developed for it is being used by users who bring their own cell systems.

We have rewritten the tomography data processing software to improve the speed by a factor of 5, which is important for the high data rates at the facility. We have implemented a high-speed CoaXPress camera with up to 2200 frames/s to capture fast dynamic phenomena.

The post-doc on this project, Max Yakovlev, has continued work on producing semi-automated segmentation and visualization pipelines that are easy for users to adapt to highly variable micro-CT data using tools such as Ilastik and Dragonfly. The main advantages of our data workflow are its flexibility, which decreases the amount of introductory material needed to enable new users to process their images, and the robustness of individual models when used on similar datasets, which reduces the amount of training data and time required to set up automated segmentation pipelines.

Science objectives for 2026-2028:

We plan to develop two new major new capabilities for the geoscience community.

The first is a new cell, based on the Heitt Mjolnir design but with ultrasonic measurement capabilities. It will be able to do both active studies to measure sound velocity and attenuation as well as passive sensing for acoustic emission studies. The former is important for correlating changes in porosity and fractures measured with X-ray imaging with the seismically observable velocity and attenuation. Passive sensing is important for determining which fractures in a sample are failing in a manner that gives rise to acoustic emission events, where those occur in the sample, and the seismic moments from these events.

The second development will be the capability to perform X-ray diffraction on the samples in the Heitt Mjolnir cell with a rapid change between the imaging mode and diffraction mode. Because the path length through the sample is large (~10 mm) it is necessary to use Soller slits to limit the diffraction volume in the sample. We have already implemented a system that allows rapid switching between diffraction using a Pilatus 1M CdTe detector, and normal tomography mode for samples at ambient conditions. What is needed is a mounting system for the Soller slits that will allow the same capabilities for samples in the Heitt Mjolnir cell.

Publications supported by this project 2023-2025 (Selection):

1. Tommaso Mandolini, Zachary Zega, Mark L. Rivers, Wenlu Zhu, "Permeability enhancement by slow faulting under high pore fluid pressure" *Geophysical Research Letters*, **2026**, <https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2025GL119145>.
2. Juliane Weber, Carolyn G. Cooke, Vitalii Starchenko, Mark Rivers, Jan Ilavsky, Rui Zhang, Michael J. Zachman, Elliot P. Gilbert, Jitendra Mata, Andrew G. Stack, Lawrence M. Anovitz, "Reaction-Induced Fracturing of Porous Carbonate Rocks during Volume-Increasing Replacement by Witherite" *Cryst. Growth Des.*, **2025**, 25f, 6604-6619, <http://doi.org/10.1021/acs.cgd.5c00522>.
3. Mark L. Brusseau, Matthew E. Narter, Gregory Schnaar, Juliana Araujo, Justin Marble, "Fluid/Fluid Interfacial Areas Measured for Different Non-Wetting/Wetting Fluid Pairs in Natural Porous Media" *Environments*, **2025**, 12, <https://www.mdpi.com/2076-3298/12/10/380>
4. Yang Li, Shubjot Singh, Giuseppe Buscarnera, "X-Ray Microtomography Measurements of Coevolving Particle Size and Shape" *J. Geotech. Geoenviron.*, **2025**, <https://ascelibrary.org/doi/10.1061/JGGEFK.GTENG-13362>
5. Mohammad Safi, Khalid A. Alshibli, Haochen Li, "Influence of Pore Water on the Fracture of Silica Sand at Particle Scale" *J. Geotech. Geoenviron.*, **2025**, 12, <https://ascelibrary.org/doi/10.1061/JGGEFK.GTENG-12607>
6. Yize Pan, Dawa Seo, Mark Rivers, Xiaohui Gong, Giuseppe Buscarnera, Alessandro F. Rotta Loria "Microscopic insights into thermal cycling effects in granular materials via X-ray microtomography" *Granul. Matter*, **2024**, 26, <https://link.springer.com/article/10.1007/s10035-024-01468-6>
7. Rui Zhang, Paul A. Bosomworth, Juliane Weber, Jan Ilavsky, Si Athena Chen, Alexis Flores-Betancourt, Elliot Paul Gilbert, Jitendra Mata, Mark L. Rivers, Peter J. Eng, Lawrence M. Anovitz, "The role of annealing and grain boundary controls on the mechanical properties of limestones and marbles" *Int. J. Rock Mech. Mining Sci.*, **2024**, <https://www.sciencedirect.com/science/article/pii/S1365160924002910?via%3Dihub>
8. Zheng Li, Alexandra N. Kravchenko, Alison Cupples, Andrey K. Guber, Yakov Kuzyakov, G. Philip Robertson, Evgenia Blagodatskaya, "Composition and metabolism of microbial communities in soil pores," *Nat. Commun.*, **2024**, <https://www.nature.com/articles/s41467-024-47755-x>
9. Douglas E. Meisenheimer, Dorte Wildenschild, "Incorporating bubble evolution and transport in constitutive relationships for quasi- and non-equilibrium two-phase flows in porous media" *Front. Water*, **2023**, <https://www.frontiersin.org/journals/water/articles/10.3389/frwa.2023.1167872/full>

Unraveling the Physics of Earthquake Precursors Using Ultrasonic Imaging and Physics-Informed Machine

Jacques Rivière, Principal Investigator

Department of Engineering Science and Mechanics, Pennsylvania State University, University Park, PA

Graduate Students: Prabhav Borate, Mengyan Ding

Collaborators: Parisa Shokouhi, Chris Marone, Derek Elsworth

Overall research goals:

The objective of this research is to discover the mechanisms of precursory changes in rock and fault properties preceding laboratory earthquakes and to improve our understanding of earthquake precursors. Novel machine learning (ML) approaches are exploited and data from laboratory experiments designed to unravel the physics of earthquake failure. The research establishes a mechanistic understanding that connects precursory changes in acoustic wave features prior to laboratory quakes to fault heterogeneity, pre-slip and associated pre-seismic fault weakening. While the main focus is on improving our understanding of precursors to laboratory quakes, upscaling of the laboratory findings to the field will also be explored and techniques applicable to tectonic faults will be developed and tested with seismic catalogs and existing data.

Significant achievements during 2023-2025:

We have developed a Physics-Informed Neural Network (PINN) model that encodes fault physics in the deep learning loss function using active ultrasonic data collected during a friction experiment where stick-slip instabilities occur, the equivalent of earthquakes in the laboratory. PINN models outperform data-driven models and significantly improve transfer learning for small training datasets and conditions outside those used in training. This offers a promising path for machine learning-based failure prediction and, ultimately for improving our understanding of earthquake physics and prediction. This work has been published in Nature Communications (Borate et al, 2023).

In a second study, a more advanced physical model that incorporates the rate and state friction (RSF) laws is included in the machine learning model. In this work, the ultrasonic amplitude is assumed to be a proxy for the state variable, which can be thought as representing the average lifetime of contacting asperities along the fault surface. In the model, frictional parameters are set up as unknown constants in the model, and we find that they can be properly retrieved. This is promising, since currently, such parameters can only be extracted under stable sliding conditions, and this opens the doors for retrieval of frictional properties at larger scale. This work has been published in Scientific Reports (Borate et al., 2024).

We also collected ultrasonic data using dense arrays of ultrasonic sensors in both active and passive modes during friction experiments to study fault heterogeneity, as detailed in the following.

For the experiments in active mode, we find that changes in wave velocity do not vary greatly from sensor to sensor (located 1 mm apart), as opposed to changes in amplitude that do vary greatly. This suggests that the latter is greatly sensitive to the fault characteristics (asperities, roughness, etc), while wave velocity is more sensitive to the stress state of the host rock. The wave amplitude is found to vary greatly from sensors to sensors, with small variation over time during the stick-slip cycles. Temporal variations along the stick-slip cycles can be studied only after normalizing the data across the array. For some locations along the fault plane, little variation in wave amplitude can be seen, suggesting spectator regions. At some locations exhibiting larger changes, we first see a decrease during elastic reloading, and then a pre-seismic increase, while other sensors exhibit the opposite. Similar datasets are currently collected where heterogeneity is more precisely controlled, with strips of fine gouge placed at regular – and known – intervals on the fault plane.

A second dataset collected with an array in passive mode (acoustic emission) has been analyzed. We use a beamforming approach to locate all pre-seismic events to the mainshocks. The

beamforming approaches has been modified to account for both P- and S-waves, as well as changes in polarity observed for the P-wavefront. We are also carefully comparing the seismic signatures of pre-seismic events, where small asperities rupture along the fault plane, with those of the mainshock, where the entire fault plane ruptures. For some mainshocks, we find impulsive sub-events within them, which corresponds to ruptures of small asperities along the fault plane. A manuscript is under review on this topic.

Science objectives for 2026-2028:

Working with newer datasets where heterogeneity along the fault plane is more precisely controlled, we aim to understand the evolution of wave amplitude with respect to fault characteristics, and in particular discovering the presence – or lack – of precursors to failure near/away from gouge asperities. This will help us establish process-based models that link heterogeneity to pre-seismic activity and precursory changes.

We also plan to use another dataset collected in active mode on gouge material, and with a softer host material (acrylic), in an attempt to reduce the critical nucleation length and capture the full nucleation process using the ultrasonic array. We have started to use a reconstruction algorithm that can account for the different wave velocities in the system (acrylic – gouge – acrylic). By doing so, we are hoping to (1) properly visualize the sheared gouge layer throughout the seismic cycles, (2) quantify heterogeneity and associated precursory changes to failure.

Finally, the physics-informed ML models previously developed will be used and adapted to newer datasets – collected with the ultrasonic array, and where heterogeneity is better controlled – to determine which precursory changes are the most predictive of laboratory quakes and help us reveal the underlying micro-physical mechanisms.

Publications supported by this project 2023-2025:

1. P. Borate, J. Rivière, S. Marty, C. Marone, D. Kifer, P. Shokouhi, “Physics informed neural network can retrieve rate and state friction parameters from acoustic monitoring of laboratory stick-slip experiments.” *Scientific Reports*, **2024**, 14, 1–16, [10.1038/s41598-024-75826-y](https://doi.org/10.1038/s41598-024-75826-y).
2. L. Gao, P. Shokouhi, J. Rivière, “Effect of Grain Shape and Relative Humidity on the Nonlinear Elastic Properties of Granular Media” *Geophysical Research Letters*, **2023**, 50(11), e2023GL103245, [10.1029/2023GL103245](https://doi.org/10.1029/2023GL103245).
3. P. Borate, J. Rivière, C. Marone, A. Mali, D. Kifer, P. Shokouhi, “Using a physics-informed neural network and fault zone acoustic monitoring to predict lab earthquakes” *Nature Communications*, **2023**, 14, 3693, [10.1038/s41467-023-39377-6](https://doi.org/10.1038/s41467-023-39377-6).

Fundamental Mechanisms of Reactivity at Complex Geochemical Interfaces

Kevin M. Rosso, Principal Investigator

Co-PI's: Mavis Boamah, Eric Bylaska, Jim De Yoreo, Eugene Ilton, Sebastien Kerisit, Drew Latta*, Ben Legg, John Loring, Sebastian Mergelsberg, Duo Song, and Xin Zhang

Pacific Northwest National Laboratory, Richland, WA; *University of Iowa, Iowa City, IA

Graduate Students: Yifu Feng, Kit Joll, Juejing Liu, Ashley McNeil, Bavan Rajan, Heath Stanfield, Guomin Zhu

Postdoctoral Fellows: Narendra Adhikari, Yuna Bae, Colin Campbell, Ping Chen, Robert Felsted, Biao Jin, Xiaoxu Li, Winnie Liu, Marie Mortensen, Maxime Pouvreau, Xinyi Shen, Tom Underwood, Xiang Wang, Sichuang Xue, Yatong Zhao, and Jianbin Zhou

Collaborators: Jochen Blumberger (UCL)

Overall research goals:

Developing affordable, reliable, and secure energy options for the nation depends on our ability to safely and routinely access the energy, mineral, water, and storage resources in the Earth's subsurface. Central to this mission is our ability to accurately predict rates of the geochemical processes, such as mineral nucleation, growth, or dissolution, that govern how the subsurface responds to change. Current geochemical kinetics models are inadequate because they are based on thermodynamic equilibria for bulk minerals and solutions at idealized conditions, neglecting the real mechanistic complexity at mineral water interfaces. Important unknowns include the impacts of mineral defects and impurities, metastable intermediates in multi-step reactions, and the development of coatings or passivation layers on mineral grains.

The program at PNNL is systematically addressing these unknowns by developing a molecular-to-mesoscale understanding of how the structure, composition, and properties of reacting mineral-water interfaces change over time. Recent accomplishments include isolating key reaction intermediates controlling carbonate and redox-mediated metal oxide mineral transformations, uncovering the interfacial nanoscale forces that align mineral particles during aggregation, and quantifying relationships between impurity type, mode of incorporation, and mineral stability. We are now building on these insights pursuing new questions to understand how nanoscale chemical and electric field gradients control mineralization fronts, and how and when amorphous silica passivation layers limit carbon mineralization rates. Enabled by team science that integrates advanced experimental, theoretical, and computational capabilities at PNNL, our discoveries provide the foundation for new, accurate rate models based on mechanistic insight.

Significant achievements during 2023-2025:

Over our prior reporting period of July 2021 to July 2025, our program led or contributed to 123 peer-reviewed papers and book chapters in top domain and high impact journals; a complete compilation can be found [here](#). Major accomplishments include: 1) Our discovery that the accelerating effect of Fe(II) on Fh transformation to more crystalline Fe(III) oxyhydroxides entails labilization of sparingly soluble Fe(III) inspired new investigations which showed that the kinetics and properties of product phases depend on its rate of accumulation to a critical concentration, consistent with principles of classical nucleation theory, and also that undercoordinated Fe(III) sites on Fh surfaces may be an important source of labile Fe(III) after Fe(II) contact.¹ 2) We have uncovered how different divalent metal impurities have distinct impacts on the reactivity of goethite, by connecting detailed information on the coordination environment of impurities obtained from AIMD-informed EXAFS to the redox potential and stability of the host phase. Our work demonstrated that the measured difference in electrochemical potential between Ni- and Zn-doped goethite arises from the difference in strain energy associated with a nearby iron vacancy.² 3) We showed that the process of mineral growth through interfacially driven nucleation and oriented attachment to form complex mesocrystals significantly influences the properties of the resulting minerals. In particular, we found that, when mesocrystals formed via crystallization by particle attachment (CPA) are exposed to undersaturated conditions, they then undergo a process of dissolution by particle detachment (DPD). Applying a

numerical generalization of the Gibbs-Thomson effect, we showed that the physical drivers of DPD are curvature and strain inherently tied to the original CPA process. Based on the generality of the model, we anticipate that DPD is widespread for both natural minerals and synthetic crystals formed via CPA.³ 4) In the nucleation of metal carbonates, including during the formation of coatings, water is known to play a controlling role in directing mineralization pathways and stabilizing metastable amorphous intermediates. Our theoretical simulations of the dependence of the X-ray PDF on water content, n , for amorphous carbonates of experimentally relevant cations ($\text{MCO}_3 \cdot n\text{H}_2\text{O}$ systems, where $\text{M} = \text{Ca}, \text{Mg}, \text{Sr}$) showed good agreement with available experimental data, thus providing evidence for a lack of significant nanoscale structure in amorphous carbonates. Additionally, the molar volumes of amorphous calcium carbonate and strontium carbonate and several intermediate compositions were well predicted by an equation parameterized using the crystal structures of hydrated carbonates. This finding demonstrates that, like for anhydrous and hydrous crystalline carbonates, the molar volumes of amorphous carbonate solid solutions are additive with respect to water content and cation size, further evidence that water is a structural component of amorphous carbonates.⁴⁻⁵ 5) We identified the cause of a unique passivation effect during the carbonation of the basalt mineral forsterite (Mg_2SiO_4) in thin water films that condense from humid supercritical CO_2 . ^{29}Si MAS-NMR spectroscopy combined with TEM and EDS data of post-reacted particles indicated that the silica is present as a Mg-depleted layer just 2-3 nm thick, too thin to serve as a physical barrier to forsterite dissolution. Detailed in situ and ex situ IR spectroscopy experiments showed that the water film thickness decreased during the same time period as when the carbonation reactivity slowed, corresponding well with water film conductance measurements. All findings pointed to passivation by a loss in the amount of adsorbed water facilitating dissolution and ion transport.⁶

Science objectives for 2026-2028:

Subtask 1: Quantifying the role of reactive intermediates governing mineral redox reaction rates; Subtask 2: Developing a framework to predict how impurities impact mineral stabilities; Subtask 3: Understanding how chemical gradients and electric fields at mineral-water interfaces control nucleation and growth; Subtask 4: Understanding the nucleation, growth, and properties of secondary coatings and their impact on mineral reactivity; Subtask 5: Understanding passivation extent arising from compositional alteration of reacting surfaces; Crosscut: Build modern web-based automation tools by developing AI-assisted workflows for all theory and computational efforts across these five program subtasks.

Selected publications supported by this project 2023-2025:

1. Liu, J.; Sheng, A.; Li, X.; Arai, Y.; Ding, Y.; Nie, M.; Yan, M.; Rosso, K. M. Understanding the importance of labile Fe(III) during Fe(II)-catalyzed transformation of metastable iron oxyhydroxides. *Environ. Sci. Technol.* **2022**, DOI: [10.1021/acs.est.1c08044](https://doi.org/10.1021/acs.est.1c08044).
2. Latta, D. E.; Mergelsberg, S. T.; Song, D.; Bylaska, E. J.; Scherer, M. M.; Popejoy, B.; Saslow, S. A.; Catalano, J. G.; Ilton, E. S. Effect of impurities on the redox properties of goethite. *Environ. Sci. Technol.* **2025**, DOI: [10.1021/acs.est.4c13480](https://doi.org/10.1021/acs.est.4c13480).
3. Zhu, G.; Legg, B. A.; Sassi, M.; Liang, X.; Zong, M.; Rosso, K. M.; De Yoreo, J. J. Crystal dissolution by particle detachment. *Nature Comm.* **2023**, DOI: [10.1038/s41467-023-41443-y](https://doi.org/10.1038/s41467-023-41443-y).
4. Mergelsberg, S. T.; Prange, M. P.; Kerisit, S. N. Additive molar volumes in amorphous ca/sr carbonate solid solutions. *J. Phys. Chem. C* **2024**, DOI: [10.1021/acs.jpcc.3c08311](https://doi.org/10.1021/acs.jpcc.3c08311).
5. Prange, M. P.; Mergelsberg, S. T.; Kerisit, S. N. Structural water in amorphous carbonate minerals: ab initio molecular dynamics simulations of X-ray pair distribution experiments. *Phys. Chem. Chem. Phys.* **2023**, DOI: [10.1039/d2cp04881g](https://doi.org/10.1039/d2cp04881g).
6. Rajan, B. P.; Mergelsberg, S. T.; Bowden, M. E.; Peterson, K. A.; Burton, S. D.; Bowers, G. M.; Wietsma, T. W.; Graham, T. R.; Qafoku, O.; Thompson, C. J.; Kerisit, S. N.; Loring, J. S. Decreasing hygroscopicity slows metal silicate carbonation under low-water conditions. *Environ. Sci. Technol.* **2025**, DOI: [10.1021/acs.est.5c01695](https://doi.org/10.1021/acs.est.5c01695).

Understanding Multicomponent Fluid Effects on Permeability Evolution in Fractures

Kevin M. Rosso, Principal Investigator

Co-PI's: Xin Zhang, Xiaoxu Li

Postdoctoral Fellows: Juejing Liu

Pacific Northwest National Laboratory, Richland, WA

Collaborators: Lu Ma (NSLS-II), Luxi Li (APS), and Wenjun Liu (APS), Mark Rivers (APS)

Overall research goals:

Flow of aqueous solutions in the Earth's subsurface is central to the development of energy and critical mineral resources on which modern society increasingly depends. Permeation of hydrothermal fluids through the small apertures of natural fractures is the basis for many primary critical mineral deposits over geologic history. Likewise, hydrothermal fluid circulation through fracture networks is the cornerstone process for extracting deep heat via natural or engineered geothermal systems (EGS). A critical factor spanning these examples is the necessity for fluid flow to be sustained at sufficient rates over sufficiently long periods of time to ultimately obtain an economically viable resource. This is particularly important to the success of EGS, where return on investment timelines of ten or more years of heat production are required. However, because these fluids are often far from equilibrium from their host rock, flow gradually redistributes minerals and metals through dissolution and precipitation reactions. This evolving microstructure in turn alters the flow pathways and the ability to sustain flow. This well-known hydrogeochemical coupling arises from the fact that solute concentrations locally at mineral-fluid interfaces are controlled by rates of advection, diffusion, and reaction. Thus, to accurately predict permeability evolution requires comprehensive treatment of the dynamics of these coupled processes.

Current pore and continuum scale numerical modeling approaches are implemented with accurate advection-diffusion mass transport equations but rely on relatively simplistic treatment of geochemical reaction rates. Unitless rate ratios (e.g., Peclet number, Damköhler number, etc.) are typical inputs to describe the balance of processes dominating the solute flux. But for realistic, compositionally complex aqueous fluids, these models fail to predict how dissolution and precipitation impact the connected porosity that controls permeability. Metal carbonates such as calcite (i.e., carbonate scale) provide a canonical example. Common divalent cations such as Mg^{2+} , Sr^{2+} , Fe^{2+} , and Mn^{2+} strongly impact nucleation, polymorphism, and the growth behavior of calcium carbonate minerals by either solubility effects or by competing with Ca^{2+} for binding sites. Recent studies also show that even at undersaturation calcite dissolution rates can be limited by an armoring effect from isomorphic substitution of calcium in surface layers. These important impacts of competitive cations, as well as anions, on the microstructural evolution of metal carbonates comprise a major source of uncertainty in reactive transport simulations designed to predict how permeability evolves during hydrothermal fluid flow.

Science objectives for 2026:

This exploratory one-year project aims to show that we can establish a mechanistic understanding of how fluid composition variables control permeability by integrating data-rich insights from in situ real-time nano-to-microscale imaging and morphology characterization of mineralization during flow with machine learning and pore-scale numerical simulations. Our objectives are to:

- 1.) Obtain a systematic understanding of how competing cations impact dissolution and growth rates of calcite single crystal surfaces at the nanoscale. Numerous studies have explored the effects of various individual cations on growth of calcite surfaces but comparatively few have examined impacts on dissolution, and no systematic relationship between cation type and growth or dissolution rates currently exists. We have an opportunity to design select experiments to connect existing observations and establish trends based on cation properties.

2.) Learn how these trends impact larger length and time scale phenomena such as the morphological evolution of polycrystalline calcite microstructures. Couple nanoscale observations to microscale reactive flow experiments using well-defined channels to specifically examine how porosity and permeability depend on the cationic composition of the fluid.

3.) Build a conceptual model that connects nanoscale mechanistic insights to the dynamics of microstructure evolution that can help inform and validate reactive transport simulations of coupled advective-diffusive-reactive processes.

To achieve our objectives, we are performing a combination of atomic force microscopy (AFM) studies with hydrothermal reactive flow studies that use microscale capillaries at synchrotron X-ray beamlines with ML-based imaging and spectroscopic data analyses. Our approach takes advantage of existing personnel, access, and team momentum in these three capability areas, and focuses on a select set of conditions that can be uniformly applied across the experiment types to ensure substantial progress within a year's duration. Specifically, we are focusing on calcite at circumneutral pH, in variably saturated air-equilibrated solutions that contain variable ratios of Mg^{2+} , Mn^{2+} , Co^{2+} , and Ni^{2+} cations. The latter cations are both geochemically relevant critical metals as well as high enough AMU to enable our hard X-ray in situ microscale studies. The team is comprised of PNNL staff Drs. Xin Zhang and Xiaoxu Li for synchrotron-based capillary hydrothermal flow experiments, and postdoc Dr. Juejing Liu for ML-enabled image and spectroscopic data analyses. These beamline experiments are being enabled through user proposals with Drs. Lu Ma (NSLS-II), Luxi Li (APS), Wenjun Liu (APS), and Mark Rivers (APS). Success of this exploratory work will demonstrate an approach that can be carried forward into more complex systems and questions relevant to fracture flow systems in real field settings

We are performing in situ multi-spectroscopic studies of calcite-centric metal carbonate dissolution and precipitation in the presence of co-cations during flow under hydrothermal conditions (up to 250 °C 65 MPa) to characterize: 1) the morphologic and crystallographic development of polycrystalline carbonate microstructures within geometrically well-defined microscale capillaries, 2) porosity and permeability changes over time resulting from precipitation and dissolution, 3) the spatial solution-chemistry evolution coupling with this precipitation and dissolution, including the pH, cation concentrations, and local coordination change of cations. The beamline measurements will be complemented by laboratory-based measurements using a novel hydrothermal flow cell under well-defined fluid pressure with UV-vis/Raman microspectroscopy.

We will apply ML-assisted data analysis methods to quantitatively extract mechanistic information from large imaging and spectroscopic datasets obtained across the nano- to microscale experiments. At the nanoscale, deep learning-based feature recognition (e.g., convolutional neural networks) will be used to identify surface step morphologies, etch pits, and compositional heterogeneity from high-resolution AFM phase contrast and topographic images. We will train segmentation models to classify growth/dissolution regimes and quantify step velocities under different ionic compositions, enabling a statistical analysis of co-cation competition effects on surface dynamics. At the microscale, we will use ML algorithms to process 2D and 3D datasets from synchrotron-based XRF, XRD, and full-field imaging. For example, unsupervised clustering of XRF and UV-vis spectral data will help identify spatial gradients in cation concentrations and pH evolution during flow. Morphological quantification of polycrystalline carbonate aggregates—including grain size, inter-granular connectivity, and pore structure—will be performed using ML-based image reconstruction and segmentation of tomographic datasets, allowing us to compute key properties such as total and connected porosity over time. In particular, we will leverage our recent success in applying multi-task deep learning models for accurate and data-efficient classification of micro-XRD mapping datasets under hydrothermal conditions. This approach enables automated reconstruction of crystallographic structure and spatial phase distribution from large-volume datasets, accelerating the analysis of time-resolved reactive flow experiments. These data-driven insights will be integrated into a conceptual framework for permeability evolution under competitive ion environments. Using outputs from image analysis and spectroscopic deconvolution, we will parameterize pore-network models and inform upscaled continuum-scale simulations of reactive transport. This multiscale coupling will allow us to directly connect cation-specific reaction dynamics to macroscopic permeability evolution under hydrothermal flow, providing a predictive framework that can be extended to more complex mineral-fluid systems.

MARBLE-C: Machine leArning Rheology of Brittle-ductiLe Experiments and Crust

Seth Saltiel, Principal Investigator

Earth and Atmospheric Sciences, Cornell University, Ithaca, NY

Co-PIs: Tushar Mittal (Penn State University), Benjamin Holtzman (Lamont Doherty Earth Observatory), Anna Barth (Strabo Engineering)

Graduate Students, Postdocs, and Research Scientists: Pascal Caraccioli Salinas, Congcong Yuan (Cornell), Kangan Li, Edo Nurshal (Penn State), Eric Beaucé (LDEO)

Overall research goals:

Geothermal energy extraction and electricity conversion is significantly more efficient as resource temperature increases, enabling cost-competitive, baseload power generation, using fewer, more energy-dense wells. However, at higher temperatures (deemed superhot when approaching water's critical point), deeper in the crust, fracturing and fluid flow become more complex and less well understood. Upon reaching the brittle-ductile transition zone, rocks start to "flow" (visco-plastic behavior), significantly altering the stresses required to engineer fracture networks. Furthermore, rocks - made of mineral aggregates - are highly variable in their mechanical properties, and it is not straightforward to develop effective rheology laws from lab datasets. This project focuses on hydrofracture and hydroshear processes in fracture networks, considering the dynamic coupling between fluid flow and stress at static temperature conditions. Machine learning (ML) offers a promising approach for developing robust, extrapolatable models to predict the rock's responses to various engineering activities. We are developing a new approach to predict material response with ML design that incorporates the laws of thermodynamics and integrates mechanical and fluid-flow, as well as seismic, data. We extensively train and test the ML model using well-controlled laboratory experiments with detailed measurements. Informed by insight into physical processes across a range of material conditions, we then apply this approach to field-scale data from actively engineered subsurface reservoirs. When properly trained, the ML model can be used in real-time to inform decision-making, such as avoiding unwanted fractures, potentially causing damaging seismicity and/or short-circuiting fluid flow, which can limit heat extraction.

Significant achievements during 2024-2025:

The project is based on the development and validation of a novel constitutive model for subsurface fracture networks undergoing coupled thermo-hydro/poro-mechanical (THM) processes. The model ensures thermodynamic consistency by using a Thermodynamics of Irreversible Processes informed Neural Network architecture (TIP-INN) to learn rheology directly from stress-strain laboratory data, utilizing the full loading path history directly. This data-driven approach does not pre-assume a rheological model and requires only limited training data. In addition to predicting the given stress-strain data, the method provides accurate predictions of the thermodynamic potentials underlying deformation without explicit training, thereby enabling more robust extrapolation and the calculation of material properties such as the elastic modulus and viscosity. We demonstrate the general applicability of this framework by using a range of loading histories across different standard rheological models, as well as laboratory experiments with complex internal-variable evolution (Li et al., *in prep*). We also test the model on laboratory deformation data of a complex porous material, demonstrating that sophisticated acoustic features are needed to capture the material's internal state. We have also successfully incorporated epistemic uncertainty quantification, enabling users to have a robust estimate of the model's accuracy when extrapolated beyond the training conditions.

Acoustic data processing work has focused on ground-truthing unsupervised ML methods to extract features from transmitted ultrasonic waveforms and spectrograms that best characterize fracture-network (or damage-distribution) configurations. These methods offer considerable promise for interpreting variations in complex, spatiotemporally varying geophysical wavefields, but so far lack

benchmarking analysis. We use synthetic and controlled-experiment waveforms to examine which features and under what conditions we can detect changes in the properties of the fracture network and the seismic source. Our initial results using supervised learning show that a single sensor's waveform is sufficient to distinguish among different damage types and damage intensities, based on the structure of the scattered coda waveforms. However, unsupervised approaches have shown limited success, and we are evaluating various time-series and spectrogram feature-generation algorithms on our benchmark for robust feature selection (Barth et al., 2025).

In order to train and test the model framework with well-controlled and detailed measurements of relevant experimental settings, we have set up a new laboratory facility (at Cornell) focused on acoustics, mechanics, and fluid flow in fracture networks. This includes the ability to 3D-print highly controlled fracture networks in transparent acrylic (a common rock analog material with much lower brittle-ductile transition temperature) and monitor their mechanical properties with high-rate acoustic and visual imaging during compressive and torsional stressing (a couple MPa) under a range of temperature (up to 350°C) and fluid pressure (up to 6000 psi) conditions. Recent experiments characterize acoustic waveforms transmitted through different fracture network configurations and their sensitivity to fluid, temperature, and stress conditions (Yuan et al., 2025). These controlled mechanical and acoustic datasets will serve as train/test data for acoustic processing and TIP-INN.

Science objectives for 2026-2028:

In the second half of our three-year project timeline, we will integrate all components to complete the validation of TIP-INN. We have begun experiments on our 3D printed fracture networks using TIP-INN to design optimal stressing time-series for the prediction of sample rheology over a range of temperatures, through the brittle-ductile transition. Using our optimized and validated acoustic data processing workflow, we will identify the acoustic features that best diagnose fracture network configuration and apply them to the experimental acoustic data as a proxy for the internal state. This will demonstrate TIP-INN's capability as a real-time rheology model for predicting the mechanical response of fracture networks. We are also working on incorporating TIP-INN into a continuum representation for reservoir modelling.

Further theoretical work has shown that TIP-INN can readily extend to fluid-flow data and facilitate the prediction of permeability evolution. In addition to stress-strain mechanical data, we can use the fluid pressure and flow rate data within the thermodynamic potentials and learn the dynamic permeability with the model, analogous to the time-dependent viscosity learned for viscoelastic models. The next project period will include numerical development to incorporate this into the neural network architecture and laboratory experiments to collect data on varying fluid pressure and flow rate, which will feed into these expanded TIP-INN capabilities.

We also plan to apply TIP-INN to field datasets and have already acquired and processed seismic catalogs from active and prospective superhot geothermal sites, using ML-based feature extraction to characterize spatial and temporal patterns in seismicity (Barth et al., 2026).

Selection of Publications and Conference Abstracts supported by this project 2024-2025:

1. Li, K., Mittal, T., Saltiel, S., Holtzman, B., "Thermodynamic Consistent Neural Networks for Constitutive Modeling: a new data driven approach", *Journal of Geophysical Research*, **in prep**
2. Barth, A., Beaucé, E., Hjörleifsdóttir, V., Saltiel, S., Mittal, T., Holtzman, B., "Seismicity Across the Supercritical-to-Steam Transition: Comparative Patterns Above Two Distinct Heat Sources in the Hengill Geothermal Field, Iceland", *Proceedings World Geothermal Congress*, **2026**
3. Barth, Holtzman, Beaucé, Mittal, Saltiel, "Detecting Changes in Seismic Source and Fracture Network Morphology with Unsupervised Machine Learning on Synthetic Wavefields", *AGU25*, **2025**
4. Yuan, C., Saltiel, S., Mittal, T., Barth, A., Beauce, E., Holtzman, B., "Seismo-Acoustic Monitoring of Hydro-Thermal-Mechanical Processes in Fractured Rock Analogs", *AGU25*, **2025**

Coseismic chemistry: Thermal maturation of organic matter at short timescales

Heather Savage, Pratigya Polissar, co-Principal Investigators

Department of Earth and Planetary Sciences, University of California, Santa Cruz

Department of Ocean Sciences, University of California, Santa Cruz

Graduate Students: Miranda Baker

Overall research goals:

Earthquakes are one of the most unpredictable and destructive of Earth processes. Whether earthquakes nucleate and continue growing into large events is dictated by conditions within the fault zone such stress, rock strength, and structural heterogeneity. However, earthquakes mostly occur deep within the Earth making direct observation impossible. Therefore, investigation of exhumed faults and faults recovered through drilling is required to study paleoearthquakes and in situ conditions along the fault during past slip. Because earthquakes dissipate some amount of energy as frictional heat, temperature in fault zones should rise dramatically during earthquakes (from hundreds up to over a thousand °C) compared to the surrounding rocks. We investigate paleoearthquake slip in fault zones, in order to better understand the conditions for earthquake nucleation and propagation, using the thermal maturity of organic matter as a novel earthquake temperature proxy. Our DOE work thus far has three major objectives: 1) development of geolipidomics, 2) creation of new instrumentation and execution of experiments that allow for the development of the kinetics of organic thermal maturation at earthquake timescales and temperatures, and 3) determination of earthquake temperature in specific faults.

Significant achievements during 2023- 2025:

Development and evaluation of geolipidomics. Lipids are useful for studying ancient environments and geologic processes because they are common and often well-preserved in sedimentary rocks. Traditional chemical analyses of these lipids focus on specific, known compounds linked to past organisms or conditions. In contrast, biological analyses regularly use a non-targeted lipidomics approach that searches for all detectable lipids to discover new biomarkers. Here we have developed “geolipidomics”, the application of analytical chemistry techniques and data processing pipelines of the biological science’s “-omics” fields to answer environmental and Earth science questions. However, these pipelines need to be tested and adapted because lipids in rocks are more challenging complex mixtures. We compared four non-targeted methods of extracting features consisting of a peak area and mass spectrum from raw gas chromatography mass spectrometry data. We used samples from hydrous pyrolysis experiments on an organic-rich shale and compared targeted molecular analyses and untargeted feature results. This comparison highlighted one method (MSHub) as the most robust computational tool able to separate the most challenging features (coeluting and spectrally identical). Exploration of the MSHub results revealed a wide number of molecular indices affected by heating and we illustrated a systematic way to identify new thermal maturity indices in this non-targeted feature space.

Development of rapid heater and new kinetics for methylphenanthrene transformation. We fabricated a unique rapid heating apparatus that can reach temperatures of 600 °C within minutes in the presence of water. Most heating apparatuses are meant to investigate longer term processes and heat up slowly, or very short timescales but without containment and hence without water as a reactant. Our work on thermal maturation during earthquakes requires earthquake conditions (heating at temperatures up to 1000 °C over a few seconds to minutes) with water-saturated samples. Our new apparatus has allowed us to conduct experiments that expand on our previous work on the kinetics of methylphenanthrenes, a common compound found in sedimentary rocks that have been buried a few kilometers. Our new experiments are in general agreement with our previous work and we are currently conducting detailed kinetics analysis of the results.

New measurements of thermal maturity on the San Andreas fault. The central section of the San Andreas fault (SAF) creeps slowly and has not hosted a large, historic earthquake. An outstanding question for seismic hazard is whether this fault section can participate during a large earthquake, connecting the seismogenic northern and southern sections and creating a San Andreas system-spanning event. Our previous work on a drillcore through the SAF at 3 km depth shows that there are parts of the fault zone with high thermal maturity, indicating hundreds of earthquakes have occurred in the creeping zone over the last few million years. However, sizable errors from biomarker-based temperature estimates on large 5-10 g bulk samples have prevented further analysis. Our new work on milligram samples taken from localized slip zones, which are tiny in volume, show that earthquake temperatures are higher than our original estimates indicating larger slip events likely occurred.

Science objectives for 2026-2028:

Identification of geolipidomics features with machine learning. A major challenge in geolipidomics is the annotation and identification of unknown features. For example, in our work to-date, only 11% of features had matches in published mass spectral databases. Annotation and/or identification is essential to discover new lipid biomarkers and apply them to geologic samples. We plan to use machine learning approaches to the inverse task of inferring molecule identity/properties from mass spectra and the forward task of predicting mass spectra from candidate molecular structures. A second challenge is the evaluation of false positives (noise identified as features) in non-targeted analysis. We plan to use supervised ML tools for post-pipeline annotation of feature tables with confidence in extracted feature areas.

Chemical kinetic analysis of thermal transformation on non-targeted features. Our geolipidomics pipeline and rapid hydrous pyrolysis experiments provide an opportunity to investigate the kinetics of molecular biomarker transformations. This requires developing a framework for kinetics analysis (equilibrium concentrations, reaction order) that is amenable to the feature tables from geolipidomics. The outcome will be (1) kinetic parameters (reaction order, activation energy and pre-exponential constants) of thousands of features and (2) relationships between molecular structure/properties and kinetic behavior. These outcomes will enable application of a wide range of new lipid biomarker indices to studies of organic matter thermal transformation.

Application of geolipidomics to the San Andreas Fault. As in other aspects of our work, the application of geolipidomics to our work on the San Andreas fault will allow us to find more features within our samples that are thermally transformed. Multiple thermal proxies will allow for more accurate determination of earthquake temperature, size, and timing.

Broader Earth science applications of the geolipidomics of thermal maturity. We plan on porting our knowledge of thermal maturation of organic material to other fields including wildfire histories and hydrocarbon contaminant mapping and transformation in the natural environment.

Publications supported by this project 2023-2025:

Use this format (Vol, Pages, are optional) and hyperlink the DOI:

1. Baker, M., P. Polissar, H. Savage, "Geolipidomics: Non-targeted Analysis of Rapidly Heated Geologic Samples with Gas Chromatography-Mass Spectrometry" *JGR – Machine Learning and Computation*, **under revision**, [10.22541/essoar.175308977.75842778/v1](https://doi.org/10.22541/essoar.175308977.75842778/v1).
2. Baker, M., P. Polissar, H. Savage, "Untargeted Analysis of Geologic Samples with Machine Learning" *International Meeting on Organic Geochemistry*, **2025**, Volume 2025, p.1 – 2, [10.3997/2214-4609.202533093](https://doi.org/10.3997/2214-4609.202533093).

Physics-based Induced Earthquake Forecasting: Process Understanding, and Hazards Mitigation

Manoochehr Shirzaei, Principal Investigator

Department of Geosciences, Virginia Tech, Blacksburg, VA, USA

Graduate Students: Guang Zhai, Grace Carlson, Chandra Ojha, Sui Tung, Mohammad Khorrami, Mahmoud Reshadati, Sonia Zehsaz

Collaborators: Michael Manga (UC Berkeley), Susanna Werth (Virginia Tech), Thomas Goebel (University of Memphis), Roshan. Koirala (University of Memphis), Grzegorz Kwiatek (GFZ Germany), Emily Brodsky (UC Santa Cruz), Trenton Cladouhos (Stoneway Geothermal), Michael Swyer (CyrQ Energy), Ole Kaven (US Geological Survey), Timothy Masterlark (South Dakota School of Mines), Herbert F. Wang (University of Wisconsin–Madison), Wei-Chuang Huang (US Navy Geothermal Program Office), Kurt L. Feigl (University of Wisconsin–Madison)

Overall research goals:

Advancing satellite Synthetic Aperture Radar Interferometry (InSAR) to monitor surface deformation driven by subsurface fluid injection, and developing coupled physics-based and data-driven frameworks to quantify the relationship between injection processes and induced earthquake occurrence, magnitude distributions, and seismic probability.

Significant achievements during 2023- 2025:

This project advanced the use of spaceborne and ground-based observations to quantify coupled hydrologic, mechanical, and seismic processes across water-stressed and energy-producing regions. We expanded the application of Sentinel-1 Interferometric Synthetic Aperture Radar (InSAR) to routinely detect millimeter-scale vertical land motion associated with elastic crustal loading, poroelastic deformation, geothermal reservoir compaction, and fault slip over large spatial domains and multi-year periods. These observations enabled direct monitoring of drought-driven water loss, groundwater depletion, and geothermal reservoir long-term deformation at resolutions not achievable with conventional hydrologic measurements.

We developed integrated multi-sensor observational frameworks combining InSAR with GNSS, in-situ pressure measurements, and seismic catalogs. This integration allowed separation of elastic loading, poroelastic deformation, and tectonic signals, providing consistent constraints from local to regional scales. In parallel, we introduced new multitemporal InSAR processing algorithms optimized for large SAR data stacks, including improved interferometric pair selection, statistically robust pixel selection, and advanced tropospheric correction. These developments significantly reduced atmospheric noise, decorrelation, and closure errors, achieving sub-centimeter agreement with independent GNSS observations.

Building on these advances, we developed a suite of physics-based and hybrid modeling tools that directly link underground fluid mass changes, surface deformation, and seismicity. We implemented inverse elastic load models to translate observed deformation into spatially resolved estimates of water storage change. This approach was extended through a novel joint inversion framework that assimilates GNSS and InSAR observations while explicitly accounting for both elastic crustal loading and poroelastic aquifer deformation, yielding high-resolution estimates of total and fluid storage change with quantified uncertainties.

To investigate recharge processes, we combined geodetic and hydrologic observations with process-based diffusion and flow models, enabling discrimination between pressure propagation and actual fluid flow across mountain-to-basin systems. For geothermal environments, we developed finite-element poroelastic and thermo-mechanical models coupled with Coulomb stress analysis to capture the effects of long-term production, fluid migration, abrupt operational

changes, thermal contraction, and aseismic fault slip on deformation and seismicity. Probabilistic stress-based modeling was further used to evaluate how anthropogenic fluid injection perturbs earthquake timing on major faults.

The integrated observational and modeling advances yielded several key scientific insights. First, we demonstrated that severe droughts propagate through coupled surface–groundwater systems, producing measurable elastic and poroelastic responses that reveal both total water loss and subsurface hydraulic connectivity. Groundwater accounted for a substantial fraction of drought-driven water loss, with spatially variable connectivity controlling delayed system responses and reducing groundwater buffering capacity during prolonged drought. Second, we established that high-mountain snowpack plays a first-order role in recharging deep basin aquifers through mountain block recharge. Observed delays between snowmelt, groundwater levels, and groundwater storage indicate that deep aquifer replenishment depends on long flow paths through fractured bedrock, a mechanism largely absent from current hydroclimate and groundwater management models. Third, we showed that induced seismicity and deformation in geothermal systems arise from multi-mechanism coupling, not solely from pore-pressure changes. Short-term seismicity spikes are driven by poroelastic stressing during rapid operational changes, whereas long-term deformation reflects aseismic fault slip and thermal contraction. Finally, we provided evidence that anthropogenic fluid injection can alter earthquake timing on major tectonic faults, underscoring the need to incorporate human-induced stress perturbations into time-dependent seismic hazard assessments.

Science objectives for 2026-2028:

The overarching objective of the proposed 2026–2028 research is to advance national-scale understanding and prediction of geothermal-system deformation and induced seismicity through integrated observation-driven and physics-informed modelling. First, we will produce the a nation-wide, high-resolution map of surface deformation at geothermal sites across the United States, leveraging long-term Sentinel-1 InSAR, GNSS, and ancillary operational data to systematically quantify deformation rates, temporal evolution, and associated uncertainties. Second, we will develop and validate artificial intelligence models that link fluid injection and production histories to surface deformation and induced earthquake occurrence, including event timing, magnitude, and size–frequency distributions. These data-driven models will be rigorously benchmarked against established physics-based models to assess predictive skill, physical consistency, and generalizability. Together, these objectives will establish a new, scalable framework for operational monitoring and forecasting of geothermal-induced deformation and seismic hazards, directly supporting safer and more sustainable geothermal energy deployment.

Publications supported by this project 2023-2025:

Selected publications include:

1. Mohammad Khorrami, Susanna Werth, Sonia Zehsaz, Manoochehr Shirzaei, “Drought-induced changes in groundwater-surface water exchange at Lake Mead area”, *Journal of Hydrology: Regional Studies*, **2025**, 1, 62, <https://doi.org/10.1016/j.ejrh.2025.102996>
2. Sui Tung, Ole Kaven, Manoochehr Shirzaei, Timothy Masterlark, Herbert F. Wang, Wei-Chuang Huang, Kurt L. Feigl, “Seismicity zoning at Coso geothermal field and stress changes from fluid production and migration”, *Earth and Planetary Science Letters*. **2024** 15, 646, <https://doi.org/10.1016/j.epsl.2024.119000>
3. Roshan Koirala, Grzegorz Kwiatek, Manoochehr Shirzaei, Emily Brodsky, Trenton Clad ouhos, Michael Swyer, Thomas Goebel, “Induced seismicity and surface deformation associated with long-term and abrupt geothermal operations in Blue Mountain, Nevada”, *Earth and Planetary Science Letters*. **2024**,1, 643, <https://doi.org/10.1016/j.epsl.2024.118883>.

Quantitative Assessment of the couplings among frictional, seismic, and transport properties of fractured rock

Parisa Shokouhi, Principal Investigator

Engineering Science and Mechanics, The Pennsylvania State University, University Park, PA

Graduate Students: Clay Wood, Prabhav Borate, Evan P. Bozek, Asef I. Sadaf, Ali Bastani Lay

Collaborators: Jacques Rivière, Chris Marone, and Derek Elsworth (Penn State), Christopher W. Johnson, Bryan J. Euser (LANL), Mark Rivers (University of Chicago)

Overall research goals:

The goal of this project is to illuminate the fundamental coupled processes governing unconventional and geothermal energy production, CO₂ sequestration, hydrogen storage and nuclear waste disposal through an integrated experimental and numerical study. Frictional and deformational properties of subsurface fractures control permeability, influence recovery rates of water, energy and fuels and impact the safety and longevity of waste isolation. There is a critical need to remotely image fractures and faults across multiple scales and quantify factors controlling fluid-injection induced seismicity. The proposed research focuses on quantitatively understanding the mechanisms behind shear reactivation and fluid-injection earthquake triggering including the evolution of the elastodynamic and flow properties prior to, during and after shear failure. To that end, we have designed and built a setup to combine measurements of frictional constitutive behavior, active-source ultrasonic, passive acoustic emissions, and fluid flow of fractured rock under stress with time-lapse in-situ synchrotron X-ray imaging and ex-situ quantitative fracture characterization. The in-situ synchrotron X-ray imaging is key in quantifying the evolution of fracture aperture and contact area under stress and in interpreting the measured frictional, elastodynamic and permeability responses. In addition, high-resolution X-ray images are used to create 'digital fractured rocks' - microstructure-informed numerical models that closely emulate the elastic, elastodynamic and hydraulic properties of fractured rock as well as micro-seismicity during shear - to eventually allow upscaling of our laboratory results and understand reservoir-scale behavior.

Significant achievements during 2023- 2025:

- Designed and fabricated a novel pressure cell that allows simultaneous in-situ X-ray imaging, ultrasonic and acoustic recording (laboratory-scale seismic and seismicity data) as well as flow measurements under both quasi-static and dynamic loading [*completed*].
- Conducted three (3) sets of sophisticated experiments (March 2023, October 2024, November 2025) at Argonne National Laboratory (ANL) that coupled synchrotron X-ray imaging and ultrasonic/acoustic testing of fractured Westerly granite and Berea sandstone samples to (i) quantitatively map the recorded ultrasonic response of fractured rock on fracture characteristics such as contact area, contact size distribution, and fracture aperture [*completed*]; (ii) understand the contribution of rock bulk versus fracture to the measured linear and nonlinear elastodynamic response of fractured rock [*data analysis near completion*]; (iii) constrain the mechanisms and acoustic precursors of fault's shear activation [*data interpretation near completion*]; and (iv) understand the mechanistic controls on fluid injection-induced fault reactivation and related acoustic precursors [*data analysis ongoing*].
- Conducted microstructure-informed numerical simulation of linear and nonlinear elastic wave propagation in 'digital fractured rock' to help interpret, expand, and upscale the laboratory-scale experimental results. Two distinct sets of numerical simulations were conducted: (i) two-dimensional X-ray images were directly used to create a numerical model of the fractured Berea sample (without considering bulk porosity) and to simulate the corresponding linear ultrasonic

response using finite element analysis [*completed*]; and (ii) in collaboration with LANL scientific and through a SCGSR fellowship (September 2025 – December 2025), conducted finite-discrete element (FDEM) simulations using HOSS (Hybrid Optimization Software Suite) to simulate the nonlinear elastodynamic response of fractured rock and investigate how it evolves with fracture aperture, contact area and its distribution considering bulk porosity [*ongoing*].

- Analyzed acoustic emissions (i.e., event detection, localization, focal mechanisms identification) and active-source ultrasonic data (i.e., ultrasonic tomography) recorded using an array of ultrasonic transducers during a fluid-induced slip experiment (conducted as a part of another project) to understand the rupture dynamics and precursors to fluid-induced fault reactivation [*data analysis near completion*].
- Developed a generalized physics-based approach to extracting features from ultrasonic/seismic data, a deep learning model for automatic feature extraction from full ultrasonic/seismic waveforms. Deep learning models for predicting laboratory earthquakes (i.e., shear failure timing and magnitude in laboratory-scale shear experiments) using the generalized physics-based features outperform those based on physics-based and data-driven features [*completed*].

Science objectives for 2026-2028:

- Continue investigating the mechanisms and precursors of fluid-injection induced seismicity under varying stress and drainage conditions. We are working on a fluid-injection slip experiment under biaxial loading condition, where hybrid active-source ultrasonic and passive acoustic emission data are recorded using an array of ultrasonic transducers.
- Quantify the contribution of bulk versus fracture to the measured linear and nonlinear elastodynamic response of fractured rock by combining experimental and numerical results.

Publications supported by this project 2023-2025 (Selected):

1. Evan Bozek, Prabav Borate, Mark Rivers, Clay Wood, Chun-Yu Ke, Lalith Sai Srinivas Pillarisetti, Colin L. Williams, Derek Elsworth, Jacques Rivière, Parisa Shokouhi. "Linear and nonlinear elastodynamic signatures of fractured rock in relation to fault characteristics inferred from in-situ synchrotron X-ray computed tomography" JGR Solid Earth, **2026**. [*fully supported; revised manuscript submitted in December 2025*]
2. Parisa Shokouhi, Asef I. Sadaf, Michail Skiadopoulos, Evan P. Bozek, Derek Elsworth, Mark Rivers, Jacques Rivière, "Microstructure-informed simulation of wave propagation in fractured rock" Proceedings of the 59th U.S. Rock Mechanics/Geomechanics Symposium, Santa Fe, New Mexico, **2025**, <https://doi.org/10.56952/ARMA-2025-0429>. [*fully supported*]
3. Clay Wood, Prabhakaran Manogharan, Andy Rathbun, Jacques Rivière, Derek Elsworth, Chris Marone, Parisa Shokouhi, "Relating Hydro-Mechanical and Elastodynamic Properties of Dynamically Stressed Tensile-Fractured Rock in Relation to Applied Normal Stress, Fracture Aperture, and Contact Area" JGR Solid Earth, **2024**, 129(8), e2023JB027676, <https://doi.org/10.1029/2023JB027676>. [*fully supported*]
4. Prabhav Borate, Jacques Rivière, Samson Marty, Chris Marone, Daniel Kifer, Parisa Shokouhi, "Physics informed neural network can retrieve rate and state friction parameters from acoustic monitoring of laboratory stick-slip experiments", Scientific Reports, **2024**, 14, 24624, <https://doi.org/10.1038/s41598-024-75826-y>. [*partially supported*]
5. Pengliang Yu, Ankur Mali, Thejasvi Velaga, Alex Bi, Jiayi Yu, Chris Marone, Parisa Shokouhi, Derek Elsworth, "Crustal permeability generated through microearthquakes is constrained by seismic moment" Nature Communications, **2024**, 15, 2057, <https://doi.org/10.1038/s41467-024-46238-3>. [*partially supported*]

Magnetic Properties of Cobalt-bearing Minerals

Sarah Slotznick, Principal Investigator

Earth and Planetary Sciences Department, Dartmouth College, Hanover, NH

Graduate Students: None (recruitment in progress)

Collaborators: Thom Chaffee (Dartmouth College), Mike Krawczynski, Zhihua Xiong (Washington University in Saint Louis), Geoffroy Hautier (Rice University), Roger Fu (Harvard), Gerald Zieg (Tintina Montana Inc.)

Overall research goals:

Cobalt is a rare but critical element for the energy transition due to its use in electrical vehicle batteries. While pure cobalt is a known ferromagnet, the magnetic properties of the more common cobalt minerals which are mined for this element are poorly understood. Most cobalt from primary ore deposits occurs in arsenide and sulfide minerals (1). Although some material science studies have analyzed the magnetic properties of cobalt in alloys and synthesized materials, natural cobalt minerals remain largely unstudied. In the geosciences, most fundamental research into magnetic phases focuses on iron oxides and sulfides due to their importance in the field of environmental and paleo-magnetism. Prior work on geologic cobalt-bearing samples simply measured natural remanent magnetization and susceptibility (2-4) for economic-geology applications in magnetic separation techniques and aeromagnetic surveying. The goals of this project are to gain a better understanding of the magnetism of cobalt minerals in the context of their natural occurrence. Therefore, we specifically will:

- 1) Perform magnetic experiments on six minerals—carrollite ($\text{Cu}(\text{Co},\text{Ni})_2\text{S}_4$), pentlandite ($(\text{Fe},\text{Ni},\text{Co})_9\text{S}_8$), linnaeite (Co_3S_4), siegenite (CoNi_2S_4), cobaltite (CoAsS), and glaucodot ($(\text{Co},\text{Fe})\text{AsS}$)—to understand their magnetic properties across a range of temperatures and grain sizes. To characterize both natural mineral occurrences with compositional variance and the behavior of compositionally pure phases, specimens will be both from museum collections and synthesized at a variety of compositions.
- 2) For minerals that retain a remanent magnetization at room-temperature, carry out first principles computations to understand the fundamental nature of the magnetism and micromagnetic modeling to probe the effects of grain size and crystal shape on magnetic domain structure, saturation strength, and remanence.
- 3) Collect and analyze the rock magnetism of multi-mineralic cobalt ore samples from the Cu-Co-Ag Black Butte Deposit, MT to test the utility of these magnetic parameters for determining the occurrence and distribution of cobalt phases in natural systems and understanding the paragenesis of Co-bearing deposits for resource prediction.

Significant achievements during 2023- 2025:

Fourteen mineral specimens of cobaltite, pentlandite, siegenite, glaucodot, linnaeite, and cattierite (CoS_2) were obtained from the Harvard Mineralogical and Gemological Museum collections. These specimens do not have paired mineralogical characterization information so preliminary compositional checks have been performed using electron dispersive spectroscopy. Synthesis experiments have begun at Washington University in Saint Louis.

Room temperature magnetic analyses (hysteresis loops, direct current demagnetization, and first order reversal curves (FORCs)) have been measured on the museum specimens at Dartmouth College. Magnetic remanence at room temperature was observed in natural samples of cobaltite, glaucodot, siegenite, and pentlandite. However, EDS spectra could not confirm the presence of cobalt in our pentlandite samples. Fully paramagnetic behavior at room temperature was observed in natural samples of cattierite and linnaeite; however, EDS analyses highlighted that the latter was multi-mineralic and did not contain stoichiometric linnaeite.

Science objectives for 2026-2028:

Obtain additional mineral specimens of cobaltite, glaucodot, and siegenite and specimens of cobaltpentlandite, carrollite, and linnaeite, which we have not yet characterized, from the Smithsonian Institution and continue synthesis experiments to produce minerals of interest including compositionally pure endmembers and ones with varying compositions. Perform the same measurements detailed above to further explore the range of magnetic properties for cobalt minerals. One additional property we plan to analyze is how much gyroremanent magnetization (GRM) and rotational remanent magnetization (RRM) these samples acquire as anomalously large GRM and RRM are a curious property of magnetic iron sulfides that can be key for identification of these phases in sediments and rocks.

Perform anisotropy experiments and data analysis in order to determine the type (e.g. easy-axis, easy-plane) and intensity of magnetocrystalline anisotropy in non-isometrically structured minerals with ferromagnetic character. Additionally, perform high-temperature experiments to determine the remanence unblocking temperatures and magnetic exchange behavior for samples of minerals with ferromagnetic character. These data will be used in conjunction to perform high resolution micromagnetic simulations using MERRILL (5). Initially we will explore the parameter space of magnetic domains within cobalt minerals with model results that can be directly compared to experimental data to provide detailed physical explanations for the observations, as well as identifying domain state transition thresholds useful for analysis of ore deposits.

The incoming PhD student will expand this experimental work by performing low temperature magnetic experiments. When the mineral is undergoing a crystallographic-structural and/or magnetic ordering transition, magnetization changes at the atomistic scale and can be another tool permitting rapid and nondestructive mineralogic identification.

Begin the modeling using first principles computations at the atomistic scale in the density functional theory (DFT) framework. The properties that will be simulated are magnetic ordering (nature of the ordering: ferro-, ferri- or anti-ferromagnetic), magnetic anisotropy and magnetization. Temperature effects can also be modeled thus our experiments will allow for testing and continued refinement of the DFT computations.

Collect multi-mineralic ore samples from drill cores of the Black Butte Deposit and begin bulk room temperature analyses on the samples. Compare with parameters obtained from mineral specimens to see if specific cobalt phases can be uniquely identified magnetically.

Publications supported by this project 2023-2025:

None

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1. Slack, J. F., Kimball, B. E., & Shedd, K. B. "Cobalt", *USGS Professional Paper*, **2017**, 1802F, <https://doi.org/10.3133/pp1802F>.
2. Dahlin, D. C., & Rule, A. R. *Magnetic susceptibility of minerals in high magnetic fields*, **1993**, 9449.
3. Krs, M., & Kropáček, V., "A contribution to the magnetic properties of natural cobaltite" *Phys. Earth Planet. Inter.*, **1987**, 46(1–3), 227–232. [https://doi.org/10.1016/0031-9201\(87\)90184-1](https://doi.org/10.1016/0031-9201(87)90184-1).
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Mechanisms of Atomic- to Pore-Scale Geochemical Processes

Andrew G. Stack, Principal Investigator

Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN

Key Personnel: Lawrence M. Anovitz (ORNL), Sang Soo Lee (Argonne), Vitalii Starchenko (ORNL), Juliane Weber (ORNL), Ke Yuan (ORNL)

Post-doctoral Researchers: Uwaila Iyare (ORNL), Bupmo Kim (Argonne), Stephanie Wong (ORNL)

Overall research goals:

The long-term, overarching goal of this project is to attain a fundamental, predictive understanding of the geochemical processes that control mineral nucleation, growth and the evolution of porosity in the subsurface, especially fracture filling/widening.

Significant achievements during 2023- 2025:

Discovered the mechanism of amorphous-to-crystalline phase change. Using in operando time-resolved synchrotron X-ray diffraction and reactive transport modeling, we demonstrated that the amorphous-to-crystalline transition for amorphous CaCO_3 occurs via a dissolution–reprecipitation mechanism with an activation energy of 63 ± 2 kJ/mol (Chen et al. 2024). We found that the presence of an impurity, Sr, inhibits the transformation to the crystalline phase.

Quantified effect on nucleation rates due to changing instrument resolution. While multiple nucleation studies report interfacial energies, the pre-factor is often not reported, yet may play a significant role during especially heterogeneous nucleation. In this study, we determined the apparent pre-factor and interfacial energy both depend on instrument resolution (You et al. 2025), so existing measurements of nucleation likely have some bias.

Probed reactivity of defects on quartz surfaces toward solutes. We expanded our work on understanding defects and non-stoichiometric terminations of quartz surfaces to show that portions of the quartz–water interface contained greater extents of adsorbed ions (Yang et al. 2024), and that the thermodynamics of ion sorption (Walker et al., in press) and proton affinities (Yuan et al. 2024) changed on defects vs. stoichiometric surface sites.

Observed crystallization pathways for two sparingly soluble minerals. We analyzed heterogeneously nucleated barite and calcite crystals using synchrotron-based Bragg coherent diffractive imaging (BCDI), high-resolution TEM, and CMD simulations (Yuan et al., accepted). The calcite contained abundant strain and numerous defects that likely formed along the twinning plane along the *c*-axis due to nonclassical nucleation (e.g., oriented aggregation or multiple nucleation events). By contrast, the barite contained little or no defects, likely because it grew by 2D island nucleation that annealed any defects.

Correlated relationship between nucleation density and pore structure. By combining small-angle neutron/X-ray scattering (SANS/SAXS), tomography, and electron microscopies, we found that volume-increasing witherite (BaCO_3) precipitation leads to rock fracturing, consistent with expectation, but nucleation of siderite (FeCO_3) causes grain boundary widening in low-porosity limestones, inconsistent with expectation (Weber et al. 2023, 2025). Our results suggest that the starting porosity plays a critical role in rock fracturing in response to replacement.

Science objectives for 2026-2028:

Our immediate goals are organized into two synergistic thrusts: **(1) Discover the influences of solution and substrate structure and dynamics on heterogeneous nucleation, and (2)**

Build an understanding of the role of internal and external strains during crystal growth.

Objectives in Thrust 1 include isolating specific processes that contribute to nucleation rates and constraining their magnitudes. This task is necessary because prior work indicates that changes in the solution and substrate composition have multiple and contradictory effects on nucleation. In Thrust 2, our objective is to establish the relationship between strain and crystal growth rate. Specifically, we are examining the effects of internal strain from impurity incorporation and external strain from crystallization pressures. These goals will be achieved by conducting in situ experiments, using advanced microscopies, acoustic methods, and neutron/X-ray scattering approaches, coupled with post hoc characterization. Simulations will include atomic-scale modeling of nucleation processes and computational fluid dynamics to understand the roles of fluid flow and thermal density fluctuations. This work will create a fundamental understanding of the mechanisms behind mineral nucleation in porous media and the forces driving the chemomechanical interplay between precipitates and the porous media in which they occur. In turn, this understanding will enable more robust predictions of subsurface processes while confronting several long-standing conceptual and practical challenges for the geosciences.

A small selection of solely supported publications supported by this project 2023-2025:

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2. Rampal, Nikhil; Wang, Hsiu-Wen; Brady, Alexander B.; Borreguero, Jose M.; Biriukov, Denys; Mamontov, Eugene; Stack, Andrew G. (2024) "Testing the Hypothesis that Solvent Exchange Limits the Rates of Calcite Growth and Dissolution." *RSC Advances*. **2024**, 14, 15743-15754. DOI: [10.1039/D4RA00565A](https://doi.org/10.1039/D4RA00565A).
3. Weber, Juliane; Cooke, C.; Starchenko, Vitalii; Rivers, Mark; Ilavsky, Jan; Zhang, Rui.; Zachman, Michael J.; Gilbert, E.; Mata, Jitendra; Stack, Andrew G.; Anovitz, Lawrence M. "Reaction-Induced Fracturing of Porous Carbonate Rocks During Volume-Increasing Replacement by Witherite." *Cryst. Growth Des.* **2025**, 25, 6604-6619. DOI: [10.1021/acs.cgd.5c00522](https://doi.org/10.1021/acs.cgd.5c00522) (Alternate Cover)
4. Weber, Juliane; Starchenko, Vitalii; Ilavsky, Jan; Allard, Lawrence F.; Mata, Jitendra; Debeer-Schmitt, Lisa; Cooke, Carolyn G.; Littrell, Kenneth; He, Lilin; Zhang, Rui; Stack, Andrew G.; Anovitz, Lawrence M. "Grain Boundary Widening Controls Siderite (FeCO_3) Replacement of Limestone (CaCO_3)." *Sci. Rep.* **2023**, 13:4581 DOI: [10.1038/s41598-023-30757-y](https://doi.org/10.1038/s41598-023-30757-y)
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8. Yuan, Ke; Rampal, Nikhil; Irle, Stephan; Criscenti, Louise J.; Lee, Sang Soo; Adapa Sai; Stack, Andrew G. "Variations in proton transfer pathways and energetics on pristine and defect-rich quartz surfaces in water: Insights into the bimodal acidities of quartz." *J. Coll. Int. Sci.*, **2024**, 666, 232-243. DOI: [10.1016/j.jcis.2024.03.144](https://doi.org/10.1016/j.jcis.2024.03.144)
9. Zhang, Rui; Bosomworth, Paul A.; Weber, Juliane; Ilavsky, J.; Chen, Si Athena; Flores-Betancourt, Alexis; Gilbert, Elliot P.; Mata, Jitendra; Rivers, Mark L.; Eng, Peter J.; Anovitz, Lawrence M. "The role of annealing and grain boundary controls on the mechanical properties of limestones and marbles." *Int. J. Rock Mech. Min. Sci.* **2024**, 183, 105926. DOI: [10.1016/j.ijrmms.2024.105926](https://doi.org/10.1016/j.ijrmms.2024.105926)

Chemo-Mechanical Modeling with NMM-Crunch-CrunchODiTi

Carl Steefel (Presenter)¹, Mengsu Hu¹, Christophe Tournassat^{1,2}

¹ Energy Geosciences Division, Lawrence Berkeley National Laboratory

² Earth and Space Science Observatory at Orléans, University of Orléans, France

Overall research goals: Chemo-mechanical processes at micro-interfacial and pore scales control mesoscale time-dependent behavior. To analyze these microscale processes, we have developed a new chemo-mechanical model by linking the NMM code (Hu and Rutqvist, 2020) for continuum and discontinuum mechanics to Crunch (Steefel et al., 2015) for reactive transport. The model can rigorously consider realistic geometry, reactive transport and kinetics, contact mechanics, and their coupling in interfacial, granular, and fractured systems. It has been applied to analyze stress-mediated dissolution (i.e., pressure solution) to provide insights for time-dependent compaction, such as salt creep (Hu, Steefel, et al., 2021, 2023). The overarching goal of this project is to substantially extend NMM-Crunch and CrunchODiTi so that they apply to a diverse range of rock types and chemo-mechanical and electrochemical processes. These include (1) extension of NMM-Crunch to simulate and predict the chemical, mineralogical, and coupled processes associated with serpentinization, and (2) continued development of the software CrunchODiTi to simulate ion transport with explicit treatment of electrical current, with application to geophysical signals and critical mineral extraction.

High-level overview

Our efforts here are built in part on the Crunch family of codes (crunch.lbl.gov). The software was developed by Steefel and others, combining flow, diffusion, equilibrium thermodynamics, and kinetic reaction modeling (Steefel et al., 2015; Steefel and Hu, 2022). CrunchFlow/CrunchTope also uses the Nernst-Planck equation to resolve the different diffusivities of the ions in solution while maintaining charge balance. NMM-Crunch and CrunchODiTi were developed in FY23-25, supported by the LBL BES program.

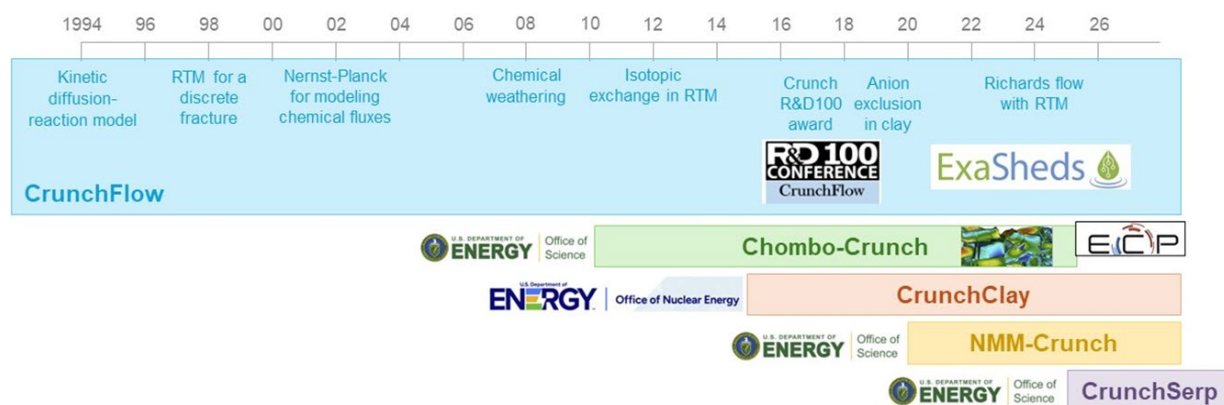


Figure 1. Brief history of Crunch family of codes (crunch.lbl.gov)

Significant achievements during 2023- 2025:

- **NMM-Crunch.** Development of NMM-Crunch and application to analyze pressure solution and provide insights for time-dependent compaction, such as salt creep (Hu, Steefel, et al., 2021, 2023) and carbonate fracture evolution.
- **CrunchODiTi.** Continued development of CrunchODiTi focused on the parallelization of the software based on message passing interface (MPI) and pure Fortran functions. The software was applied to 1) ion migration in clay-rich rock (Tournassat et al., 2023; Tertre et al., 2024; Sarsenbayev et al, 2025), 2) the interpretation of geophysical electrical signals in clay-rich rock, and 3) electrokinetic in situ leaching of copper and critical minerals from sulfide ore.
- **CrunchSerp and initial development of NMM-CrunchSerp.** CrunchSerp was calibrated with the use of batch and meter-scale laboratory experiments at T and P. A new unified

thermodynamic rate law for stress-mediated mineral dissolution (pressure solution) and crystallization pressure has been implemented and applied to serpentinization.

Science objectives for 2026-2028

- To implement new reactions and chemo-mechanical processes such as volumetric expansion, reaction-induced fracturing, and stress-mediated dissolution in NMM-CrunchSerp for rigorous treatment of serpentinization. The model will be validated with laboratory experiments on both serpentinization and the periclase → brucite reaction.

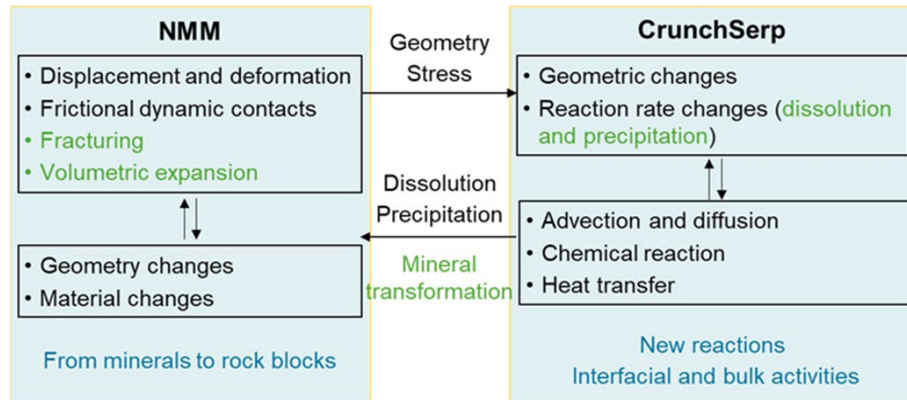


Figure 2. NMM-CrunchSerp framework (Processes in green are new capabilities to be added).

- To apply the features for modeling electrical current in CrunchODiTi to the electrokinetic leaching and critical mineral extraction from sulfide ores.

Publications supported by this project 2023-2025

- Tournassat, Christophe, Carl I. Steefel, Patricia M. Fox, and Ruth M. Tinnacher. "Resolving experimental biases in the interpretation of diffusion experiments with a user-friendly numerical reactive transport approach." *Scientific Reports* 2023, 13(1), 15029.
- Mengsu Hu, Carl I. Steefel, Jonny Rutqvist, and Benjamin Gilbert. "Microscale THMC modeling of pressure solution in salt rock: impacts of geometry and temperature." *Rock Mechanics and Rock Engineering* 2023, 56, 7071-7089.
- Tertre, Emmanuel, Thomas Dabat, Jingyi Wang, Sébastien Savoye, Fabien Hubert, Baptiste Dazas, Christophe Tournassat, Carl I. Steefel, and Eric Ferrage. "Influence of salinity gradients on the diffusion of water and ionic species in dual porosity clay samples." *Journal of Contaminant Hydrology* 2024, 264, 104357.

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- Steefel, Carl I., C. Anthony J. Appelo, Bhavna Arora, Diederik Jacques, Thomas Kalbacher, Olaf Kolditz, Vincent Lagneau et al. "Reactive transport codes for subsurface environmental simulation." *Computational Geosciences* 19, no. 3 (2015): 445-478.
- Steefel, Carl I., and Mengsu Hu. "Reactive transport modeling of mineral precipitation and carbon trapping in discrete fracture networks." *Water Resources Research* 2022, 58(9), e2022WR032321.
- Mengsu Hu, and Jonny Rutqvist. "Microscale mechanical modeling of deformable geomaterials with dynamic contacts based on the numerical manifold method." *Computational Geosciences* 2020, 24(5), 1783-1797.
- Mengsu Hu, Carl I. Steefel, and Jonny Rutqvist. "Microscale mechanical-chemical modeling of granular salt: Insights for creep." *Journal of Geophysical Research: Solid Earth* 2021, 126(12), e2021JB023112.
- Sarsenbayev, Dauren, Christophe Tournassat, Carl I. Steefel, and Haruko M. Wainwright. "Building confidence in models for complex barrier systems for radionuclides." *Proceedings of the National Academy of Sciences* 2025, 122(27), e2511885122.

Stable hydrogen isotopes as tracers of H₂ reactivity in geological systems

Daniel Stolper, Principal Investigator

Dept. of Earth and Planetary Science, University of California Berkeley, CA

Energy Geoscience Division, Lawrence Berkeley National Laboratory, Berkeley, CA

Postdocs: Flora Hochsheid (UC Berkeley/LBNL), Andrew Turner (UC Berkeley/LBNL)

Collaborators: Ben Gilbert (LBNL), Markus Bill (LBNL), Piotr Zarzycki (LBNL)

Overall research goals:

H₂ is a critical molecule in the cycling of electrons in numerous geologic systems ranging from high-temperature igneous systems to rocks experiencing hydrothermal alteration to low temperature sediments. Beyond its importance in geological systems, H₂ is actively being explored as a resource for energy production in the US including by exploiting naturally occurring processes. Our goal here is to use isotope geochemistry as a way to measure and identify the fundamental reactions occurring between H₂ and common geological materials at geologically relevant conditions

Isotopic measurements of hydrogen and deuterium in H₂ have been conducted since the discovery of deuterium by Harold Urey in the 1930s. However, constraints at the precision and accuracy achievable using modern geochemical techniques are lacking but necessary for the interpretation of the variations D/H ratio of molecular hydrogen in nature. For example, before this work, there remained significant uncertainty on the distribution (i.e., fractionation) of D/H between H₂ and liquid water at <100°C temperatures. Exchange rates of hydrogen isotopes between H₂ and liquid water are known to be pH dependent, but the rates have only been measured quantitatively and usefully at one acidic pH. There are limited constraints on the equilibrium partitioning of HD/H₂ in the gas vs. aqueous phase. And finally, little is known about the isotope geochemistry of H₂ generation from natural systems such as during serpentinization. Put another way, how isotopes partition between gaseous and aqueous hydrogen and liquid water during dissolution, the rates of exchange, and final equilibrium values are not well constrained. The goal of this project is to measure these fundamental properties in the laboratory using modern techniques and, where possible, compare our measurements to state-of-the-art theoretical chemical calculations.

Significant achievements during 2023- 2025:

For published work, we conducted what is currently the highest level theoretical study in geochemistry on the partitioning of hydrogen isotopes between H₂ and H₂O along with other common small organic H bearing species (Korol et al., 2025). This work involved the calculation of high-accuracy potential energy surfaces at the CCSD(T) level (specifically CCSD(T)-F12A with the aug-cc-pVTZ basis set) along with explicit inclusion of anharmonicity using Path Integral Monte Carlo techniques and corrections to the Born-Oppenheimer approximation. We additionally conducted catalysed measurements of the equilibrium fractionation factor between H₂(g)-H₂O(l) in the laboratory from 3-90°C and provided an experimentally grounded calibration of this fractionation factor as a function of temperature (Hochsheid et al., 2025).

In terms of unpublished work, we have (i) created a methodology to measure isotopic partitioning between H₂(g)-H₂(aq) in the laboratory and have conducted preliminary measurements from 10-45°C. We have additionally compared this measurement to theoretical calculations. (ii) We have measured the isotope exchange rate of hydrogen between dissolved H₂ and liquid water in a single phase from 25-90°C at a low and high pH and quantified for the first time the pH effect for exchange. (iii) We have designed an experimental approach to measure the hydrogen isotope effect for H₂ generated from iron bearing minerals in liquid water and demonstrated it has a large isotope effect consistent with H₂ generation from suspected serpentinization systems. These results along with the published work will be presented in a poster at the meeting.

Science objectives for 2026-2028:

Our goals are to finish and publish our results on the work discussed above in terms of hydrogen isotope fractionation factors for $H_2(g)$ - $H_2(aq)$, exchange rates of hydrogen isotopes between hydrogen and water including at an additional pH and set of temperatures, and results of the iron oxidation+ H_2 production work.

Our focus over the next few years will be to introduce and apply a new tracer of these processes, D_2 , as it has measurably different properties from HD (the main carrier of deuterium at natural abundance). We will constrain rates of exchange of D_2 vs H_2 with water for the first time, differences in solubility, and distinct isotopic fractionations. The goal is that the combined measurements of D_2 and HD vs. H_2 will allow us to distinguish processes occurring in nature that D/H ratios alone cannot identify.

Publications supported by this project 2023-2025:

1. Korol, Roman, Andrew C. Turner, Apurba Nandi, Joel M. Bowman, William A. Goddard III, and Daniel A. Stolper. "Stable isotope equilibria in the dihydrogen-water-methane-ethane-propane system. Part 1: Path-integral calculations with CCSD(T) quality potentials." *Geochimica et Cosmochimica Acta* **2025**, 396, 71-90. [10.1016/j.gca.2025.02.028](https://doi.org/10.1016/j.gca.2025.02.028).
2. Hochscheid, Flora, Andrew C. Turner, Noam Lotem, Markus Bill, and Daniel A. Stolper. "Experimental determination of hydrogen isotopic equilibrium in the system $H_2O(l)$ - $H_2(g)$ from 3 to 90° C." *Geochimica et Cosmochimica Acta*, **2025**, 394, 368-382, [10.1016/j.gca.2025.02.029](https://doi.org/10.1016/j.gca.2025.02.029)

Next Generation Synchrotron X-ray Methods and Tools for Quantifying Chemical Complexity at Mineral-Fluid Interfaces

Joanne E. Stubbs, Principal Investigator

Center for Advanced Radiation Sources, The University of Chicago, Chicago, IL

Co-Investigators: Peter J. Eng and Stephen R. Sutton, Center for Advanced Radiation Sources, The University of Chicago, Chicago, IL

Overall research goals:

GeoSoilEnviroCARS (GSECARS) operates Sector 13 at the Advanced Photon Source (APS) for the benefit of the Earth and environmental science community, providing access to state-of-the-art synchrotron X-ray instrumentation, software, and knowledgeable staff who support both novice and experienced users in bringing their science to fruition. This project focuses on the surface and interface program at GSECARS with four principal objectives targeting chemical and structural complexity at mineral-fluid interfaces: (1) Expanding the landscape of mineral-water interface problems probed via crystal truncation rod (CTR) scattering; (2) Improving capabilities for mapping reciprocal space in 3D; (3) Exploiting the extraordinary brightness and coherence of the APS Upgrade (APS-U); and (4) Improving data reduction and analysis strategies for interfacial X-ray methods. Science topics include adsorption chemistry of rare earth elements (REE), atomic scale structures of uraninite (UO_2) ageing products, redox-driven pyrite (FeS_2) surface chemistry, diaspore (AlOOH) surface structure under varying chemical conditions, and complex structural analysis of carbonate minerals.

Significant achievements during 2023-2025:

April 2023 marked the beginning of the APS Upgrade during which time the storage ring was rebuilt to enhance both brightness and coherence by multiple orders of magnitude. We used this once-in-a-generation opportunity to make numerous enhancements to the GSECARS beamlines, including improvements supported by this award to serve the surface and interface program. These included replacing the primary sample rotation axis and translation stages on the 13-ID-C diffractometer with an axis that rotates >20 times faster with an order-of-magnitude smaller positional error as well as a hexapod that enhances rigidity and supports improved sample alignment. These tools will dramatically improve our reciprocal space mapping capabilities. Both of the GSECARS beamlines that support surface and interface experiments have resumed user support, with some installation and commissioning activities ongoing. Measurements using the new source have included projects investigating REE adsorption on iron oxide and carbonate minerals, the surface chemistry of barite (a scale-forming mineral in oil and gas pipelines), and UO_2 aged > 2 years in oxidizing environments. During this period, we have substantially upgraded our data reduction software, implementing a major transition to modern, maintainable Python code. Numerous publications have resulted from collaborations enabled by this award (selected examples below), with more in various stages of preparation and review.

Science objectives for 2026-2028:

The current award period extends through June 2026. Between now and then we will complete installation of the new mirror system for 13-ID-C, enabling submicron focal spots suitable for coherent scattering measurements that leverage APS-U. Replacement translation stages for the 13-BM-C diffractometer have been procured and we anticipate placing the final orders soon for a system that will enhance travel ranges, stability, and flexibility for the sample positioning system there. Software improvements will be finalized. Additional measurements will be made for the UO_2 project, completing a data set that will enable description of the surface structures of three crystallographic

surfaces at various stages of oxidation and their relationships to one another. Collaborators working on REE adsorption, barite, and other projects will return to the beamlines. Multiple manuscripts are close to publication, including two led by Anna Wanhala (former postdoc supported by this award) describing the surface chemistry of pyrite under suboxic conditions (collaboration with Gilbert group at LBL; recently submitted to Langmuir) and the effects of pH on the surface structure and chemistry of diasporite (collaboration with Anovitz group at ORNL; in final stages of preparation).

Selected publications supported by this project 2023-2025:

1. Yihang Fang, Sang Soo Lee, Greg J. Ledingham, Joanne E. Stubbs, Peter J. Eng, and Jeffrey G. Catalano, "Complex Adsorption Behavior of Neodymium and Ytterbium on Structurally-Distinct Alumina Surfaces", *Environ. Sci. Technol.*, **2025**, 59, 3972-3981, <https://doi.org/10.1021/acs.est.4c10140>
2. Peng Yang, Jacquelyn N. Bracco, Gabriela Camacho Meneses, Ke Yuan, Joanne E. Stubbs, Mavis D. Boamah, Matthew Brahlek, Michel Sassi, Peter J. Eng, Matthew G. Boebinger, Albina Borisevich, Anna K. Wanhala, Zheming Wang, Kevin M. Rosso, Andrew G. Stack, and Juliane Weber, "Carbonation of MgO Single Crystals: Implications for Direct Air Capture of CO₂", *Environ. Sci. Technol.*, **2025**, 59, 3484-3494, <https://doi.org/10.1021/acs.est.4c09713>
3. Gabriela Camacho Meneses, Juliane Weber, Raphaël P. Hermann, Anna Wanhala, Joanne E. Stubbs, Peter J. Eng, Ke Yuan, Albina Y. Borisevich, Matthew G. Boebinger, Tingting Liu, Andrew G. Stack, and Jacquelyn N. Bracco, "Inhibition of Reaction Layer Formation on MgO(100) by Doping with Trace Amounts of Iron", *J. Phys. Chem. C.*, **2025**, 129, 3457-3468, <https://doi.org/10.1021/acs.jpcc.4c06311>
4. Amanda Dorfman, Anna K. Wanhala, Sang Soo Lee, Peter J. Eng, Joanne E. Stubbs, Lexi Kenis, and Jacquelyn N. Bracco, "Lead Adsorption and Desorption at the Barite (001) Surface in the Presence of EDTA", *ACS ES&T Water*, **2025**, 5, 264-273, <https://doi.org/10.1021/acsestwater.4c00836>
5. Anna K. Wanhala, Luke D. Gibson, Joanne E. Stubbs, Radu Custelcean, Cole Hexel, Peter J. Eng, Vyacheslav S. Bryantsev, and Andrew G. Stack, "Structure of the Basträsite (001) Surface by Crystal Truncation Rod X-ray Diffraction and Ab Initio Molecular Dynamics: Implications for Separations of a Rare Earth Ore Mineral", *J. Phys. Chem. C.*, **2023**, 127, 21150-21160, <https://doi.org/10.1021/acs.jpcc.3c05905>
6. Si Athena Chen, Juliane Weber, Vitalii Starchenko, Peter J. Eng, Joanne E. Stubbs, Hsiu-Wen Wang, Tingting Liu, Tyler L. Spano, Bryan C. Chakoumakos, and Andrew G. Stack, "Real-Time Atomic-Scale Structural Analysis Resolves the Amorphous to Crystalline CaCO₃ Mechanism Controversy", *Cryst. Growth Des.*, **2024**, 24, 5027-5038, <https://doi.org/10.1021/acs.cgd.4c00245>
7. Jacquelyn N. Bracco, Gabriela Camacho Meneses, Omar Colón, Ke Yuan, Joanne E. Stubbs, Peter J. Eng, Anna K. Wanhala, Jeffrey D. Einkauf, Matthew G. Boebinger, Andrew G. Stack, and Juliane Weber, "Reaction Layer Formation on MgO in the Presence of Humidity", *ACS App. Mater. Interfaces*, **2024**, 16, 712-722, <https://doi.org/10.1021/acsami.3c14823>
8. Amanda Dorfman, Anna Wanhala, Peter Eng, Joanne Stubbs, Omar Colon, Malik Donetian, and Jacquelyn N. Bracco, "Inhibition of Strontium Adsorption and Desorption by Ethylenediaminetetraacetic Acid at the Barite (001)–Water Interface", *J. Phys. Chem. C.*, **2023**, 127, 9049-9058, <https://doi.org/10.1021/acs.jpcc.3c01092>
9. Julia Neumann, Jessica Lessing, Sang Soo Lee, Joanne E. Stubbs, Peter J. Eng, Maximilian Demnitz, Paul Fenter, and Moritz Schmidt, "Y(III) Sorption at the Orthoclase (001) Surface Measured by X-ray Reflectivity", *Environ. Sci. Technol.*, **2023**, 57, 266-276, <https://doi.org/10.1021/acs.est.2c06703>

Pathways, Controls and Signatures of Hydrogen Production in Serpentinization

Drew Syverson (University of North Carolina at Charlotte), Benjamin Gilbert (LBNL), Mengsu Hu (LBNL), Daniel Stolper (UC Berkeley), Jill Banfield (UC Berkeley), John Christensen (LBNL), Michael Manga (UC Berkeley), Carl Steefel (LBNL), Christophe Tournassat (LBNL and University of Orléans), and Piotr Zarzycki (LBNL)

Graduate Students: Yarong Qi (UC Berkeley)

Postdoctoral Research Scholar: Yanling Wang (UNCC)

Overall research goals:

The overall goal of our research is to determine the fundamental chemical, mechanical and microbial processes that determine the feasibility of harnessing geologic hydrogen from natural or stimulated serpentinization. The goals of the collaborative tasks described below are:

1. Establish the molecular mechanism of the hydrogen evolution reaction during serpentinization
2. Establish and model how solution chemistry controls the products of serpentinization and the overall rate and yield of hydrogen generation.
3. Establish the microbial metabolic processes that cycle hydrogen, iron and carbon in serpentinizing systems
4. Develop isotopic and geochemical indicators of the pathway and conditions of hydrogen generation to interpret laboratory experiments and the rock record

A high-level overview:

Serpentinization comprises a suite of mineral reactions with water that transform magnesium- and iron-rich silicate rocks into hydrated phases, principally the serpentine polymorphs. Hydrogen production accompanies the release of iron(II) and the oxidation of iron coupled to the reduction of water or protons. The molecular pathway of this hydrogen evolution reaction (HER), however, remains unknown. We are combining laboratory research and *ab initio* studies to test hypotheses for the rate-limiting step. Moreover, iron redox reactions occur simultaneously with mineral transformations that liberate or capture iron and that are strongly influenced by solution chemistry. We employ batch and flowthrough reactions, informed by isotopic data, to establish how mineral transformations control iron redox reactions. Following subsurface serpentinization, numerous biogeochemical processes can transform hydrogen coupled to carbon and iron cycles. We seek field and metagenomic data on this control on available hydrogen.

Significant achievements during 2023-2025:

- Demonstrated that solution chemistry, pH and dissolved silica controls molecular hydrogen generation rates
- Identified pathways for mineral surface catalysis of hydrogen production
- Developed a $^{29}\text{SiO}_2(\text{aq})$ tracer to accurately quantify serpentinization rates to link hydrogen generation to mineral transformation
- Implemented synchrotron X-ray absorption spectroscopy for elemental and oxidation state records of hydrogen generation from laboratory and field samples
- Identified field sites in California for geochemical and microbiological studies of hydrogen production and transformation

Science objectives for 2026-2028:

This project will establish the roles of iron geochemistry and biogeochemistry on the generation and loss of hydrogen from serpentinization.

Develop and use silicon, magnesium and oxygen isotopes to track the pathway of serpentinization in the laboratory in geologic settings.

Publications supported by this project 2023-2025:

This research was initiated in 2025 and several papers are in progress but none are published.

AI Assisted Neural Network Quantum Dynamics Simulations of Stress-Corrosion and Carbon Sequestration

Priya Vashishta, Principal Investigator

Collaboratory for Advanced Computing and Simulations, Department of Chemical Engineering & Materials Science, Department of Physics, and Department of Computer Science, University of Southern California, Los Angeles, CA 90089

Graduate Students: Logan Yamamoto

Postdoc: Nitish Baradwaj

Overall research goals:

I. Allegro-FM: Equivariant Foundation model for exascale simulations. Large scale simulations will be carried out for stress corrosion cracking and damage on silica from shock induced nano bubble collapse, and CO₂ sequestration.

II. Nanoconfined water and effect of confinement dimensionality. Dielectric constant of water and self-diffusion are investigated for parallel plate, cylindrical and spherical geometries.

II. Exascale neural network quantum molecular dynamics (NNQMD) simulations of CO₂ sequestration and molecular mechanisms of mineralization of magnesium silicates. We will investigate reaction dynamics and carbonation pathways for CO₂ sequestration.

Significant achievements during 2023- 2025:

- **Foundational model for exascale scale geosciences simulations:** An AI Foundation Model

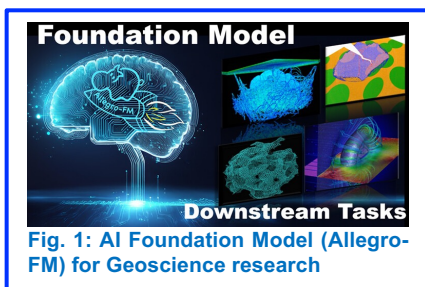


Fig. 1: AI Foundation Model (Allegro-FM) for Geoscience research

(Allegro-FM) for geosciences is developed that is versatile for geosciences simulations for diverse downstream tasks covering 89 elements in the training data sets, figure 1. The Allegro-FM foundation model is capable of exascale simulations. Allegro-FM scales up to multi-billion-atom systems with an efficiency of 97.5% on the exascale supercomputer at Argonne Leadership Computing Facility.

Research Details: An E(3) equivariant neural network architecture (Allegro) is trained using a set of large-scale

organic and inorganic materials datasets merged by Total Energy Alignment (TEA) framework. Allegro-FM exhibits excellent agreements in describing structural, mechanical, and thermodynamic properties, while exhibiting emergent capabilities for structural correlations, reaction kinetics, mechanical strengths, fracture, and solid/liquid dissolution, for which the model has not been trained.

- **Nanoconfined water – dielectric constant and diffusion:** We have conducted MD simulations to compute the effect of geometric confinement on the dielectric constant of water. Planar, cylindrical, & spherical geometries are considered as shown in figure 2.

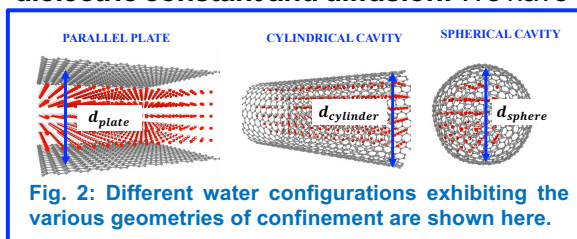


Fig. 2: Different water configurations exhibiting the various geometries of confinement are shown here.

Research Details: The dielectric constant was computed using the fluctuation formula within the canonical ensemble, capturing dipole moment fluctuations over time. Our simulation results indicate a significant reduction in the dielectric constant at confinement lengths near 2 nm, with a monotonic recovery as the confinement becomes less severe as shown in figure 3. This trend is consistent across all

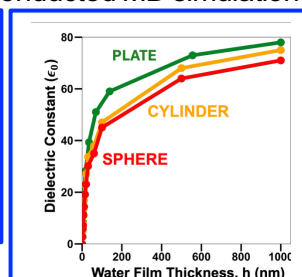


Fig. 3: Dielectric constant of water as a function of water film thickness confined in a plate, cylinder and sphere geometries.

three geometries. We found that the dimensionality of confinement has a profound effect on the properties of water.

- **Molecular mechanisms of CO₂ sequestration in MgSiO₃:** We have performed QMD simulations to investigate the atomistic mechanisms of CO₂ mineralization in MgSiO₃, figure 4.

Research Details: QMD trajectories reveal that the carbonation proceeds through the formation of reaction intermediates. We find the formation of water, which supports the original hypothesis by Seifritz, that water merely acts as a catalyst.

Science objectives for 2026-2028:

- **Billion atom simulations of nanobubble collapse on a silica surface:** Researchers have been studying the effects of cavitation and there are a number of unanswered questions: What are the characteristics of microturbulence during bubble collapse? Is there a limiting factor to the damage due to nano jets? What is the extent of chemical damage leading to the mechanical damage? How does the extent of damage scale with size of the bubble? We use the silica-water system to study the factors affecting

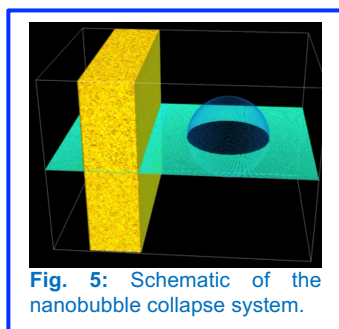


Fig. 5: Schematic of the nanobubble collapse system.

mechanical as well as chemical damage due to shock-induced collapse of nanobubble. Schematic of the simulation setup is shown in figure 5.

- **Exascale neural network quantum dynamics simulations for reaction pathways and molecular mechanisms of CO₂ sequestration in MgSiO₃:** We will do large-scale neural network quantum molecular dynamics simulations for carbonation reactions of CO₂ and H₂O on Mg₂SiO₄ and Ca₂SiO₄ surfaces, figure 6.

- *Publications supported by this project 2023-2025 (Selected):*

1. Ken-ichi Nomura,* Shinnosuke Hattori,* Satoshi Ohmura, Ikumi Kanemasu, Kohei Shimamura, Nabankur Dasgupta, Aiichiro Nakano, Rajiv K. Kalia, and Priya Vashishta, "Allegro-FM: Toward an Equivariant Foundation Model for Exascale Molecular Dynamics Simulation", *Journal Physical Chemistry Letters*, 2025, 16, 6637-6644, doi: [10.1021/acs.jpclett.5c00605](https://doi.org/10.1021/acs.jpclett.5c00605).

2. Shinnosuke Hattori, Kohei Shimamura, Ken-ichi Nomura, Aiichiro Nakano, Rajiv K. Kalia, Priya Vashishta, "Beyond Scaling: Chemical Intuition as Emergent Ability of Universal Machine Learning Interatomic Potentials" *Nature Communications*, 2026, in press to be published.
3. Nitish Baradwaj, Logan Yamamoto, Hind Aljaddani, Tyler Zheng, Ruru Ma, Ankit Mishra, Aiichiro Nakano, Rajiv K. Kalia, and Priya Vashishta, "Dielectric constant of nano-confined water and the effect of dimensionality of nano-confinement", manuscript under preparation for publication.
4. Logan Yamamoto, Nitish Baradwaj, Aiichiro Nakano, Rajiv K. Kalia, and Priya Vashishta, "Silica – water interface and maximum water coverage of silica surface", Manuscript under preparation and will be submitted for publication soon.

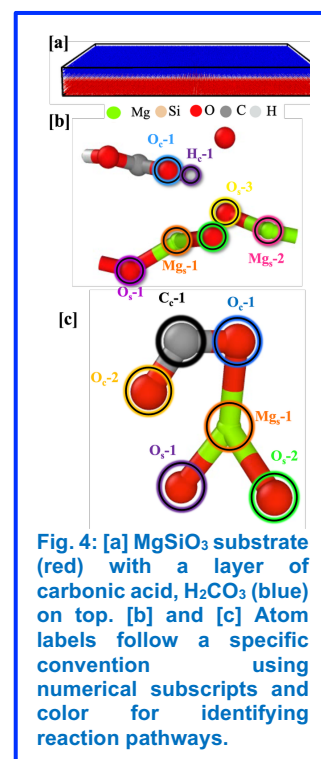


Fig. 4: [a] MgSiO₃ substrate (red) with a layer of carbonic acid, H₂CO₃ (blue) on top. [b] and [c] Atom labels follow a specific convention using numerical subscripts and color for identifying reaction pathways.

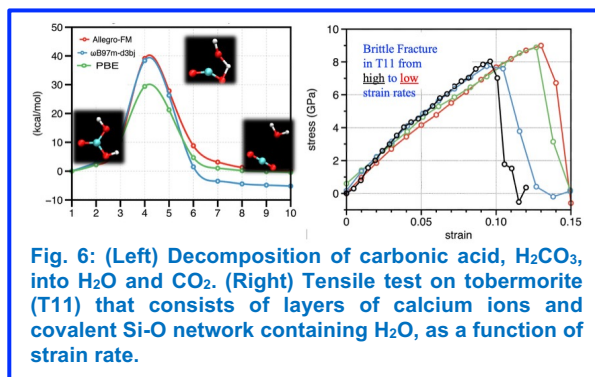


Fig. 6: (Left) Decomposition of carbonic acid, H₂CO₃, into H₂O and CO₂. (Right) Tensile test on tobermorite (T11) that consists of layers of calcium ions and covalent Si-O network containing H₂O, as a function of strain rate.

Elucidating Hydrothermal Fracture Flow, Rheology, and Transport Phenomena

Hari S. Viswanathan, Principal Investigator

Earth and Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM

Graduate Students: Jacob Hampton, Qiang Xiong; Postdoctoral Researchers: K.C. Bijay, Lawrence Boampang, Yerkezhan Madenova

Collaborators: Luke P. Frash, Jeffrey D. Hyman, Daniel O'Malley, Matthew R. Sweeney, Wenfeng Li; key co-authors include Javier E. Santos, J. William Carey, Nathan J. Welch, Meng Meng.

Overall research goals:

Fractured rock hosts a large fraction of U.S. energy, water, and critical-mineral resources, yet predictive control of permeability, reactivity, and induced seismicity remains limited because fractures evolve dynamically under coupled thermal–hydrological–mechanical–chemical (T-H-M-C) processes. This project integrates (i) in situ high-pressure/high-temperature true-triaxial and direct-shear experiments with dynamic imaging, (ii) AI methods that fuse sparse, heterogeneous field observations into a Transparent Earth model, and (iii) high-performance discrete-fracture-network simulations (dfnWorks) to quantify how fracture growth, rheology, and reactive transport govern flow and transport in hydrothermal systems.

Significant achievements during 2023–2025:

Key outcomes since the previous PI meeting include:

- Identified crack-parallel stress (T-stress) as a previously underappreciated control on hydraulic-fracture branching and network complexity, improving mechanistic prediction of stimulated permeability.
- Demonstrated laboratory “fracture caging” in critically stressed shear faults, achieving high recovery of injected fluid and preventing large critical rupture events—an actionable pathway for induced-seismicity mitigation.
- Developed the Senseiver transformer architecture to reconstruct global fields from sparse, multi-modal observations, directly addressing the dominant uncertainty bottleneck in conditioning DFN models to field data.
- Advanced AI for fracture and failure prediction (e.g., foundation-model approaches and differentiable sensor-placement strategies) to improve generalization across materials, loading paths, and data sparsity regimes.
- Quantified how multi-scale aperture heterogeneity and network intensity jointly control anomalous flow channelization and solute-transport tails in 3-D fracture networks, enabling more robust upscaling and uncertainty quantification.
- Led an integrative review of carbon mineralization in fractured mafic and ultramafic rocks, synthesizing state-of-the-art experimental and modeling tools and clarifying priority mechanisms governing mineralization pathways.

Science objectives for 2026–2028:

During 2026–2028 we will pursue three tightly coupled thrusts, aligned with our integrated experiment–AI–DFN workflow:

- Task 1 – Injection-induced seismicity and fracture stability: use repeatable shear-block and triaxial direct-shear experiments to test cyclic versus constant/stepped injection protocols, identify precursor signatures for seismic slip, and quantify the roles of high-temperature/high-stress

slickenside formation and proppant–water–rock interactions on frictional strength and permeability. Develop scaling relationships for fracture caging and critical-slip onset.

- Task 2 – AI-enabled Transparent Earth model: develop an attention-based, multi-modal sparse mixture-of-experts (SMoE) framework that fuses heterogeneous data while embedding differentiable physics. Enable few-shot adaptation for new sites and output probabilistic fracture parameters and boundary conditions to condition DFN simulations.
- Task 3 – Physics-based T-H-M-C DFN simulation and upscaling: extend dfnWorks-based modeling to incorporate stress-dependent aperture evolution and reactive dissolution/precipitation processes (including proppant degradation/transport where relevant). Validate against laboratory datasets, perform ensemble uncertainty quantification, and translate findings to geothermal stimulation, subsurface waste disposal, and mineralization pathways.

Publications supported by this project 2023–2025:

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- Yerkezhan Madenova, Luke P. Frash, Wenfeng Li, Meng Meng, Bijay KC, Jacob Hampton, Qiang Xiong, and Hari S. Viswanathan, “Fracture caging in a lab fault to prevent seismic rupture during fluid injection,” *Geothermics*, 2025, 126, 103206, DOI: 10.1016/j.geothermics.2024.103206.
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- Matthew R. Sweeney, Jeffrey D. Hyman, Daniel O’Malley, Javier E. Santos, J. William Carey, Philip H. Stauffer, and Hari S. Viswanathan, “Characterizing the Impacts of Multi-Scale Heterogeneity on Solute Transport in Fracture Networks,” *Geophysical Research Letters*, 2023, 50(21), e2023GL104958, DOI: 10.1029/2023GL104958.
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Further Experiments in Slow Dynamic Elasticity

Richard Weaver, Principal Investigator

Department of Physics, University of Illinois, Urbana, IL

Collaborator: J Y Yoritomo, Naval Research Laboratory, Washington DC

Overall research goals:

To understand the microphysical mechanisms behind slow dynamics (SD) in granular solids using ultrasound experiments on simplified structures, especially the single bead system. Tests of SD under complex conditioning, at early times and in vacuum, varying humidity and heat, with various surface treatments, and varying pump protocols are particularly valued.

Significant achievements during 2023- 2025:

We have shown that log(time) relaxations begin no later than order (msec).

We have developed and applied a digital lock-in method for measuring fast changes in materials.

We have conducted a parametric study exploring the effect of sundry pump parameters (strain amplitude, pump frequency, pump duration) on Slow Dynamic nonlinear elasticity in samples of sandstone, concrete and the single bead. In all these systems we find that SD response is linear in strain amplitude (as most people had accepted), independent of frequency (not measured elsewhere), and that it continues far longer than the pump duration (in contrast to predictions of most models that it continues only for a time of order the pump duration.)

We have introduced a new model that seamlessly and quantitatively describes both the loss of stiffness and its subsequent log-linear recovery. It is well supported by our measurements.

The figures below show (1) the laboratory system for measuring SD loss of stiffness and recovery in a sandstone prism, and (2) our confirmation that recovery slope scales linearly with pump amplitude and (3) our study showing SD recovery as independent of pump frequency

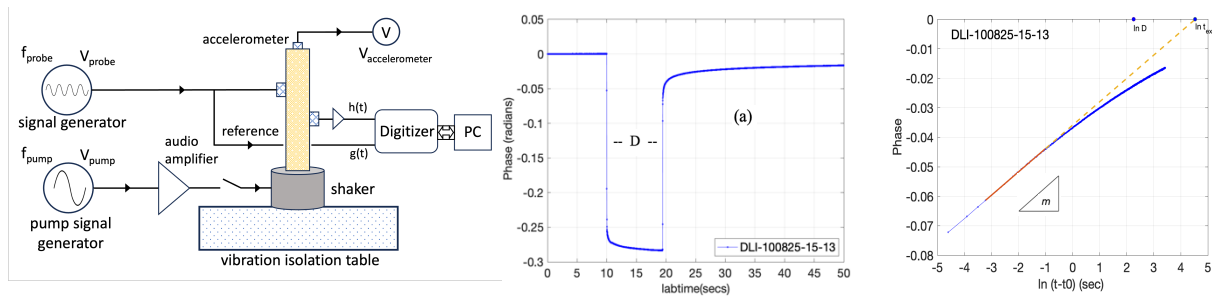


Fig 1] a) A sketch of the laboratory lock-in system for studying modulus loss during and recovery after an oscillating pump strain. b) A typical measurement consisting of 10 sec of reference followed by 10 sec of pumping, followed by 30 seconds of recovery. c) The recovery vs log(time)

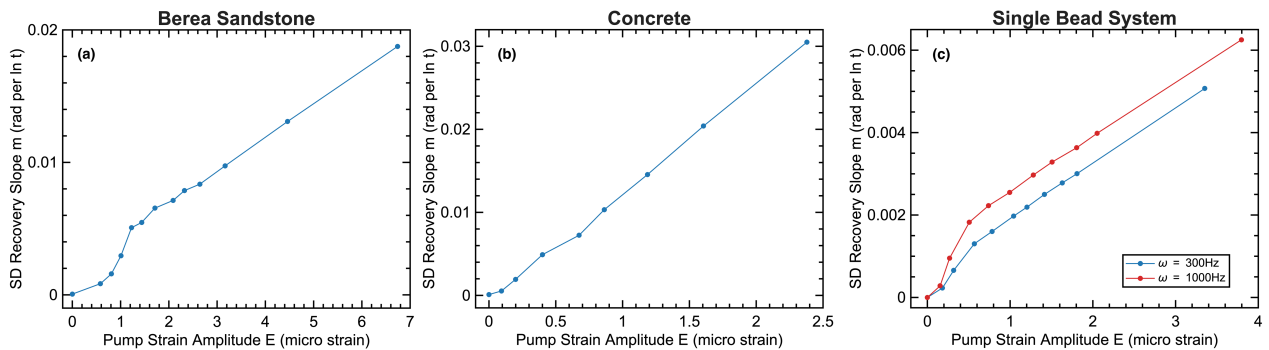


Fig 2] Recovery slope m is observed in all systems to be proportional to pump strain amplitude.

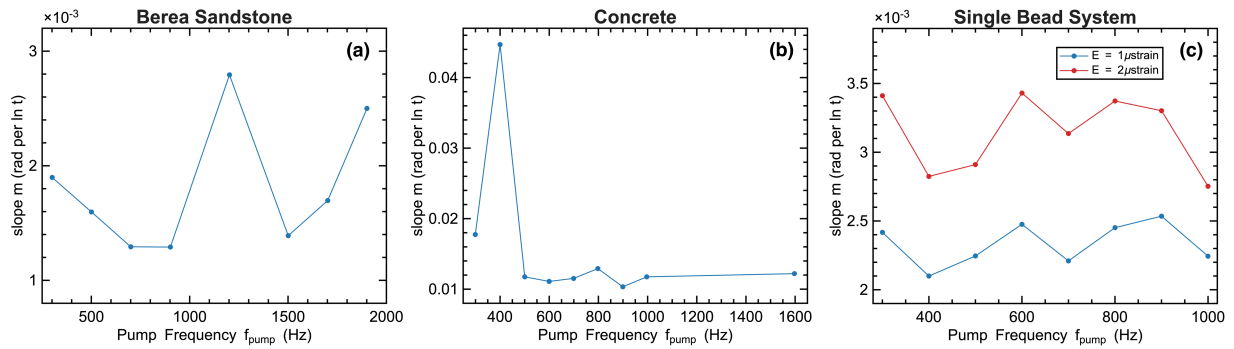


Fig 3] Challenging measurements of the effect of pump strain frequency f have established that any dependence is small, and inconsistent with predictions of scaling with f or f^2 as some phenomenological models would have it. f -dependent local deviations in excess of noise in Berea and concrete (see above) are due to lateral resonances (e.g. bending or torsion) in the prism.

Science objectives for 2026-2028:

Our parametric study[1] has raised two perplexing issues. 1) It appears that there is no simple quantitative model for damage and recovery that predicts both the frequency-independence of the SD recovery slope (Fig 3 above) and the sense of the striking pump-domain “Bowtie loops” observed by others. It appears that some frequency dependence to the damage rate is required to explain the Bowtie loops, and yet our recoveries seem to suggest otherwise. We wish to reconcile these opposing observations. Likewise, 2) We ask what is the significance of the, usually tiny ($O(\text{nsec})$) and highly variable between systems, apparent values of τ_{min} (defined as the lower limit of the relaxation spectrum) extracted from our measurements of SD damage and recovery? We have conjectured that their small values, and the bowtie-loop senses, both indicate the presence of another anomalous nonlinear elasticity mechanism, one that acts during the pump phase and contributes to the pump-domain loss of stiffness, but recovers very quickly at pump-off, before being detected by measurements such as ours that begin at a few msec.

Hence we will be modifying our laboratory set-up to measure the Bowtie loops. Our ability to control pump frequency at fixed pump amplitude, which has not been attempted elsewhere, may allow better insight. A thorough study of the bowtie loops, their senses and their offsets, will be undertaken.

We will also be further varying pump protocols: Can we quantitatively show that there must be internal hidden variables? (for example “non-equilibrium strain” or defect density as functions of relaxation time τ .) This could be revealed by demonstrating that SD recovery rate is not just a function of instantaneous SD damage, but also its history.

Publications supported by this project 2023-2025:

1. Richard L Weaver and John Y Yoritomo, "A Parametric Study of Slow Dynamic Nonlinear Elasticity with Comparisons to Models" (2026) in preparation.
2. John Y Yoritomo and Richard L Weaver " Slow dynamic nonlinear elasticity during and after conditioning, a unified theory and a lock-in probe" J Mechs Phys Solids (2025) **200** 106149 <https://doi.org/10.1016/j.jmps.2025.106149>
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DE-FG02-ER15637 Anisotropy and Residual Stress in Crustal Rocks

Hans-Rudolf Wenk, Principal Investigator

Dept. Earth and Planetary Science, UC Berkeley, Berkeley, CA

Graduate Students: Amartya Kattamalavadi, Michelle Devoe, Eloisa Zepeda

Collaborators: Sven Vogel (LANL), John Grimsich (UCB), Lung Chan (UCB), Andrey Yakovenko (APS), Nobumichi Tamura (ALS), Rebecca Stokes (USGS)

Overall research goals:

The properties of rocks are related to their geological history, creating mineral assemblages with particular microstructures. In this project we are using advanced techniques such as electron microscopy, synchrotron x-ray and neutron diffraction and modeling, with emphasis on better understanding of rock properties such as strength and anisotropy. One focus has been on the transition of shales to slates, discovering that some slates display highest crystal alignment of any materials, exceeding textures in rolled metal foils. Recently we were surprised to find that organic slates, with a significant carbon content have much weaker preferred orientation compared with adjacent inorganic slates. Preferred orientation in sheet-silicate-rich rocks is significant for interpreting observed seismic anisotropy in the Earth. We also explored residual stress in rocks subjected to high stress during tectonic deformation, earthquakes, explosive structures and recently unique microstructures during meteorite impact, similar to conditions during nuclear explosions.

Significant achievements during 2023- 2025:

- Results of the research have been presented in 7 journal publications (see below)
- Eloisa Zepeda and Michelle Devoe, partially supported by this project, finished their PhD at UCB

Science objectives for 2026-2028:

- The PI became first involved in clay anisotropy through a research project with Seiji Nakagawa of LBNL in 2007 (Geoph. 72 E69) and has since published over 20 papers on the subject due to their fascinating microstructures and anisotropy relevant e.g. to oil exploration (Seiji joined us again in #7 after 19 years). Most amazing has been to discover the extreme crystal alignment in slates, recrystallizing from shales, with alignments over 100 times random like no other material, including metals. Such alignment was documented in the Sierra Nevada (CA). Northern Spain and China (#6). However, while conducting this research, we became aware that carbonaceous slates (argillites) adjacent to regular slates have surprisingly low anisotropy and we are currently documenting this for samples from California, Spain and Taiwan. Neutron diffraction at LANL has become indispensable in this project because of the high scattering power of carbon relative to X-rays.
- A second project is residual strain in deformed rocks preserved by crystal lattice distortion and measured with microbeam synchrotron X-rays. Also here we have a long history with many publications (starting with Am. Min. 94, 1059, 2009). We have advanced the methodology and are applying it to hydrothermal explosion structures as documented in the Apennines, Hong Kong (Chan, Alvarez, J. Str. Geo. 174, 2023) and Eastern California. Measuring distortion of quartz in these samples not only provides information about strain magnitudes but also strain directions. We tested the residual strain method with rolled titanium, #1). Recently we used the microfocus Laue technique to explore in situ fracturing and crack healing in calcite (#7).
- A project related to large stress started on a fieldtrip in spring 2025 to Meteor Crater near Flagstaff. We are investigating samples of ejecta which show extreme variations in temperature, pressure and stress from particles of original dolomite and quartz transforming to siliceous glass, olivine and clinopyroxene, with particles of FeNi from the Canyon Diablo meteorite. The research relies mainly on electron microscopy (imaging, elemental spectroscopy and backscatter diffraction) to

identify phases and their history with temperatures reaching ~2500°C and 10 GPa transforming dolomite and quartz in Permian sediments to olivine, clinopyroxene and stishovite. Similar conditions are observed in nuclear explosions (#4).

Publications supported by this project 2023-2025:

1. Devoe, Michelle; Tamura, Nobumichi; Wenk, Hans-Rudolf. "Residual strain orientation in rolled titanium sheet determined with synchrotron X-ray microdiffraction". *J. Appl. Cryst.*, **2023**, 56, 135-142, doi 10.1107/S1600576722011311.
2. Schmitt, Matthew, M.; Savage, Daniel, J.; Yeager, John, D.; Wenk, Hans-Rudolf; Lutterotti, Luca; Vogel, Sven C. "Texture measurements on quartz single crystal to validate coordinate systems for neutron time-of-flight texture analysis". *J. Appl. Cryst.*, **2023**, 56, 1764-1775, doi 10.1107/S1600576723009275.
3. Vasin, Roman; N., Kunz, Martin; Wenk, Hans-Rudolf; Zepeda-Alarcon, Eloisa. "Crystallographic texture formation in Fe-9wt%Si alloy during deformation and phase transition at high pressure". *Geophys. J. Int.* **2023**, 234, 790-806, doi10.1093/gji/ggad099.
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Fundamental Controls on Capillary Trapping of scCO₂ in Geologic Storage

Dorthe Wildenschild, Principal Investigator

School of Chemical, Biological and Environmental Engineering, Oregon State University, OR

Graduate Students: Daniel Enebe, Yussif Seidu

Collaborators: Anna Herring, University of Tennessee Knoxville

Overall research goals:

Recent research has shown that when multiple fluids are present within a porous medium, capillary trapping of the nonwetting fluid (e.g. carbon dioxide, CO₂) can be significantly enhanced via cyclic injection schemes; i.e. multiple repeated consecutive injections of water and nonwetting phase. The project goal is to explore the potential mechanisms of enhanced trapping during cycling -- including fluid property variation, surface chemistry alterations, and clay fines migration -- and determine how these mechanisms manifest across length scales, from the pore-scale to the core-scale. The use of CO₂ as a nonwetting fluid for these experiments provides a unique opportunity to control and manipulate fluid properties (viscosity, interfacial tension, density), due to the proximity of the CO₂ critical point to common subsurface storage reservoir temperature and pressure conditions. Exploration across fluid properties and sample types will enable delineation of potential trapping enhancement processes such as cyclic injection strategies e.g. [1], and support further exploration of a hypothesis that suggests that the connectivity (topology) of the CO₂ after initial injection is linked to trapping efficiency after subsequent brine invasion.

This project is a collaborative project: a broad range of “screening” core-flooding experiments are being conducted at the University of Tennessee, Knoxville (UTK); these results are analyzed to identify select conditions to undertake full 3D microCT imaging experiments conducted at Oregon State University (OSU). Our goals are: (1) establish a robust OSU-UTK collaboration with complementary project foci; (2) conduct cyclic supercritical CO₂-brine injection experiments under a range of pressure and temperature conditions to determine saturation and pressure trends during cycling in three sample types (quarry sandstone, synthetic, reservoir sandstone); and (3) quantify in precise detail the amount/efficiency and connectivity of trapped CO₂ under the various conditions targeted based on the UTK findings; and (4) explore the role of CO₂-phase connectivity after initial injection to test the hypothesis and findings of earlier studies e.g. [3] that suggest that the topology of the CO₂ after initial injection, dictates the amount eventually trapped as residual phase, and thus the trapping efficiency.

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Significant achievements during 2023- 2025:

The project was initiated in Fall 2024 and operates as a collaborative experimental effort between the University of Tennessee, Knoxville (UTK), and Oregon State University (OSU). In support of the project's micro-CT imaging-focused objectives at OSU, two OSU-based Graduate Research Assistants (Enebe and Seidu) conducted ambient-condition experiments using sintered glass beads as a porous media analog, with Soltrol 220 and water representing supercritical CO₂ and subsurface brine, respectively. The purpose of this preliminary experimental phase was to investigate optimal fluid-flow control strategies, utilize quantitative metrics (Euler number χ) to assess existing CO₂ connectivity hypotheses, and develop image analysis workflows relevant to subsequent high

pressure–temperature core-flooding experiments. Results from these ambient-condition experiments were presented by Daniel Enebe at the International Society for Porous Media (InterPore) Annual Conference in Albuquerque, NM, May 19–22, 2025.

Insights gained from these initial ambient experiments informed the transition to high pressure–temperature CO₂–brine core-flooding experiments currently underway at OSU. Rebuilding and improving a high pressure-temperature experimental setup (used at OSU in a previous BES-GEO funded project) took several months in early-mid 2025 to make sure the system operates safely and produces reliable results.

The high pressure-temperature experiments target select pressure–temperature conditions ranging between 1250–1750 psi and 20–40 °C and utilize sandstone core samples to directly investigate pore-scale trapping mechanisms under conditions relevant to subsurface carbon storage.

Though the analysis of high pressure–temperature imaging data at OSU is still ongoing, preliminary ambient-condition experiments revealed tentative pore-scale trends:

- Higher flow rates appeared to reduce post-drainage nonwetting-phase connectivity, potentially enhancing residual trapping.
- A weak inverse relationship was also observed between trapping efficiency and the Euler characteristic in packed glass beads, suggesting reduced connectivity corresponds to increased immobilization.

Science objectives for 2026-2028:

The objectives for 2026–2028 focus on advancing a mechanistic understanding of CO₂ capillary trapping through controlled pressure–temperature experimentation, cyclic injection, and pore-scale imaging and quantification. The study will expand the range of pressure–temperature conditions (liquid and supercritical CO₂) and porous media types to isolate the effects of fluid state, pore structure, and surface characteristics on trapping behavior. Quantitative comparisons will be conducted across six pressure–temperature conditions and multiple porous media while maintaining constant capillary pressure and capillary number. Analyses will prioritize first-cycle behavior, followed by multi-cycle evaluation to assess trapping evolution. Comparisons will include:

- Non-wetting phase saturations, topology, connectivity, interfacial area, and curvature derived from X-ray imaging
- Differential pressure and saturation response during CO₂–brine cyclic injection
- Pore-to-core scale scaling using region-of-interest and full-core imaging

Subsequently, “Novel Strategy” experiments will be conducted as informed by earlier analysis to validate optimal injection strategies (e.g., specific cyclic schemes and flow rates) for maximizing CO₂ trapping in specific rock types.

Publications supported by this project 2023-2025:

Due to a very complex experimental build, we do not yet have sufficient data to submit manuscripts for peer-reviewed journal publication, but there are several planned for the coming year as we now have a fully operational high pressure-temperature experimental system.

1. Daniel Enebe, Simone Baumgartner, and Dorthe Wildenschild, “Optimization of Geologic Carbon Sequestration: Effect of Flowrate on Non-wetting Phase Connectivity”, International Society for Porous Media Annual Conference Albuquerque, NM, May 19–22, 2025, Conference abstract, <https://events.interpore.org/event/56/attachments/1242/2271/InterPore2025%20Book%20of%20Abstracts.pdf>
2. Shukrah A-Olabiyi, Christopher Allison, Daniel Enebe, Dorthe Wildenschild, Anna Herring, “Evolution of CO₂ Saturation under Multicycle Drainage–Imbibition on Bentheimer Sandstone.” In preparation for submission to *International Journal of Greenhouse Gas Control*.
3. Daniel Enebe, Yussif Seidu, Shukrah A-Olabiyi, Christopher Allison, Anna Herring, Dorthe Wildenschild, “The Effect of Force Balance and Topology on Trapping Efficiency During Subsurface Supercritical CO₂ Injection and Storage”. In preparation for submission to *International Journal of Greenhouse Gas Control*.

Building a Framework to Understand Transition Metals' Behavior in Euxinic Conditions

Jie Xu, Principal Investigator

School of Molecular Sciences, Arizona State University, Tempe, AZ

Graduate Students: Rachel Phillips (graduated in 2023), Tan Truong

Postdocs: Andrew Weitz (06/2023–04/2024)

Collaborators: Oliver Muleler (SLAC National Accelerator Laboratory), Morgane Desmau and Roman Chenikov (Canadian Light Source), Muammar Mansor (University of Tuebingen, Germany), Michael Hochella (*Emeritus* Professor, Virginia Tech), Benjamin Brugman and Alexandra Navrotsky (Arizona State University)

Overall research goals:

This project examines how transition metals interact with reduced sulfur species (e.g., HS^- , S_x^{2-}) in low-temperature aqueous systems, with the goal of identifying dominant reaction pathways and the controls on the formation, transformation, and persistence of metal–sulfur phases. Two objectives guide the work. First, we quantify how molybdate, vanadate/vanadyl, and Mn(IV) oxyhydroxides react with sulfide under controlled conditions, including the influence of co-occurring metal cations and redox active couples (e.g., $\text{Fe}^{2+}/\text{Fe}^{3+}$, $\text{Ni}^{2+}/\text{Ni}^{3+}$, $\text{Cu}^+/\text{Cu}^{2+}$) that can modify redox balance, coordination environments, and precipitation pathways. Second, we develop a structure–reactivity framework linking the electronic structure of resulting (oxy)sulfide phases to crystal chemistry, kinetic stability, and reactivity by integrating new experimental constraints with prior theoretical and computational results.

Significant achievements during 2023-2025:

Prior to 2023, our work examined how transition-metal cations (Fe, Cu, Ni, Co) react with HS^- and S_x^{2-} to form metastable sulfide precipitates whose short-range structure is governed by nucleation kinetics, defect chemistry, non-stoichiometry, and metal–sulfur covalency (TM d–S 3p hybridization), often yielding poorly crystalline or nanocrystalline products that subsequently transform. From 2023 to 2025, we focused on transition-metal oxyanion–sulfide systems (molybdate and vanadate), where thiolation, condensation/polymerization, and coupled redox reactions generate evolving thio-oxo speciation and, ultimately, amorphous or nanophase (oxy)sulfide solids. To resolve these materials, we combined X-ray photoelectron spectroscopy (XPS), X-ray absorption spectroscopy (XANES/EXAFS), and pair distribution function (PDF) analysis to constrain local coordination and intermediate-range order. In the molybdate–sulfide system, a key finding is that Mo(VI) undergoes stepwise thiolation to thiomolybdates, with faster conversion at lower pH and, under circumneutral conditions, in the presence of $\text{Fe}^{2+}/\text{Ni}^{2+}$ and/or freshly precipitated FeS/NiS. Subsequent reduction and condensation of thiolated Mo are consistent with FeS/NiS-associated electron transfer and sulfur–sulfur bond formation within evolving Mo–S motifs, yielding oligomeric Mo–S species and ultimately amorphous Mo–S and mixed Fe–Mo–S precipitates. In Mo–Cu–sulfide systems, we observed distinct Mo–S–O frameworks relative to Mo–Fe–S, consistent with slower electron-transfer kinetics associated with CuS-containing clusters. Together, these results constrain abiotic Mo sequestration pathways in sulfidic environments and provide a basis to evaluate the stability and potential catalytic reactivity of largely amorphous Mo-bearing sulfide phases.

Science objectives for 2026-2028:

For 2026–2028, we will complete mechanistic studies of vanadate transformation under sulfidic conditions, with and without common transition-metal cations (e.g., Fe, Cu, and Co). Under mildly acidic to circumneutral conditions, vanadate can act as an oxidant for sulfide, potentially producing S^0 that is captured as dissolved polysulfides when HS^- is in excess. Under more strongly sulfidic conditions and/or in the presence of freshly precipitated metal sulfides (e.g., FeS), V may be reduced to V(IV) and further to surface-associated V(III), leading to rapid sequestration on sulfide mineral

surfaces and/or incorporation into mixed amorphous metal–sulfur solids. We will use controlled sulfidic conditions and metal sulfide nanoparticle formation as tools to constrain apparent pH–Eh stability fields and to resolve abiotic sequestration pathways (surface association versus mixed-solid formation). A parallel set of experiments in Mn-(transition metal)-sulfide systems will provide comparative constraints on Mn fate under sulfidic conditions. Further, we will start to characterize the magnetic properties of the M-Mo-sulfide solid phases, aiming to connect the atomic structures with underlying physicochemical properties. In parallel, we will complete a review titled “Metal–Sulfur Bonding in Sulfide Minerals and Related Nanostructures: Electronic Structure, Spectroscopy, and Theory,” synthesizing advances since Vaughan and Rosso (2006).

Publications supported by this project 2023-2025:

Use this format (Vol, Pages, are optional) and hyperlink the DOI:

1. Jie Xu, Geng Yan, Benjamin Brugman, Logan Leinbach, Peng Zuo, Jared Matteucci, Hongwu Xu, Alexandra Navrotsky, “Pressure-Induced Ordering and Mesoporosity in Amorphous Magnesium Carbonate” *Crystal Growth & Design*, **2025**, 25, 10261-10267.
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Isotopic Signatures of Fluid-Rock Interactions Close to and Far from Equilibrium

Piotr Zarzycki, John Christensen, Donald DePaolo, Benjamin Gilbert, Harry Lisabeth, Daniel Stolper, Michael Whittaker, Principal Investigators

Energy Geoscience Division, Lawrence Berkeley National Laboratory, Berkeley, CA

Graduate Students: Yarong Qi (UC Berkeley), Zsolt Marka (UC Berkeley), Ziman Wu (UC Berkeley).

Postdocs: Flora Hochsheid (LBNL/UC Berkeley), Andrew Turner (LBNL/UC Berkeley)

Collaborators: Roman Korol (UdeS, QC, Canada), Drew Syverson (UCN Charlotte, NC)

Overall research goals:

The isotopic composition of geological materials has been studied since the 1940s. Today, virtually every element in the periodic table that has multiple isotopes is measured as part of geochemical studies. Isotope geochemistry is anchored in and linked to basic chemistry, as isotopes influence bond energies that, in turn, are reflected in both equilibrium and kinetic processes. Measurements of isotopic substitutions have long served as tracers of chemical mechanisms and tests for theoretical calculations in chemistry. The application of isotopic measurements grounded in chemical theory has led to key geochemical discoveries, ranging from the reconstruction of Earth's climate history to the oldest evidence of life on Earth.

This project focuses on measuring and calculating the isotopic signatures of chemical reactions that occur between minerals, water, and solutes. Our goal is to obtain a new level of understanding of the molecular mechanisms associated with mineral growth and solute speciation. We use the carbonate system as our example study system given its importance both in fundamental geochemical studies and applications across a range of Earth Science areas. We are specifically conducting experiments and theoretical calculations to examine whether condensed phase theory is capable of matching both the accuracy and precision of modern geochemistry and thus is useful to systems and conditions that experiments cannot simulate.

Significant achievements during 2023- 2025:

Our work commenced in October 2025 and is grounded in prior published work supported by the BES project. This includes, for example, measurements and calculations of hydrogen isotope effects between small inorganic and organic molecules (Korol et al., 2025; Turner et al., 2025; Hochsheid et al. 2025, Eldridge et al., 2023), consideration of mineral growth in confined systems (Legg et al., 2025) the importance of complexation models in geochemistry (Zarzycki et al., 2025), and mineral dissolution rates at near chemical equilibrium (DePaolo and Zhang, 2024)

As our new Thrust has begun over the past few months, we have developed computational pipelines and experimental protocols for high-accuracy predictions and measurements of isotopic fractionation factors. Here, we illustrate our approach by presenting the preliminary experimental and computational results for Ca, O, and C fractionation between carbonate minerals and between minerals and fluids under near-equilibrium conditions.

Science objectives for 2026-2028:

We will focus on systems in which carbonate and dissolved carbonate species participate in isotopic exchange reactions across a range of temperatures and conditions. This includes high-level computations and corresponding laboratory measurements of isotopic exchange between gas phase molecules and aqueous solutions (e.g., CO₂, H₂, H₂O), aqueous solution and mineral phase (e.g., divalent cations, carbonate and phosphate ions exchange between carbonate mineral surfaces and solutions at varying pH, pressure, and temperature conditions), and finally isotopic equilibrium

of carbonate mineral phases (e.g., calcite, aragonite, magnesite, rhodochrosite, siderite). We employ a range of measurements and associated calculations including bulk isotope compositions (e.g., $^{44}\text{Ca}/^{40}\text{Ca}$ and $^{18}\text{O}/^{16}\text{O}$), systems with multiple isotopes such as determinations of ^{16}O , ^{17}O , and ^{18}O , and finally clumped isotopes with multiple rare substitutions (e.g., $^{13}\text{C}^{16}\text{O}_2^{18}\text{O}$). This range allows us to probe a variety of different aspects of the energetics of systems and thus assumptions embedded in various theoretical calculations.

Our specific goals are as follows. We will precipitate dissolved inorganic carbon from solution at a range of pH and temperature to measure isotopic properties described above and compare with high-level models. We will grow carbonate minerals in chemostatic systems under a range of growth rates and trace element contents and measure isotope effects and compare to models of growth. We will recrystallize carbonate minerals at ultra slow rates using a compaction system and determine equilibrium isotopic and chemical partitioning between mineral and fluid. Illustrative examples of this future work will be given in the poster.

Publications supported by this project 2023-2025:

1. Korol, Roman, Andrew C. Turner, Apurba Nandi, Joel M. Bowman, William A. Goddard III, and Daniel A. Stolper. "Stable isotope equilibria in the dihydrogen-water-methane-ethane-propane system. Part 1: Path-integral calculations with CCSD(T) quality potentials." *Geochimica et Cosmochimica Acta* **2025**, 396, 71-90. [10.1016/j.gca.2025.02.028](https://doi.org/10.1016/j.gca.2025.02.028).
2. Turner, Andrew C., Roman Korol, Markus Bill, and Daniel A. Stolper. "Stable isotope equilibria in the dihydrogen-water-methane-ethane-propane system. Part 2: Experimental determination of hydrogen isotopic equilibrium for ethane-H₂ from 30 to 200° C and propane-H₂ from 75 to 200° C." *Geochimica et Cosmochimica Acta* **2025**, 396, 91-106. [10.1016/j.gca.2025.02.033](https://doi.org/10.1016/j.gca.2025.02.033).
3. Hochscheid, Flora, Andrew C. Turner, Noam Lotem, Markus Bill, and Daniel A. Stolper. "Experimental determination of hydrogen isotopic equilibrium in the system H₂O (l)-H₂ (g) from 3 to 90° C." *Geochimica et Cosmochimica Acta*, **2025**, 394, 368-382, [10.1016/j.gca.2025.02.029](https://doi.org/10.1016/j.gca.2025.02.029).
4. Eldridge, Daniel L., Andrew C. Turner, Markus Bill, Mark E. Conrad, and Daniel A. Stolper. "Experimental determinations of carbon and hydrogen isotope fractionations and methane clumped isotope compositions associated with ethane pyrolysis from 550 to 600° C." *Geochimica et Cosmochimica Acta* **2023**, 396, 235-265. [10.1016/j.gca.2023.06.006](https://doi.org/10.1016/j.gca.2023.06.006).
5. Legg, Benjamin A., Yandi Hu, Ayumi Koishi, and Michael Whittaker. "Pathways for Nucleation and Growth in Confined Spaces and at Interfaces." *Elements* **2025**, 21, 1, 40-45. [10.2138/gselements.21.1.40](https://doi.org/10.2138/gselements.21.1.40).
6. DePaolo, Donald J., and Shuo Zhang. "Reaction order of near equilibrium calcite dissolution: Uncertainties and ambiguities of isotopic tracer methods." *Geochimica et Cosmochimica Acta* **2024**, 379, 233-243. [10.1016/j.gca.2024.06.004](https://doi.org/10.1016/j.gca.2024.06.004).
7. Zarzycki, Piotr, Johannes Lützenkirchen, and Benjamin Gilbert, eds. "Surface Complexation Models." *Reviews in Mineralogy and Geochemistry* **2025**, 91A. [10.2138/rmg.2025.91A.00](https://doi.org/10.2138/rmg.2025.91A.00).

The Effect of Surface Forces on Subcritical Crack Growth and Macroscopic Failure of Carbonates in Aqueous solutions

Yida Zhang, Principal Investigator

Department of Civil, Environmental and Architectural Engineering, University of Colorado Boulder, Boulder, CO.

Graduate Students: Hooman Dadras (CUBoulder), Mohammadali Behboodi (CUBoulder).

Collaborators: Seiji Nakagawa (LBNL), Benjamin Gilbert (LBNL), Harry Lisabeth (LBNL), Piotr Zarzycki (LBNL).

Overall research goals:

The central hypothesis of this research is that crack growth kinetics can be predicted by focusing on two key physio-chemical attributions, namely the alternation of surface forces due to the intrusion of sorptive species and the transport of these species along the crack. We further hypothesize that the chemical-mediated time-dependent compaction, failure, and localization of porous rocks can be understood in the same vein by resolving the surface energetics and chemistry during new surface area creation (due to microcracking, grain crushing, etc.). Specific research goals are: 1) To establish a mechanistic theory for subcritical crack growth (SCG) in calcite exposed to aqueous solutions by properly modeling surface forces and ion transport under nanoconfinement; and 2) To upscale the SCG analysis to inform continuum-scale modeling of carbonate rocks and to investigate the potential for deformation banding induced by changes in pore fluid chemistry.

Significant achievements during 2023- 2025:

Build upon our Surface Force based Fracture Theory (SFFT) (Eskandari-Ghadi et al., 2022), we have successfully enriched the mass-transport solver to account for the varying crack opening along the crack. The new transport solver is based on a finite difference algorithm. The nanoconfinement-dependent fluid viscosity and permeability are now inherently embedded in the Fick's law. The transport solver is successfully coupled with the other components of SFFT and can now produce more realistic fluid pressure profile during crack growth and healing. Significant effort has been devoted to revise the SFFT code to perform reduced stress intensity (i.e., healing) simulations. We borrowed elements from contact mechanics, specifically, the Lagrange multiplier method, to prevent crack faces from overlapping with each other. This, combined with the Barenblatt cohesive crack framework, can provide a working code to mimic the virgin cracking, healing, and recracking behaviors of brittle solids. We are continuing our close collaboration with LBNL to test and validate our model hypotheses using double-torsion (DT) experimental data on calcite and glass materials. One manuscript is under preparation to be submitted to Journal of the Mechanics and Physics of Solids (JMPS).

The adaptation of SFFT for calcite SCG simulation is progressing smoothly and rapidly. We have implemented a new surface force model that explicitly resolves the Van der Waals, electrical double layer, and hydration forces. The attractive force due to covalent bonds is now incorporated separately through a constant term in the fundamental linear elastic fracture mechanics (LEFM) equations. The model is now subjected to intense calibration and parametric studies for us to understand the role of different surface forces and ion transportation on the predicted SCG curves. The manuscript is now published in JGR-Solid earth.

We have started working on a chemomechanical continuum model for carbonate rocks. The key idea of the model is to use specific surface area as the single Internal State Variable (ISV) to represent the development of damage and grain breakage in the solid matrix. The advantage of this choice is that surface energy, as the thermodynamic conjugate of specific surface area, be naturally incorporated into the resulting constitutive relations. Therefore, the effect of chemical composition of

the pore fluid automatically enters the model through surface energy to control the yielding and hardening responses of the material. We are currently developing this surface-based damage model to ensure it captures the fundamental hardening and softening behavior of carbonate rocks sheared under varying mean effective stress levels, while remaining simple enough to enable analytical and numerical bifurcation analyses for studying chemically induced strain localization.

Science objectives for 2026-2028:

Finalizing and submitting the manuscript on "Interfacial fluid dynamics controls the subcritical crack growth and spontaneous crack healing of brittle solids".

Preparing a manuscript on "Spontaneous crack healing: experimental observation and thermodynamics" in collaboration with LBNL.

Fully develop a surface-based damage theory that capable of capturing the stress-strain behavior, creep, delayed and failure of carbonate rocks. Then conduct Rudnicki and Rice (1975)'s bifurcation analysis to probe the condition for shear banding and compaction banding of carbonate rocks at various stress and pore chemistry conditions.

Publications supported by this project 2023-2025:

1. Hooman Dadras, Mehdi Eskandari-Ghadi, Seiji Nakagawa, Benjamin Gilbert, and Yida Zhang, "Complex dependence of calcite crack kinetics on salinity: the role of DLVO and hydration forces", *Journal of Geophysical Research: Solid Earth*, **2025**, 130, e2025JB031332, <https://doi.org/10.1029/2025JB031332>
2. Mohammadali Behboodi, and Yida Zhang, "Interfacial Evolution Explains the Complex Swelling-Shrinkage Responses of Porous Materials from Vacuum-Dry to Full Liquid Saturation." *Journal of the Mechanics and Physics of Solids*, **2025**, 106425, <https://doi.org/10.1016/j.jmps.2025.106425>
3. Mohammadali Behboodi, Mehdi Eskandari-Ghadi, Wenjie Xia, and Yida Zhang, "An analytical model for the bending and reaction force of hygroscopic bilayers upon water adsorption." *International Journal of Solids and Structures*, **2025**, 309, 113191, <https://doi.org/10.1016/j.ijsolstr.2024.113191>
4. Shubjot Singh, Yuxuan Wen, Wanqi Yu, Efrain A. Rondinel Oviedo, Julie A. Korak, Giuseppe Buscarnera, Yida Zhang, "Long-term Creep and Permeability Evolution of Tailings Sand at Elevated Stresses", **2026**, in press.

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- Rudnicki, J.W., Rice, J., 1975. Conditions for the localization of deformation in pressure-sensitive dilatant materials. *Journal of the Mechanics and Physics of Solids* 23, 371–394.

Seismic Foundation Model for Integrated Subsurface Monitoring

Weiqiang Zhu, Principal Investigator

Department of Earth and Planetary Science, University of California, Berkeley, CA

Graduate Students: Haoyu Wang

Collaborators: Chun Zhang, Junhao Song, Richard Allen (UCB), Zhongwen Zhan (Caltech)

Overall research goals:

Monitoring subsurface processes for energy applications faces multiple challenges: the rapidly growing volume of seismic data from modern sensor networks, the complexity of integrating multiple recording systems with different characteristics, and the difficulty of understanding complex induced seismicity patterns. Artificial Intelligence (AI) offers a promising solution to these challenges by enabling us to leverage information from multiple recording systems to improve earthquake detection and characterization. This project aims to develop a seismic foundation model that jointly learns from diverse seismic data sources, including conventional seismic networks, dense nodal arrays, borehole sensors, and fiber-optic distributed acoustic sensing, to detect previously missed microseismic signals and provide comprehensive insights into subsurface processes for geological energy applications, particularly enhanced geothermal systems and carbon storage.

Significant achievements during 2023- 2025:

(This is the first PI meeting for the project.)

Science objectives for 2026-2028:

Our objectives for 2026-2028 include: (1) Model Development: Design a unified transformer-based foundation model architecture for multi-modal seismic data processing. (2) Dataset Building: Curate a large multi-modal training dataset from seismic networks, dense nodal arrays, borehole arrays, and distributed acoustic sensing deployments. (3) Model Training: Train and validate the foundation model using self-supervised learning and multi-task learning strategies to enable robust performance across diverse monitoring scenarios.

Publications supported by this project 2023-2025:

(No publications yet.)

Evolution of pore structure and rock strength in subsurface fluid-rock systems

Wen-lu Zhu, Tommaso Mandolini, Travis Hager, Zack Zega, and Megan Ryan

Department of Geology, University of Maryland College Park

The safety and efficiency of many subsurface practices depend on accurate assessments of fluid flux and distribution. At various depths, the pore structure and permeability of rocks vary continuously under mechanical, thermal, and chemical stresses. Consequently, it is essential to quantify the spatial and temporal evolution of fluid flux and distribution. Additionally, fluid-rock interactions can profoundly modify the strength and deformation behaviors of subsurface rocks. Therefore, understanding the coupling and feedback between chemical reactions and mechanical deformation is crucial in energy-related subsurface research.

Our experimental investigation aims at elucidating the physical and chemical processes that control fracture propagation and fault permeability under subsurface conditions. Using synchrotron X-ray microtomographic imaging, we quantified the three-dimensional (3D) pore geometry and permeability structure of the fault core, damage zone, and wall rock of experimentally deformed porous rocks. Our results show that, under the same effective stress, faulting style, fault morphology, and fault permeability structure vary significantly across different pore-fluid pressures. Under high pore-fluid pressure, unstable fault propagation is inhibited, and the resulting fault zone is more diffuse and exhibits a much higher intensity of microcracking and grain comminution. Significant enhancements in permeability and permeability anisotropy are observed in fault zones formed under high pore-fluid pressures. These results provide new insights into the evolution of fracture network stability and permeability in enhanced geothermal reservoirs.

To quantify the feedback between chemical reactions and mechanical deformation, we also conducted in.situ microtomography experiments and measured the microstructural evolution of silicate rocks reacting with carbon-rich brines. We quantified the spatial and temporal evolution of pore structure and quantified the rate and extent of reaction-induced fractures. We found that the precipitation of reaction products is sensitive to carbon concentrations. Consequently, the rate of reaction-induced fracturing varies significantly in brines with different carbon content. These data enable us to address questions such as how the rate and extent of mineral carbonation change with time, and under what conditions reaction-induced fracturing occurs, which are critical pieces of information currently missing for developing an efficient strategy for carbon sequestration through reactive fluid injection.

*Attendee
List*

First Name	Last Name	Email Address	Company Name
Mohammad Javad	Abdolhosseini Qomi	mjaq@uci.edu	University of California, Irvine
Lawrence	Anovitz	anovitzlm@ornl.gov	Oak Ridge National Laboratory
Sourabh	Apte	sva@oregonstate.edu	Oregon State University
Polly	Arnold	pla@lbl.gov	Lawrence Berkeley National Laboratory
Lauren	Beckingham	leb0071@auburn.edu	Auburn University
Yehuda	Ben-Zion	benzion@usc.edu	University of Southern California
Gregory	Beroza	beroza@stanford.edu	Stanford University
Ian	Bourg	bourg@princeton.edu	Princeton University
Eric	Boyd	eric.boyd@montana.edu	Montana State University
Jacquelyn	Bracco	jbracco@qc.cuny.edu	Queens College, CUNY
Emily	Brodsky	brodsky@ucsc.edu	University of California, Santa Cruz
Giuseppe	Buscarnera	g-buscarnera@northwestern.edu	Northwestern University
Jeffrey	Catalano	catalano@wustl.edu	Washington University in St. Louis
Abdullah	Cihan	acihan@lbl.gov	Lawrence Berkeley National Laboratory
Maarten	De Hoop	mdehoop@rice.edu	Rice University
Donald	DePaolo	djdepaulo@lbl.gov	Lawrence Berkeley National Laboratory
Patricia	Dove	dove@vt.edu	Virginia Tech
Paul	Fenter	fenter@anl.gov	Argonne National Laboratory
Benjamin	Gilbert	bgilbert@lbl.gov	Lawrence Berkeley National Laboratory
Pupa	Gilbert	pgilbert@lbl.gov	University of Wisconsin, Madison and Lawrence Berkeley National Laboratory
Thomas	Goebel	thw.goebel@gmail.com	University of Memphis
Alexander	Gysi	alexander.gysi@nmt.edu	New Mexico Institute of Mining and Technology
Amanda	Haes	amanda.haes@science.doe.gov	U.S. Department of Energy, Basic Energy Sciences
Jesse	Hampton	jesse.hampton@wisc.edu	University of Wisconsin-Madison
Mark	Harrison	tmark.harrison@gmail.com	University of California, Los Angeles

Reza	Hedayat	hedayat@mines.edu	Colorado School of Mines
Anna	Herring	aherri18@utk.edu	University of Tennessee Knoxville
Tuan	Ho	taho@sandia.gov	Sandia National Laboratories
Benjamin	Holtzman	benh@mit.edu	Massachusetts Institute of Technology
Mengsu	Hu	mengsuhu@lbl.gov	Lawrence Berkeley National Laboratory
Ryan	Hurley	rhurley6@jhu.edu	Johns Hopkins University
Nicole	Hurtig	nicole.hurtig@nmt.edu	New Mexico Institute of Mining and Technology
Anastasia	Ilgen	agilgen@sandia.gov	Sandia National Laboratories
Eugene	Ilton	eugene.ilton@pnnl.gov	Pacific Northwest National Laboratory
Christopher	Johnson	cwj@lanl.gov	Los Alamos National Laboratory
Young-Shin	Jun	ysjun@wustl.edu	Washington University
Shaina	Kelly	sak2280@columbia.edu	Columbia University
Sebastien	Kerisit	sebastien.kerisit@pnnl.gov	Pacific Northwest National Laboratory
Aaditya	Khanal	aak4074@utulsa.edu	University of Tulsa
Matthew	Kohn	mattkohn@boisestate.edu	Boise State University
Tony	Ladd	tladd@che.ufl.edu	University of Florida
Sang Soo	Lee	sslee@anl.gov	Chemical Sciences and Engineering Division
Benjamin	Legg	benjamin.legg@pnnl.gov	Pacific Northwest National Laboratory
Kevin	Leung	kleung@sandia.gov	Geochemistry and Geomechanics Department
Harrison	Lisabeth	hlisabeth@lbl.gov	Lawrence Berkeley National Lab
Craig	Lundstrom	lundstro@illinois.edu	University of Illinois, Urbana- Champaign
Gail	McLean	gail.mclean@science.doe.gov	U.S. Department of Energy, Basic Energy Sciences
Tushar	Mittal	tmittal@psu.edu	Pennsylvania State University
Olufemi	Olorode	folorode@lsu.edu	Louisiana State University and A&M College
Daniel	O'Malley	omalled@lanl.gov	Los Alamos National Laboratory
Pratigya	Polissar	ppolissa@ucsc.edu	University of California, Santa Cruz

Laura	Pyrak-Nolte	ljpn@purdue.edu	Purdue University
Cornelia	Rasmussen	crasmussen@utexas.edu	University of Texas at Austin
Alex	Rinehart	alex.rinehart@nmt.edu	New Mexico Institute of Mining and Technology
Mark	Rivers	rivers@cars.uchicago.edu	University of Chicago
Jacques	Rivière	riviere@psu.edu	Pennsylvania State University
Kevin	Rosso	kevin.rosso@pnnl.gov	Pacific Northwest National Laboratory
Seth	Saltiel	sas697@cornell.edu	Cornell University
Heather	Savage	hsavage@ucsc.edu	University of California, Santa Cruz
Viviane	Schwartz	viviane.schwartz@science.doe.gov	U.S. Department of Energy, Basic Energy Sciences
Parisa	Shokouhi	parisa@psu.edu	Penn State
Pauline	Simonnin	pauline.simonnin@pnnl.gov	Pacific Northwest National Laboratory
Sarah	Slotznick	sslotz@dartmouth.edu	Dartmouth College
Duo	Song	duo.song@pnnl.gov	Pacific Northwest National Laboratory
Andrew	Stack	stackag@ornl.gov	Oak Ridge National Laboratory
Carl	Steefel	CSteefel@lbl.gov	Lawrence Berkeley National Laboratory
Daniel	Stolper	dstolper@berkeley.edu	University of California, Berkeley
Joanne	Stubbs	stubbs@cars.uchicago.edu	The University of Chicago Center for Advanced Radiation Sources
Ana	Suzana	asuzana@anl.gov	Argonne National Laboratory
Drew	Syverson	dsyverso@charlotte.edu	University of North Carolina, Charlotte
Priya	Vashishta	priyav@usc.edu	University of Southern California
Hari	Viswanathan	viswana@lanl.gov	Earth and Environmental Sciences Division
Laura	Waters	laura.waters@nmt.edu	New Mexico Institute of Mining and Technology
Hans-Rudolf	Wenk	wenk@berkeley.edu	University of California, Berkeley
Dorthe	Wildenschild	dorthe@engr.orst.edu	Oregon State University
Philip	Wilk	Philip.Wilk@science.doe.gov	U.S. Department of Energy
Jie	Xu	jiexu10@asu.edu	Arizona State University
Ke	Yuan	yuank@ornl.gov	Oak Ridge National Laboratory

Piotr	Zarzycki	ppzarzycki@lbl.gov	Lawrence Berkeley National Laboratory
Yida	Zhang	Yida.Zhang@Colorado.edu	University of Colorado, Boulder
Weiqiang	Zhu	zhuwq@berkeley.edu	University of California, Berkeley
Wen-lu	Zhu	wzhu@umd.edu	University of Maryland, College Park