Mineralization and Metamaterial Contrast Agent Monitoring in Geologic Carbon Storage Reservoirs

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To facilitate carbon storage monitoring and verification, we are investigating the low frequency (seismic) properties of injectable framework nanoparticles to be used as seismic metamaterial contrast agents. These injectates are low-frequency absorptive acoustic metamaterials exhibiting anomalous sound transmission loss and tunable resonance. This acoustic metamaterial technology can be used along with traditional seismic imaging techniques to enable transformational monitoring of subsurface CO_2. We will also present results from an in-revision Review manuscript (Holliman Jr. et al. 2022, Materials Advances) that discusses the broad field of metamaterials, including seismic metamaterials. We also report forced oscillation core test experiments to observe the effects of our metamaterial contrast agent on extensional seismic wave attenuation (Q^{-1}_E) and the Young's modulus (E) dispersion in rock samples by comparing dry, water-saturated, and contrast agent-saturated Berea sandstone samples. We validate our results using the Kramers-Kronig (K-K) relation, relating Q^{-1}_E and E through causality. In addition, we are performing a comprehensive K-K data analysis for published low-frequency forced oscillation studies. This involves extracting and analyzing compiled literature data, helping predict seismic responses in a broad range of lithologies and conditions. Lastly, we discuss new results for carbon storage via rapid mineralization in basalts at the Wallula Pilot Project, which were enabled by renewed focus on post-injection collected sidewall cores. Recent observations and new characterization of the samples has revealed exotic carbonate mineral compositions and zonation while clarifying the paragenetic sequence of mineral dissolution-precipitation reactions.

CO_2 Mineralization at the Wallula Basalt Sequestration Pilot Project

Wallula Basalt Sequestration Pilot Project injection site (~850 m) where 50 sidewall cores were recovered; many contained carbonate minerals resulting from the CO_2-basalt interactions

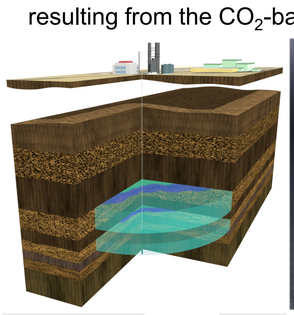
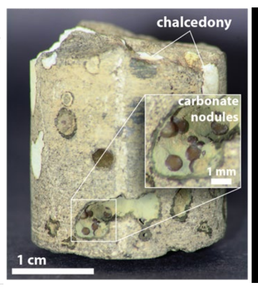
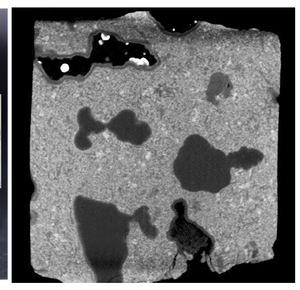


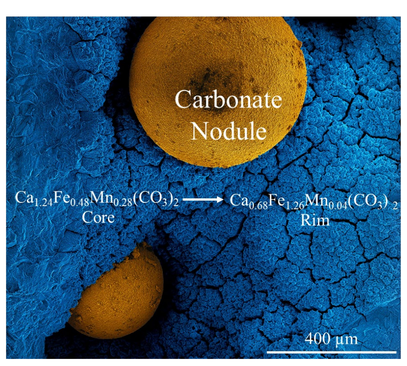
Diagram of injection site



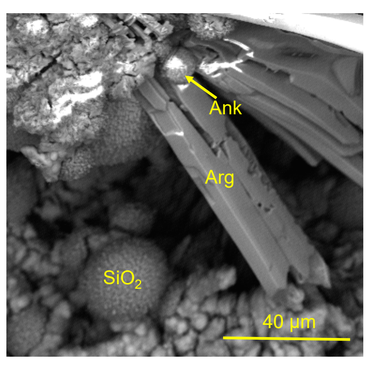
Sidewall core extracted from injection site



XMT analysis of a sidewall core containing carbonate nodules



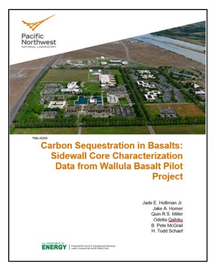
Zoned carbonate (siderite-ankerite) nodules rich in Ca, Fe, and Mn found within several sidewall cores

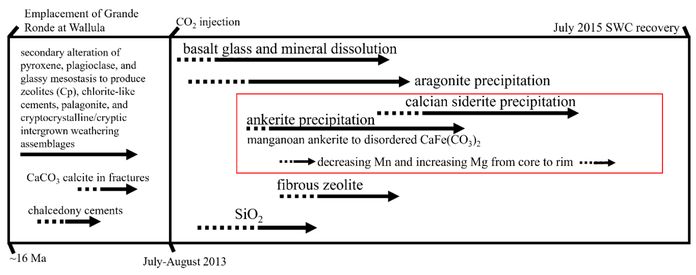


Amorphous silica is also proximal to the carbonate nodule (Ank) and aragonite (Arg) intergrowths.

Outcomes:

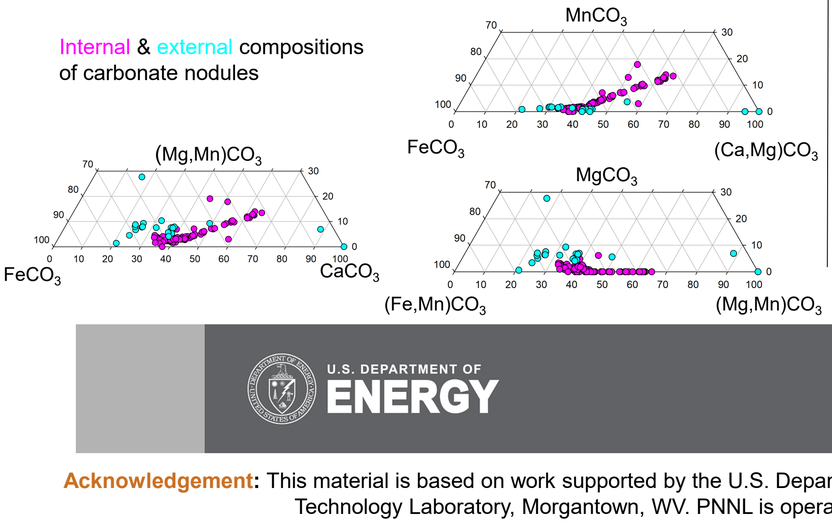
- Polites et al. 2022, Exotic Carbonate Mineralization Recovered from a Deep Basalt Carbon Storage Demonstration, In Revision at ES&T
- Horner et al. 2022, Intertek Basalt Core Analysis Report, PNNL Report 30940
- Holliman Jr. et al. 2022, Carbon Sequestration in Basalts: Sidewall Core Characterization Data from Wallula Basalt Pilot Project, PNNL Report 32848
- Depp et al., Pore-scale Microenvironments Control Anthropogenic Carbon Mineralization Outcomes in Basalt, In Review at ACS Earth & Space Chemistry



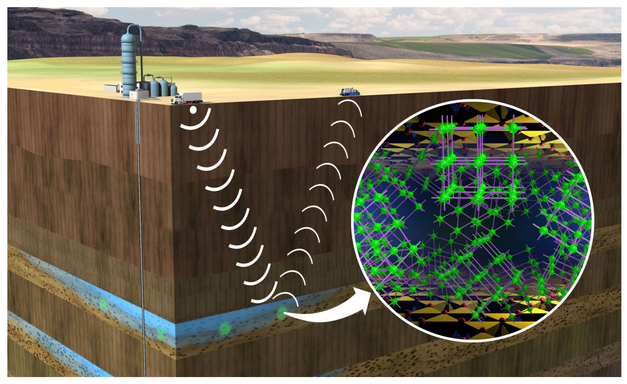


Overall mineralization paragenesis for the Grande Ronde basalt at the Wallula Basalt Carbon Storage Pilot Project. Dashed arrows denote uncertain initiation and duration. The red-framed box specifies that the compositions of the carbonate precipitates are consistent with ankerite and siderite.

Internal & external compositions of carbonate nodules

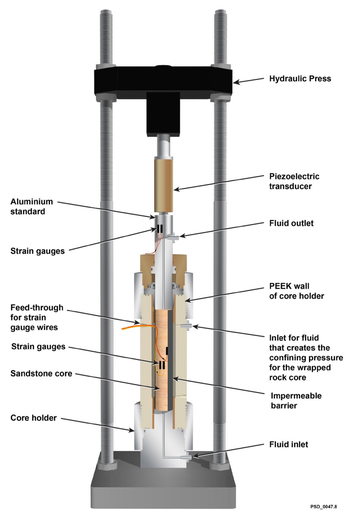


Seismic Contrast Agents for Enhanced CO_2 Monitoring

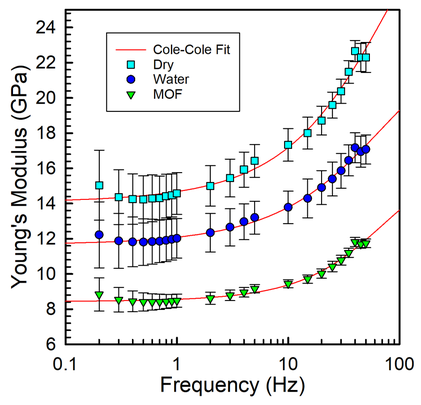


- New CO_2 monitoring technology using seismic metamaterial contrast agents in combination with seismic imaging techniques to create a reservoir-scale periodic structure

PNNL forced oscillation testing of injectable contrast agent nanofluids used to parameterize forward seismic modeling



Forced oscillation apparatus for measuring the elastic properties (Young's modulus, E) and extensional attenuation (Q^{-1}_E) in Berea sandstone subject to seismic oscillations

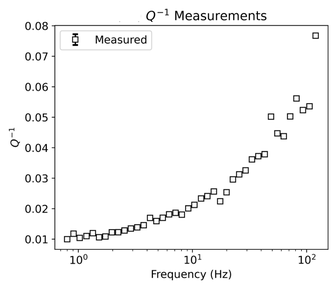


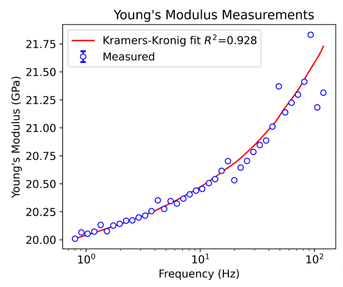
Berea sandstone in dry, water-saturated, and MOF nanofluid-saturated conditions exhibits a significant decrease in Young's modulus (E)

First quantitative analysis of broad forced oscillation literature via the Kramers-Kronig (K-K) relations to verify the causality between E and Q^{-1}_E

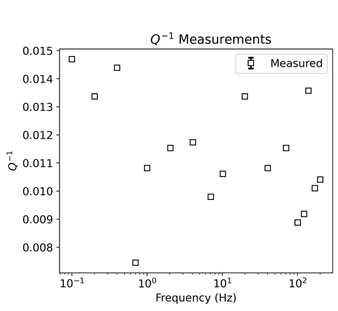
Forced oscillation datasets encompass a wide range of experimental conditions such as:

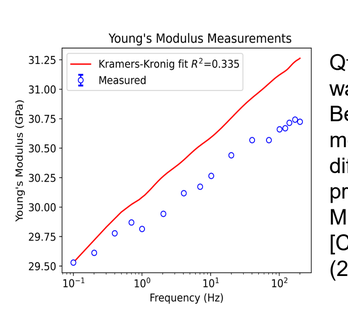
- Rock type & attributes (i.e., porosity, permeability, etc.)
- Saturating fluid (e.g., $scCO_2$, oil, brine)
- Differential pressure
- Boundary conditions (drained or undrained)
- Frequency
- Temperature





Q^{-1}_E and E for a water-glycerin-saturated Berea sandstone measured at a differential pressure of 9.86 MPa [Tisato et al. (2021)]

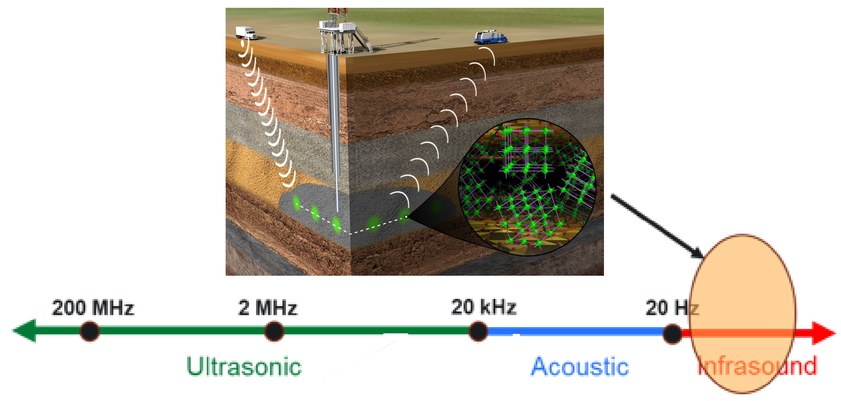




Q^{-1}_E and E for a water-saturated Berea sandstone measured at a differential pressure of 10 MPa [Chapman et al. (2022)]

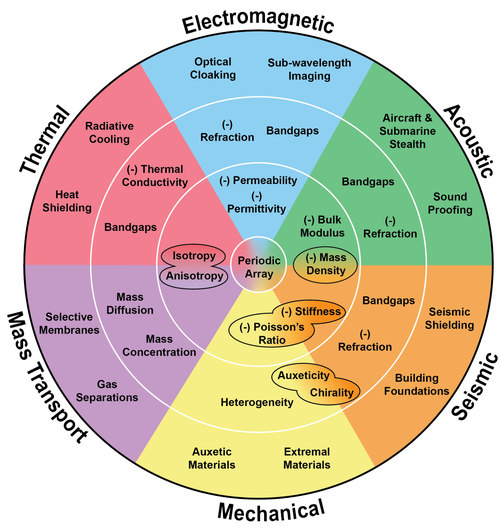
- Of the 286 datasets analyzed via the K-K relations, 87 (30.4%) produced R^2 fits of 0.8 or greater
- Ongoing analyses of the broad dataset will be used to parameterize predictive geophysical models that supplement our experimental campaign

Extending Principles of Metamaterial and Metastructure Design to the Subsurface

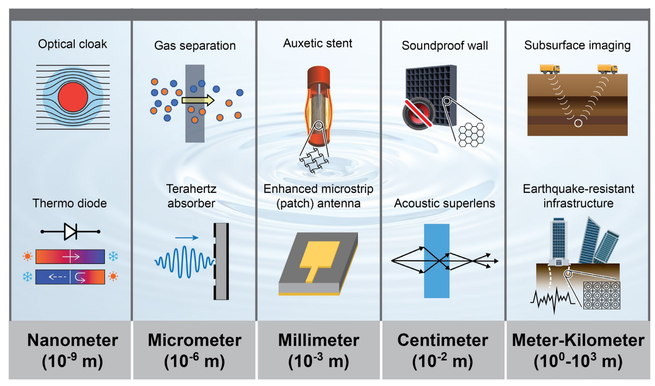


- Injectable metamaterials are key to imposing a reservoir-scale periodic structure for seismic wave manipulation
- Elastic metamaterials for seismic manipulation are less developed than acoustic and ultrasonic metamaterials
- The metastructure design would comprise of resonant periodic structures in the subsurface and increase resolution for better delineation of spatial relationships of fluids and pore/fracture networks

Understanding Metamaterials in a Broader Context



- Metamaterials are artificial materials composed of unit cells that work collectively to produce unusual, unique physical properties not found in natural materials or traditional composite materials
- Holliman Jr. et al. (2022, in revision) has prepared a Review Article covering the origin, current state of progress, and emerging directions for metamaterials



- Metamaterials have expanded over the last two decades to cover multiple physical domains (i.e., electromagnetism, acoustics, seismology, classical mechanics, mass transport, and thermodynamics)
- Metamaterials have a wide range of sizes and applications depending on the operating wavelength

Overall Highlights

- Continued analysis of world-unique sidewall core sample suite reveals carbon mineralization outcomes and pathways and will enable commercial scale carbon storage implementation
- Transformational carbon storage reservoir monitoring via injectable metamaterial contrast agents
- Driving forward the emerging field of seismic metamaterials to enhance monitoring of subsurface fluids and structures
- Key outcomes include three *in review/revision* manuscripts and two technical reports

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