Nanoparticle Injection Technology for Remediating Leaks of CO_2 Storage Formation

Project Number DE-FE0026514

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U.S. Department of Energy

National Energy Technology Laboratory

Mastering the Subsurface Through Technology Innovation, Partnerships and Collaboration:
Carbon Storage and Oil and Natural Gas Technologies Review Meeting

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Project Overview:
Objectives and Methodology

Project Goal: Develop new technology to repair wellbore leakages

Combination of a nanoparticle injection technique with the simultaneous extraction of harmful ions (e.g. chlorides) out of the leaking area.

Objective 1: Development of the injection technology for leakage repair.

- Electro-migration test unit
- Select healing agents
- Wellbore test system
- Evaluate effectiveness

Objective 2: Development of a new numerical simulation model that can simulate and predict the performance of the new wellbore repair technology.

- Model particle injection
- Model ionic removal

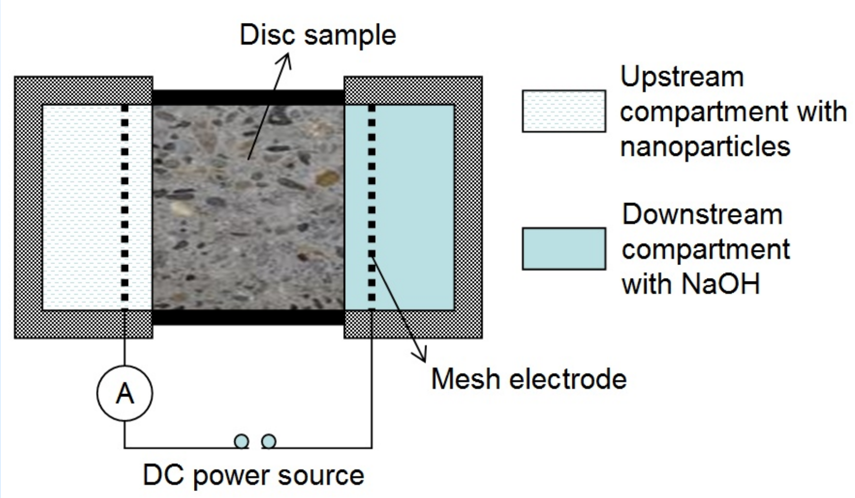
Concept Review

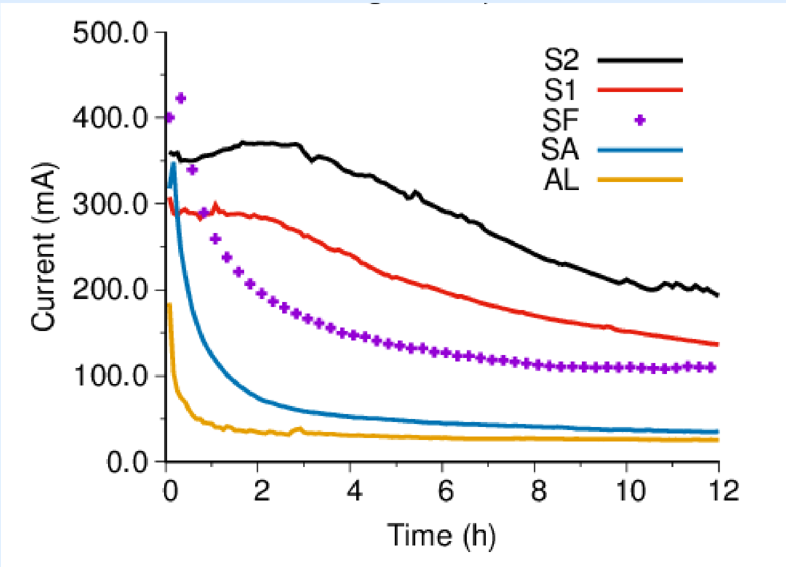
Electrochemical repair techniques are used for repairing reinforced concrete structures. We have developed this technology for repairing well cement.

Ionic exchange:

- High voltage
- Reverse anode and cathode regions
- Ions diffuse, causing gradient

Task 2: Electro-migration test unit

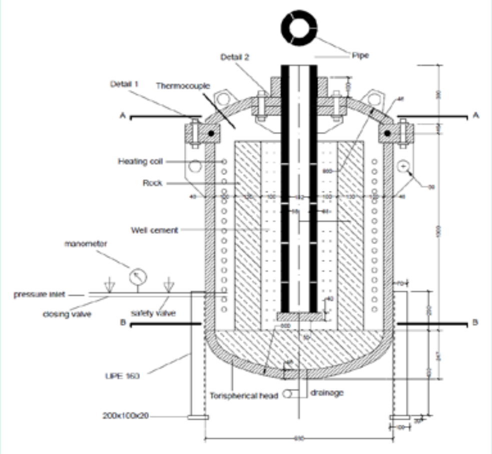




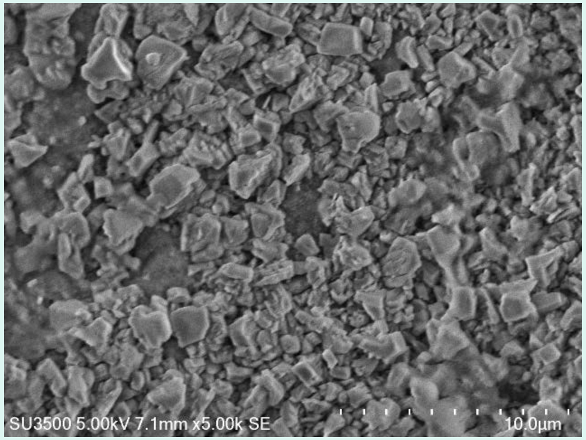
Presentation Outline

Task 3: Selection of healing agents

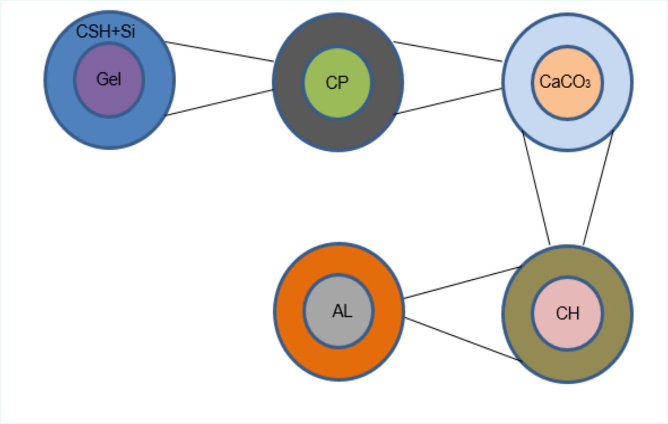
Task 4: Wellbore test system



Task 5: Evaluate effectiveness



Task 6: Numerical modeling



Task 3: Selection of healing agents

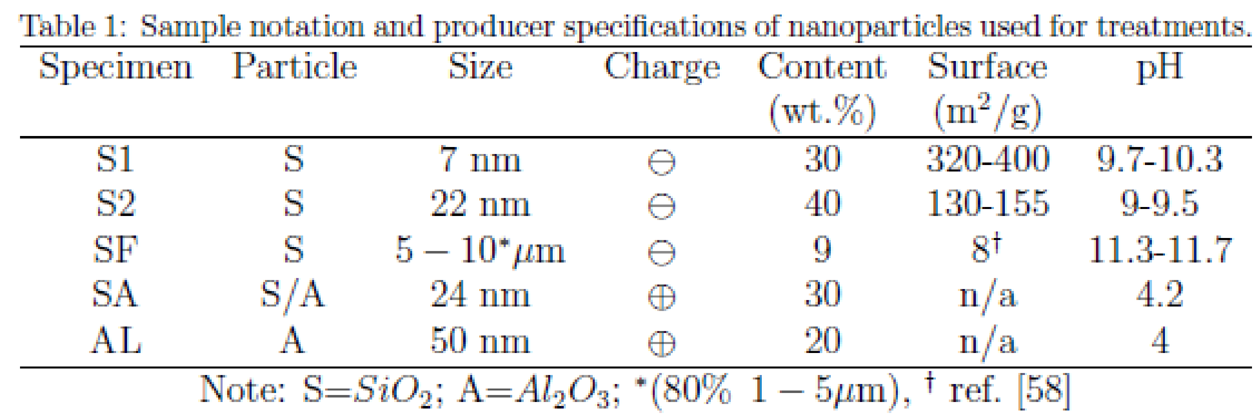


Table 1: Sample notation and producer specifications of nanoparticles used for treatments.

Specimen	Particle	Size	Charge	Content (wt.%)	Surface (m^2/g)	pH
S1	S	7 nm	⊖	30	320-400	9.7-10.3
S2	S	22 nm	⊖	40	130-155	9-9.5
SF	S	$5-10^{*}\mu m$	⊖	9	$8^{\dagger}$	11.3-11.7
SA	S/A	24 nm	⊕	30	n/a	4.2
AL	A	50 nm	⊕	20	n/a	4

Note: S=SiO_2; A=Al_2O_3; *(80% $1-5\mu m$), † ref. [58]

7nm nanosilica
22nm nanosilica
Fumed silica
Nanoalumina coated nanosilica
50nm nanoalumina

Extensive electro-migration tests:

- 50mm thick cylindrical specimens made of class-G oil well cement with 42% porosity.
- During the injection, the electric current was monitored and the total charge passed through the samples was logged.
- Two kinds of samples were tested, non-aged and aged. Aging was performed as thermal load (90°C lasting for 8 days) which caused severe cracking.

Task 3: Selection of healing agents

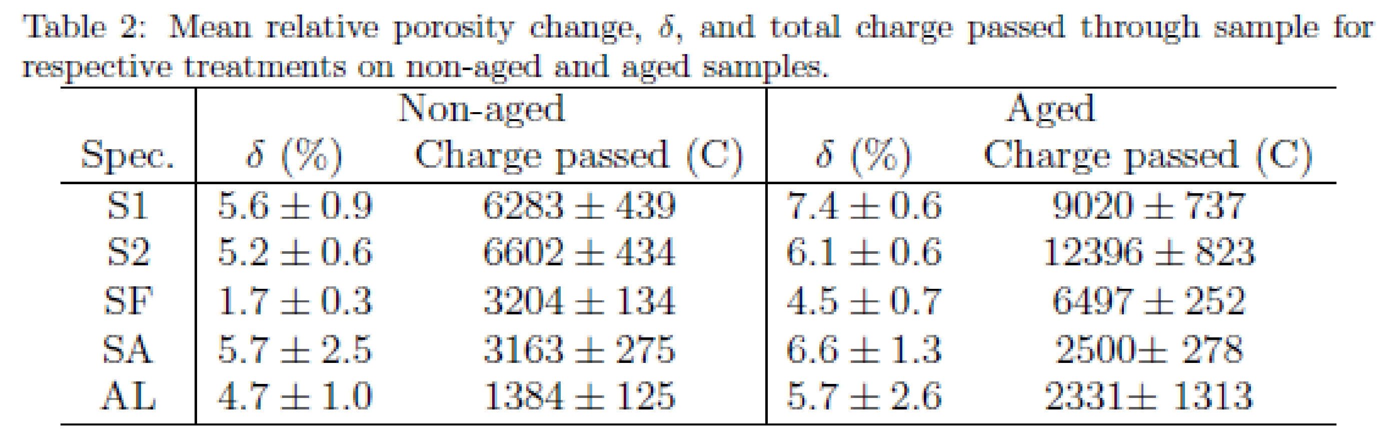
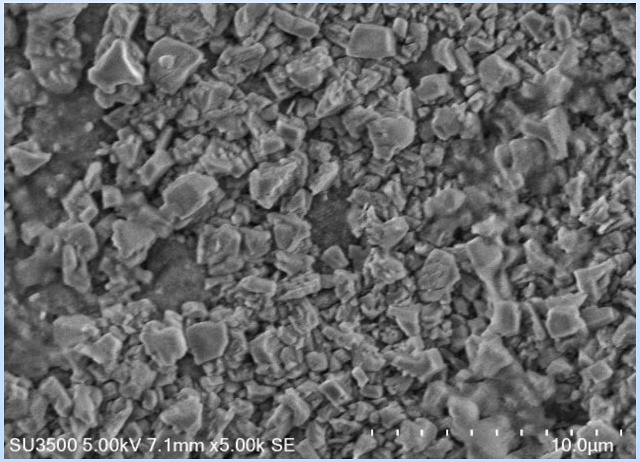


Table 2: Mean relative porosity change, δ, and total charge passed through sample for respective treatments on non-aged and aged samples.

	Non-aged		Aged	
Spec.	δ (%)	Charge passed (C)	δ (%)	Charge passed (C)
S1	5.6 ± 0.9	6283 ± 439	7.4 ± 0.6	9020 ± 737
S2	5.2 ± 0.6	6602 ± 434	6.1 ± 0.6	12396 ± 823
SF	1.7 ± 0.3	3204 ± 134	4.5 ± 0.7	6497 ± 252
SA	5.7 ± 2.5	3163 ± 275	6.6 ± 1.3	2500 ± 278
AL	4.7 ± 1.0	1384 ± 125	5.7 ± 2.6	2331 ± 1313

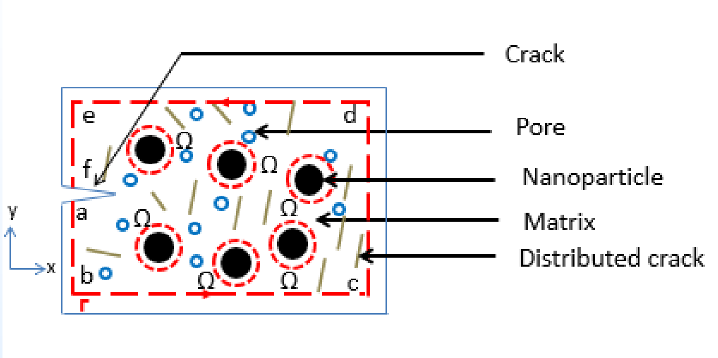


Key insights:

- Injection effectiveness based on current drop and particle agglomeration
- 7nm nanosilica was the most effective anionic particle
- Fumed silica is the most efficient cationic particle

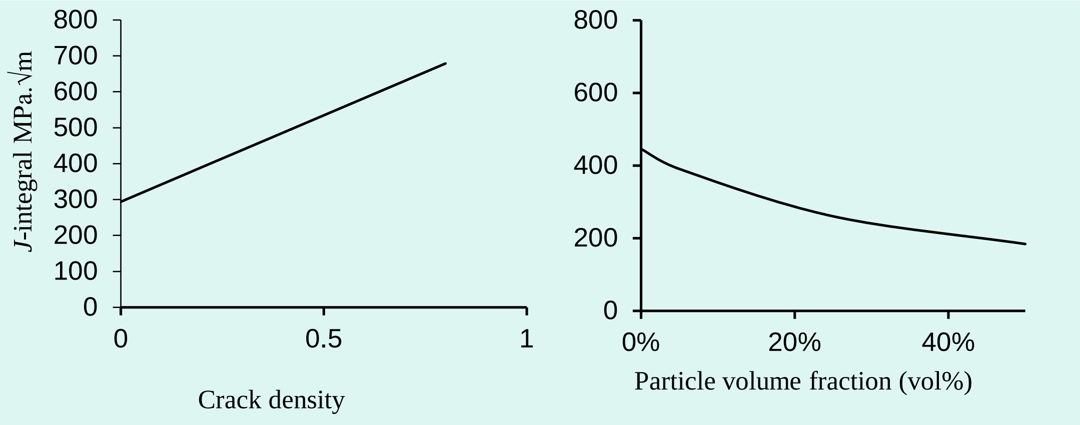
Surface agglomeration based on particle charge, size, and pH.

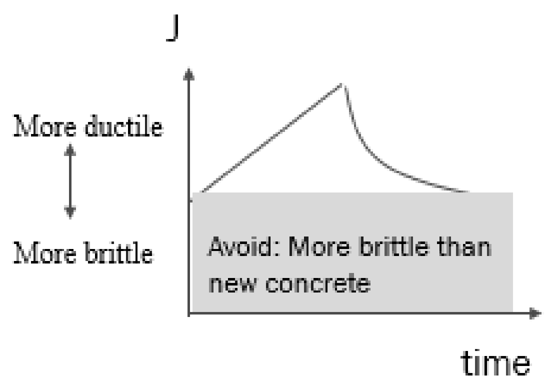
Task 3: Selection of healing agents



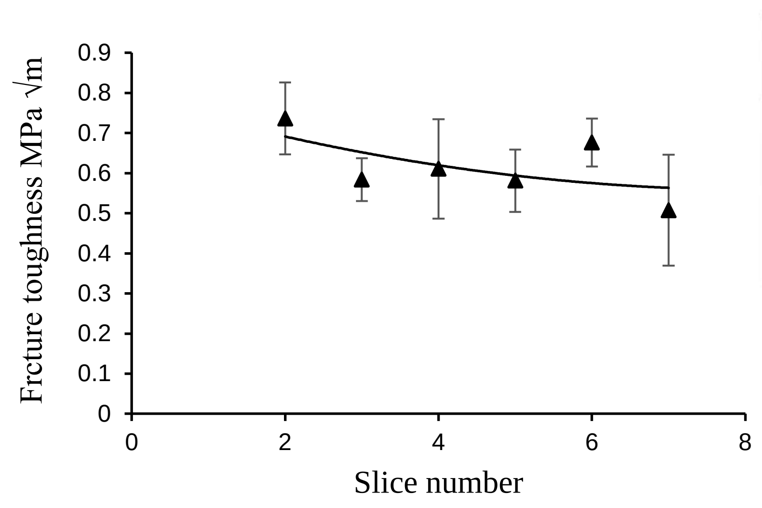
Fracture mechanics equation for cracking potential (toughness)

Theoretical behavior:





Experimental verification:

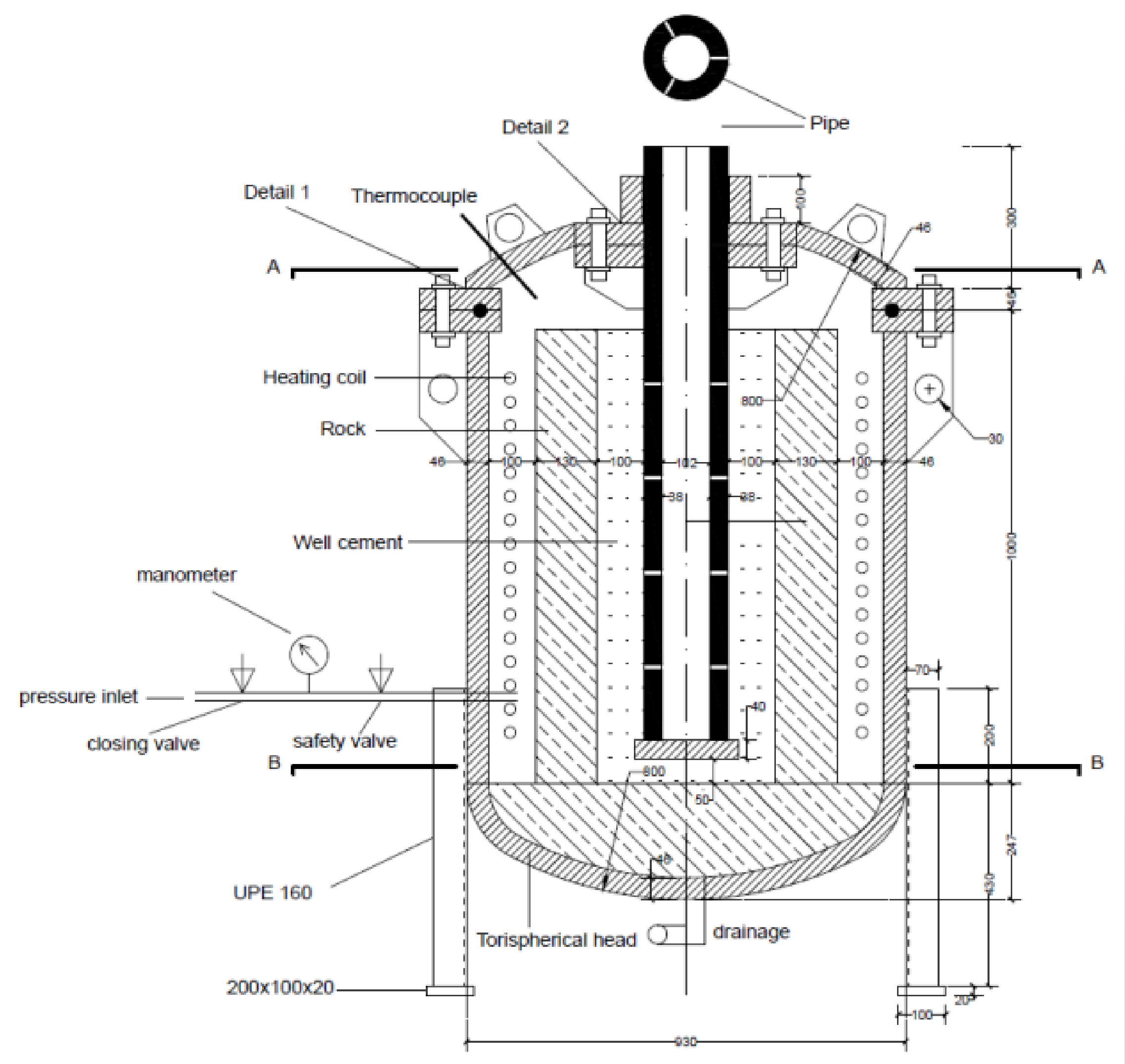


Task 4: Wellbore test system

Goal: develop a small-scale prototype wellbore testing system; and use it to simulate the real environment in the field.

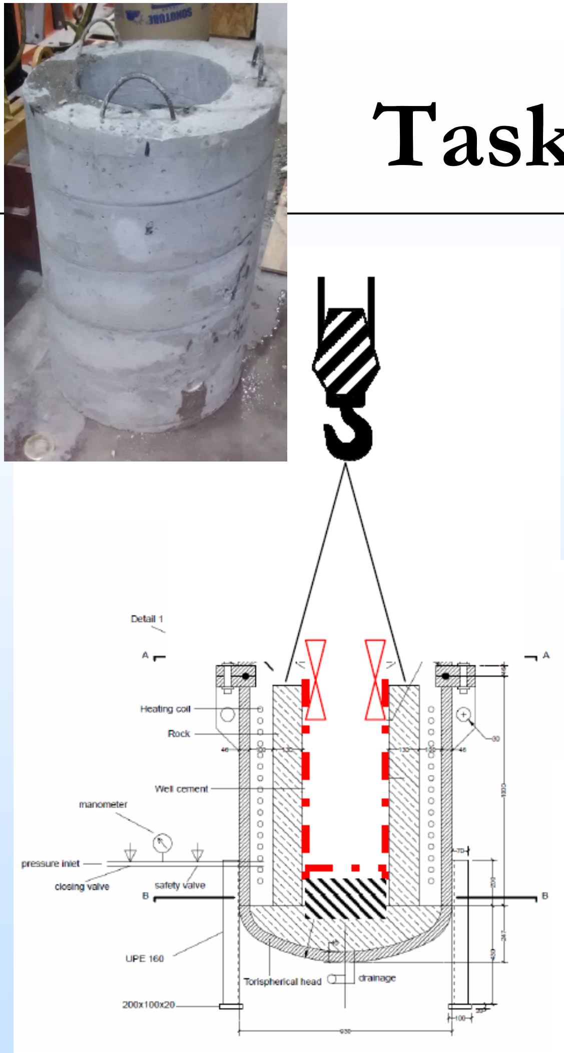
- 9 foot tall chamber
- 30 MPa internal pressure
- Heating up to 150°C
- Standard well pipe

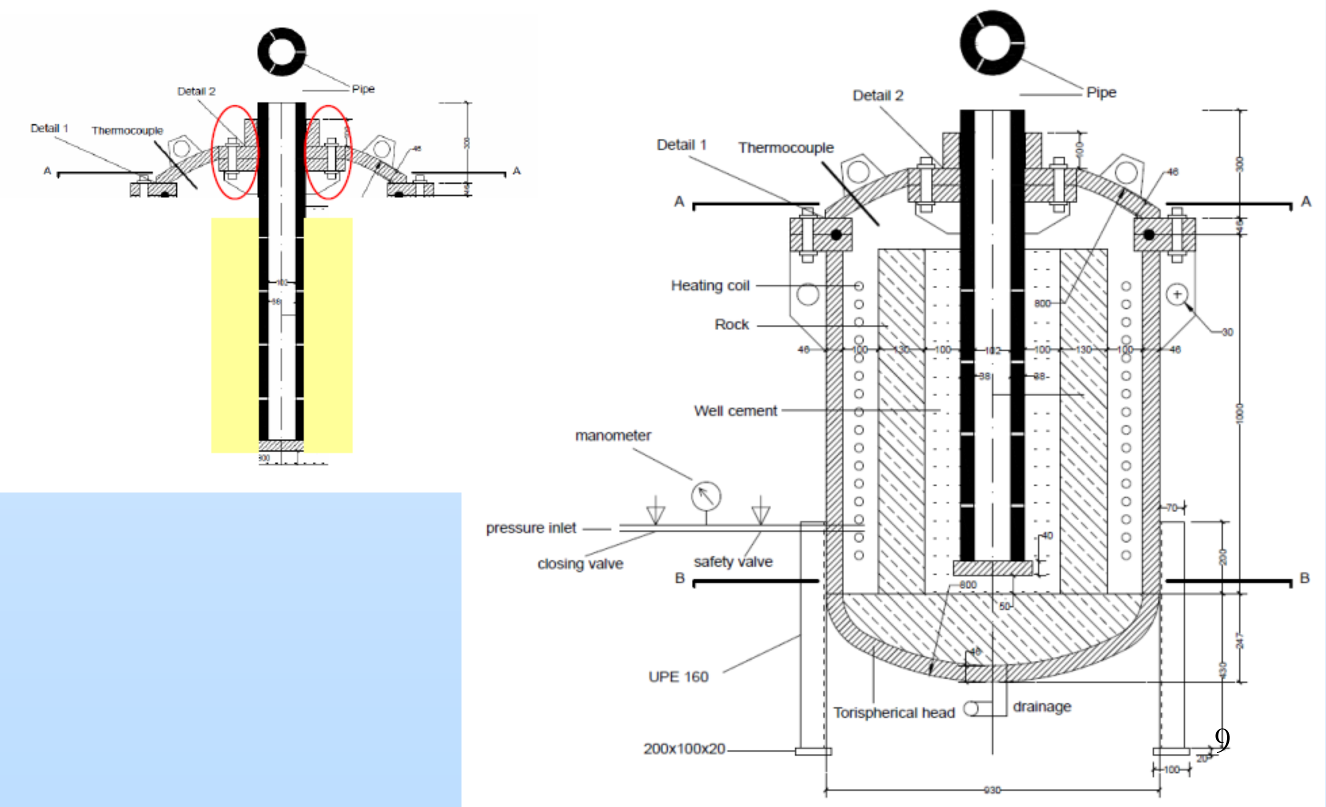
Design is complete, currently under production.



Task 4: Wellbore test system

1. Place an aged concrete cylinder inside the vessel using a crane
2. Install wall and bottom (plastic) separator.
3. Insert steel casing and lid assembly
4. Stress bolts
5. Connect mud-jacking pump and fill annulus with cement through the tube
6. Fill with brine and pressurize
7. Conduct particle injection test





Task 5: Evaluate effectiveness

- SEM-EDX measurements (morphology, elemental composition)
- light microscopy (morphology, cracking, large pores)
- BET (usually N_2 or H_2O adsorption, 1-1000nm, surface area, distribution)
- overall porosity change

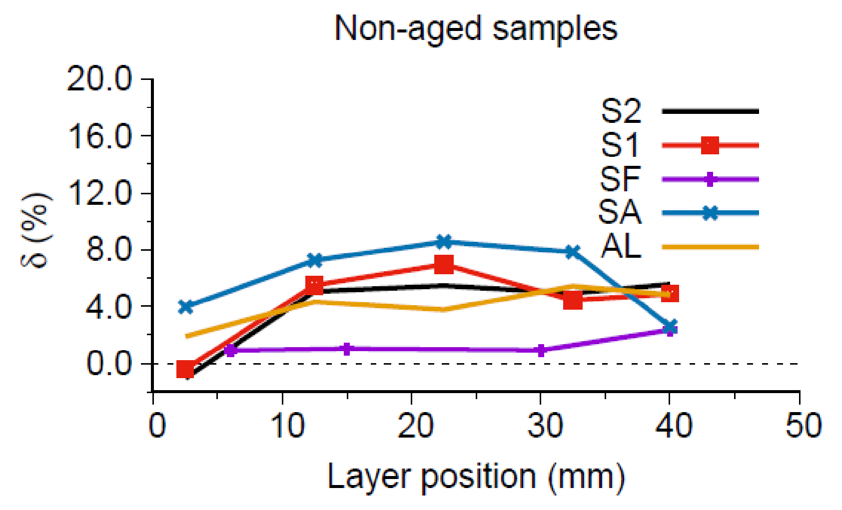
Total porosity

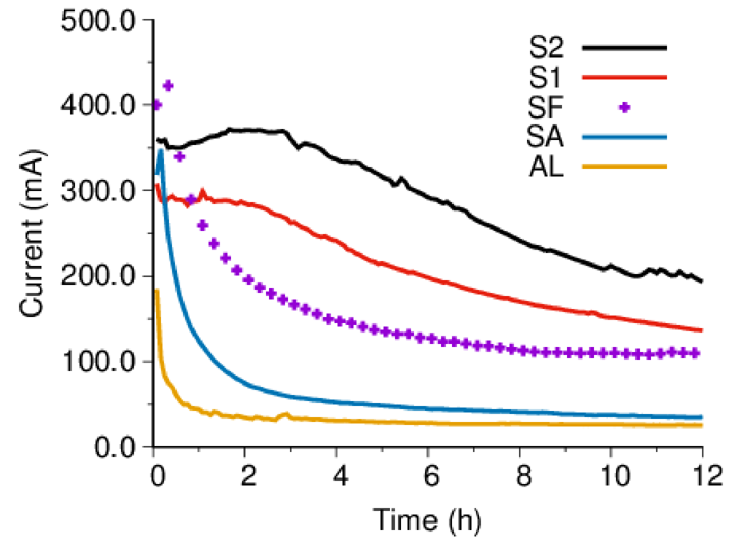
$$\Phi = \frac{w_{SSD} - w_D}{\rho V_s},$$

Rel. porosity change

$$\delta = \frac{\Phi_{untr} - \Phi_{tr}}{\Phi_{untr}},$$

Task 5: Evaluate effectiveness



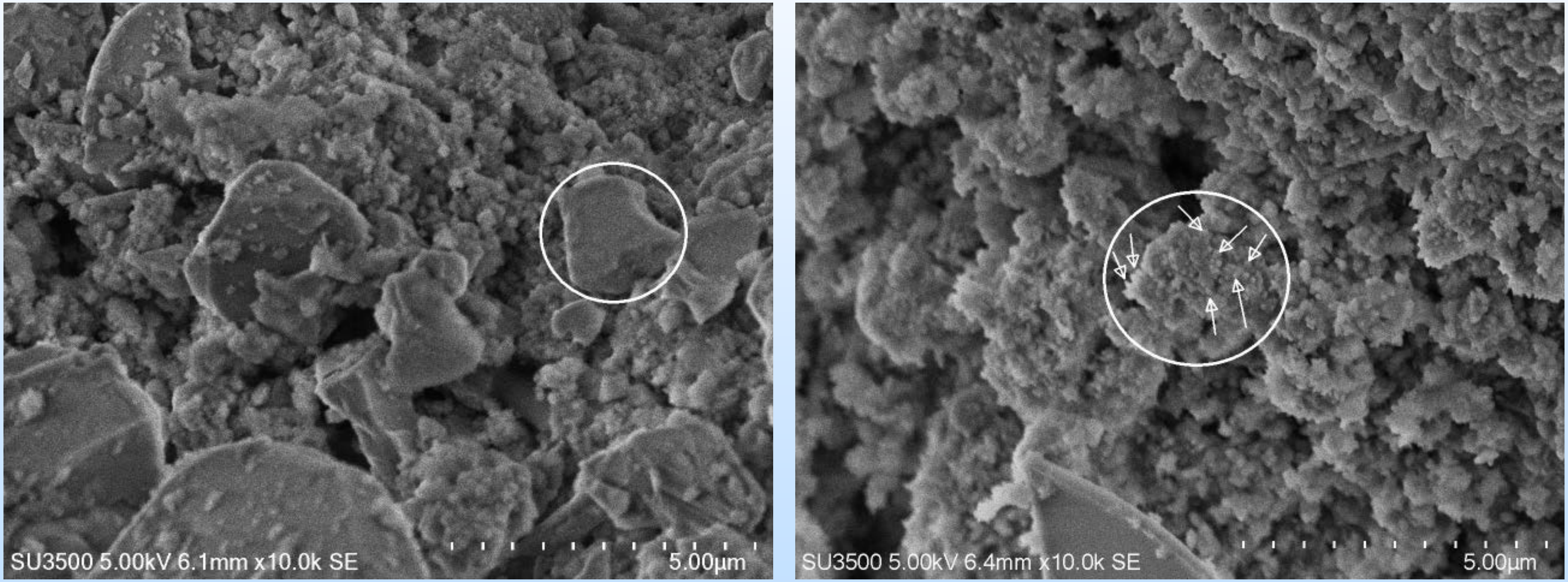


Indications of effectiveness:

- Consistent lower porosity near particle source
- Reduced porosity across the entire cylinder for particles with faster injection rates.

Task 5: Evaluate effectiveness

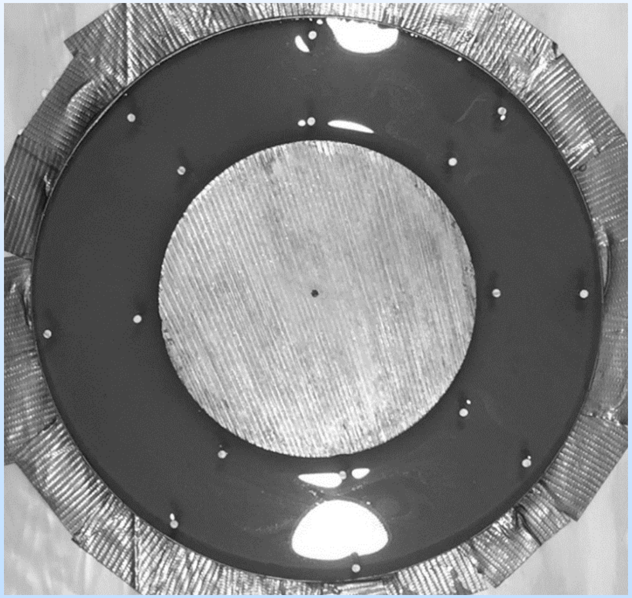
Deposition on cracked surface

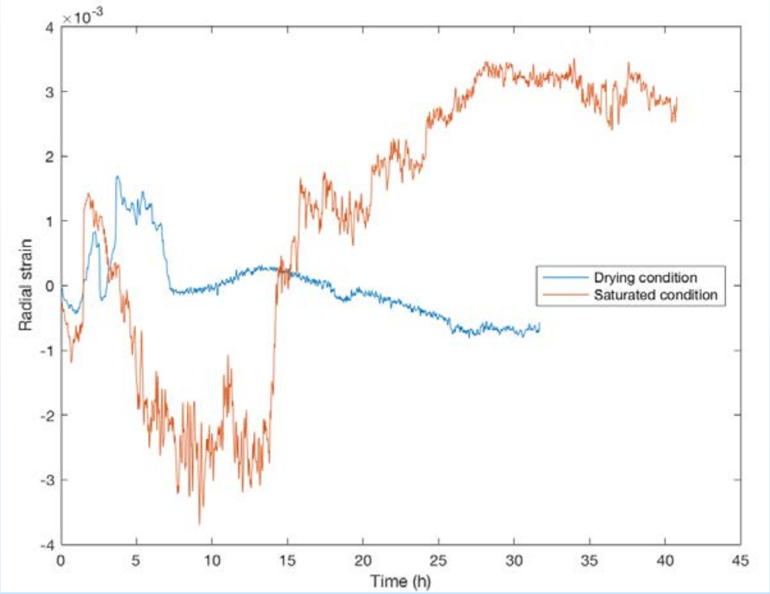


Task 5: Evaluate effectiveness

The repair effectiveness depends on the leakage paths.
Study crack development and structure to understand how particles will enter the leakage network.

Digital image correlation study of well cement ring test.



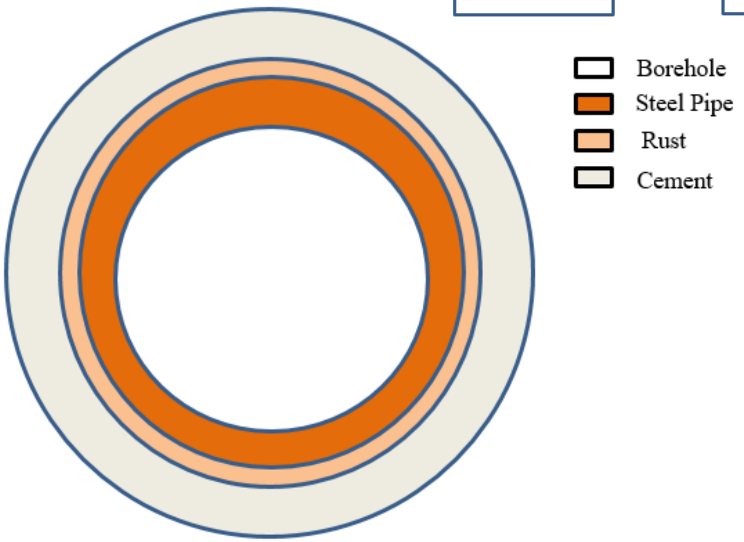


Vertical delamination crack is sensitive to stiffness of steel sheath:
lower modulus → cracking at 2 weeks
higher modulus → cracking at 1.5 days

Task 6: Numerical modeling

Rust Penetration

Intact → Rust → Stimulate → Crack → Failure



Pressure Change after the Crack Appeared (Timoshenko and Fictitious Crack Model)

Interface Pressure

$$= f_t \frac{(R_o^2 - R_{cr}^2)}{(R_o^2 + R_{cr}^2)} \frac{R_{cr}}{R_i} + \frac{1}{R_i} \int_{R_i}^{R_{cr}} f_t \left(\frac{2EG_c - \pi R_{cr} f_t^2}{2EG_c - \pi r f_t^2} \right) dr$$

Calculate service life

$$T_{service} = T_{rust} + T_{stimulate} + T_{crack}$$

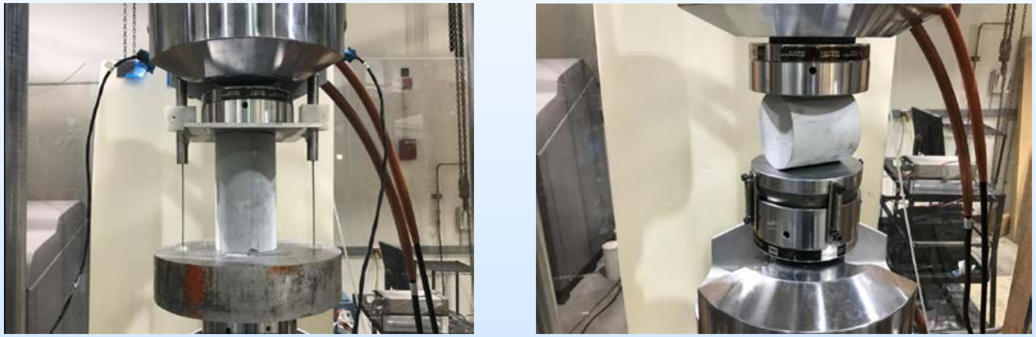
Comparison of Simulated Results with Experimental Study (Liu and Weyers 1988)

Case	R_i (mm)	R_o (mm)	E (Mpa)	f_t (Mpa)	i_{corr} ($\mu A/cm^2$)	Δt_{rust} (year)	
1	8	34.88	27000	3.3	3.75	0.72	0.67
2	8	56	27000	3.3	2.41	1.84	1.84
3	8	78.3	27000	3.3	1.79	3.54	3.54

Exp. result

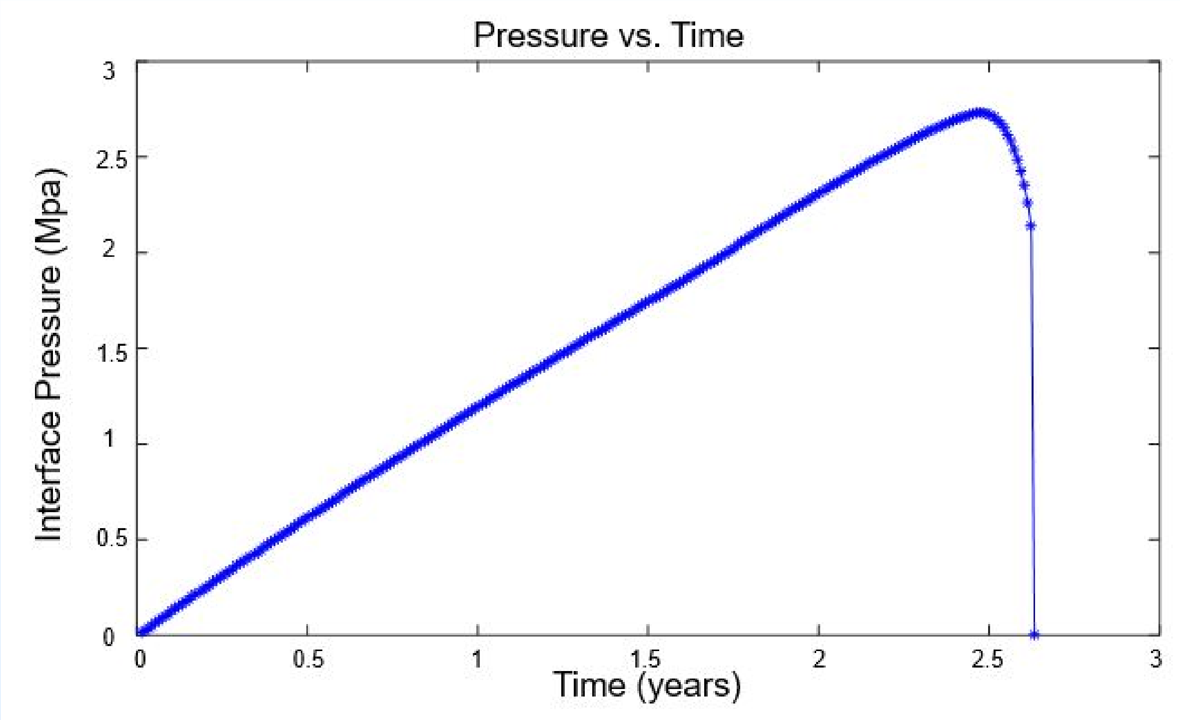
Task 6: Numerical modeling

Input Material Parameters of Well Cement



	Fracture Energy (N/mm)	Elastic Modulus (Mpa)	Tensile Strength (Mpa)	Porosity (%)
Well Cement	0.0863	8968.5	2.38	40
Concrete	0.02	27000	3.3	40

Task 6: Numerical modeling



Condition	Time (years)
Crack Initiation	1.63
Peak Pressure	2.48
Completely Crack	2.68

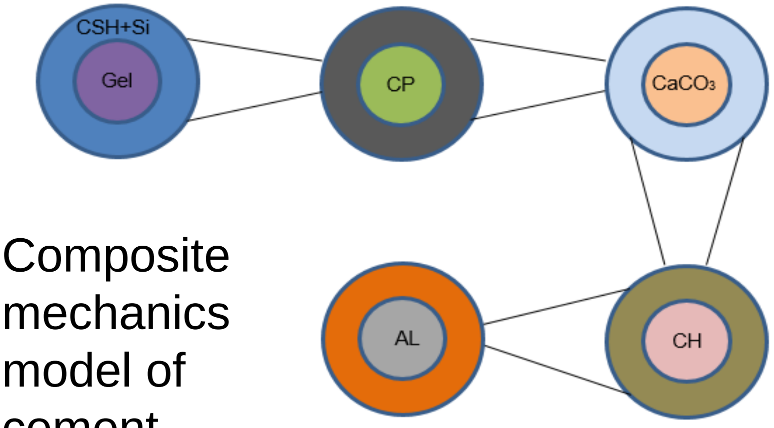
Task 6: Numerical modeling

Well Cement Carbonation

Leaking CO_2 interacts with cement

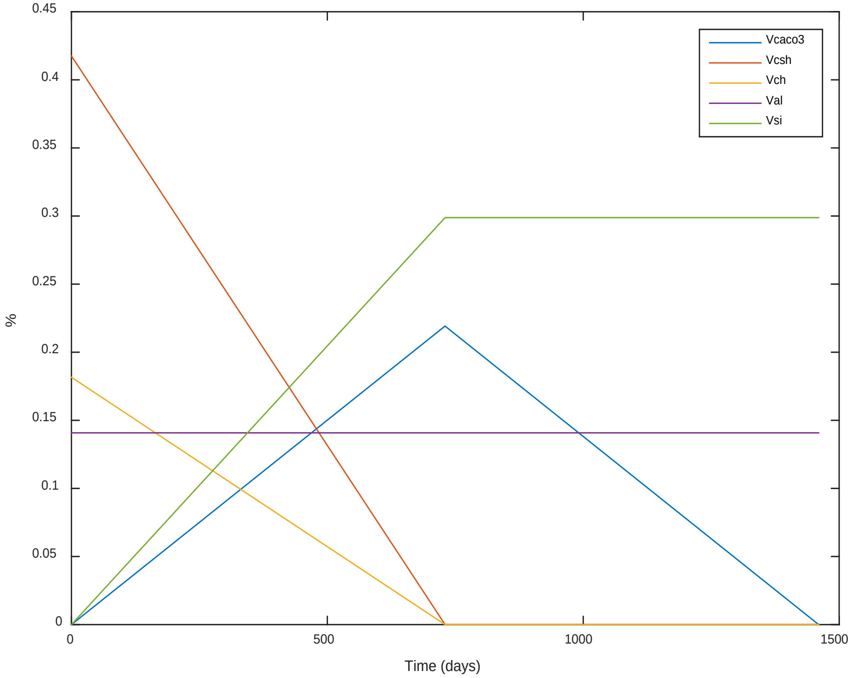
$$E_{eff} = E_{matrix}\left(1+\frac{f_{inclusion}}{\left(\frac{1-f_{inclusion}}{3}\right)+\left(\frac{1}{\frac{E_{inclusion}}{E_{matrix}}-1}\right)}\right)$$

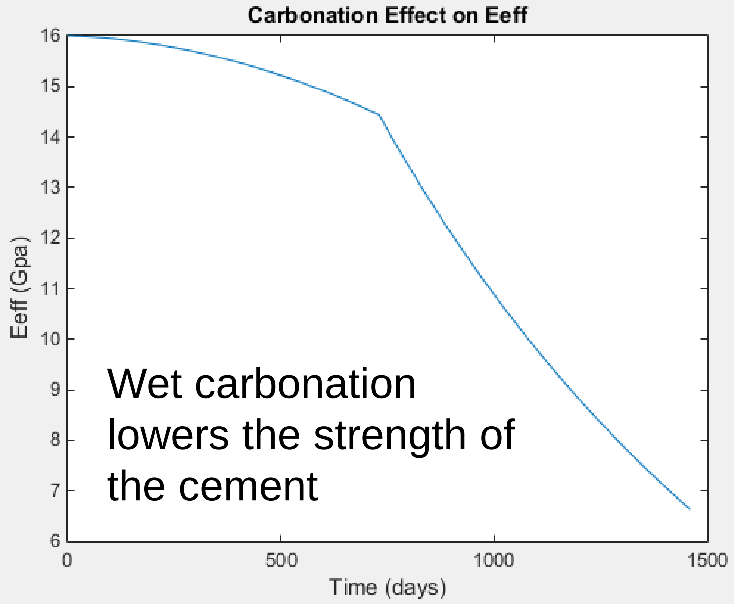
Effective modulus



Composite mechanics model of cement

Cement Composition Change due to Carbonation





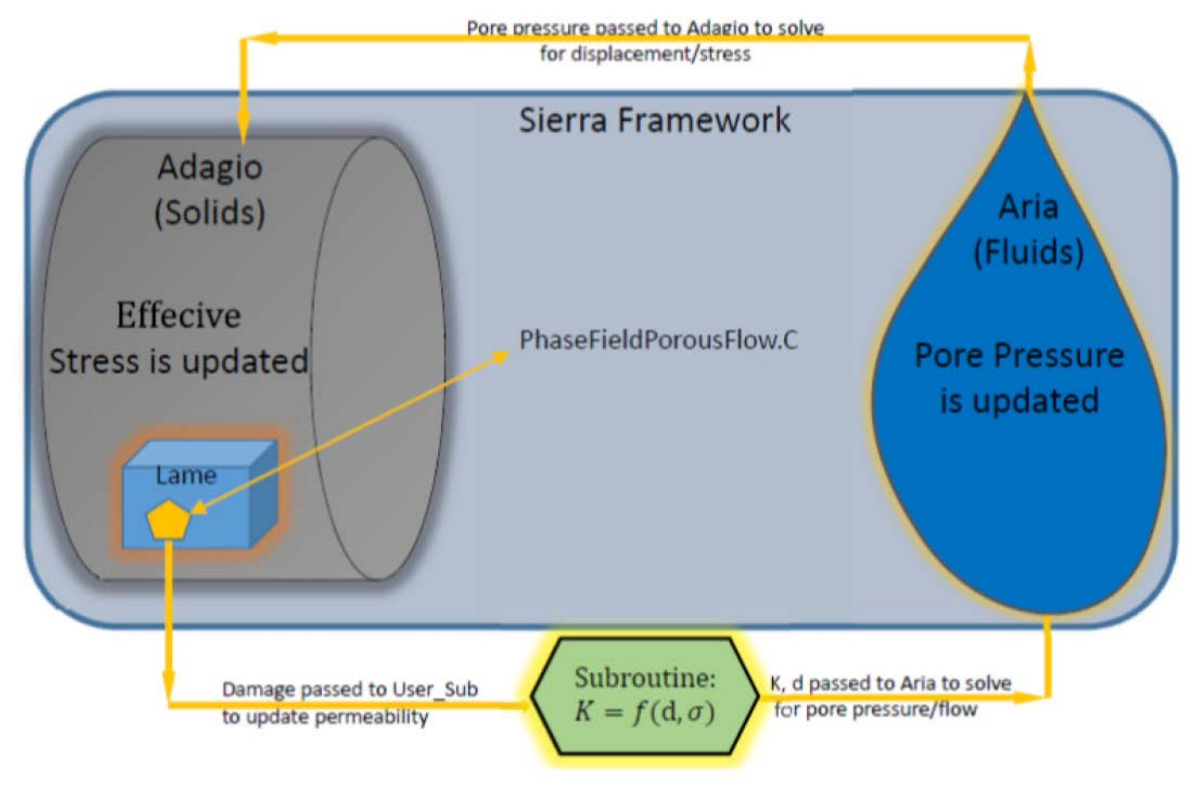
Wet carbonation lowers the strength of the cement

Task 6: Numerical modeling

Goal: Model fracture initiation on a field scale

Computationally couple the multi-phase flow of fluids through poro-elastic media and fracture propagation

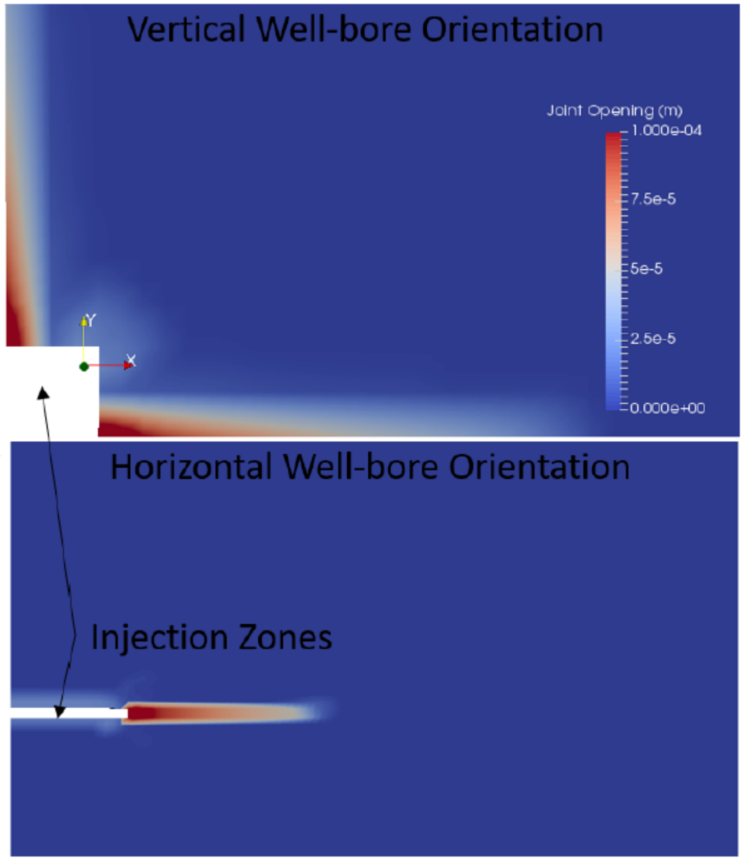
Loose Two-way Coupling of Multi-Physics using Arpeggio

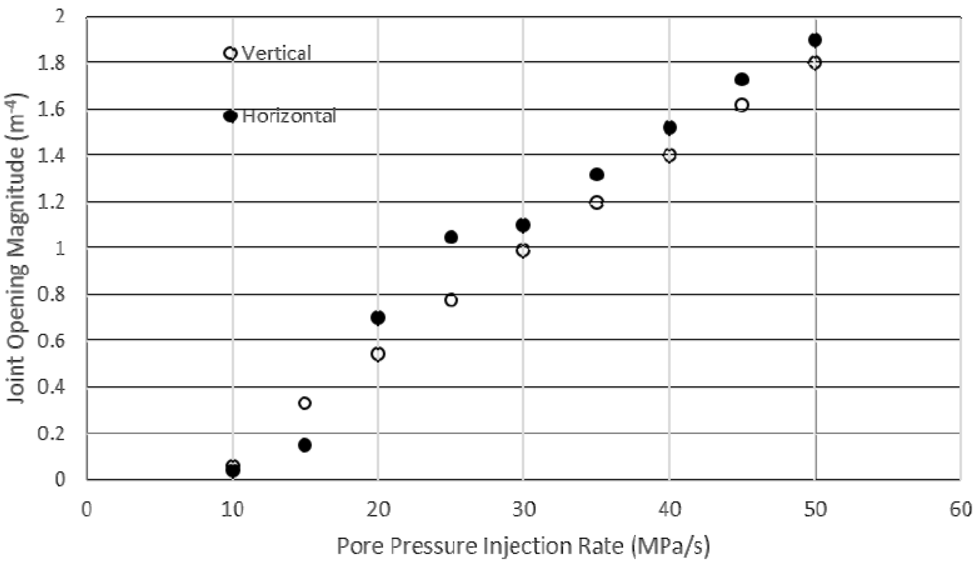


Introduce phase field modeling into Sandia framework.

Nonlinear material model for rock

Task 6: Numerical modeling





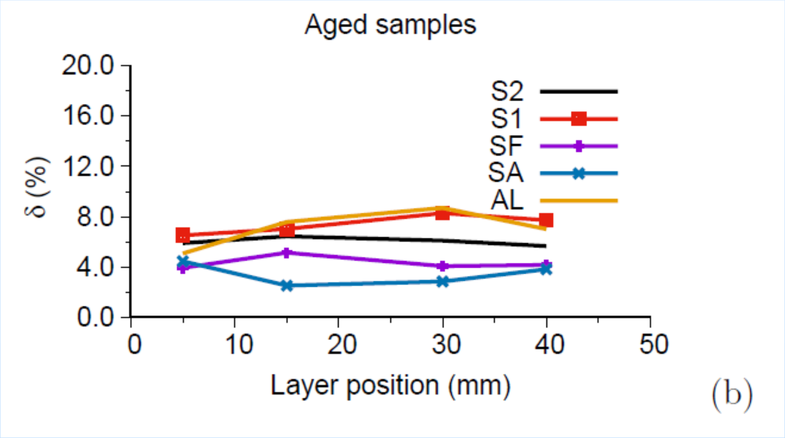
Next step: implement 2 phase flow framework (brine and CO_2), study introduction of nanoparticles into the fluid.

Accomplishments to Date

- Selected best healing agent.
- Completed bench-scale electro-migration testing.
- Conducted the design of a small scale wellbore test system.
- Studied different testing methods to measure porosity distribution in well cement.
- Experimental comparison has been achieved for numerical models of species transport, carbonation, and rust penetration.
- Introduced phase field modeling capabilities into Sandia’s field scale fracture model.

Lessons Learned

- Particle agglomeration can impede their movement through the leakage pathways
- Leakage pathways are very sensitive to the stiffness of the steel pipe, i.e.. the deterioration state of the wellbore is an important parameter for this repair technology
- Need to consider bigger particles for aged samples



Synergy Opportunities

The injection method may be used for sealing (healing) agents other than nano- and micro-particles.

In the current session

P2 – Using mineral precipitation method.

P3 – Using microbially-induced calcite precipitation.

P7 – Using nanocomposite materials for wellbore seal repair

Other sessions

Applications of nanoparticles for *hydraulic fracturing*

The evaluation methods and the simulation models may also be used for the technology.

Project Summary

- Key Findings
  - The technology performs as expected on the bench scale platform.
  - The best healing agents have been identified as 22 nm nanosilica when using this repair technology.
  - We have the tools to model rust penetration, carbonation, stiffness development of the cement, and fracture initiation due to fluid pressure.
- Next Steps
  - Construct and test technology in the large scale lab chamber.
  - Correlate repair technology control parameters to different types of leakage networks.
  - Model the complete repair process.

Appendix

These slides will not be discussed during the presentation, but are mandatory.

Benefit to the Program

The overall goal of the project is to develop a new technology that can repair cement casing leakage of the wellbore and reduce the risk of steel corrosion. The leakage problem will be solved by injecting nanoparticles electrochemically so the cement materials will be densified, and the corrosion risk will be reduced by removing some of the harmful ions in the system.

Program goals addressed:

- Develop and validate technologies to ensure 99 percent storage permanence;
- Develop technologies to improve reservoir storage efficiency while ensuring containment effectiveness.

Benefit to the Program

Project benefits:

- Development of advanced materials and methods that have the ability to prevent or remediate detected leaks in complicated environments under a variety of pressure, temperature, and chemical conditions to ensure CO2 permanence within the storage formation;
- Theoretical and numerical models to demonstrate potential long-term (i.e., at least 50 years) feasibility and effectiveness of the new technology.

Project Overview

Goals and Objectives

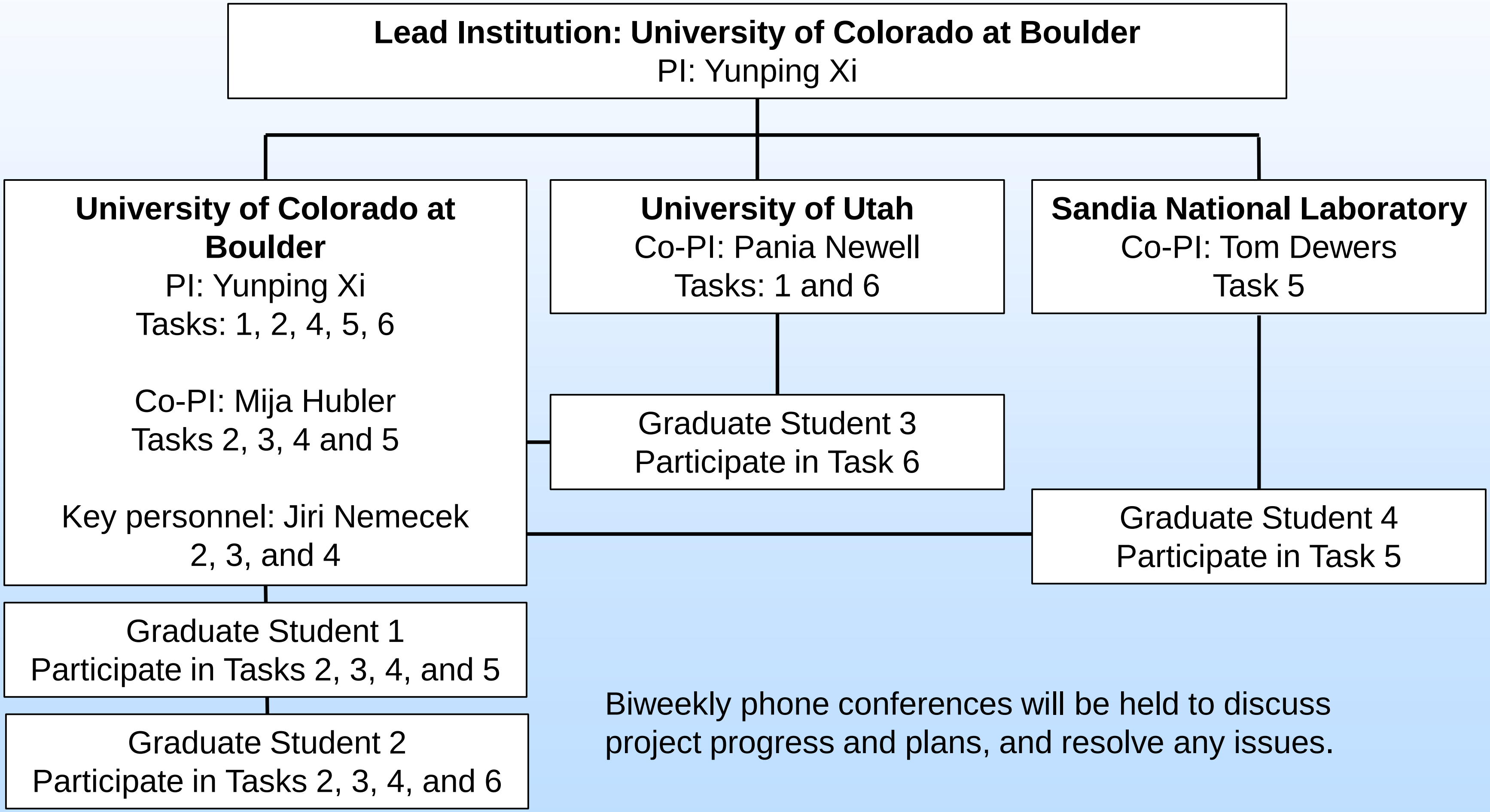
Objective 1: Development of the injection technology for leakage repair

Success criteria: we will seal artificially damaged samples and evaluate their mechanical properties and ultrasonic properties to reveal improvement.

Objective 2: Development of a new numerical simulation model that can simulate and predict the performance of the new wellbore repair technology

Success criteria: we will compare numerical results with experiments for validation.

Organization Chart



Lead Institution: University of Colorado at Boulder
PI: Yunping Xi

University of Colorado at Boulder
PI: Yunping Xi
Tasks: 1, 2, 4, 5, 6

Co-PI: Mija Hubler
Tasks 2, 3, 4 and 5

Key personnel: Jiri Nemecek
2, 3, and 4

University of Utah
Co-PI: Pania Newell
Tasks: 1 and 6

Sandia National Laboratory
Co-PI: Tom Dewers
Task 5

Graduate Student 3
Participate in Task 6

Graduate Student 4
Participate in Task 5

Graduate Student 1
Participate in Tasks 2, 3, 4, and 5

Graduate Student 2
Participate in Tasks 2, 3, 4, and 6

Biweekly phone conferences will be held to discuss project progress and plans, and resolve any issues.

Gantt Chart

		Year 1: Budget Period 1				Year 2: Budget Period 2				Year 3: Budget Period 3			
Task #	Task	1st Qtr	2nd Qtr	3rd Qtr	4th Qtr	1st Qtr	2nd Qtr	3rd Qtr	4th Qtr	1st Qtr	2nd Qtr	3rd Qtr	4th Qtr
1	Project Management, Planning and Reporting												
1.1	Project Management Plan	▨ PMP											
1.2	Project Planning and Reporting	▨	▨	▨ Presentation	▨ Report	▨	▨	▨	**Report**				**Final Report**
2	Development of an electro-migration unit system and testing							**Presentation**					
2.1	Development of an electro-migration unit system	▨	▨	▨	▨	▨	▨						
2.2	Testing with the electro-migration unit system	▨	▨	▨	▨	▨	▨						
3	Selection of Healing agents									**Report**			
3.1	Nanoparticle testing	▨	▨	▨	▨	▨	▨	▨					
3.2	Nanoparticle based slurry testing	▨	▨	▨	▨	▨	▨	▨					
3.3	Selection of healing agents	▨	▨	▨	▨	▨	▨	▨					
4	Small-scale wellbore test system												**Presentation**
	Design and construction of test system	▨	▨	▨	▨	▨	▨	▨					
5	Evaluation of the effectiveness of the technology												**Report**
5.1	Strength, stiffness, and transport properties		▨	▨	▨	▨	▨	▨					
5.2	Microscopic study		▨	▨	▨	▨	▨	▨					
5.3	Fracture testing and analysis		▨	▨	▨	▨	▨	▨					
6	Numerical modeling and verification												
6.1	Numerical modeling for ionic removal		▨	▨	▨	▨	▨	▨					
6.2	Numerical modeling for injection of healing agents				▨	▨	▨	▨					
6.3	Numerical modeling of fracture using Kayenta and Sierra Mechanics		▨	▨	▨	▨	▨	▨					**Model**

Bibliography

Journal paper
Nemecek, J., Li, L., and Xi, Y. (2017) “Electrokinetic Nanoparticle Injection for Remediating1 Leaks in Oil Well Cement”, submitted to *Construction and Building Materials*, reviewed and under revision.

Al Wakeel S., Nemecek J., Linfei L., Xi Y., & Hubler M. (2017) “Effect of particles injection on the fracture toughness of cracked well cement.” International Journal of Greenhouse Gas Control. Under review.

Conference presentation with paper
Culp, D., Newell, P., Tupek, M., Hubler, M. (2017) Two-way Coupling of fracture and fluid flow using a phase-field model. ARMA June 28th San Francisco, CA.

Conference presentation
Xi, Y., Hubler, M., Dewers, T., Newell, P. (2017) Nanoparticle Injection Technology for Remediating Leaks of CO_2 Storage Formation, Nanotech Conference in Washington DC, May 17, 2017.

Hubler, M., Li, L., Mahnaz, S., Xi, Y., Dewers, T., and Newell, P. (2016) Electrochemical nanoparticle injection technology for remediating leaks.” Carbon Capture, Utilization, and Storage Conference, June 14th, Tysons, VA.