Cement Self-Healing: Understanding and Enhancing Performance Under Reactive Conditions

CO_2 Storage Conference 2025

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Introduction and objectives

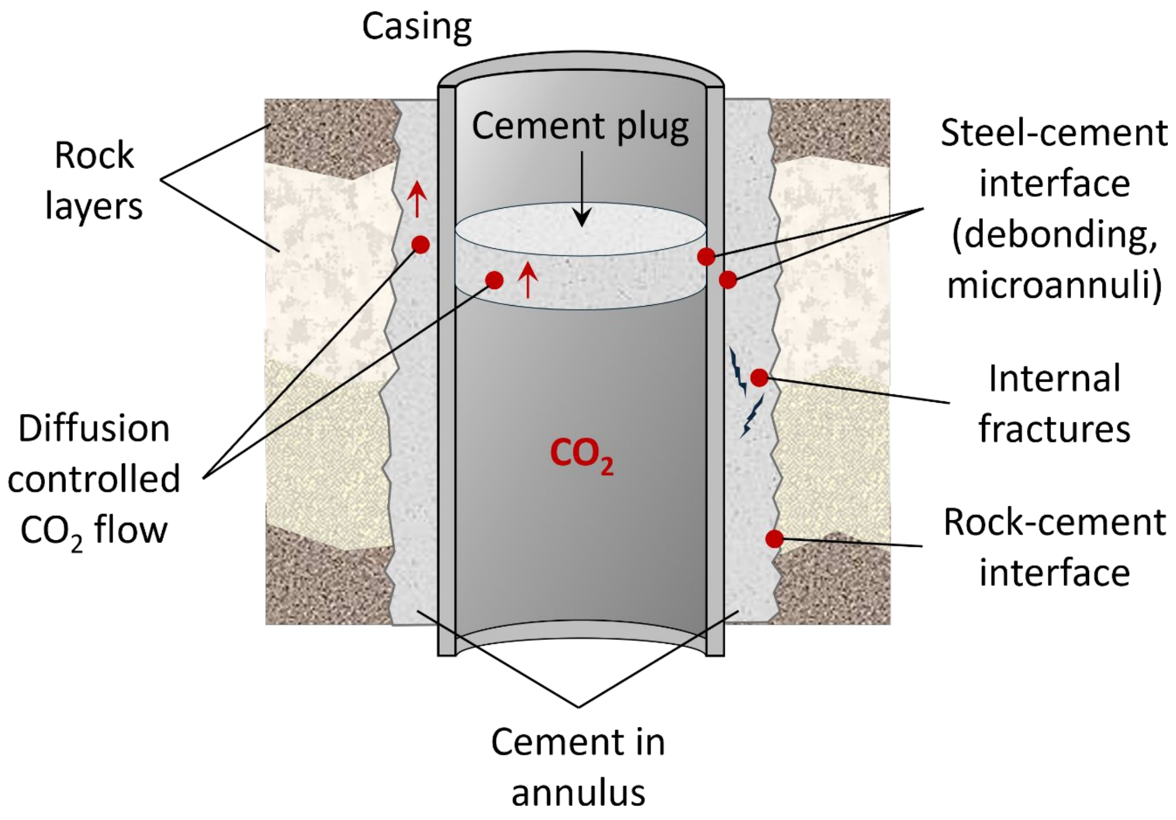
- In CS context cement is a barrier against upward leakage of CO_2-rich fluids
- Compromised well integrity
  - Physically: fractures, debonding and micro-annuli
  - Chemically: degradation (carbonation and bicarbonation)
- Possible beneficial effects – cement self-healing
- Key research priority to obtain efficient sealing against CO_2 leakages - maintain a low-permeability barrier preventing vertical or lateral gas migration

Project objectives:

1. Understanding of self-healing mechanisms of wellore cement under CO_2 exposure conditions: critical parameters influencing self-healing

2. Develop systematic experimental protocols suitable for further practical applications

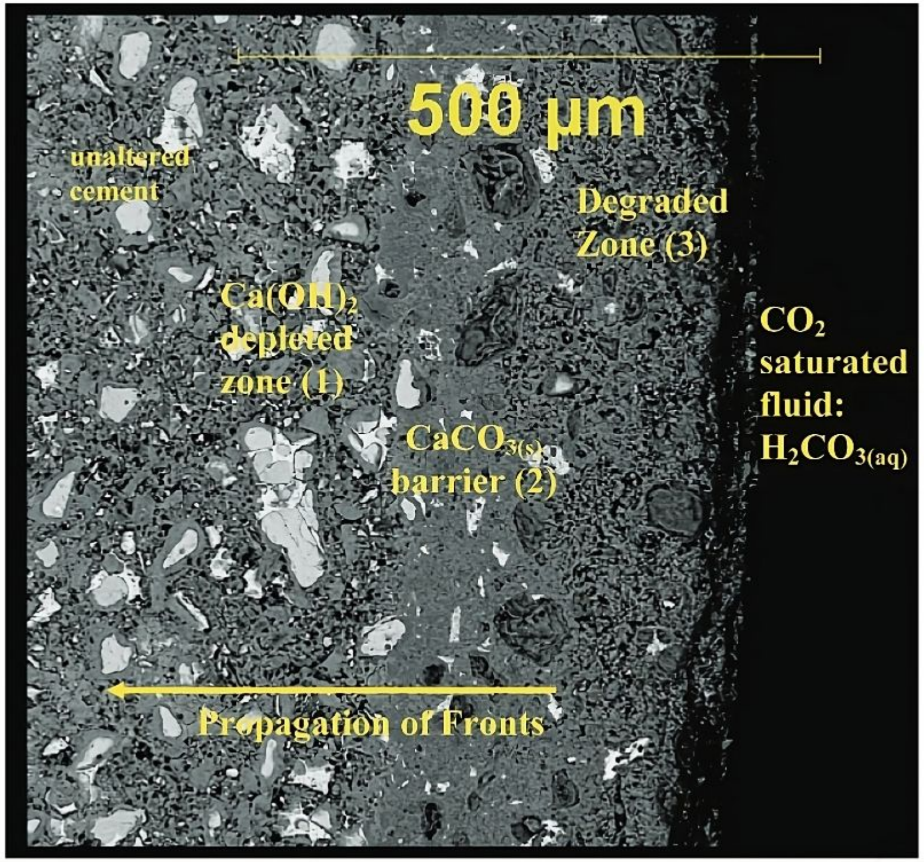
Fractures, debonding and micro-annuli in wellbore



Possible CO_2 propagation in a cased wellbore

Carbonation of intact neat wellbore cement

Formation of reaction zones in cement exposed to CO_2-saturated brine



Kutchko et al., 2008. Rate of CO_2 attack on hydrated class H well cement under geologic sequestration conditions

Zone 3 – Silica-gel (outermost, porous, weak).

After prolonged attack, carbonic acid dissolves previously formed $CaCO_3$ and decalcifies C-S-H, leaving an amorphous SiO_2-rich paste with high porosity and poor integrity.

Zone 2 – Carbonate precipitation (denser “barrier”)

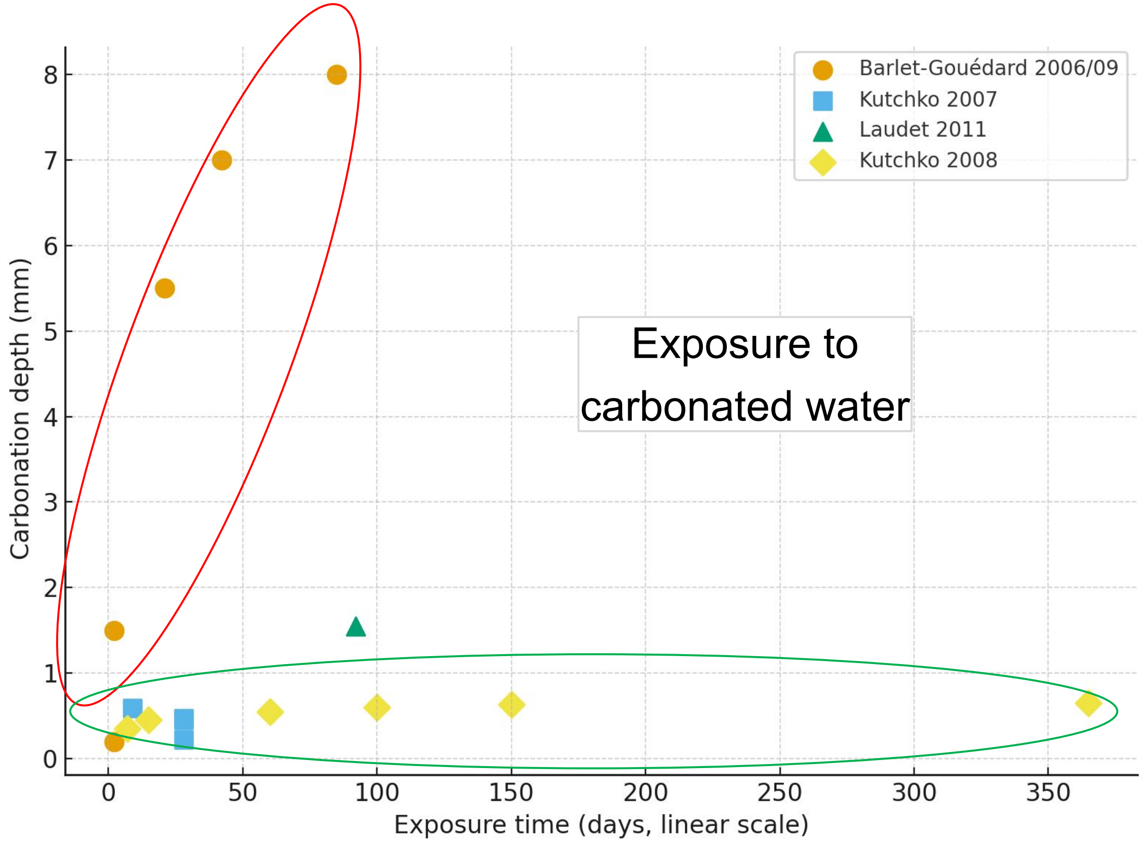
Ca^{2+} released by portlandite dissolution reacts with HCO_3^- to precipitate $CaCO_3$ within pores. This infilling reduces permeability, increases local strength, and acts as a diffusion-slowing barrier.

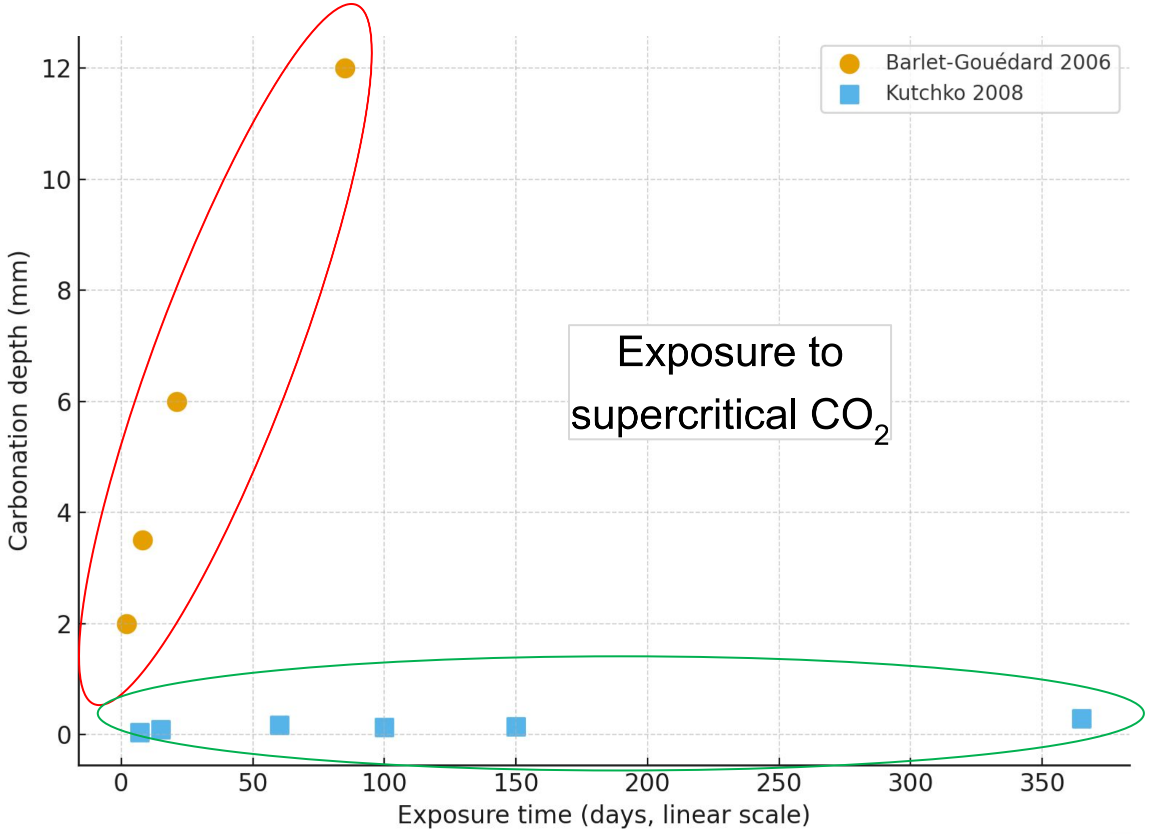
Zone 1 – Portlandite-depleted zone (CH leached)

The first step of CO_2 attack is dissolution of Portlandite ($Ca(OH)_2$) feeding Zone 2 calcite precipitation. Behind Zone 1 lies unaltered cement.

Carbonation of intact neat wellbore cement

Static exposure of neat wellbore cement to CO_2

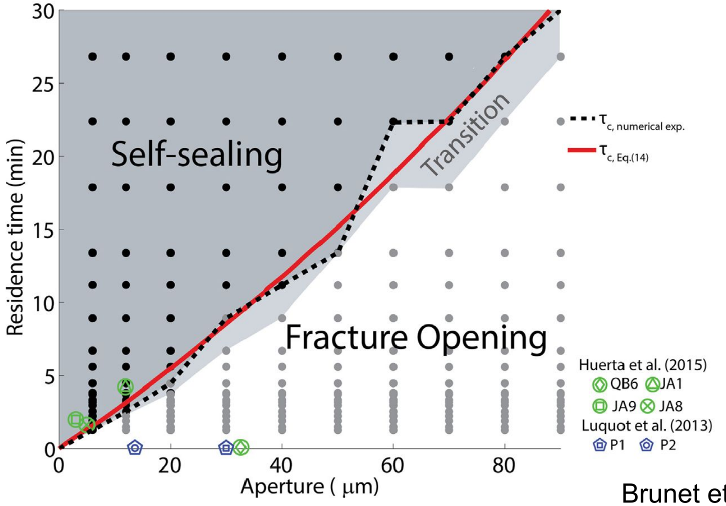




Carbonation of fractured wellbore cement

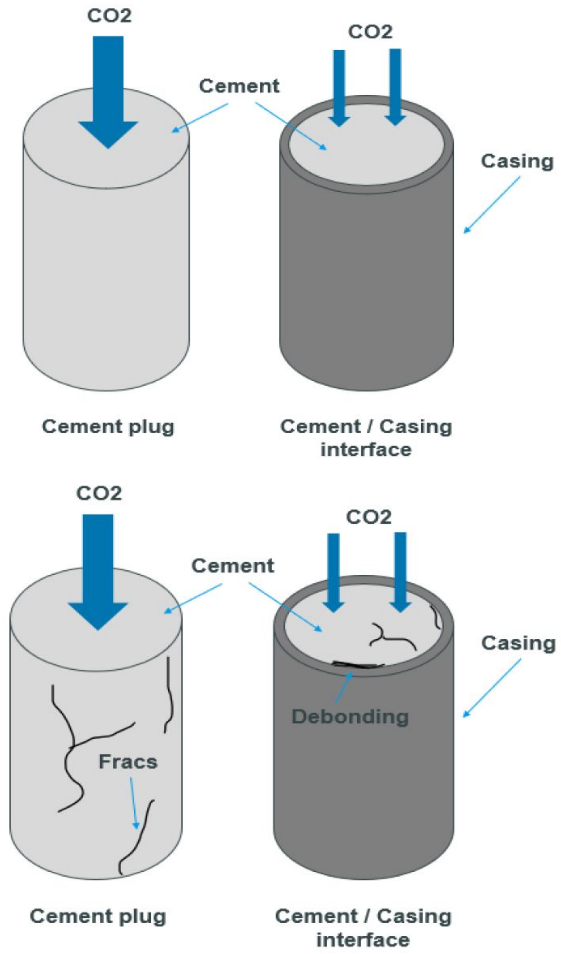
Fracture or debonding exposed to CO_2

- Fresh cement surfaces are exposed and CO_2-rich fluids
- Shift from slow diffusion in the matrix to faster advection along the fracture
- Near the fracture surface, CO_2-rich water dissolves portlandite and leaches Ca^{2+}
- As the fluid moves and mixes → $CaCO_3$ becomes supersaturated and starts to precipitate – "self-healing"
- Precipitation of calcite → denser fracture infill, can partially close the fracture aperture and reduce permeability
- The reactive transport modeling has shown a relation between the aperture size and residence time where the interplay between the sealing and opening can be determined.



Brunet et al., 2016

Intact and fractured/debonded cement samples



Self-healing mechanisms

Reaction Type	Formula	Effect on Healing	Applicable to (Neat cement/ Additives)
Portlandite carbonation	$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$	Positive: Fills pores/cracks, reduces permeability, increases strength	Neat cement; enhanced with Ca-rich additives
Bicarbonation and $CaCO_3$ dissolution	$CaCO_3 + H_2CO_3 \rightarrow Ca(HCO_3)_2$ $CaCO_3(s) + CO_2(aq) + H_2O \rightleftharpoons Ca^{2+} + 2HCO_3^-$.	Negative/Mixed: Leaches Ca^{2+}, increases porosity; may enable further precipitation if buffered	Neat cement; mitigated by pozzolans
Continued hydration of Tricalcium Silicate (C_3S)	$C_3S + H_2O \rightarrow C\text{–}S\text{–}H + Ca(OH)_2$	Positive: Forms additional C-S-H, bridges defects	Neat cement; accelerated with nanomaterials
Continued hydration of Dicalcium Silicate (C_2S)	$C_2S + H_2O \rightarrow C\text{–}S\text{–}H + Ca(OH)_2$	Positive: Similar to C_3S, but slower; contributes to long-term healing	Neat cement
C-S-H decalcification (Silica gel formation)	$C\text{–}S\text{–}H + H^+ \rightarrow$ Si-rich gel $+ Ca^{2+}$	Mixed: Forms protective silica layer; excessive can lead to porosity increase	Neat cement; stabilized by silica-based additives
Pozzolanic reaction	$Ca(OH)_2 + SiO_2 + H_2O \rightarrow CaO \cdot SiO_2 \cdot H_2O$ (C-S-H)	Mixed: Consumes $Ca(OH)_2$, forms denser C-S-H, resists leaching	Pozzolanic additives
Geopolymer Reaction (Aluminosilicate Gel)	$Na_2SiO_3 + Al_2O_3 + H_2O \rightarrow$ Na-aluminosilicate gel	Positive: Forms robust gel matrix, reduces carbonation depth	Geopolymer additives
Nano-Enhanced C-S-H Formation	SiO_2 (nano) $+ Ca(OH)_2 \rightarrow$ Nano-C-S-H	Positive: Accelerates densification, fills nano-pores	With nanomaterials
Bio/enzymatic $CaCO_3$ (MICP/EICP)	$CO(NH_2)_2 + 2H_2O \rightarrow 2NH_4^+ + CO_3^{2-}$; $Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3(s)$	Positive: Fast crack filling, densification of leakage pathways	With bacteria or enzymes

Parameters affecting self-healing

Self-healing potential

Cement composition and properties

- Intrinsic cement chemistry
- Degree of cement hydration
- Physical properties of dry cement

Water availability for carbonation

- Exposure to carbonated water
- Exposure to $scCO_2$
- Exposure to humid CO_2

Cement sample characteristics

- Sample length, reactive surface area
- Heterogeneities
- Fracture/debonding sizes
- Fracture geometry

Curing conditions

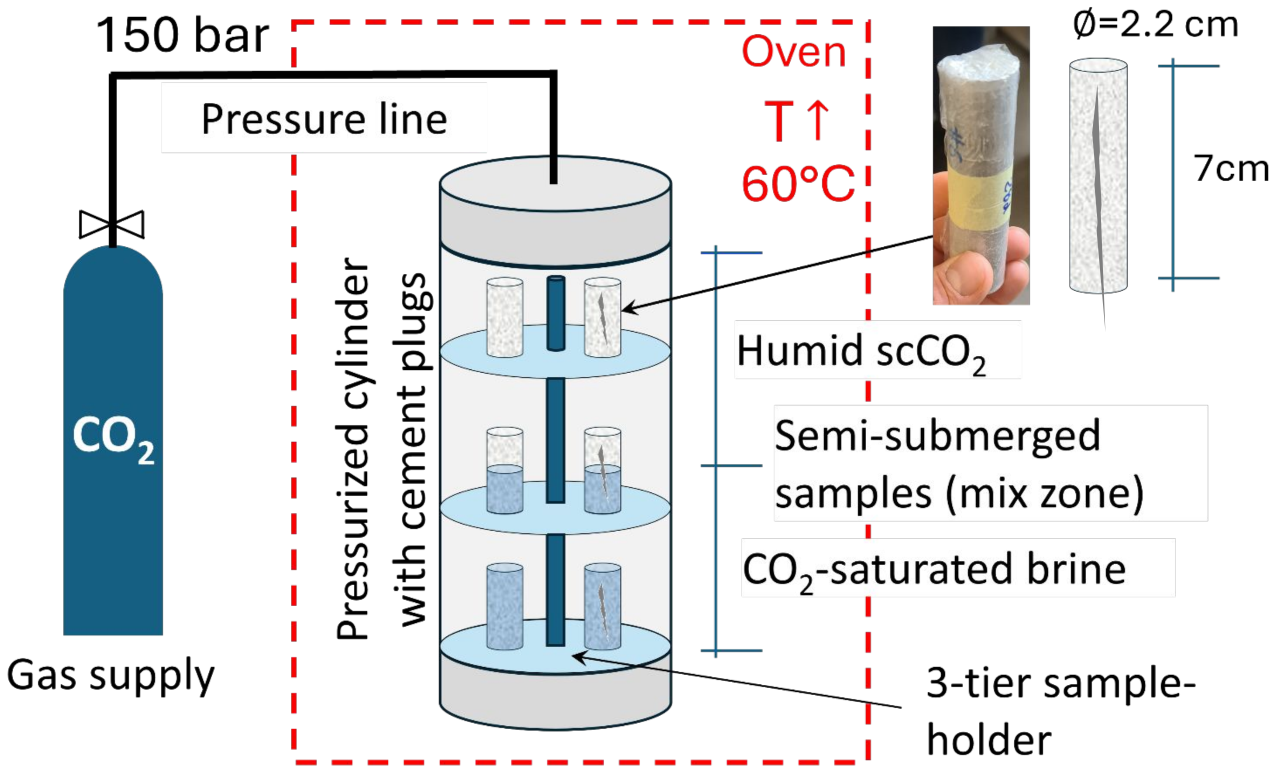
- Temperature and pressure
- Humidity / water availability
- Curing duration

Exposure conditions

- Static vs. flow conditions
- Temperature
- Applied differential pressure (ΔP)
- Residence time (flow rate)
- Duration of exposure, flooding

Static vs dynamic CO_2 exposure

Experimental protocol for soaking static CO_2 exposure tests



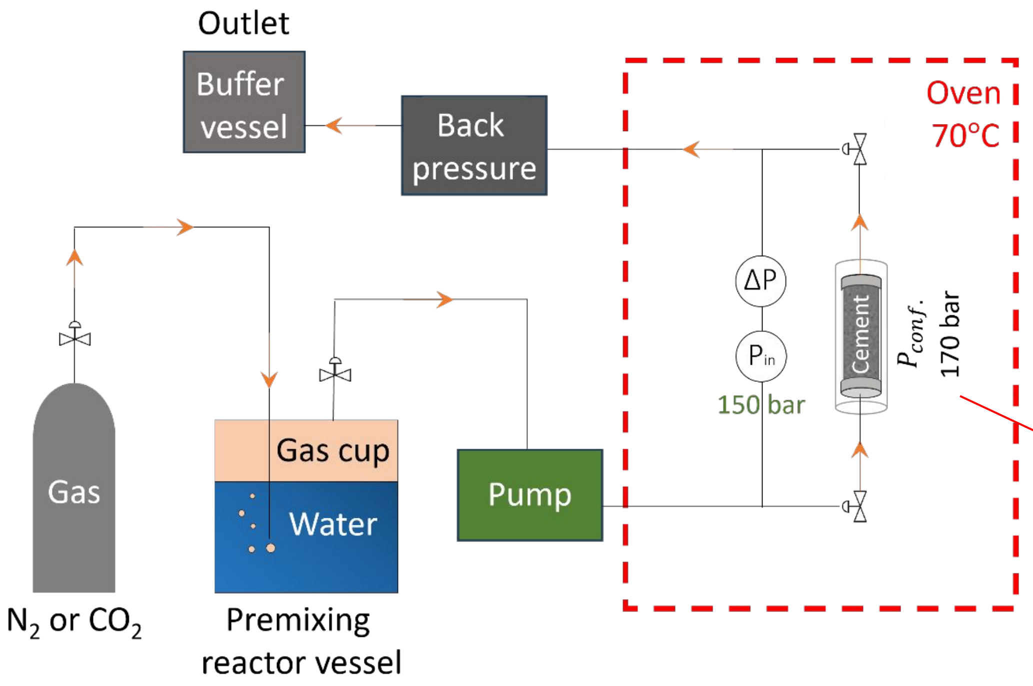
Fractured neat class G cement sample

Preparation of experimental setup

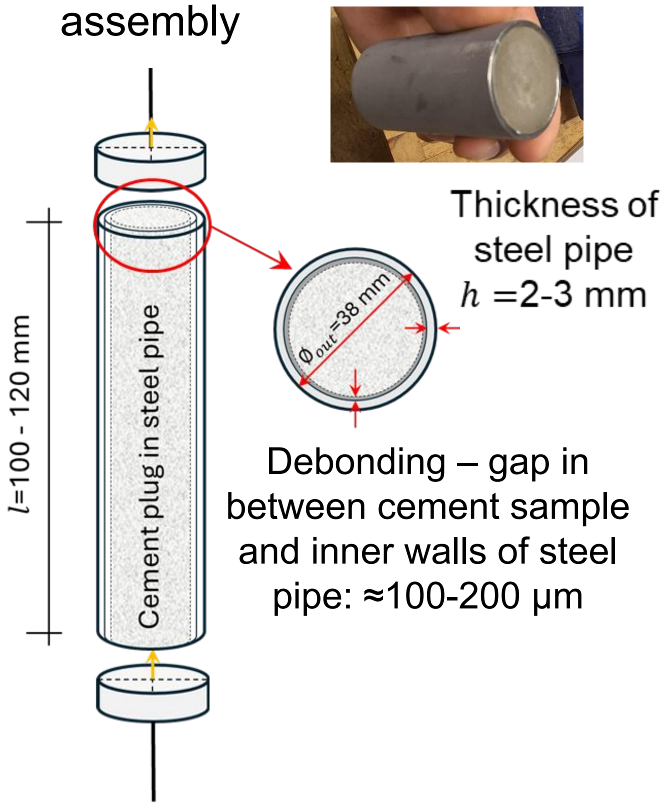
Static vs dynamic CO_2 exposure

Experimental protocol for dynamic CO_2 exposure tests

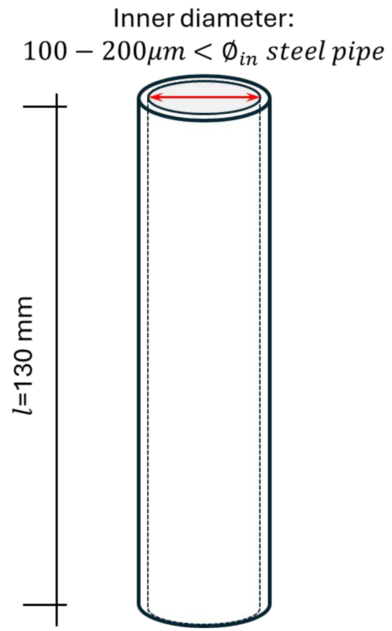
Prototype of experimental setup



Steel-cement assembly

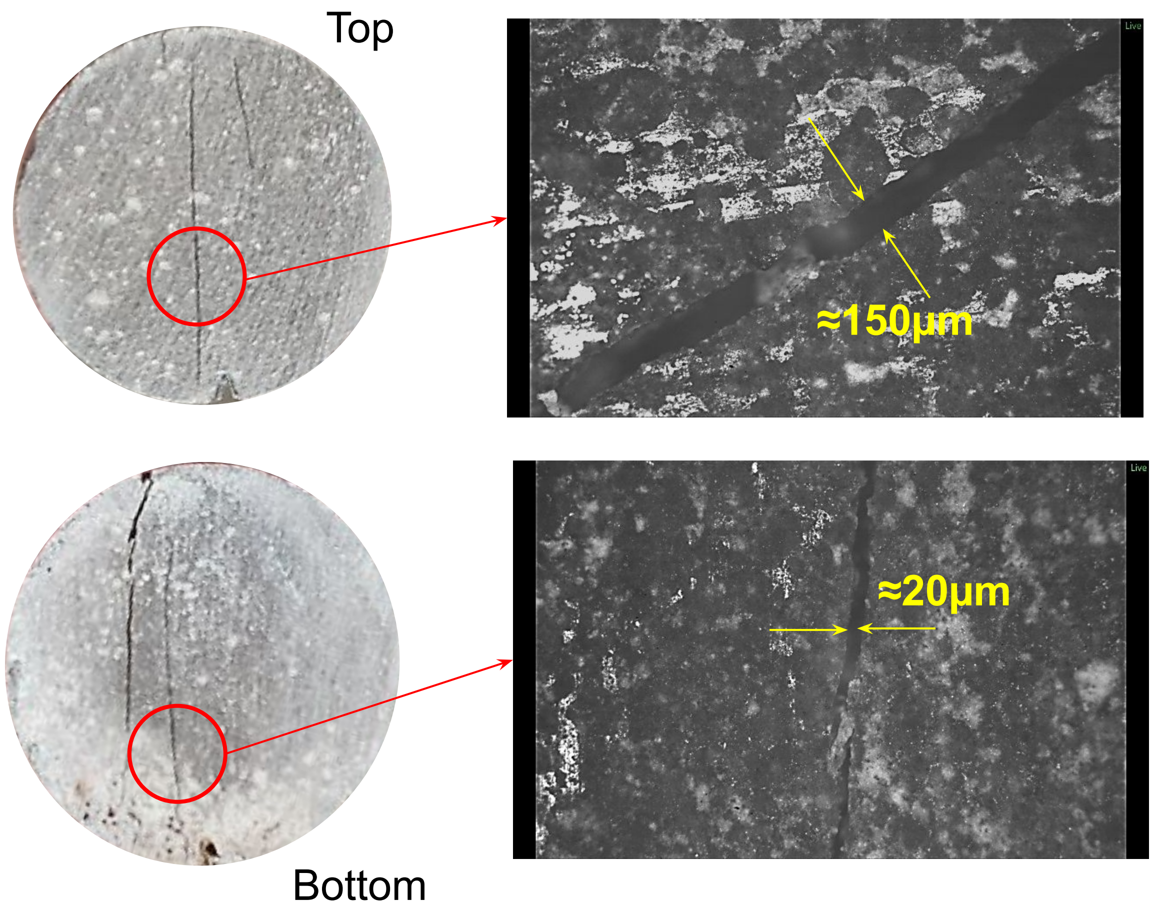


Molds for casting cement

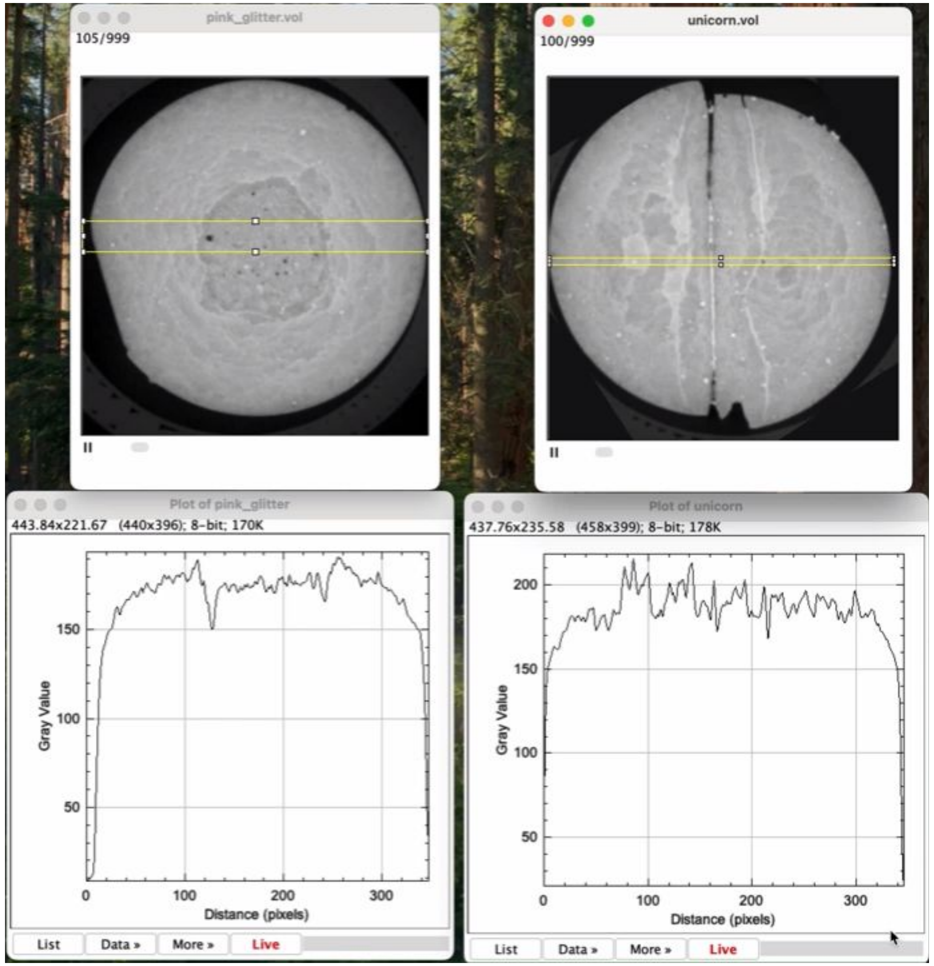


Preliminary results

Fractured G cement sample before exposure



CT scanning of fractured plug after 1 month CW exposure



Final remarks

- Cement self-healing via carbonation is promising, especially for microcracks (<20 μm), but healing of larger fractures remains uncertain.
- Engineered additives (e.g., pozzolans, nanomaterials, microbes) enhance healing but face scalability and cost challenges.
- Standardized testing, long-term durability data, and field validation are needed to ensure reliable sealing in CCS operations.
- Reactive transport modeling can be used to gain insights on the dynamic geochemical interactions in cement media through a long-term process.

Thank you for listening – Any questions?

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- **Acknowledgment**
  This work is part of a collaborative project on "Self-healing cement for long-term CO_2 applications", supported by **TotalEnergies**.