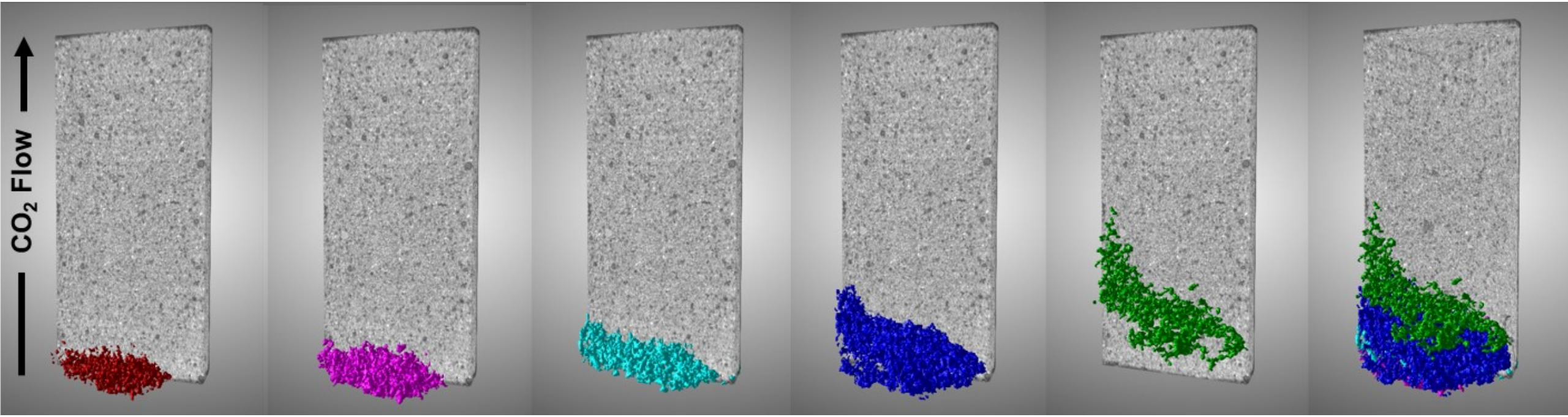
Understanding CO_2 Transport and Carbonate Formation in Portland Cement-Based Materials Using X-Ray Micro-CT

NATIONAL ENERGY TECHNOLOGY LABORATORY

Dr. Laura E. Dalton

Research Scientist
Research & Innovation Center



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Disclaimer

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Authors and Contact Information

Laura Dalton[1,2], Karl Jarvis[1,2], Dustin Crandall[1], Moe Pour-Ghaz[3]

[1]National Energy Technology Laboratory, 3610 Collins Ferry Road, Morgantown, WV 26507, USA

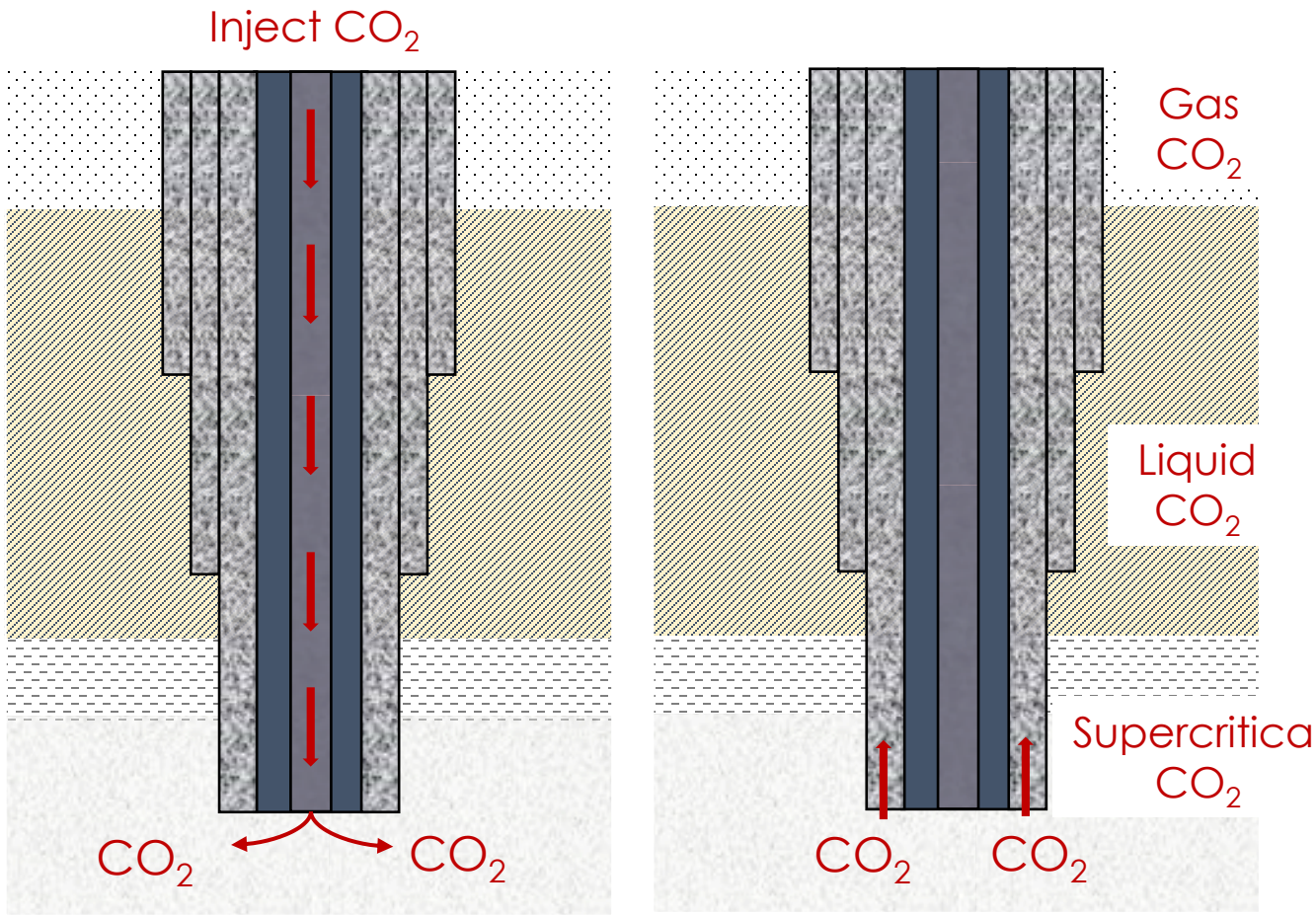
[2]NETL Support Contractor, 3610 Collins Ferry Road, Morgantown, WV 26507, USA

[3]North Carolina State University, 915 Partners Way, Raleigh, NC 27606, USA

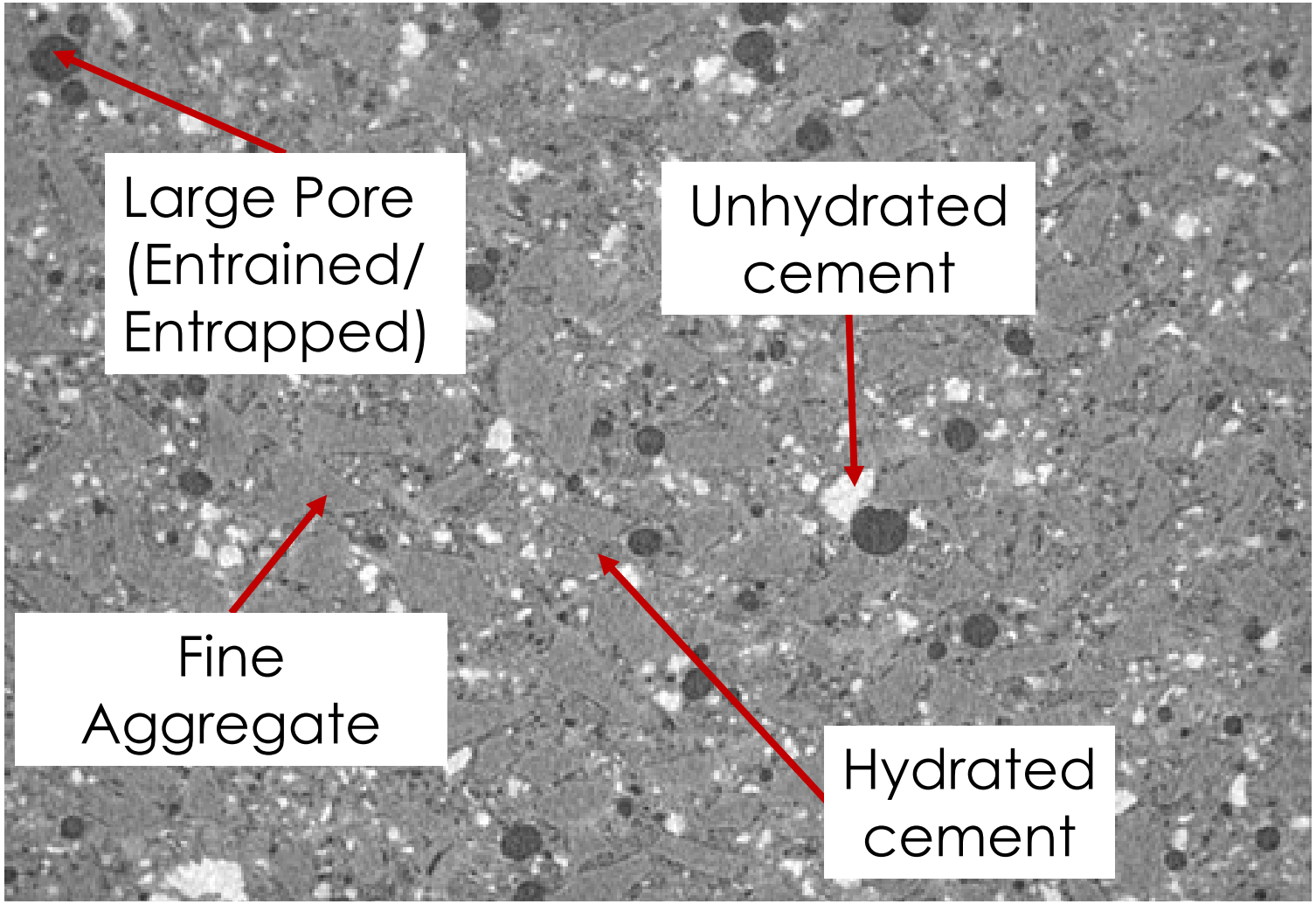
Research Motivation

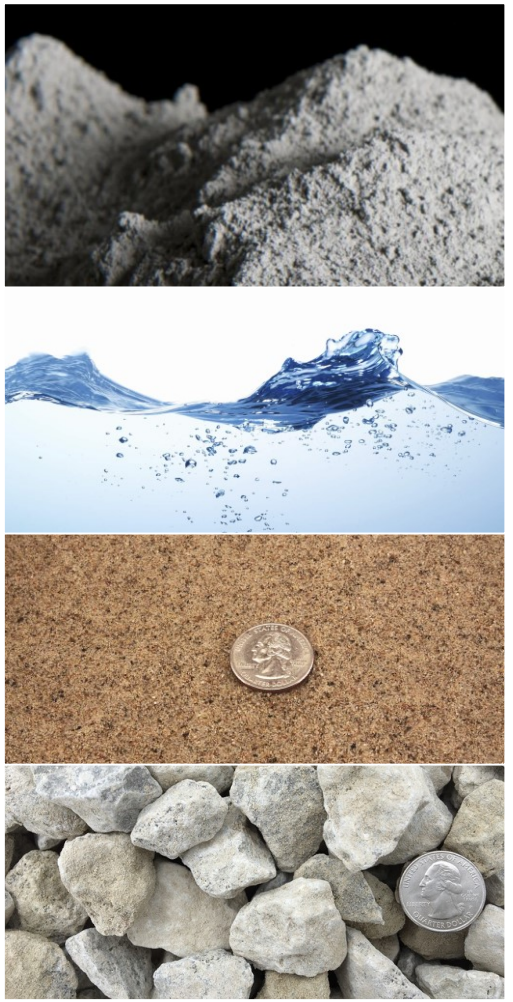
- More than 1.7 million wells in U. S.
- Steel and cement-based materials
- Used for long-term storage of CO_2
- CO_2 is known to leak over time
- Structure service life and durability

Carbon Capture & Storage (CCS) Well



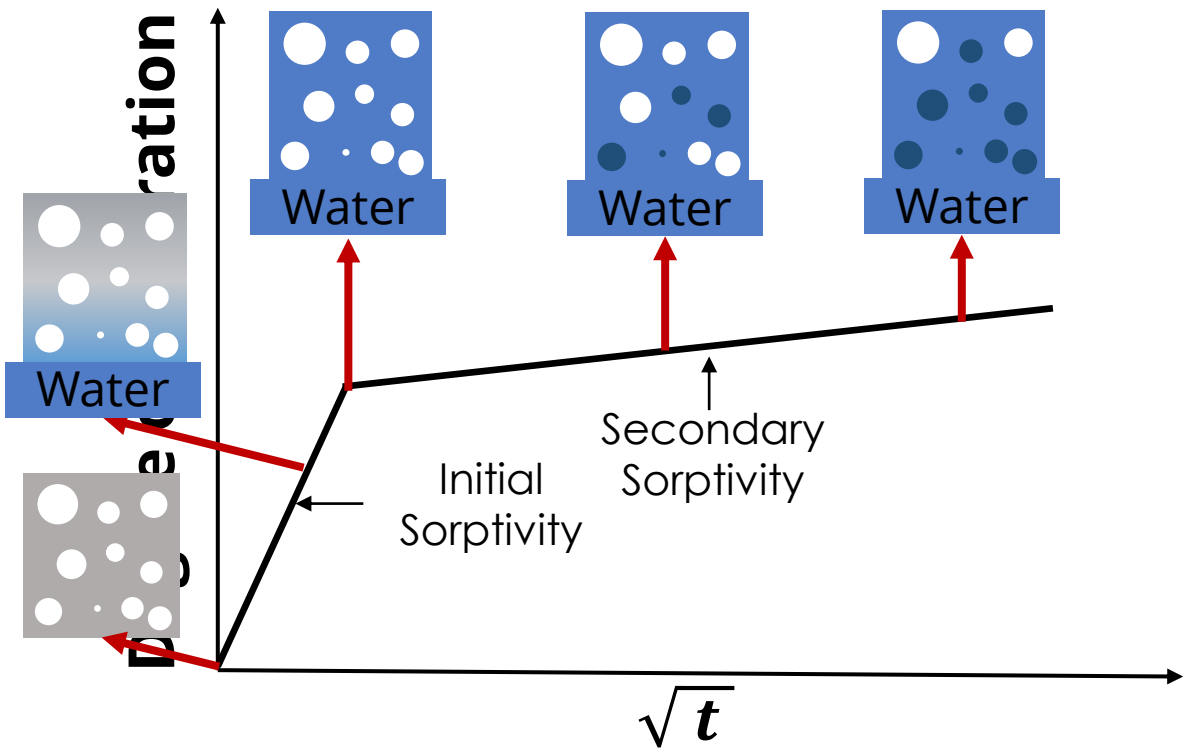
Background on Cement

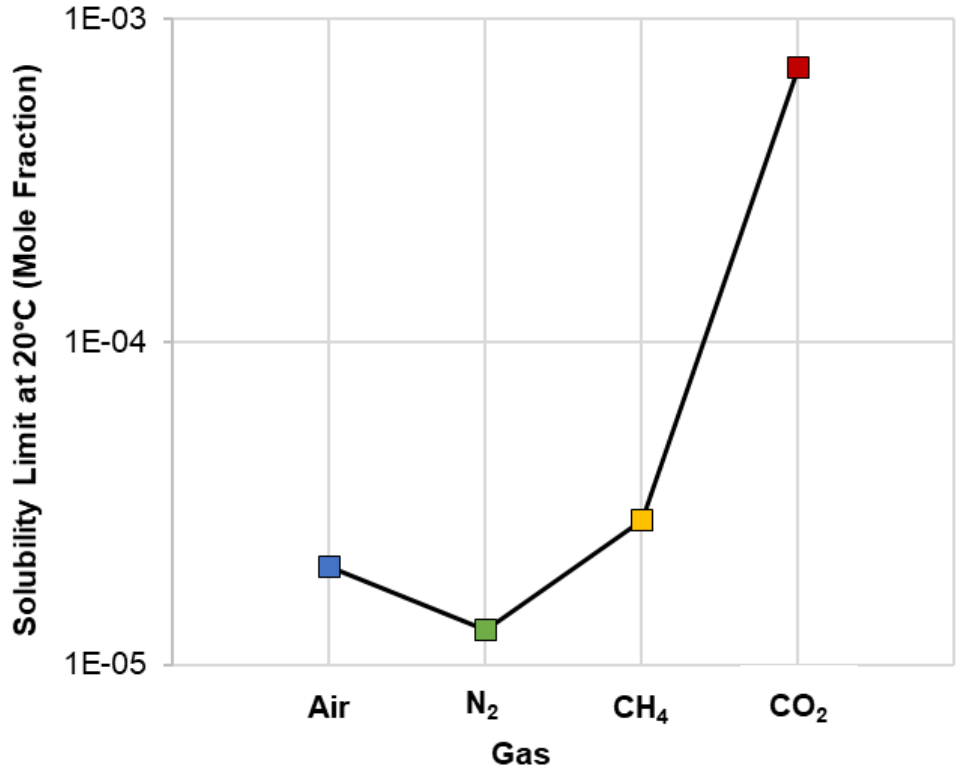




Small Pores = gel and capillary pores are sub-resolution

Background and Research Question 1



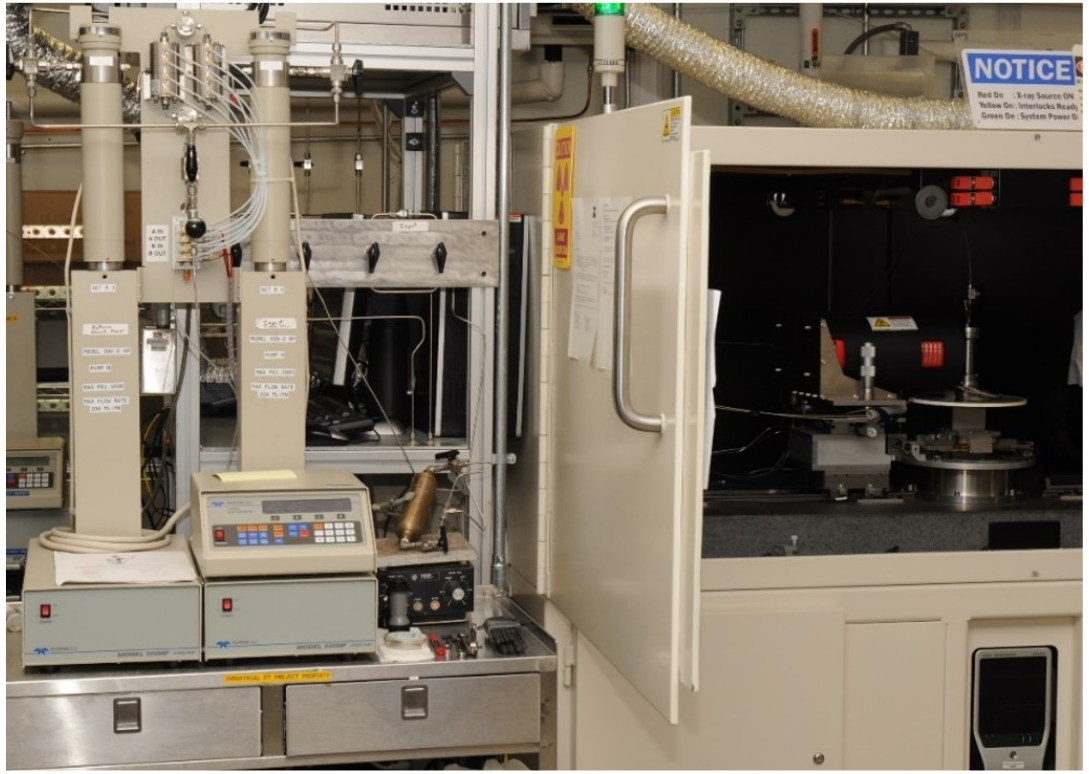


- **Research Question 1: Does the solubility of a gas phase influence the absorption of water in cement-based materials?**

Methods: Experimental Overview

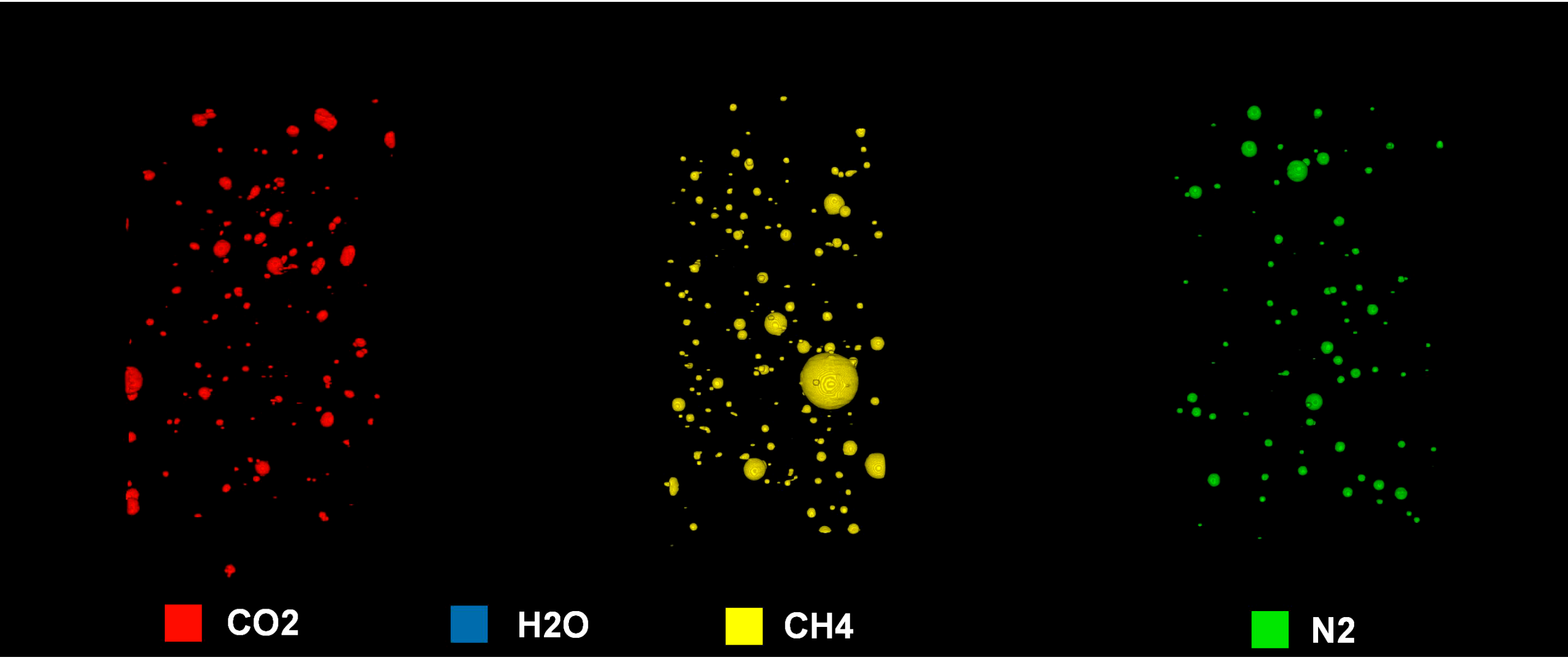
- How can we measure?

1. Air-entrained mortar (~8% air), w/c = 0.50
2. Specimen 0.25-inch (6.35 mm) diameter
3. Purge with different gas phases – CO_2, CH_4, and N_2
4. Bring water in contact with mortar
5. Use X-ray micro-CT to capture changes in large voids

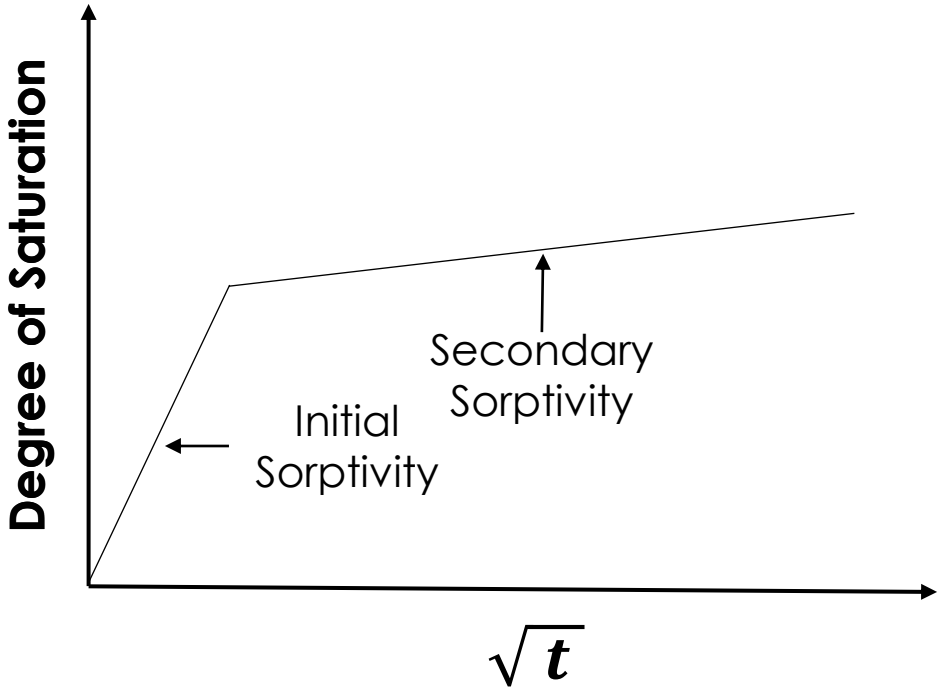


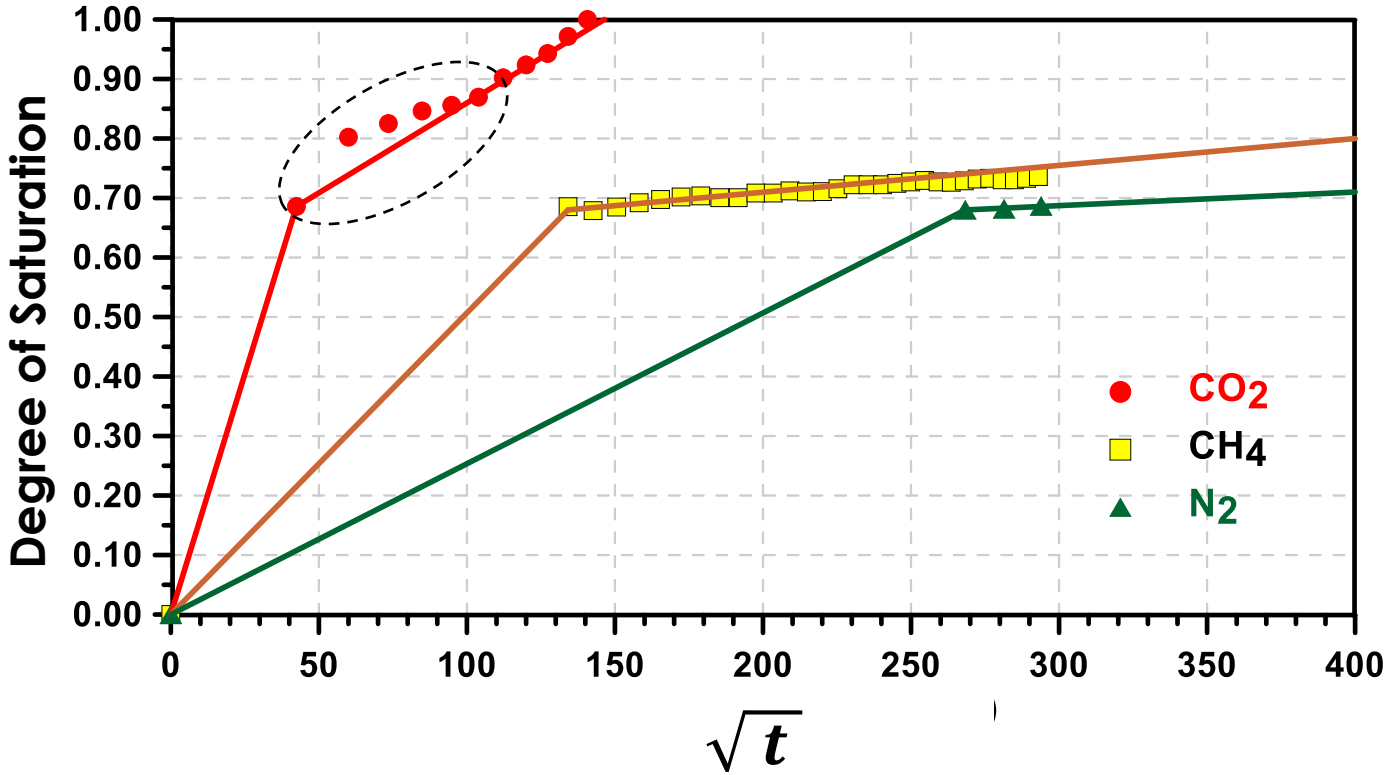
Engineered Flow-Through System and Micro-CT Scanner (Located at the National Energy Technology Laboratory)

Results: Secondary Sorption Evolution

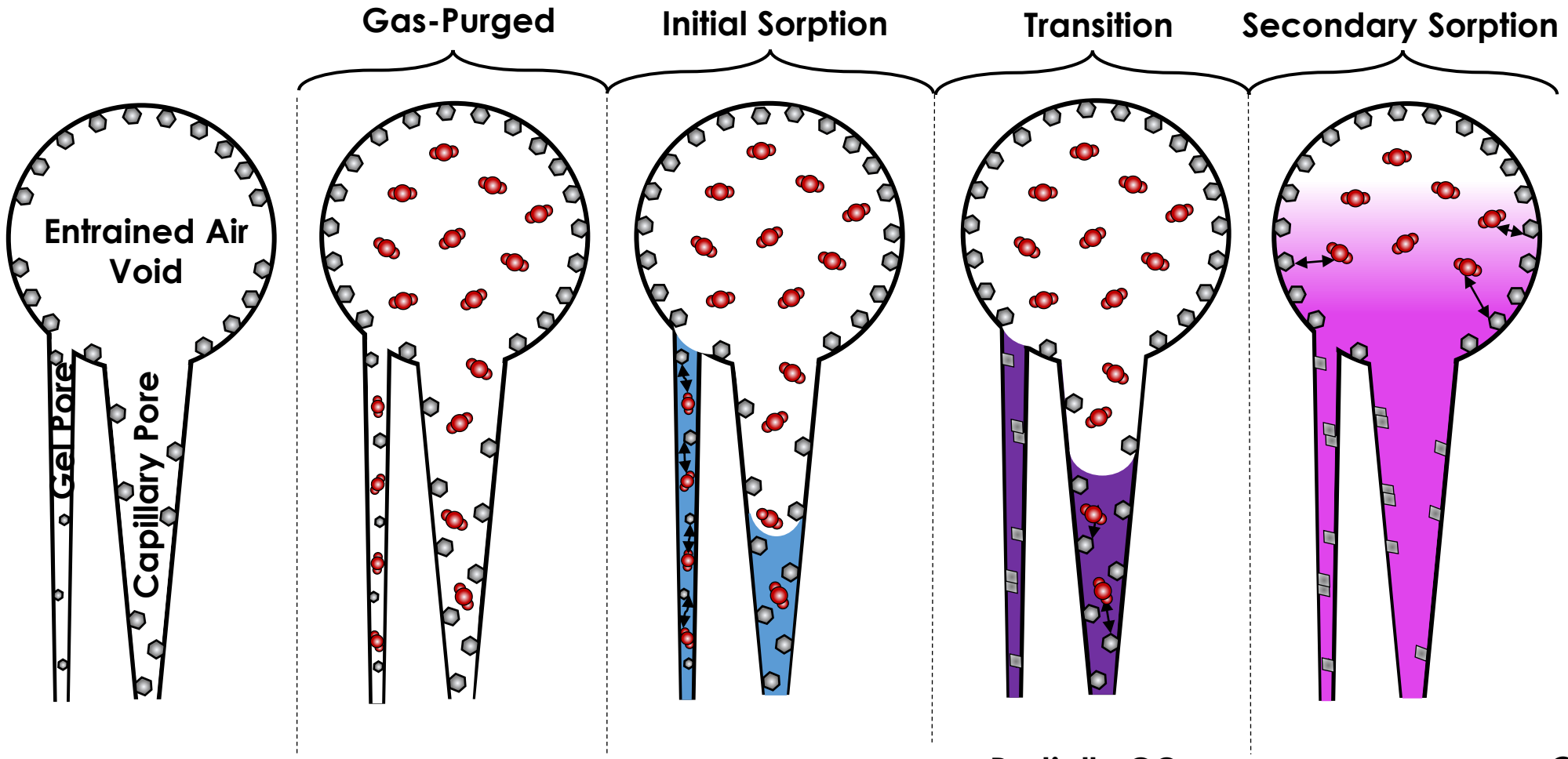


Results: Sorptivity Plots





Results: CO_2 Sorption Model



Model not to scale:

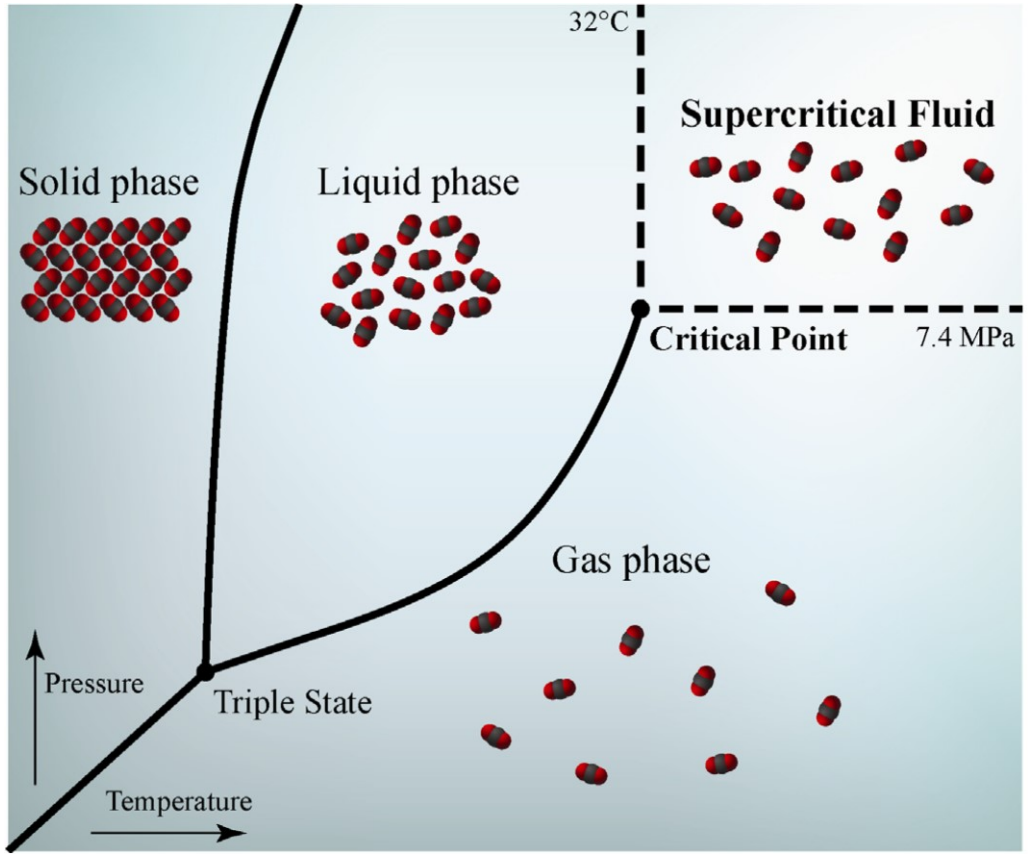
CO_2 molecule diameter = 0.33 nm

Gel pore diameter = ~10 nm

$Ca(OH)_2$ CO_2 H_2O Partially CO_2-Saturated H_2O $CaCO_3$ (Calcite) CO_2-Saturated H_2O

Background and Research Question 2

- Extensive literature on natural carbonation (0.04% CO_2 in atmosphere); Studied for more than 100 years
- Limited research on carbonation in CCS structures and reactive transport
- Supercritical CO_2 has the density of a liquid, but behaves like a gas
- **Research Question 2: What happens if we have variable moisture in a cement-based material exposed to high concentrations of CO_2?**



Carbon dioxide pressure-temperature phase diagram

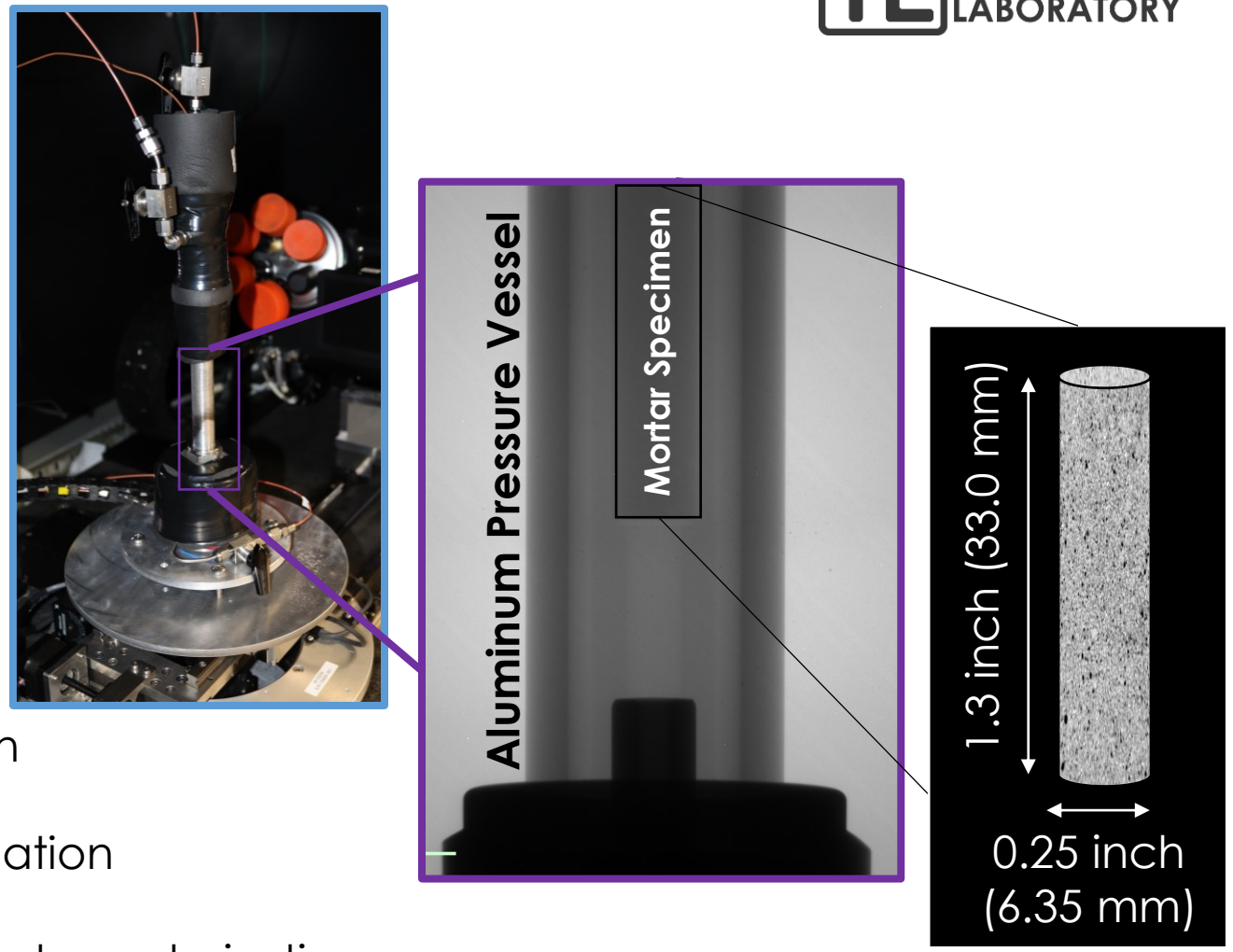
Methods: Experimental Overview

Variables:

- Material: mortar, w/c = 0.60
- Degree of Saturation (DOS): 0, 50, 100 %
- CO_2 Fluid State: Gas (Gas), Liquid (Liq), and Supercritical (SC)

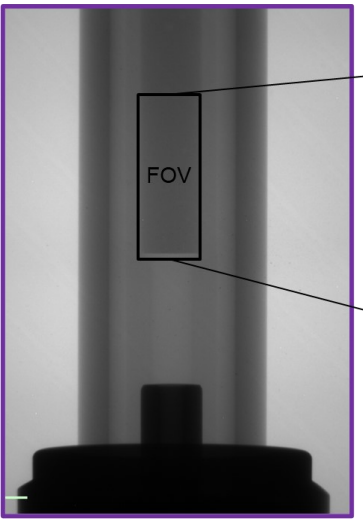
Experimental:

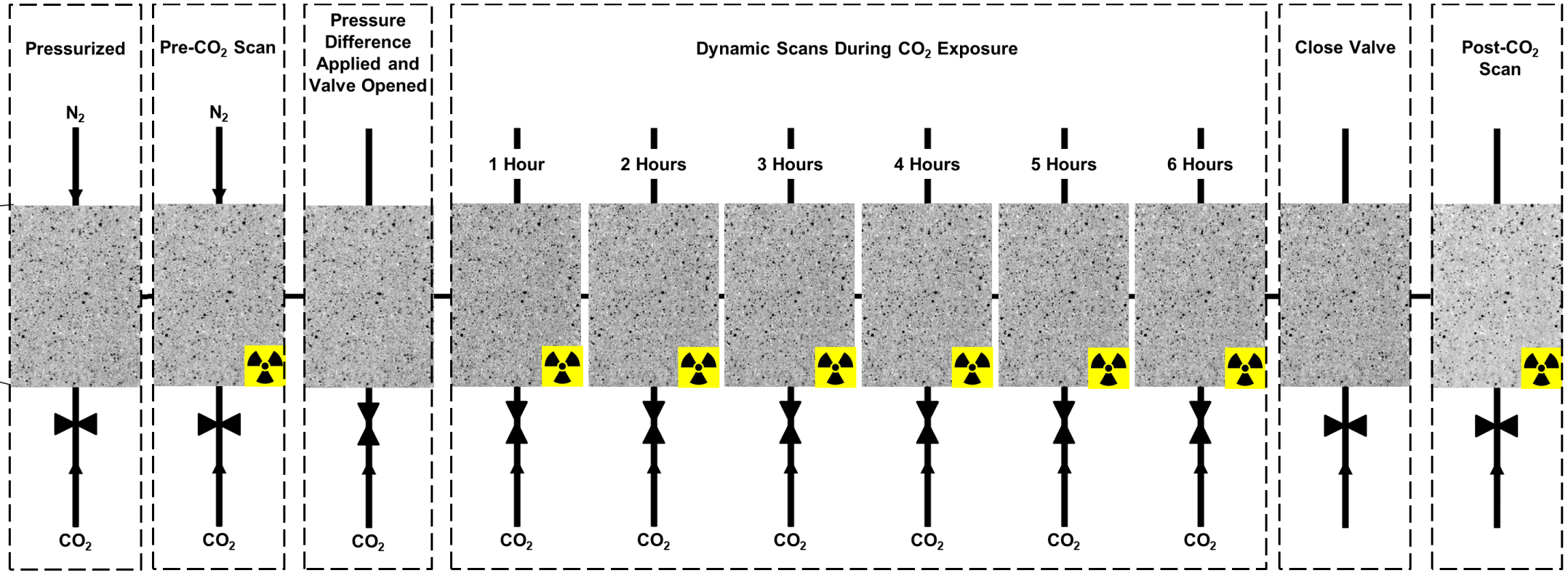
- *In situ* experiments using X-ray micro-CT → relative rate of transport
- Post CO_2-exposure X-ray CT → higher resolution
- Thermogravimetric analysis → quantify carbonation
- Vapor sorption analyzer → sub-resolution pore characterization



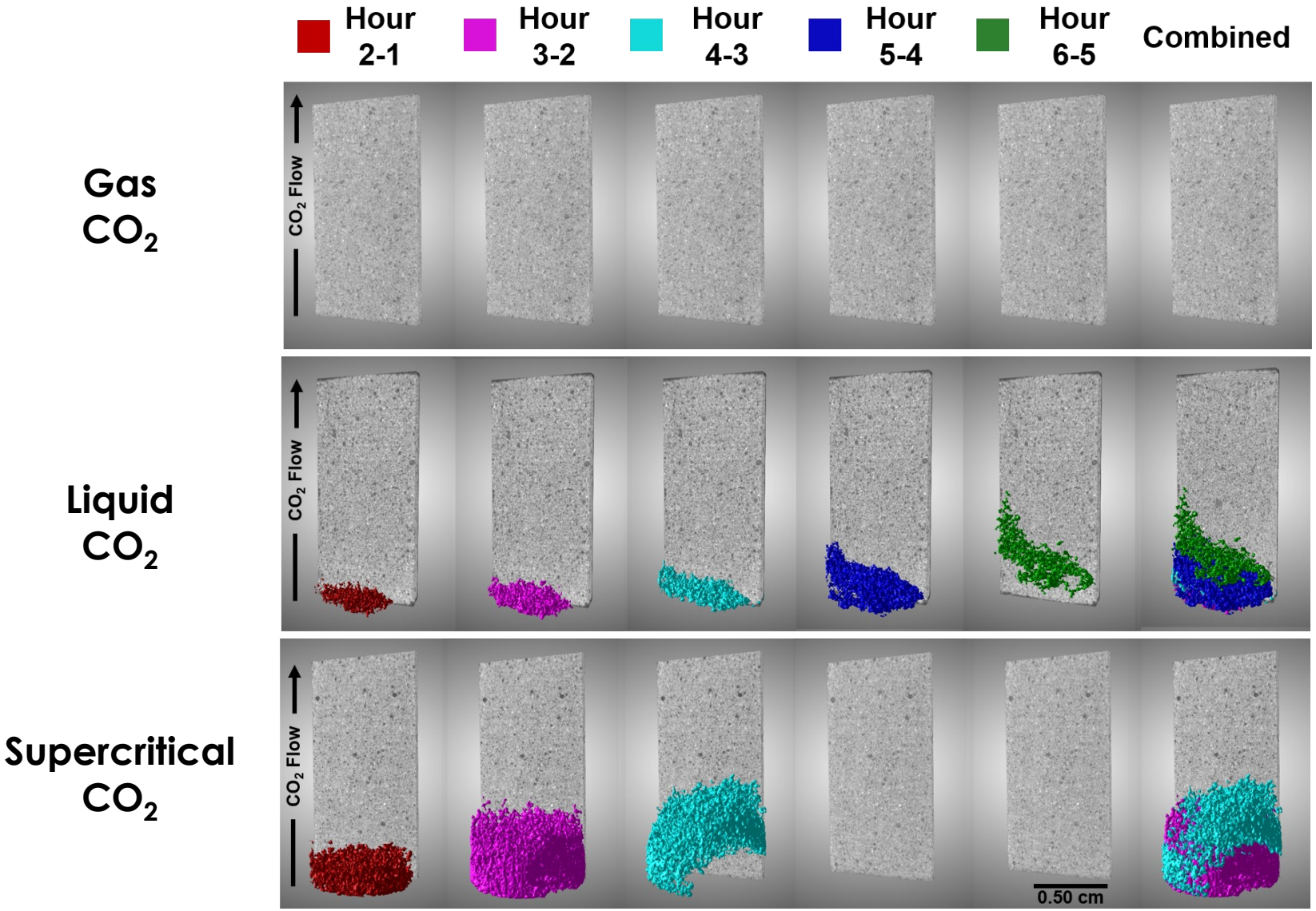
Methods: X-Ray CT Experimental Overview

Field of View (FOV)

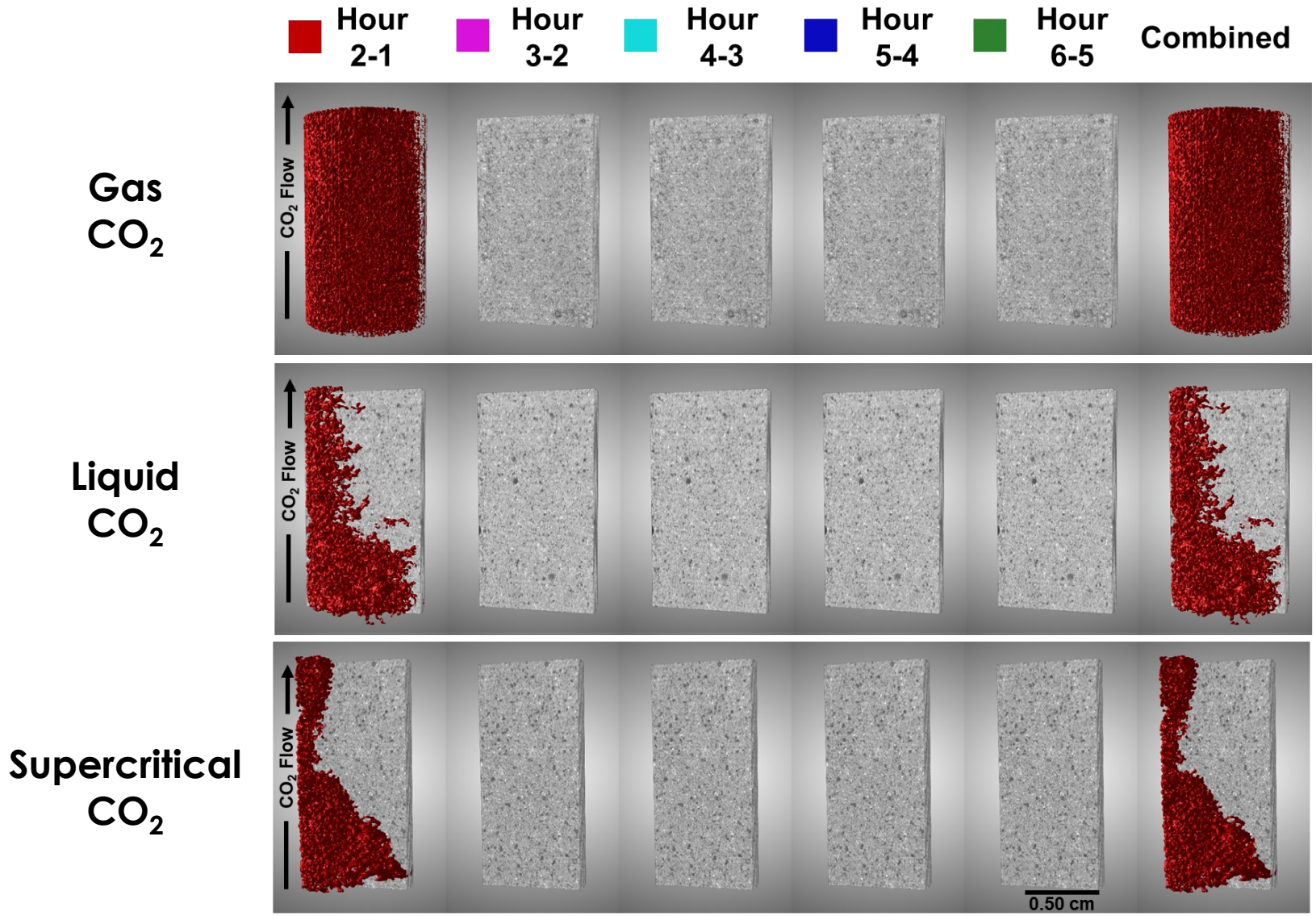




Results: 0% DOS Carbonation Front Evolution

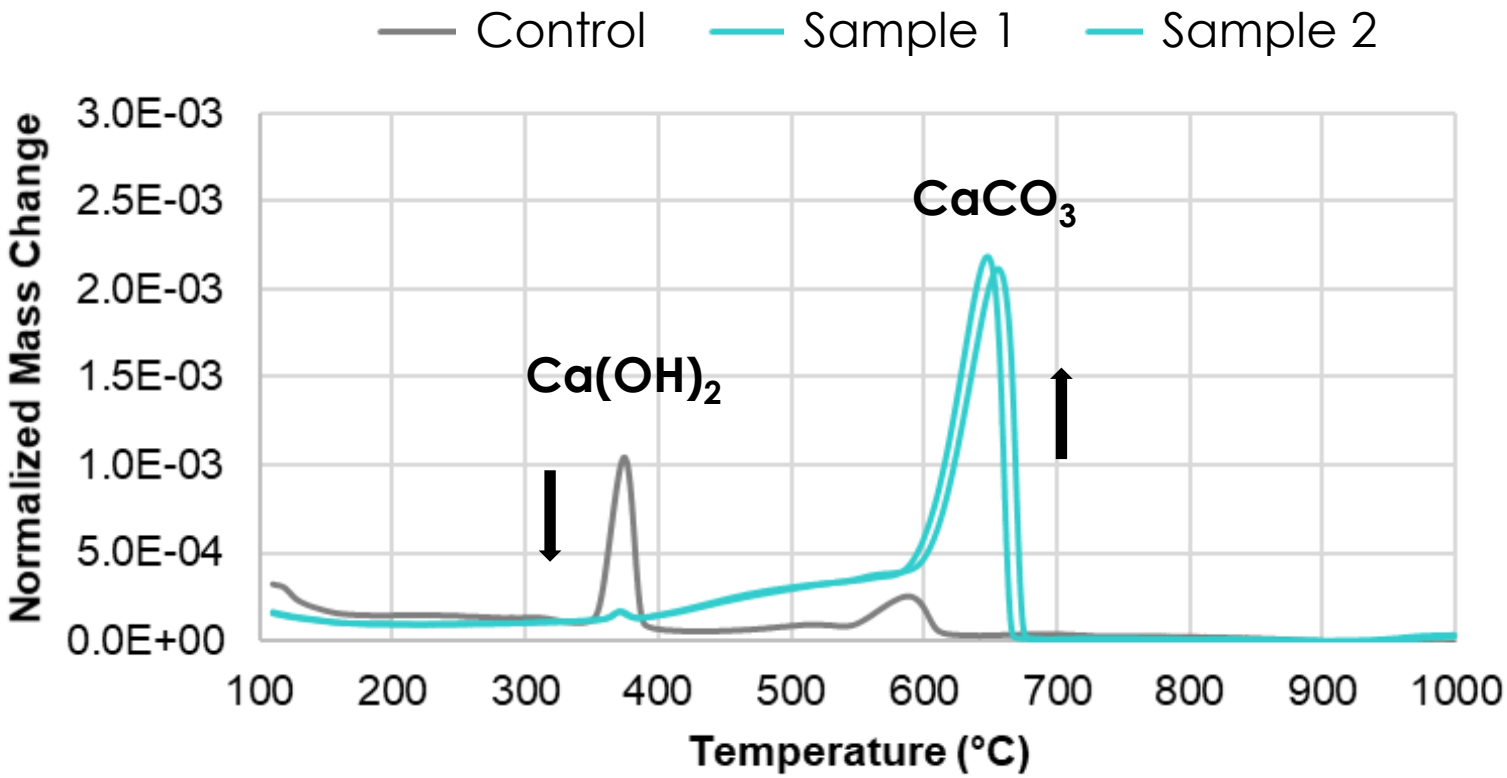


Results: 50% DOS Carbonation Front Evolution



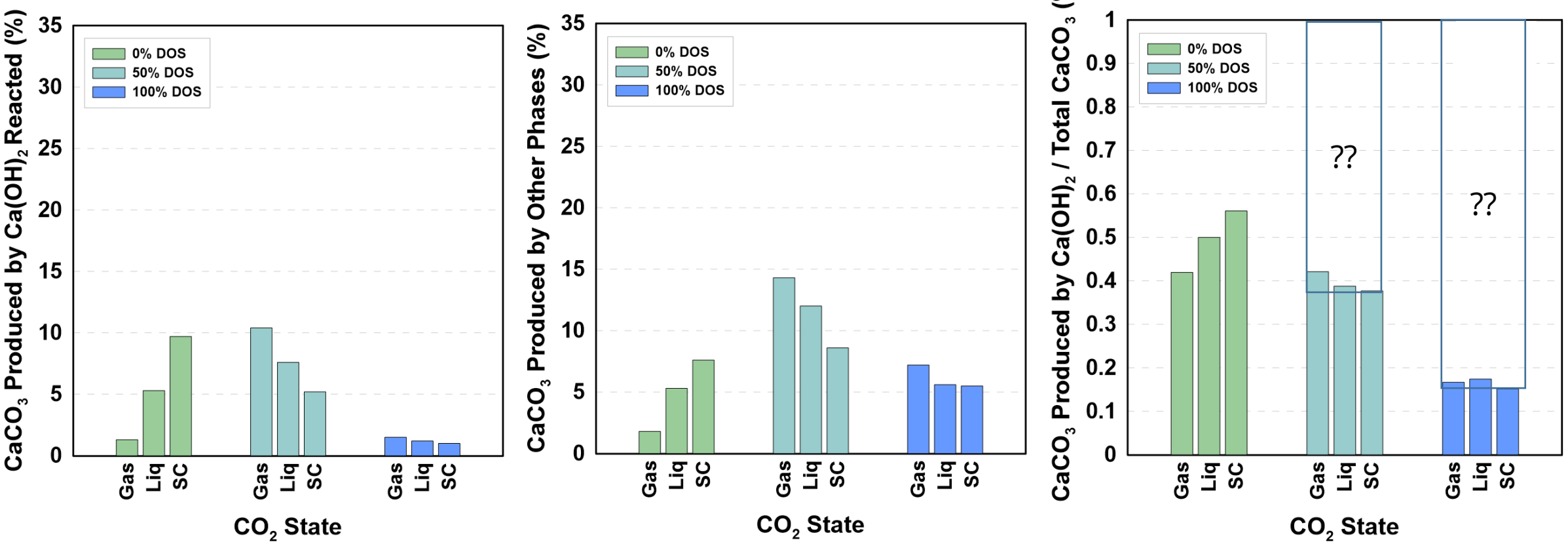
Methods: Thermogravimetric Analysis

- Thermogravimetric analysis (TGA) consists of sensitive scale in a high temperature oven
- Different compounds decompose at different temperatures



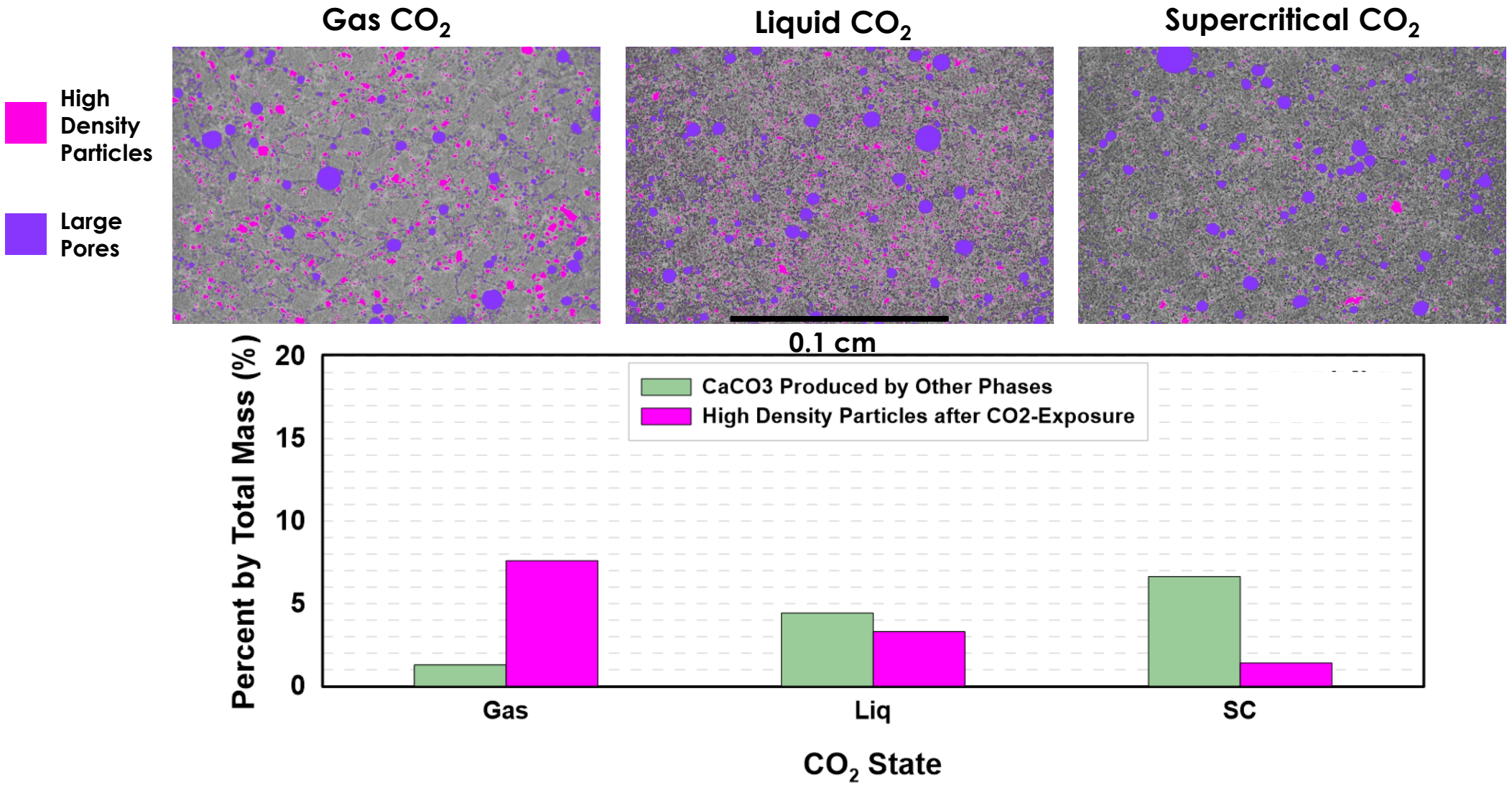
Results: Thermogravimetric Analysis

$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$



Takeaways: phases other than $Ca(OH)_2$ carbonate and phases other than $Ca(OH)_2$ produce more carbonates as degree of saturation is increased

Results: 0% DOS High Resolution Analysis



Concluding Remarks

- **Does the solubility of a gas phase influence the absorption of water in cement-based materials?**
  - Solubility of the gas phase is important in mass transport and affects the transport mechanisms involved
  - When gas CO_2 resides in the pores, reactive secondary sorption of water is nonlinear
  - Dalton, L. E., Jarvis, K., and Pour-Ghaz, M. The effect of gas phase solubility on the secondary sorption in portland cement mortar observed by X-ray CT. *Transp Porous Med* 133, 397-411 (2020).
- **What happens if we have variable moisture in a cement-based material exposed to high concentrations of CO_2?**
  - Carbonates form at expedited rate when pores are at 50% DOS and phases other than $Ca(OH)_2$ are carbonating
  - As CO_2 leaks into a CCS well, localized areas may carbonate leading to inhomogeneous microstructure
  - Dalton, L. E., Crandall, D., & Pour-Ghaz, M. (2022). Supercritical, liquid, and gas CO2 reactive transport and carbonate formation in portland cement mortar. *International Journal of Greenhouse Gas Control*, *116*, 103632.

Acknowledgement

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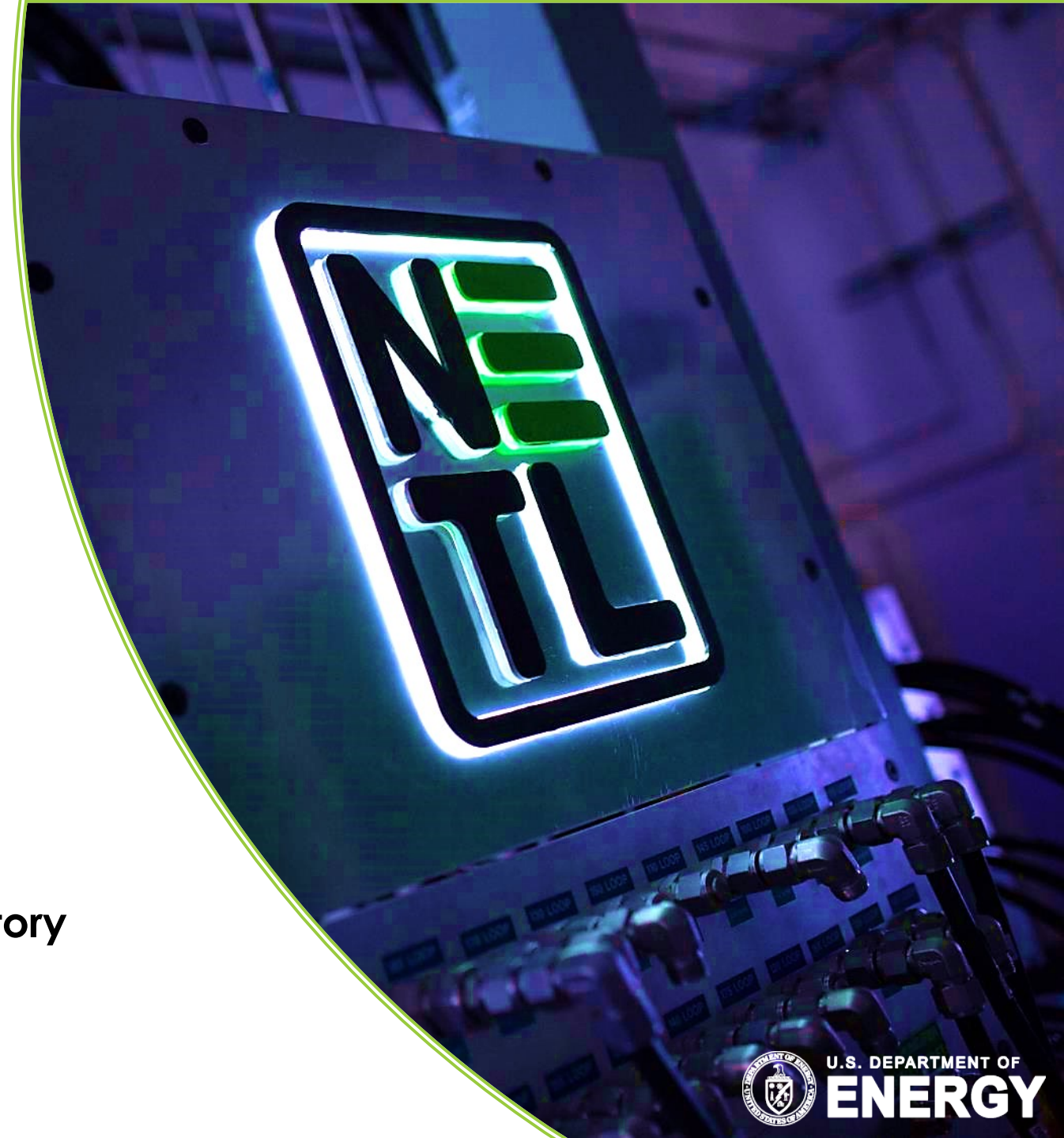

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