Cementitious Barriers Partnership

G. Flach, C. Langton, R. Seitz, S. Marra, H. Burns, SRNL
D. Kosson (Presenter), K. Brown, S. Mahadevan, CRESP, Vanderbilt U.
A. Garrabrants, J. Arnold, S. Sarkar, CRESP, Vanderbilt U.
H. Van der Sloot*, J.C.L. Meeussen, R. Comans, ECN (NL)
D. Esh, M. Furman, J. Phillip, US NRC
E. Garboczi, K. Snyder, J. Bullard, NIST
E. Samson, J. Marchand, SIMCO Technologies, Inc. (CA)

DOE Performance Assessment Community of Practice
Technical Exchange
25-26 May 2011

Environmental Management
safety ❖ performance ❖ cleanup ❖ closure
www.em.doe.gov

**Hans van der Sloot Consultancy* after 1-1-2010

Cementitious Barriers in Waste Management

CERCLA Disposal Cell

Disposal Engineered Trench

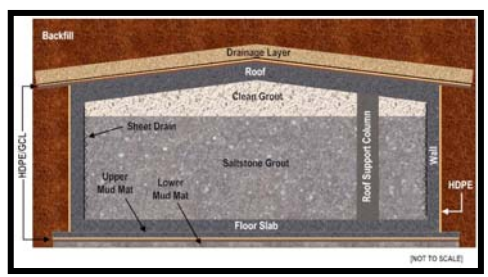
LAW Disposal in Vaults

LLW Disposal Grouted in Vault

D&D

Large Facility Closure

Tank Integrity & Closure



Saltstone Vault Disposal

Environmental Management
safety ❖ performance ❖ cleanup ❖ closure

www.em.doe.gov

Presented by Martin J. Letourneau, DOE-EM Performance Assessment Community of Practice

Print

Close

Cementitious Engineered Barriers

- Used to enhance environmental performance of waste disposal units
  - Mobile, short- and long-lived radionuclides
  - Humid and possibly semi arid sites
  - Shallow to moderately deep aquifers
- Legacy waste, facilities, and environmental clean-ups
  - **Tank Closures** (SRS and Hanford)
    - Salt waste (SRS)
    - HLW tank integrity assessment & closure (Hanford, SRS, Idaho)
  - **D&D, In-Situ Decommissioning** (Reactors, Canyons)
  - **CERCLA Remediation:** (Soil and Groundwater Remediation)

Cementitious Barrier Performance Needs

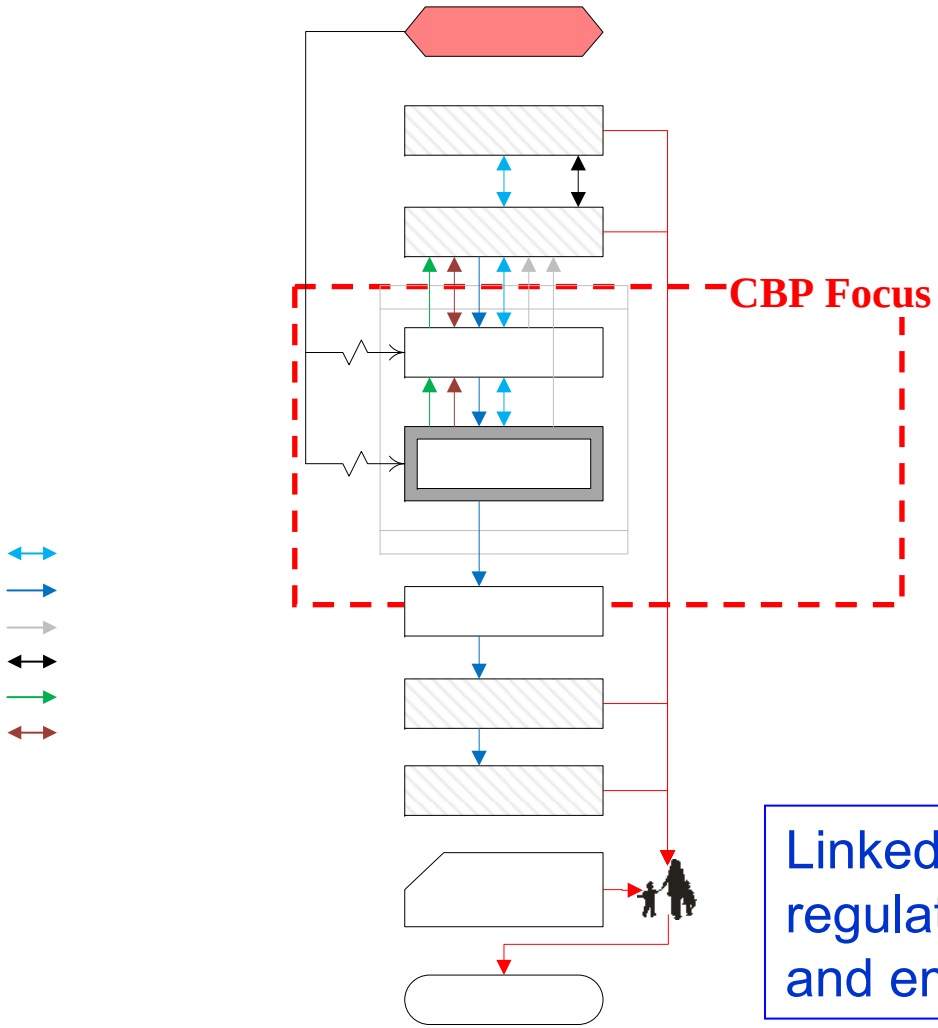
- Engineered Barrier Performance is incorporated into
  - DOE Performance Assessments
  - CERCLA Risk Documents (EPA, DOE, States and public)
- Engineered Barrier Performance is needed for
  - Management of existing waste disposal facilities
  - New waste disposal site selection decisions and facility designs
  - New waste treatment flow sheets
  - Next generation DOE and Commercial nuclear facility design and licensing (DOE and NRC)

CBP Project Goal

- Develop a reasonable and credible set of tools to predict structural, hydraulic and chemical performance of cement barriers used in nuclear applications over extended time frames (e.g., up to or >100 years for operating facilities and >1000 years for waste management).
  - Mechanistic / Phenomenological Basis
  - Parameter Estimation and Measurement
  - Boundary Conditions (physical, chemical interfaces)
  - Uncertainty Characterization

Regulatory Credibility is Essential

CBP Focus: Engineered Barriers & Source Terms



CBP Focus:

- Cementitious materials performance as part of engineered system and their interfaces with natural system
- To provide near field source term
- Uncertainty approach being developed to be broadly applicable to PA and design process

Linked with current PAs, regulatory evaluation tools (GoldSim®) and emerging DOE tools (ASCEM)

Technical Strategy / Approach

- **Coordinated experimental and computational program**
  - Focus on key aging and degradation mechanisms effecting performance
    Sulfate attack, Carbonation, Oxidation, Leaching, Cracking
  - Conceptual model improvement – mechanistic basis for radionuclide mobility and material durability
  - Uncertainty Assessment including Bayesian/Stochastic methodologies
- **Experimental program**
  - Define test methods and parameter measurements
  - Model validation – Exposure testing and field testing including cores
- **Modeling program**
  - Long-term extrapolation of performance based on field scenarios
  - Hydraulic, chemical and mineralogical changes (time, conditions)
  - Radionuclide retention and release and near-field fate and transport

Technical Strategy / Approach

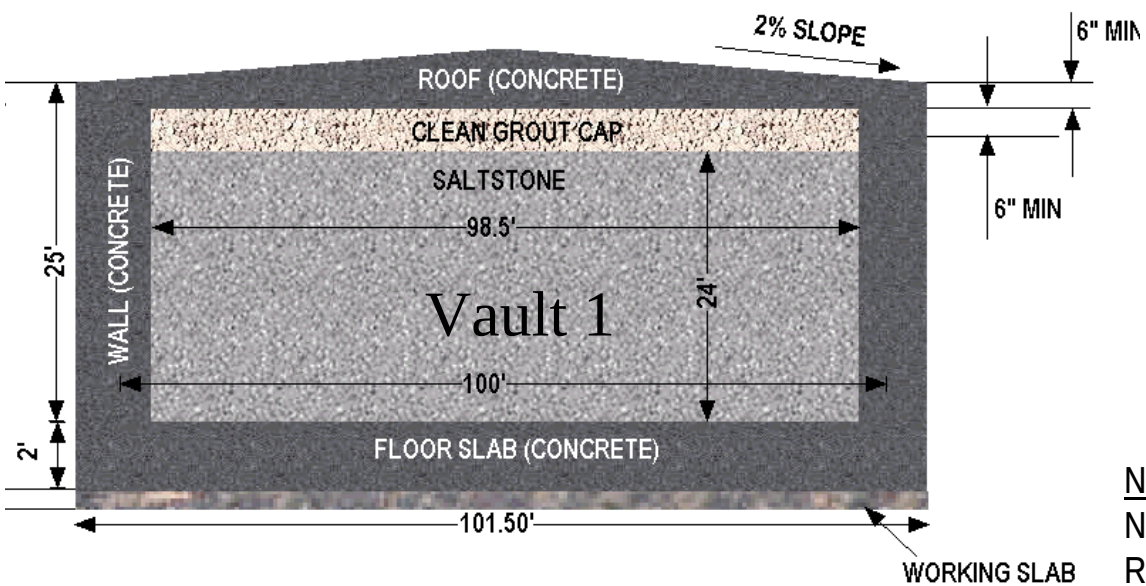
- **Reference Cases – provide basis for comparison and demonstration of CBP tools**
  - Cementitious waste form in concrete vault with cap
  - High-level waste tank integrity & closure
  - D&D and closure of nuclear processing facilities (e.g., reactors & canyons)
  - Grouted vadose zone contamination
  - Spent fuel pool
- **Reference Materials** – surrogate LAW cementitious waste form, reducing grout, reinforced concrete
- **Extension/enhancement of existing tools –**
  - STADIUM® (service life & durability)
  - LeachXS™/ORCHESTRA (leaching & chemical evolution)
  - CEMHYD3D/THAMES (transport properties)
  - GoldSim – Integration & uncertainty assessment platform

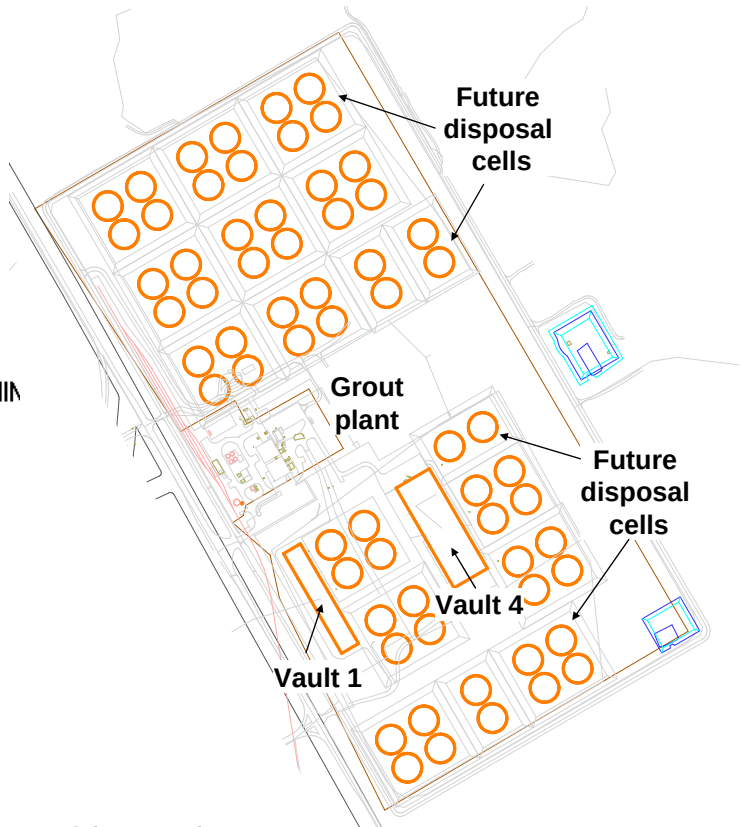
CBP Members

- **Department of Energy – Office of Environmental Management**
  - Scenarios & Key Uncertainties
  - Primary end-user
- **Nuclear Regulatory Commission**
  - Scenarios & Key Uncertainties
  - Primary end-user
- **Savannah River National Laboratory**
  - PA Interface
  - Model Integration
  - Cracking Scenarios
- **National Institute of Standards and Technology**
  - THAMES – Microstructure Evolution & Transport Properties
- **SIMCO Technologies, Inc.**
  - STADIUM – Physical & Hydraulic Performance
- **Energy Research Centre of the Netherlands**
  - LeachXS/ORCHESTRA – Chemical Performance & Constituent Release
- **Vanderbilt University/CRESP**
  - Chemical Performance & Constituent Release (experimental)
  - Uncertainty Analysis Framework
  - Model Integration

Contaminant Retention in Cementitious Materials: Savannah River -- Saltstone Disposal Facility

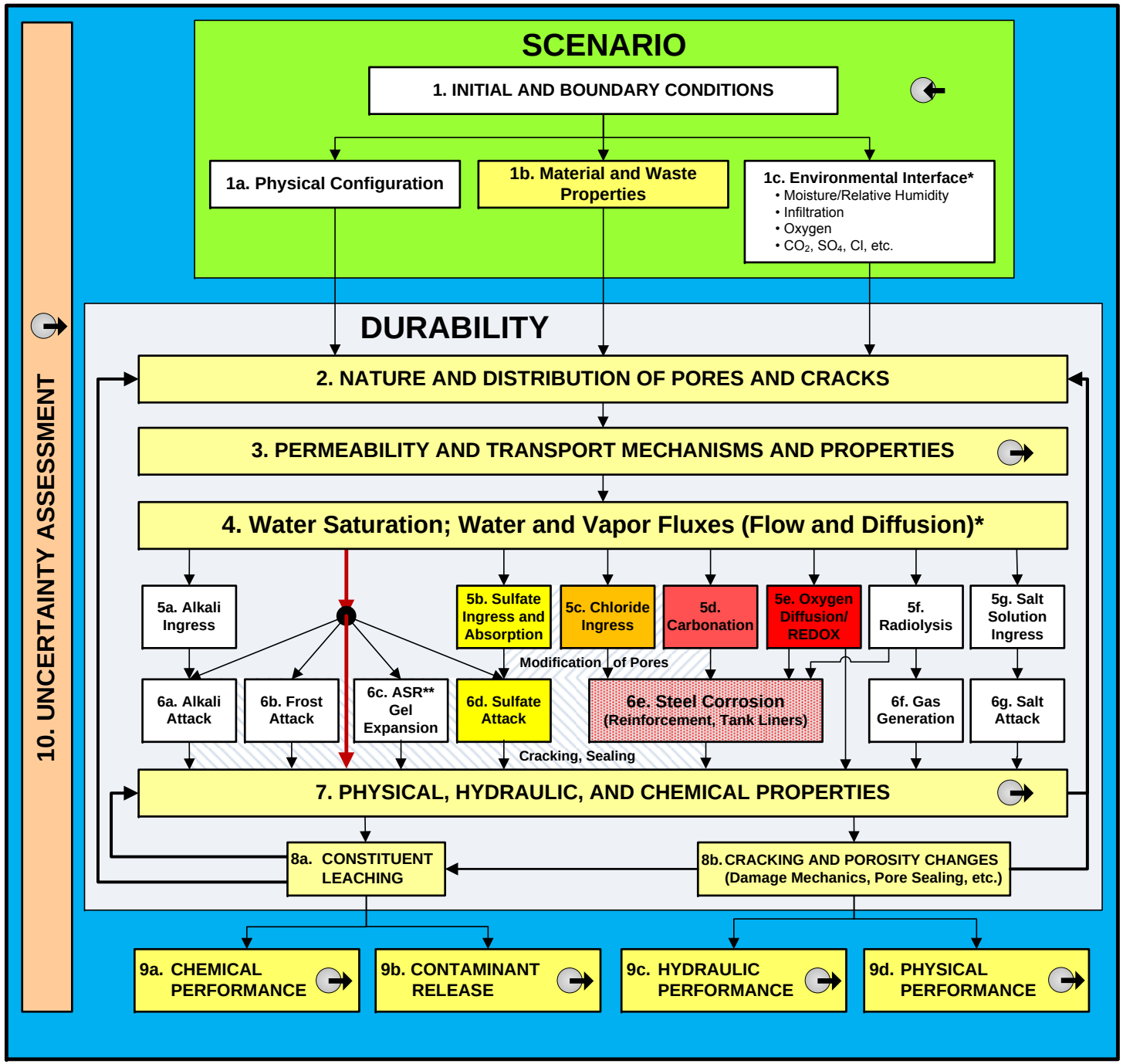
- Salt liquid waste mixed with dry grout to form "Saltstone"
- Blast furnace slag added to Saltstone grout and vault concrete to create reducing conditions





Notable species
Nitrate, Tc-99, I-129 and Ra-226 ingrowth from Th-230

Specifications, Properties, and Phenomena for the Evaluation of Performance of Cementitious Barriers



Print

Close

CBP Example Problem
Salt Waste Disposal System Integrity

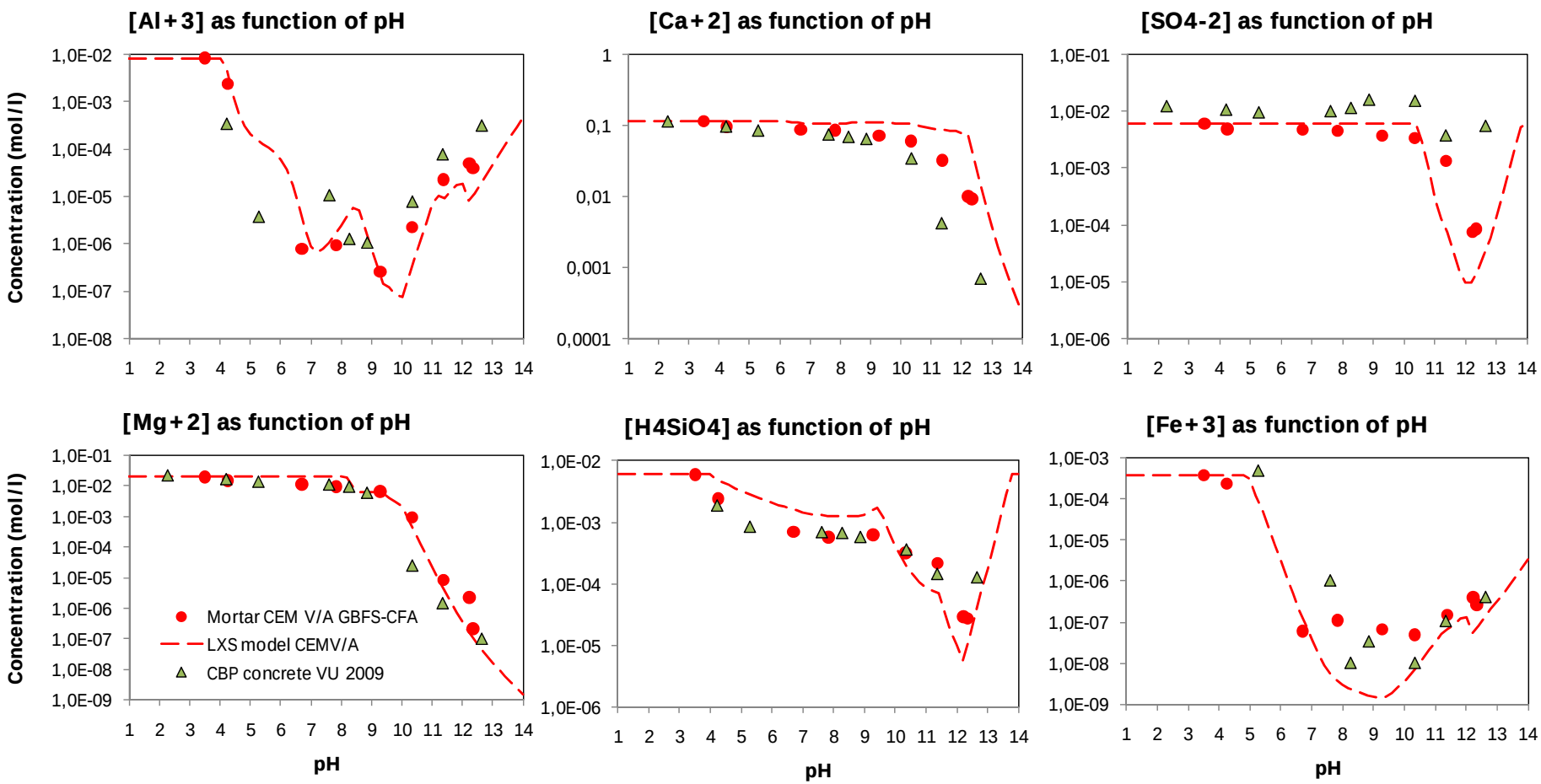
- **Potential concerns**
  - **Sulfate** from waste form may degrade vault concrete
  - **Oxygen** ingress may accelerate Tc-99 release
- **Knowledge Gaps**
  - Current concrete formulations are complex and exposure scenarios are not typical of usual environmental conditions
    - Portland cement/slag cement/fly ash and modifiers are used to create reducing and low permeability concrete
    - Complex chemical interface (between waste form and vault)
  - Rates of degradation and threshold sulfate concentrations unknown
  - Mechanistic model needed to evaluate specific design scenarios
  - Experimental data needed for model calibration & validation

CBP Example Problem
Salt Waste Disposal System Integrity

➢ Approach

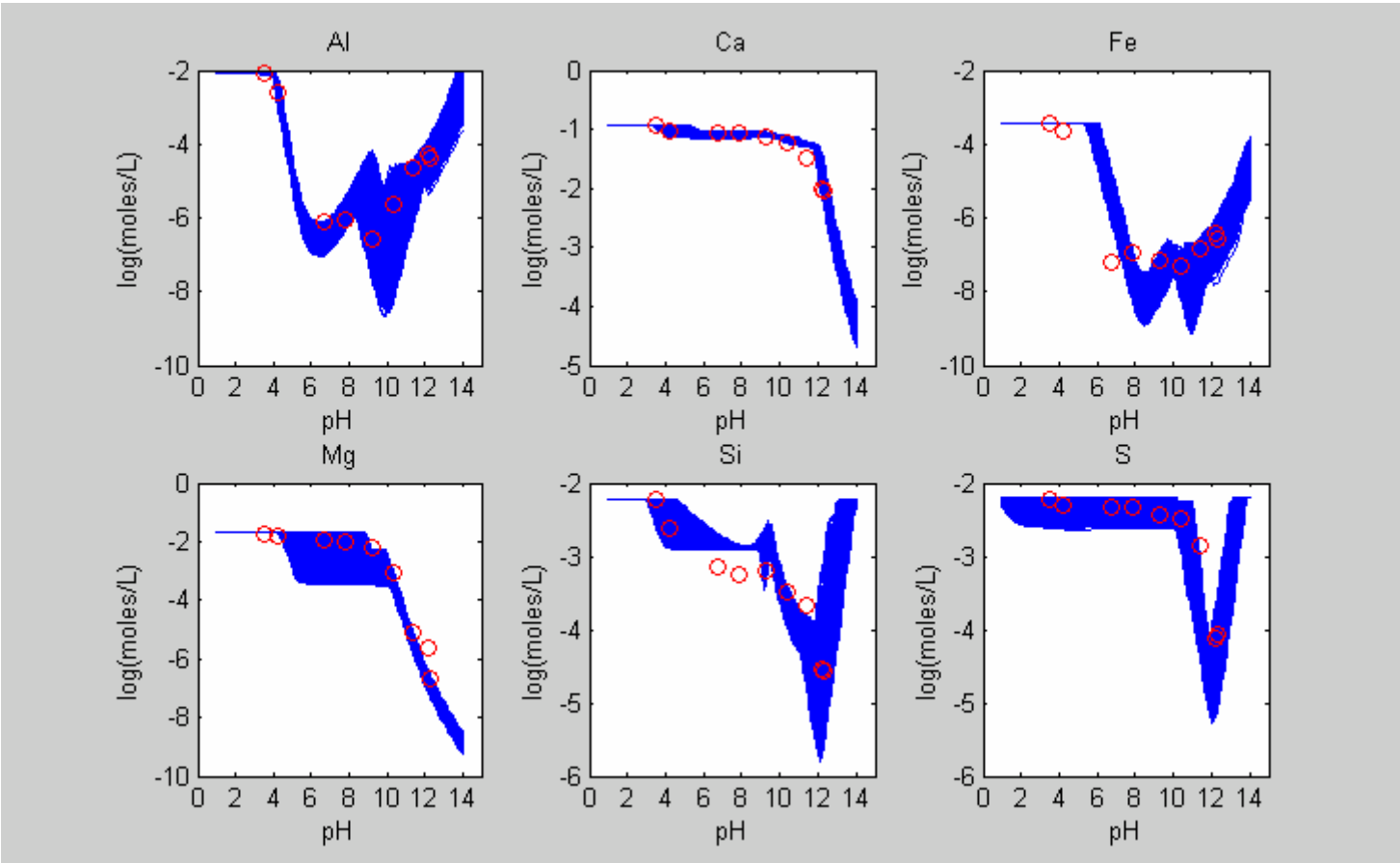
- Define and validate thermodynamic model for vault concrete
- Define conceptual model
- Adapt reactive transport model and couple with damage mechanics – captures impact of change in material structure (from cracking and porosity change) on transport phenomena
- Gather and augment experimental data for model parameterization and validation
- Model scenarios and assess uncertainties

Comparison of Cement Data and Thermodynamic Model Predictions



Experimental data from USEPA draft Method 1313

Parameter Uncertainty – Calibration of Thermodynamic Model Parameters

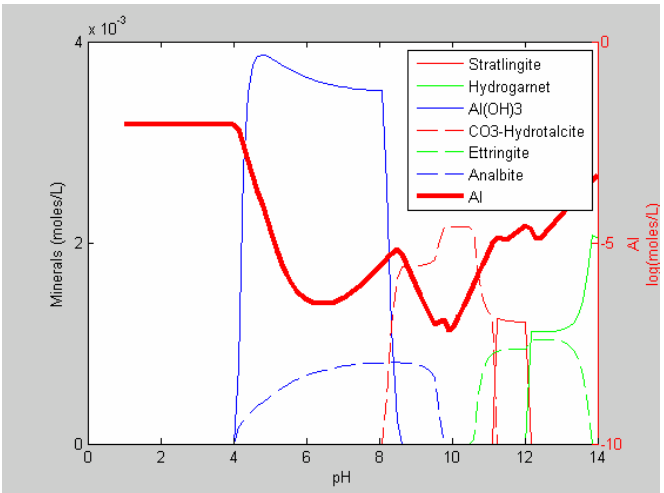


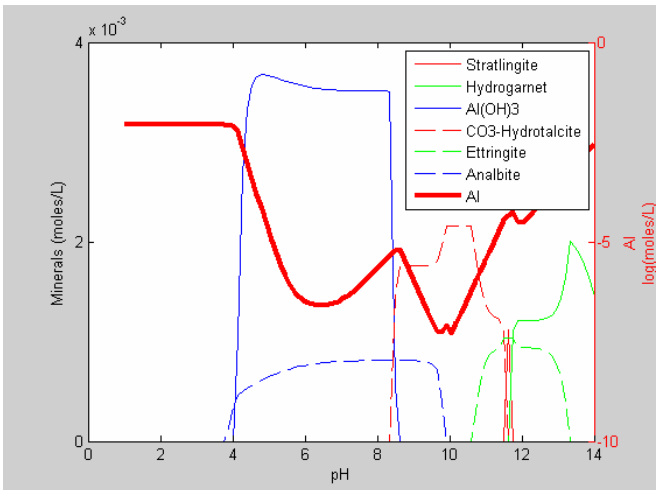
Data and Model uncertainty considered

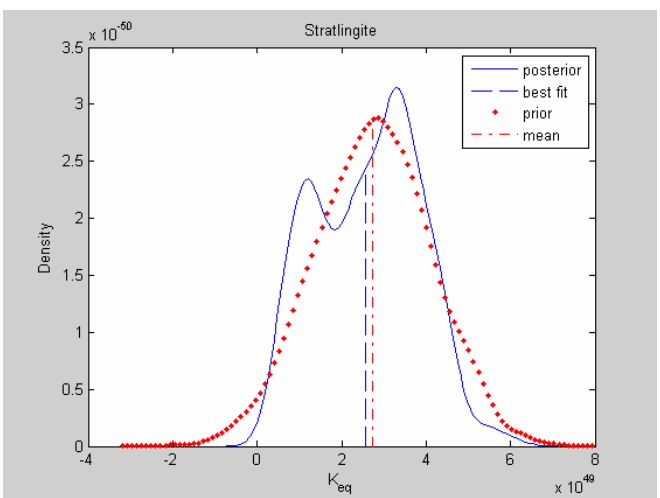
Uncertainty Reduction in Thermodynamic Model Parameters

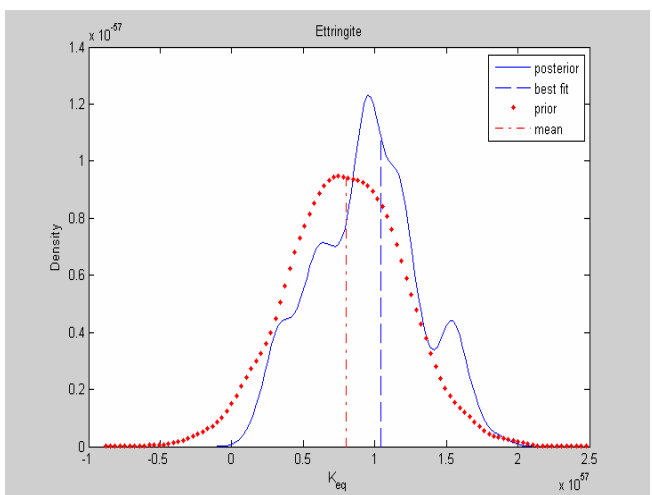
Prior | Best Fit

Al



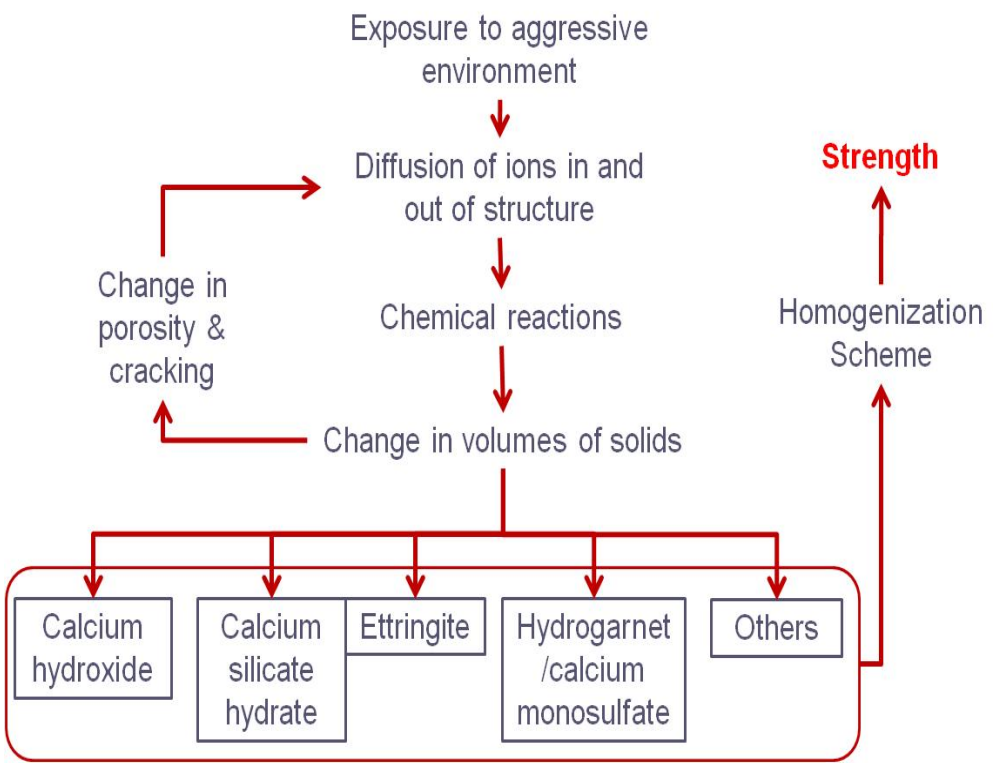




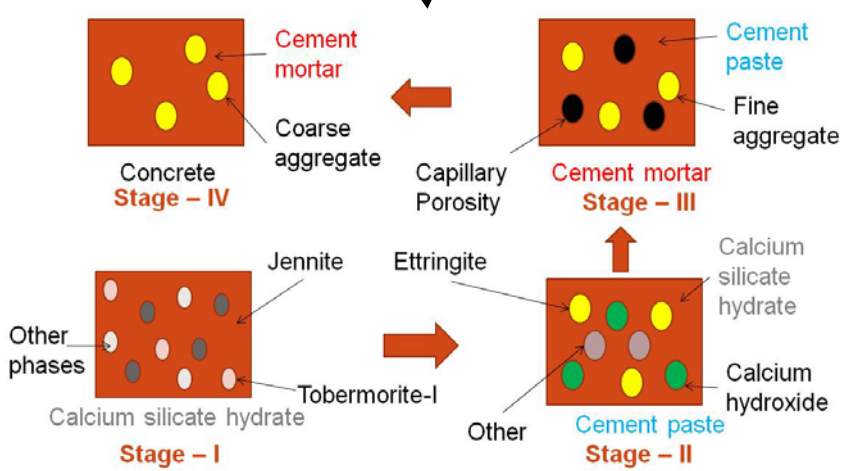


Most prominent changes: Stratlingite, hydrogarnet and ettringite

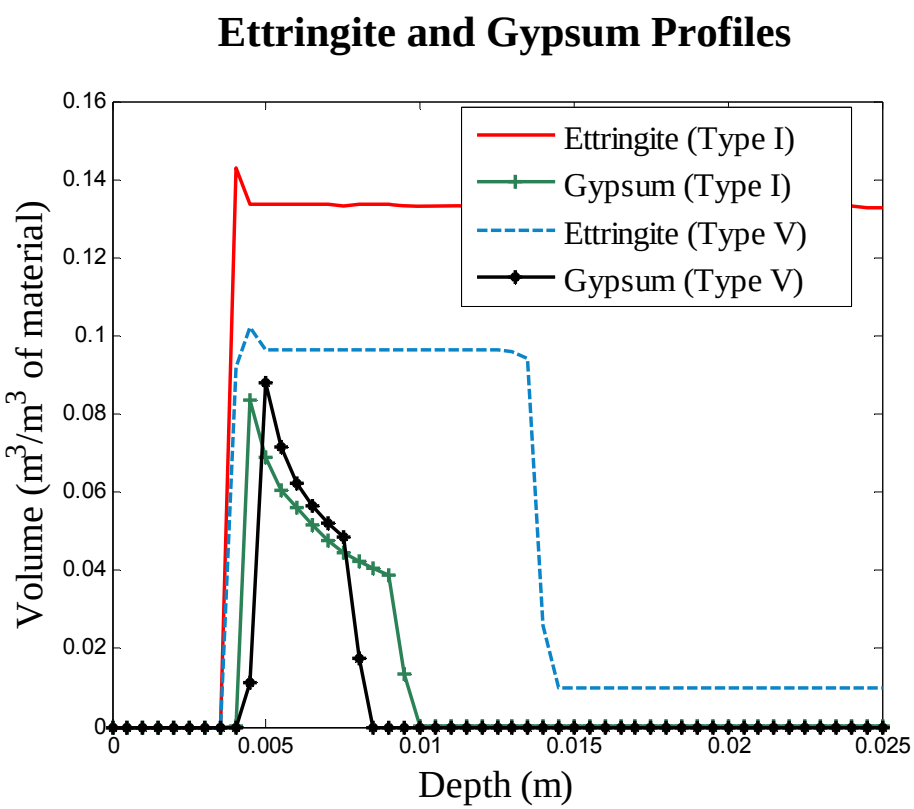
Change in Mechanical Properties due to Chemical Reactions

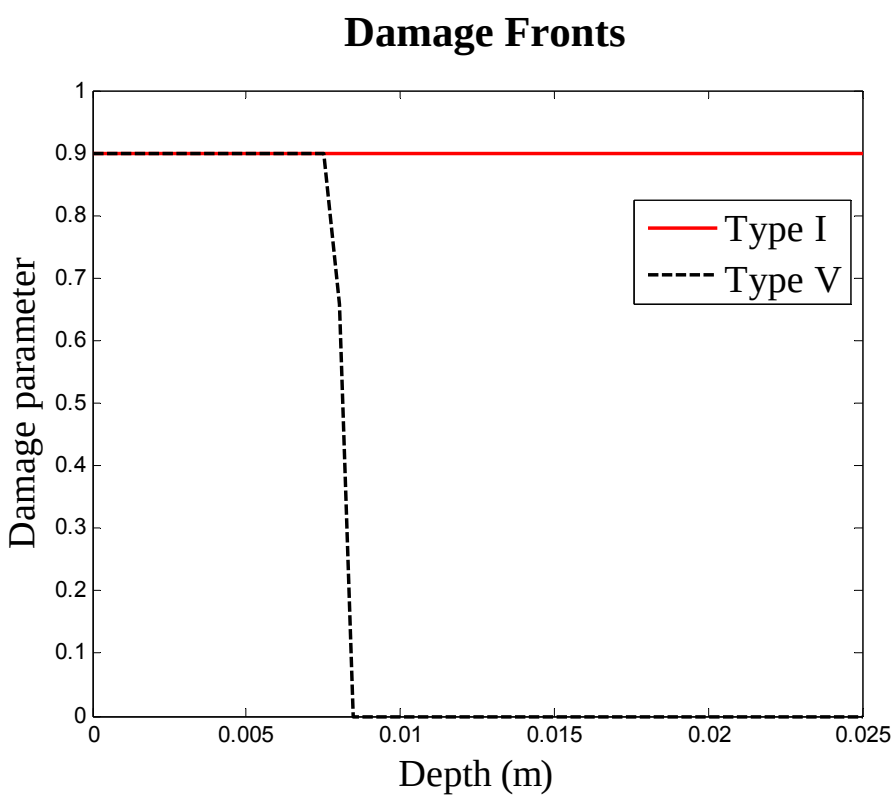


- Elastic moduli of solid phases obtained from the literature



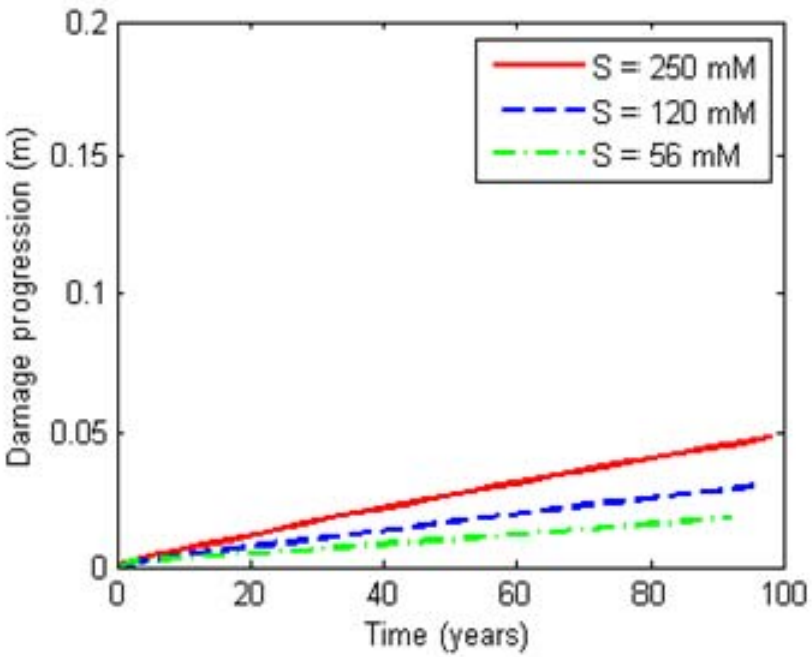
Influence of Cement Type on Damage

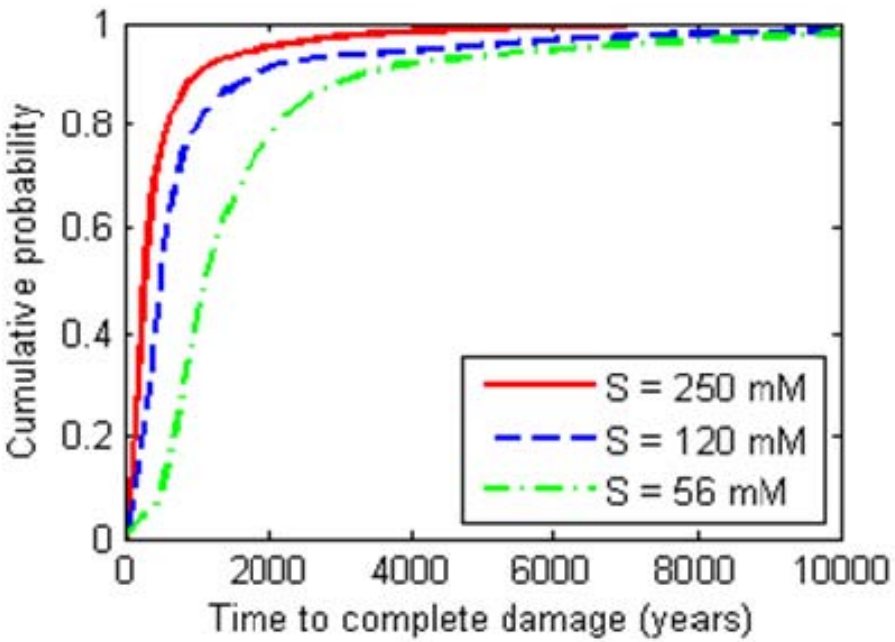




- Damage depends on both ettringite and gypsum formation; primary damage observed from ettringite for Type I and from gypsum for Type V cements.

Sensitivity Analysis – Effects of Sulfate Concentration Boundary Condition on Damage Progression in Example Concrete

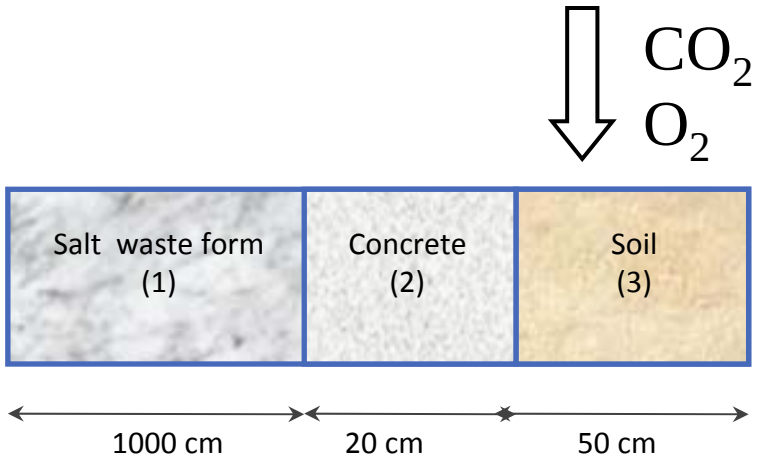




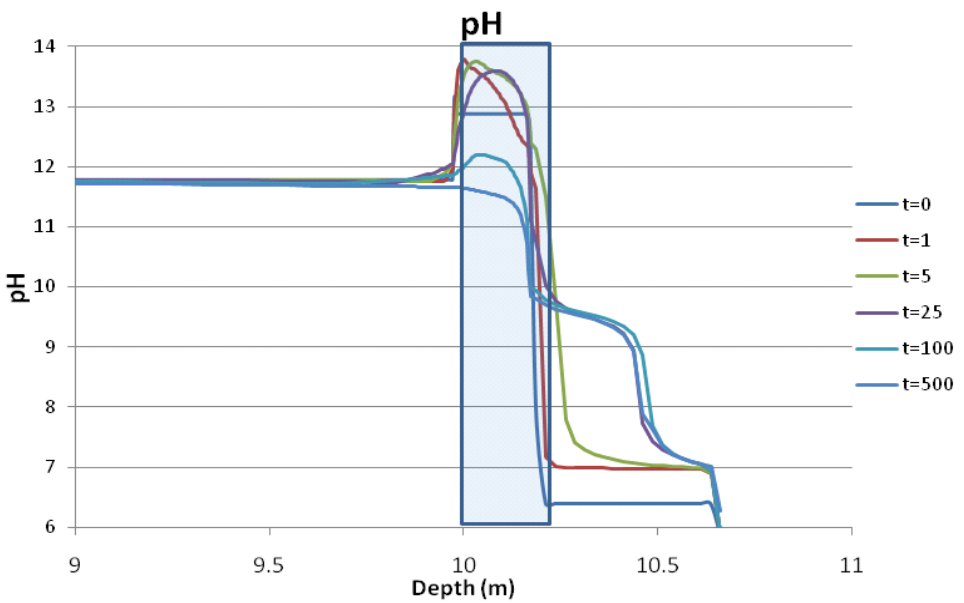
CBP Example Problem
Salt Waste Disposal System Integrity

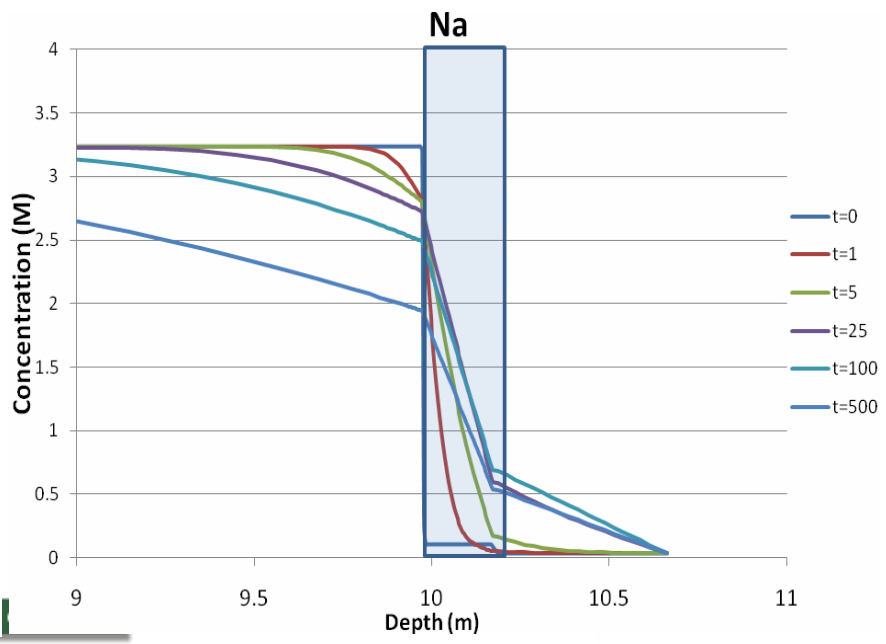
- **Summary of Results for Sulfate Attack**
  - Ability to model sulfate attack and damage as a function of concrete type (cement type, physical properties) and external sulfate concentration
  - Probabilistic analysis for model and parameter uncertainty
  - Resulting models and parameters can be used for evaluation of a range of materials and scenarios
- **Impact**
  - Allows selection of design parameters and materials to insure long-term durability and meeting performance goals
  - Results can be integrated into existing performance assessment fate and transport models

CO_2 and O_2 Ingress Challenge
3-Layer Reference Scenario



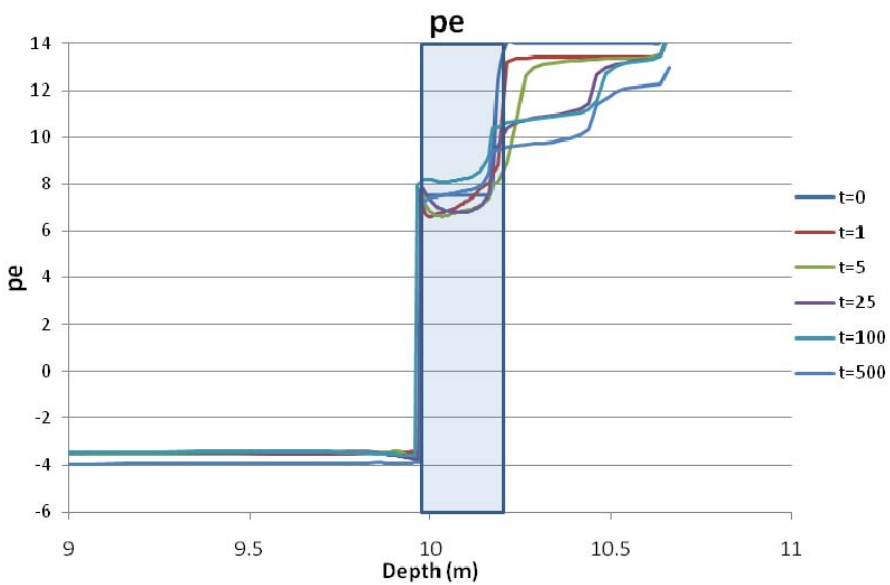
- 3-Layer, 1-D diffusion model for reactive substances
- CO_2 and O_2 influx in soil layer proportional to partial pressure difference air-soil.

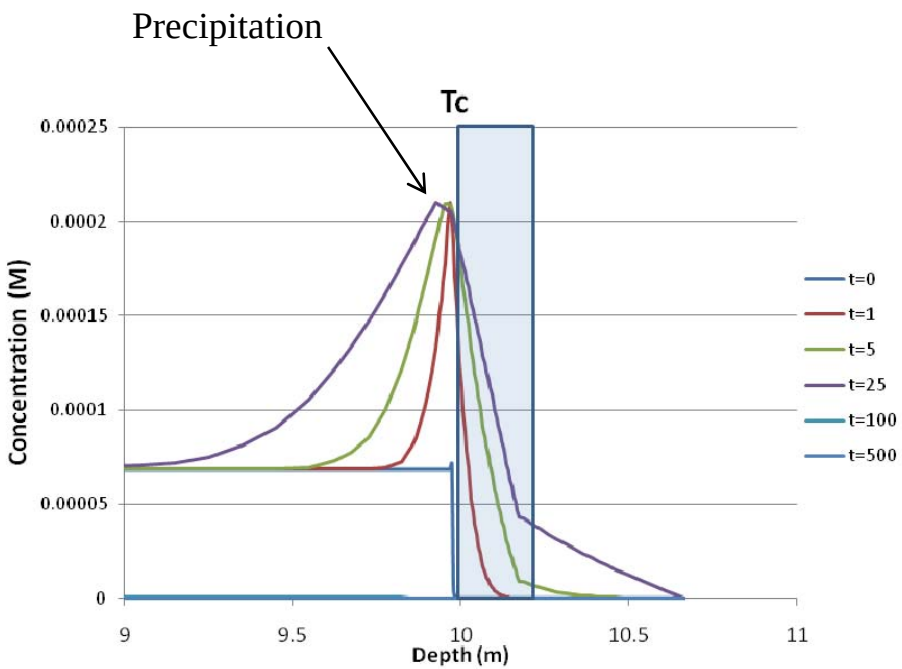




Print Close

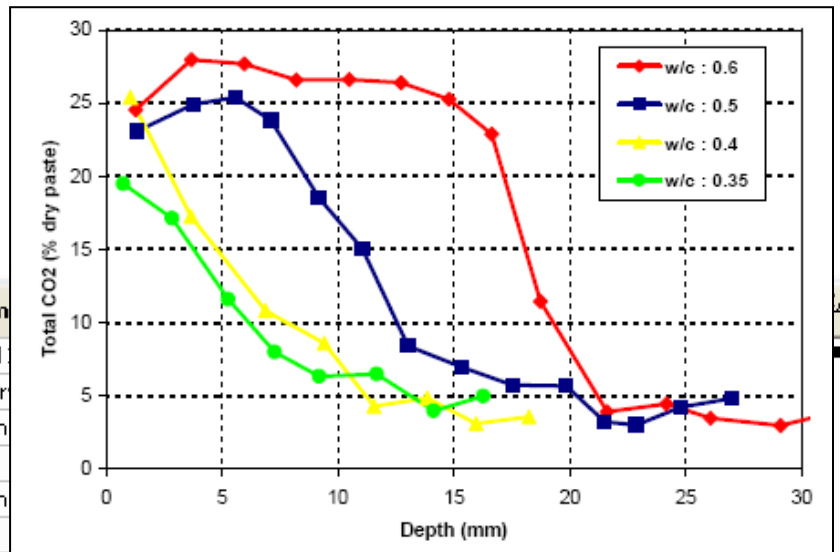
Effect of O_2 Ingress on REDOX Gradient Evolution & Tc-99 Release



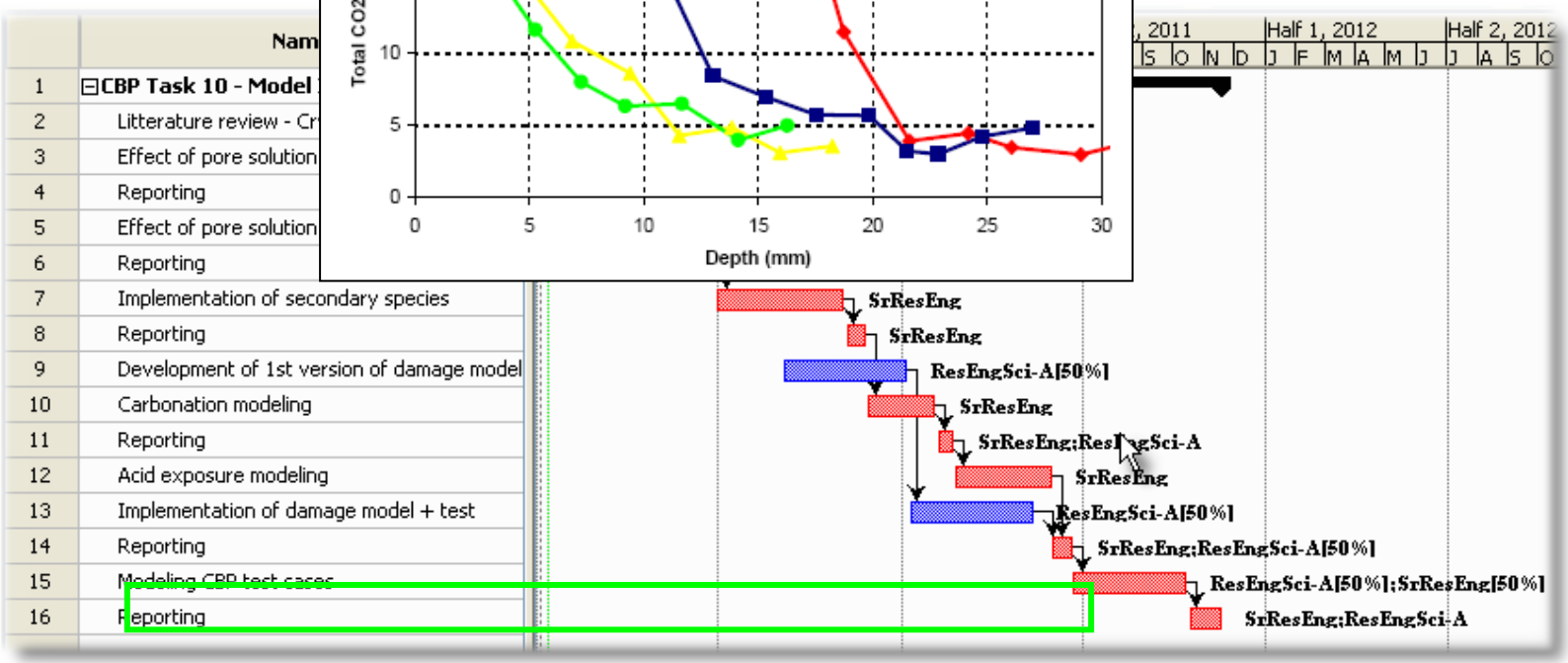


CO_2 Ingress & Carbonation Modeling for Tank Integrity and Closure Scenarios

All CBP Partners Provide Unique Data Sources

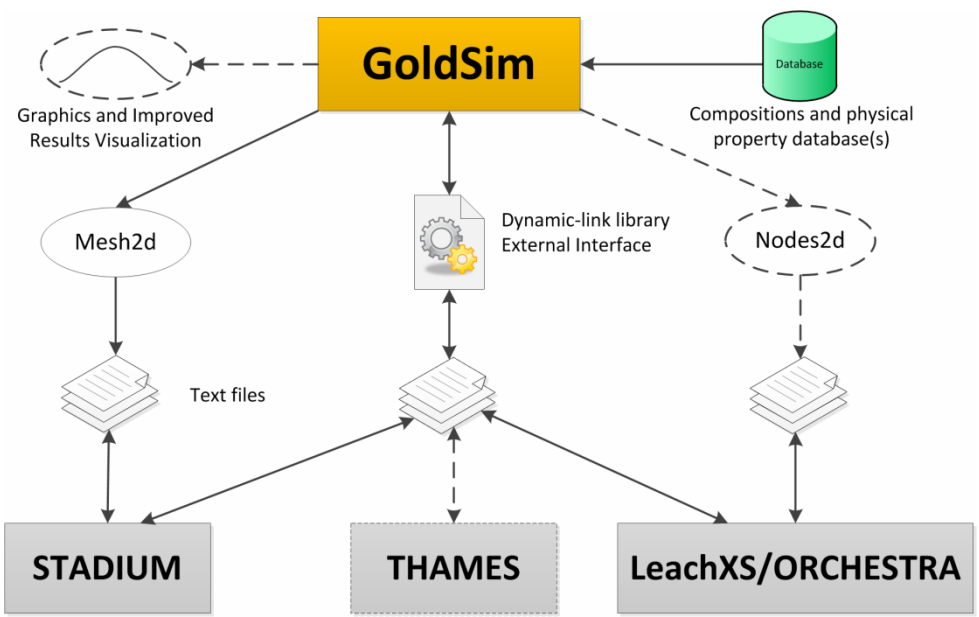


SIMCO Tech., Inc. experimental results for validation



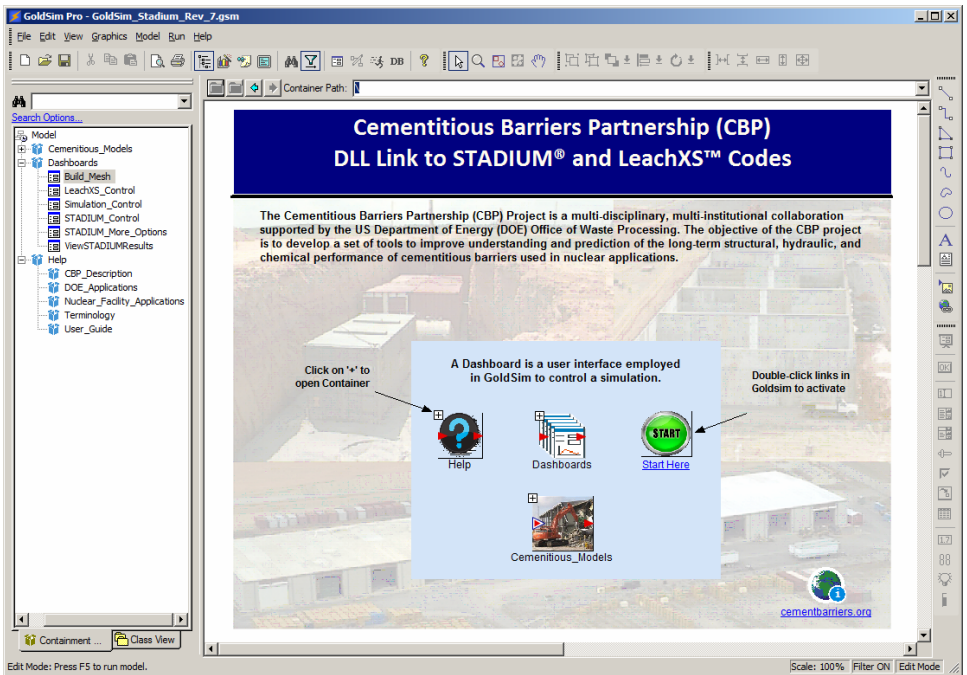
CBP Partner Code Integration

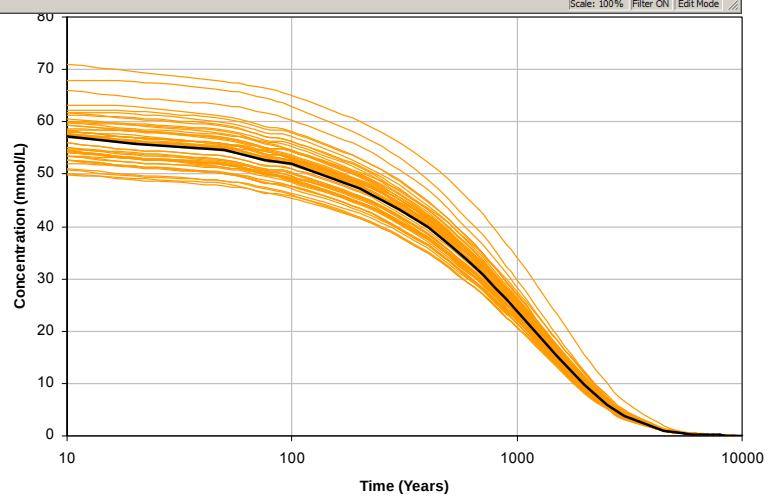
- Software integration objectives:
  - Provide a common unified interface to CBP partner codes through a GoldSim GUI
  - Provide a wrapper for probabilistic uncertainty/sensitivity analysis (e.g., Monte Carlo)
  - Couple LeachXS/ORCHESTRA, STADIUM and THAMES in a synergistic manner
  - Connect to broader systems-level environmental assessment models



CBP Partner Code Integration

- GoldSim graphical user interface to STADIUM and LeachXS/ORCHESTRA
- Implement sub-models for different scenarios
- Demonstration of user interface for deterministic and Monte Carlo analyses
- Integrates with current platforms and system models used by NRC and PA developers





Environmental Management
safety ❖ performance ❖ cleanup ❖ closure

Print Close

CBP Next Steps

- **Integration of CBP models with GoldSim Platform**
  - Demonstrated for STADIUM and LeachXS/ORCHESTRA in FY 2010
  - First licensing (sulfate attack in GoldSim) expected during FY 2011
- **Extension of CBP Models and Data to Other Phenomena**
  - Oxidation front analysis demonstrated during FY 2010
  - Coordination with ASCEM initiated in FY 2010
  - Reference material characterization completed in FY2011
  - Database for evaluation of cementitious materials released (FY 2011)
  - Impact on constituent and radionuclide release (leaching, K_ds) initiated in FY 2010
  - Impact of cracking scenarios initiated in FY 2011
  - Carbonation ingress analysis planned to be initiated in FY 2011 (e.g., tank integrity & closure scenarios)
- **Exposure Testing for Model Confirmation**

Acknowledgements

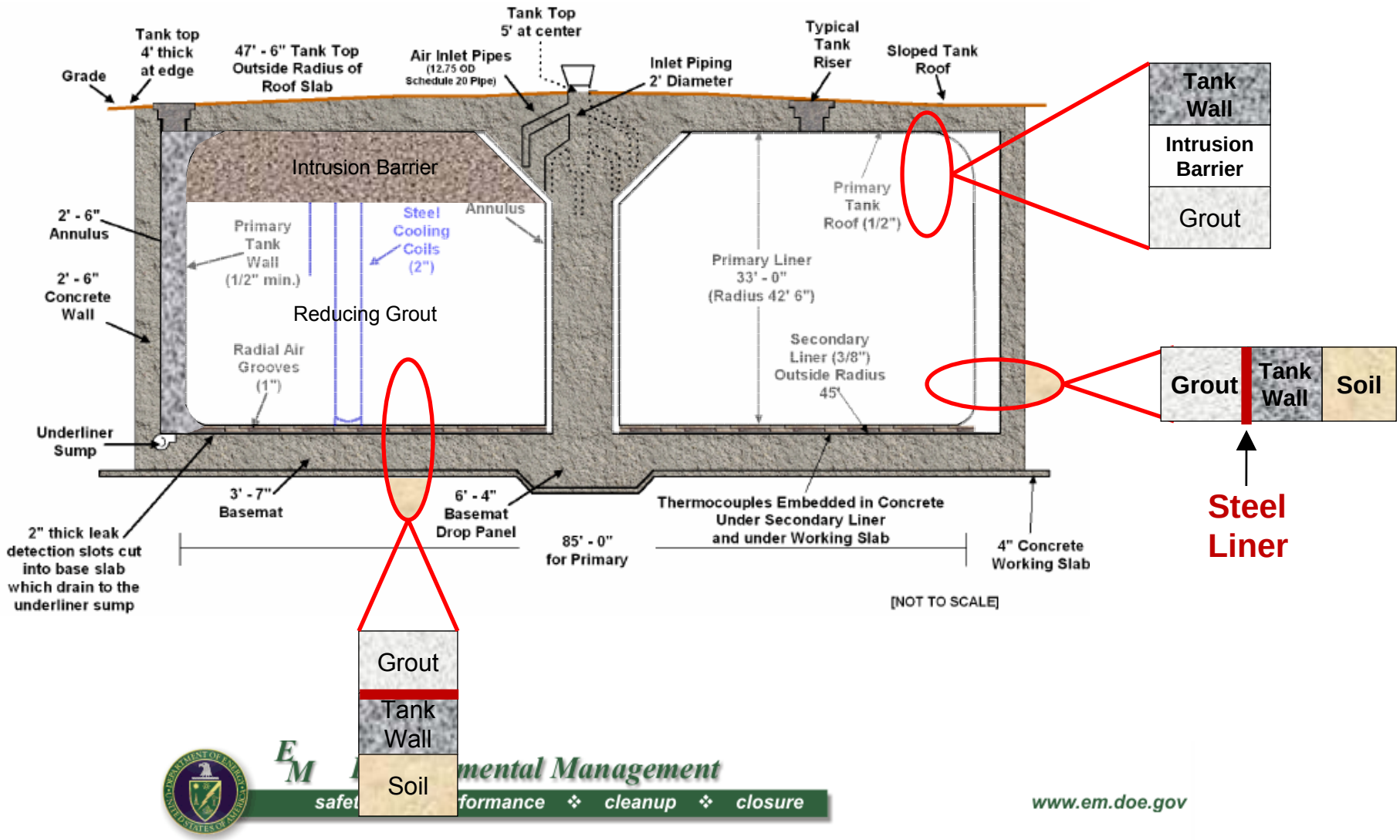
D. Esh, M. Furman, J. Phillip, US NRC
S. Mahadevan, A. Garrabrants, J. Arnold, S. Sarkar, CRESP/Vanderbilt U.
H. Van der Sloot, J.C.L. Meeussen, R. Comans, ECN (NL)
E. Garboczi, K. Snyder, J. Bullard, P. Stutzman, NIST
E. Samson, J. Marchand, SIMCO, Inc. (CA)
R. Seitz, S. Marra, H. Burns, SRNL

DOE-EM Project Manager: Pramod Mallick
CBP Project Support Provided by EM-30

Contact Information
SRNL : Christine Langton (803) 725-5806
Greg Flach (803) 725-5195
CRESP: David Kosson (615) 337-5889
Kevin G. Brown (615) 343-0391
www.cementbarriers.org

Supporting Overheads

Type IIIA Tank – Conceptual Closure Model



Print

Close