PROBABILISTIC DURABILITY ANALYSIS OF CEMENTITIOUS MATERIALS UNDER EXTERNAL SULFATE ATTACK

S. Sarkar[1], S. Mahadevan[1], J.C.L. Meeussen[2], H. van der Sloot[2], D. S. Kosson[1]

[1]Vanderbilt University

[2]Energy Research Centre of The Netherlands

Consortium for Risk Evaluation with Stakeholder Participation

CRESP III

DOE-EM
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Presentation Outline

- Motivation
- Numerical Model Framework
- Model Calibration and Validation
- Example of Deterministic Durability Analysis
- Examples of Sensitivity Analysis
- Sources of Uncertainty
- Uncertainty Quantification
- Example of Probabilistic Durability Analysis
- Conclusion

Motivation

Current DOE approach

- No tool available for assessment of progressive damage of the containment structures for low level nuclear wastes
- Undamaged to complete damaged state at selected/assumed times

→ **Need for a mechanistic model**

  - Able to predict rate of degradation of a particular structure under specific boundary conditions
  - Needs to be calibrated and validated

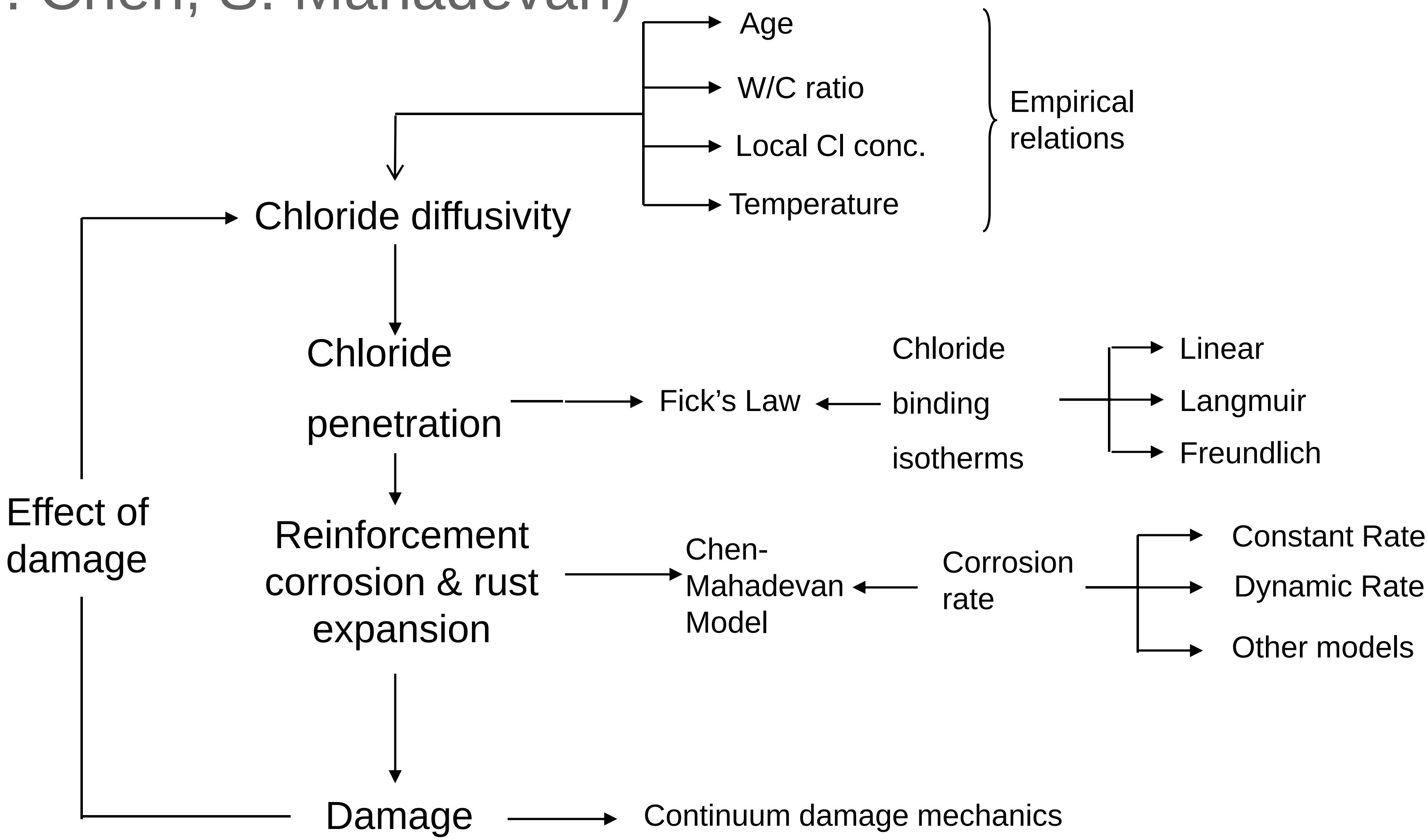
Important degradation phenomena

- Chloride attack : reinforcement corrosion, cracking
- Sulfate attack : expansive product formation, cracking
- Carbonation : reinforcement corrosion
- Leaching: loss of strength

Examples of Concrete Degradation

Courtesy of F. Sanchez

Previous Work on Chloride Attack
(D. Chen, S. Mahadevan)

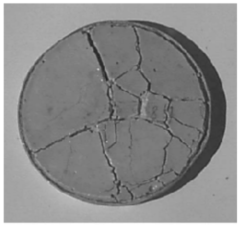


Multi-physics analysis implemented in ANSYS finite element software

Sulfate Attack on Cementitious Materials

Effects of Sulfate Attack

- Expansion
  - Gypsum formation
  - Ettringite Formation
- Loss of Strength
  - Cracking
  - CSH deterioration



Cracking

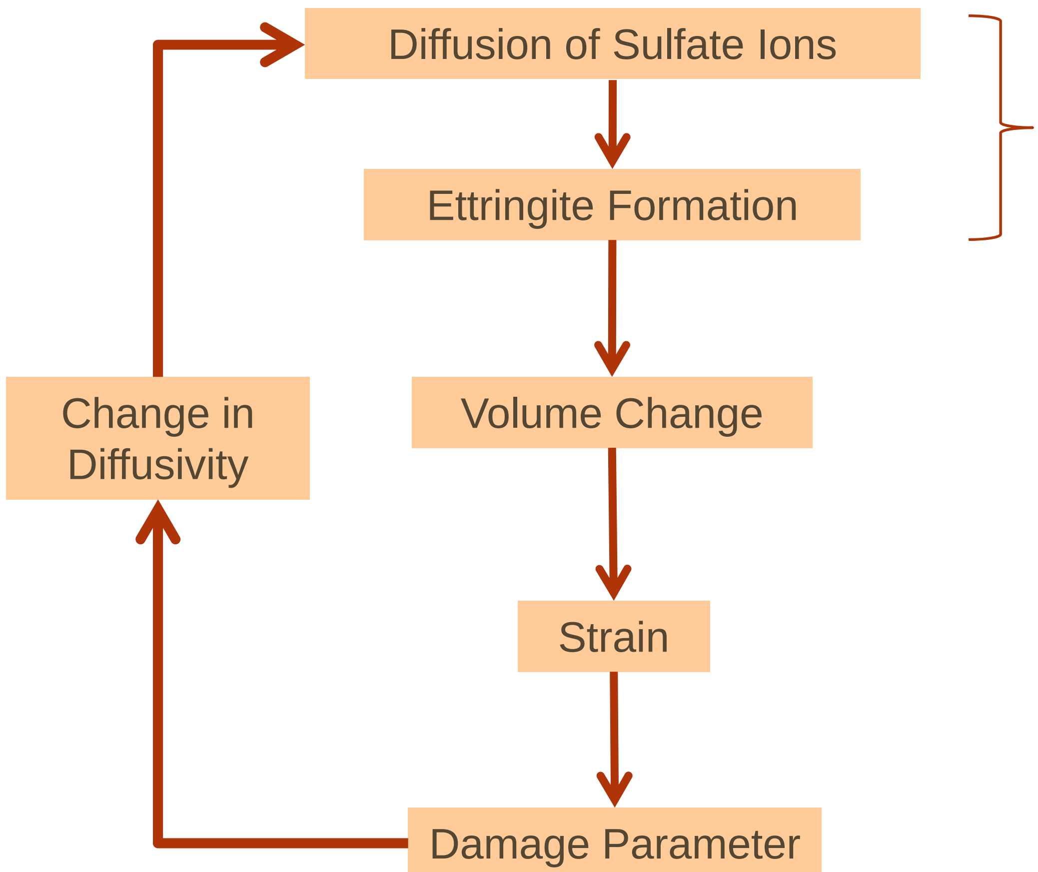
Spalling

Objectives

- Develop a numerical model to assess response of the structure (durability and contaminant release) under sulfate attack
- Develop probabilistic framework to incorporate various sources of uncertainty in durability analysis

Components of Past Numerical Models

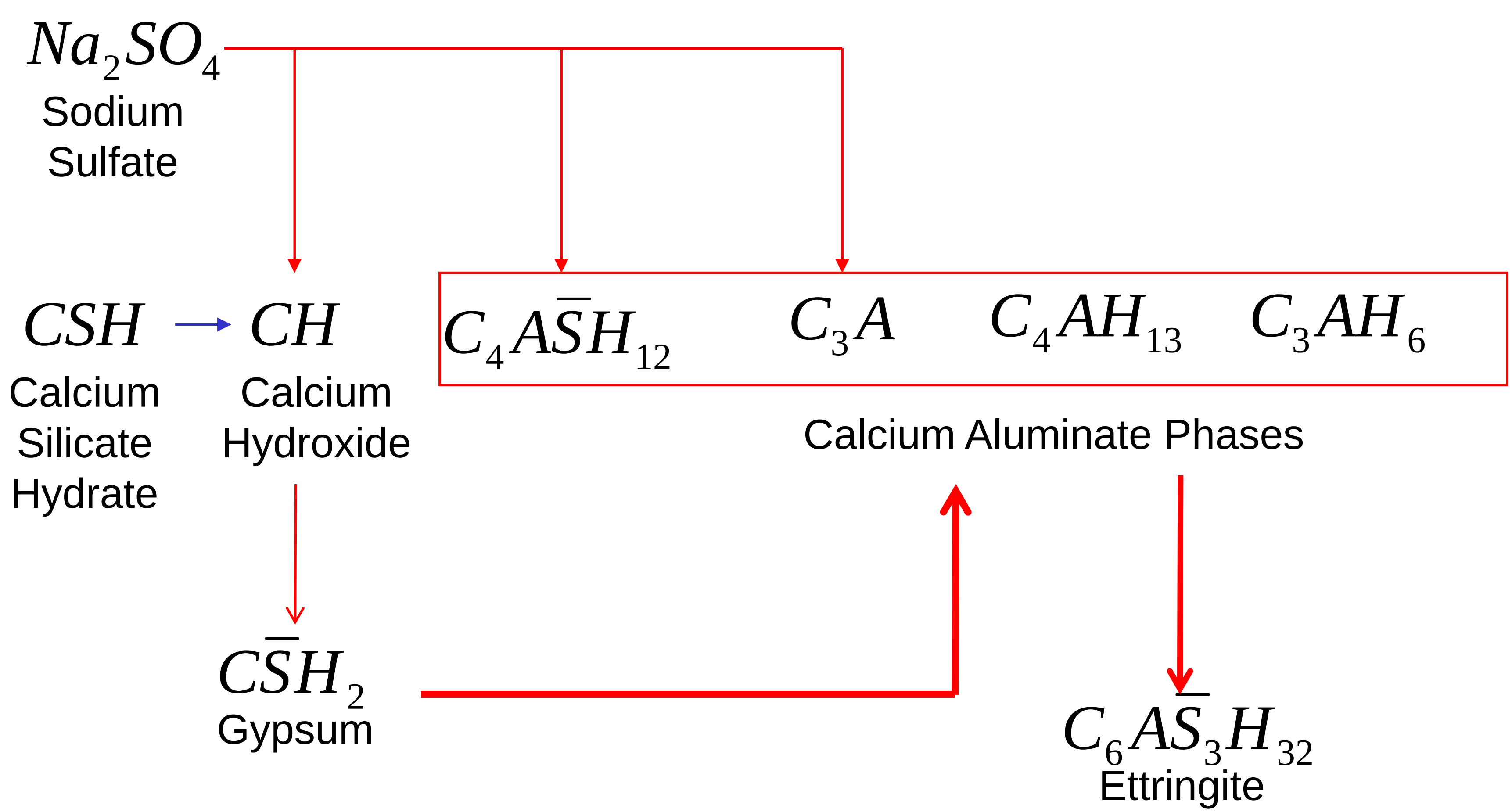
(Krajcinovic *et al.*, 1992, Tixier and Mobasher, 2003, Basista and Weglewski, 2008)



Need to improve diffusion and chemical reaction models

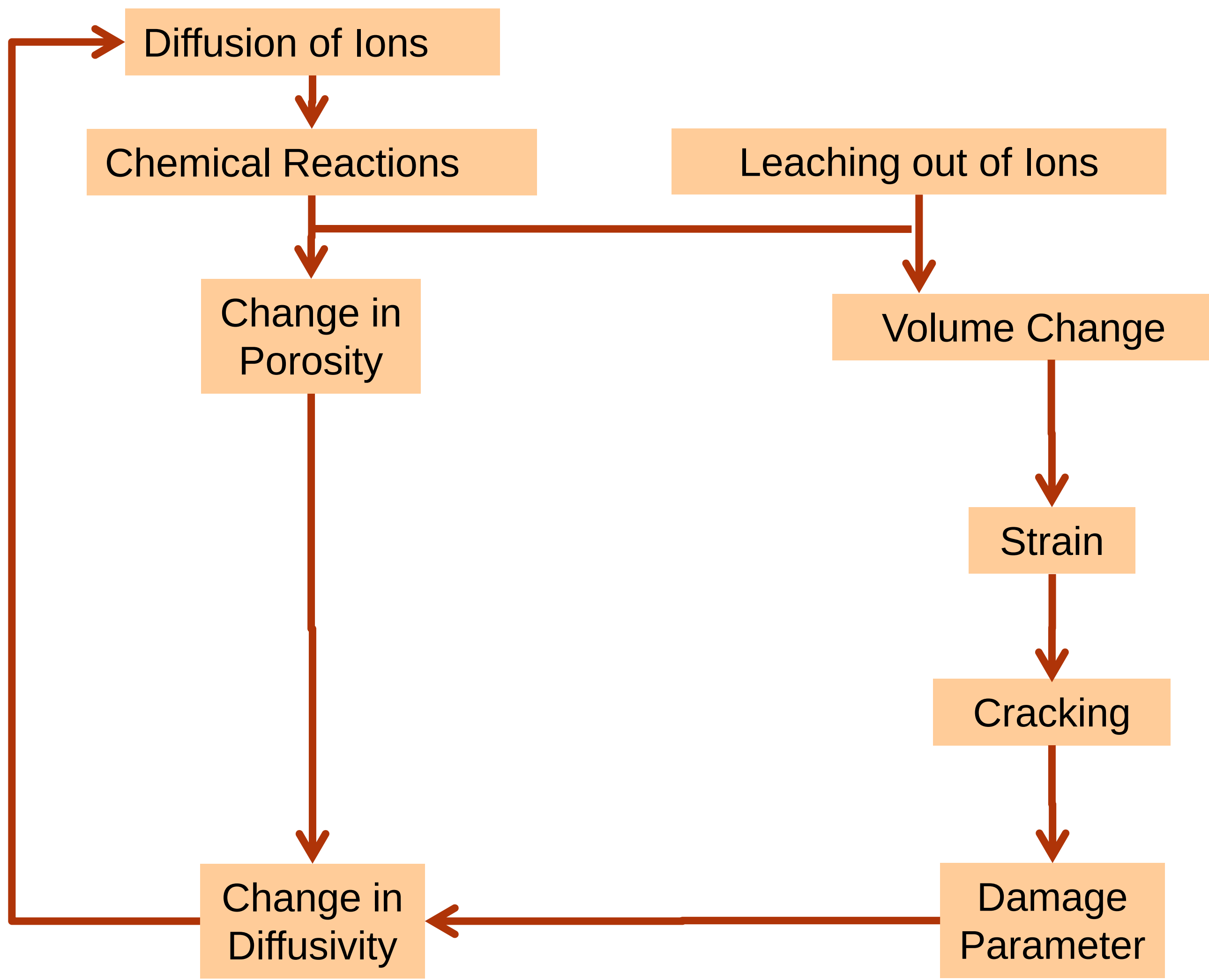
Samson *et al.*, 1999-2009: Detailed diffusion and chemical reaction model combined with effect of porosity changes on diffusion; but not structural damage due to cracking

Example of Chemical Reactions due to Sulfate Attack



Effects : expansion, cracking and strength loss

Numerical Model Framework



Diffusion and Chemical Reactions

- Governing Equation for Diffusion

(saturated porous material under isothermal condition)

Modified Davies Equation (Samson *et al.*, 1999)

$$\frac{\partial(\varphi c)}{\partial t} = \frac{\partial}{\partial x}\left(\frac{D_0 \varphi}{\tau}\left(\underbrace{\frac{\partial c}{\partial x}}_{\text{concentration gradient}} + \underbrace{c\frac{\partial \ln \gamma}{\partial x}}_{\text{chemical activity gradient}}\right)\right)$$

- Chemical Reactions
  - Available quantities of ions: pHStat test results (LeachXS database)
  - Potential solid phases: Identified by comparing results of pH-dependent leaching tests and simulations with different solid phase mineral sets using ORCHESTRA (by ECN)
  - Calculation of liquid-solid equilibrium and solid phase distribution using ORCHESTRA

Strain Development and Change in Porosity and Tortuosity

- Volume Change

$$\overline{\Delta V_s} = (V_{\text{products}} - V_{\text{reactants}}) - b\varphi$$

Fraction of porosity available (Tixier and Mobasher, 2003)

- Strain

(homogeneous and isotropic material)

$$\varepsilon = \frac{\Delta \overline{V_s}}{3}$$

- Porosity Change

$$\varphi_{\text{new}} = \varphi_{\text{original}} - (V_{\text{products}} - V_{\text{reactants}})$$

- Tortuosity change (Samson *et al.*, 2007)

$$\tau_{\text{new}} = \tau_{\text{original}} \exp((\varphi_{\text{original}} - \varphi_{\text{new}}) * 4.3 / \text{paste volume})$$

Damage Accumulation

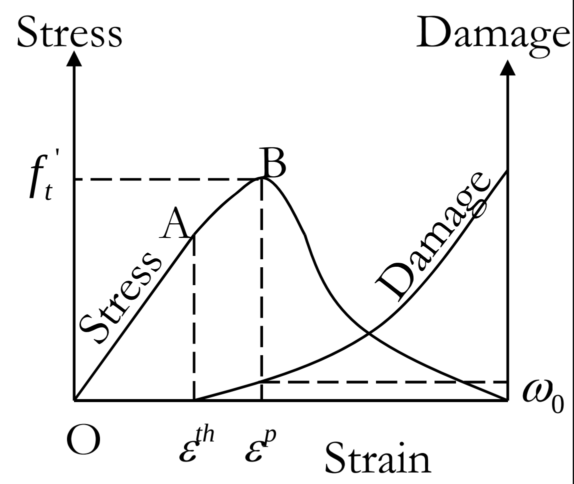
- **Nonlinear Ascending Region**

(Karihaloo, 1995, Budiansky and O'Connell, 1976)

Empirical relation between crack density and strain

$$C_d = k\left(1 - \frac{\varepsilon^{th}}{\varepsilon}\right)^m$$

k, m calibrated from stress-strain diagram



Damage parameter

$$\omega \approx \frac{16}{9} C_d$$

- **Nonlinear Descending Region**

(Nemat-Nasser and Hori, 1993)

Fracture Mechanics

$$\frac{\sigma}{f_t^{'}} = \sqrt{\frac{\tan(\pi\omega_0/2)}{\tan(\pi\omega/2)}} \text{ and } \frac{w}{w_0} = \frac{\sigma}{f_t^{'}}\left(\frac{\log(\sec(\pi\omega/2))}{\log(\sec(\pi\omega_0/2))}\right) - 1$$

Change in Material Properties

- **Mean Field Regime** (dilute concentration of cracks)

Assumption: randomly oriented penny-shaped cracks scattered in a homogeneous matrix (Salganik, 1974)

$$D = \frac{D_0}{\tau}\left(1 + \frac{32}{9} C_d\right)$$

Assumption: Linear relations (Krajcinovic *et al.*, 1992)

$$E = E_0\left(1 - \frac{16}{9} C_d\right) \text{ and } \nu = \nu_0\left(1 - \frac{16}{9} C_d\right)$$

- **Percolation Regime** (spanning cluster of cracks and macro-cracks)

(Stauffer, 1985 and Krajcinovic *et al*., 1992)

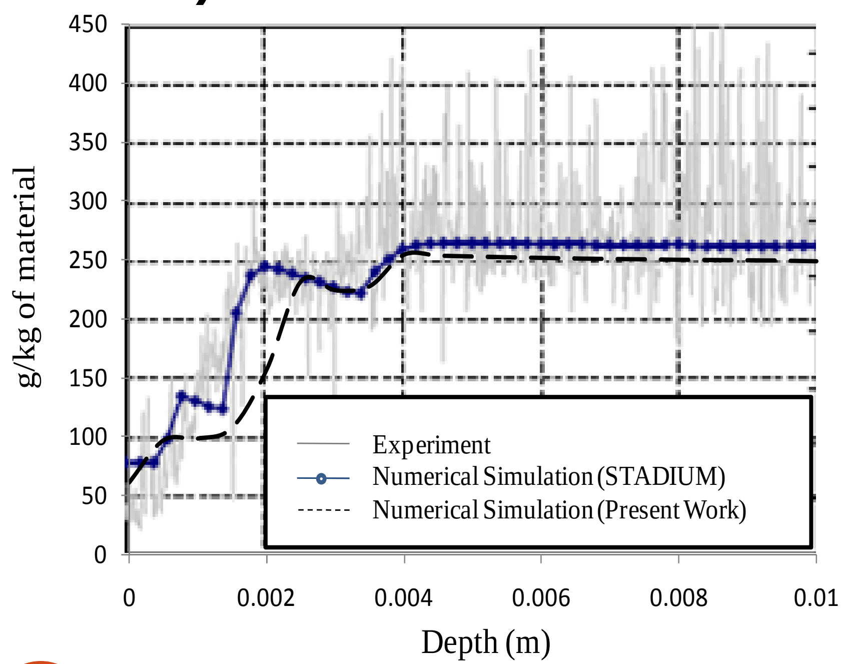
$$D = \frac{D_0}{\tau}\left(1 + \frac{32}{9} C_d\right) + \frac{D_0}{\tau}\frac{\left(C_d - C_{dc}\right)^2}{\left(C_{dec} - C_d\right)}$$

- Relation between elastic moduli and damage still needs investigation
- Mean-field regime relations assumed by Krajcinovic *et al*., 1992

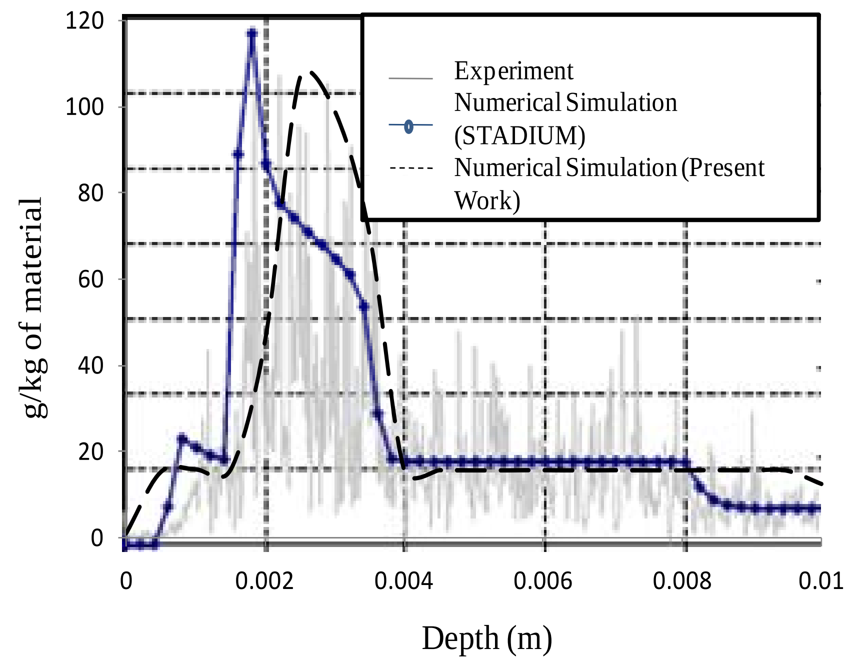
Model Calibration and Validation

- 7 cm x 20 mm CSA type 10 cement paste sample
- 50 mmol/L of Na_2SO_4 solution in 30 L tank
- External solution pH : 10.3
- 7 day renewal of solution
- Only one face exposed
- Porosity : 0.52
- Calibration parameter: tortuosity (= 18) and b (= 0.3)

Model calibrated with experimental results after 3 months and validated against experimental results after 1 year (Samson et al., 2007)

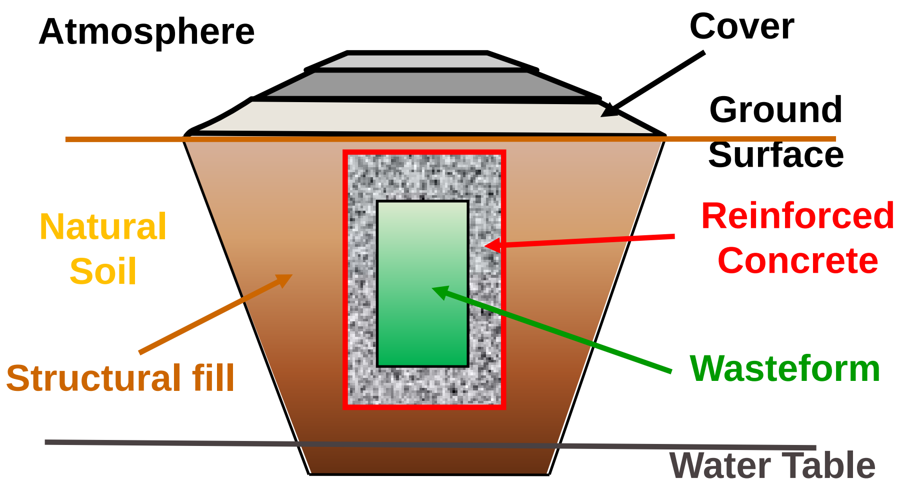


Calcium Profile after 1 year

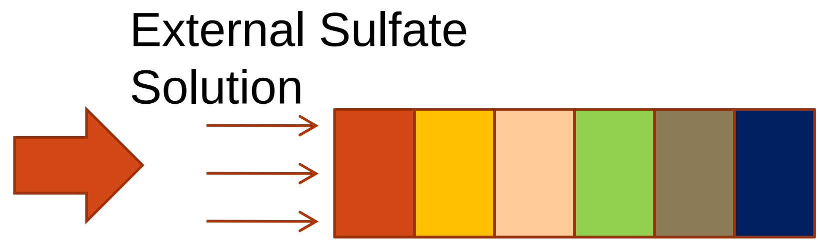


Sulfur Profile after 1 year

Example of Deterministic Durability Analysis



Low Level Waste Disposal



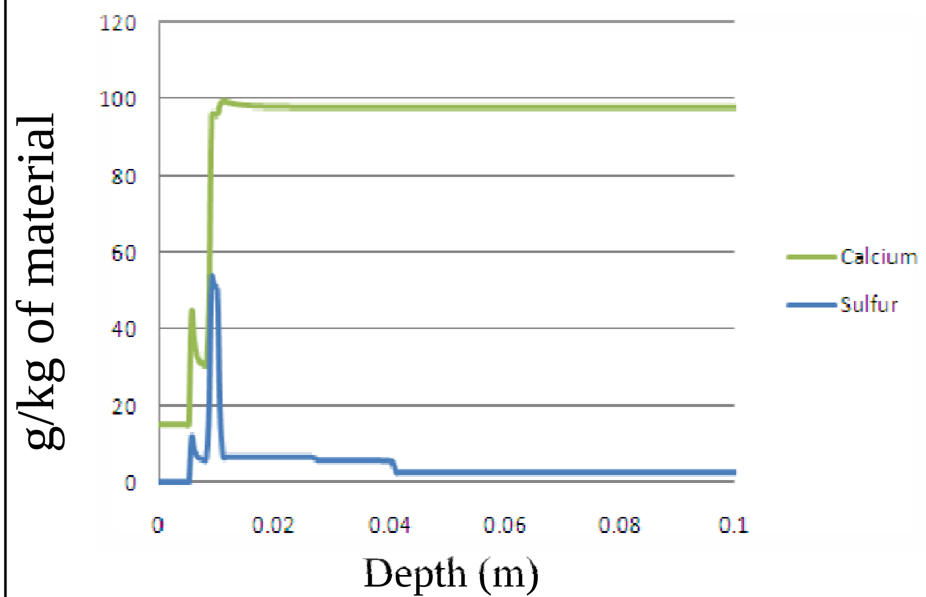
1D Idealization of vault wall

Model Specifications

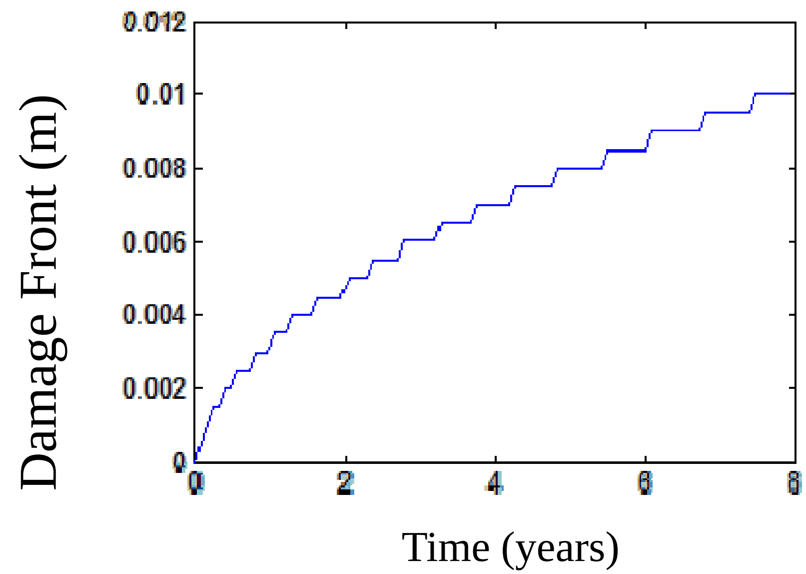
- US Type I cement with w:c:s mass ratio 0.5:1:3
- 0.35 M sodium sulfate solution (constant boundary condition)
- Length of the structure 20 cm (divided into 200 nodes with varying mesh size)
- Porosity 0.15, Tortuosity 50, fraction of available porosity 0.5
- Simulation performed for 8 years

Example of Deterministic Durability Analysis

Calcium and Sulfur Profiles

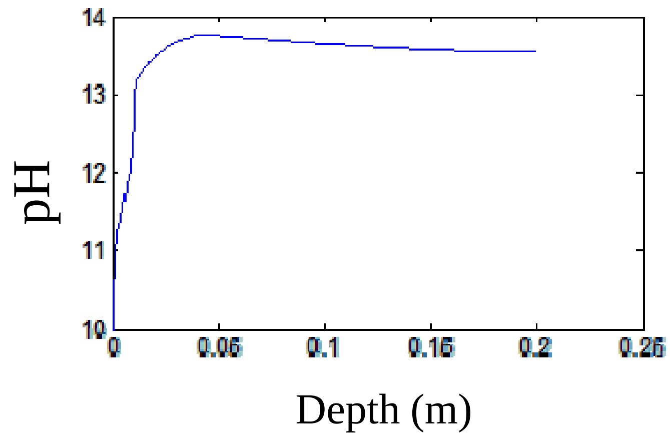


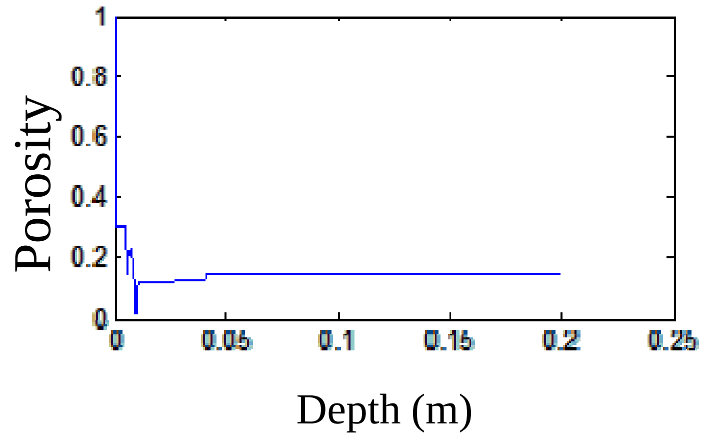
Damage Front Progression

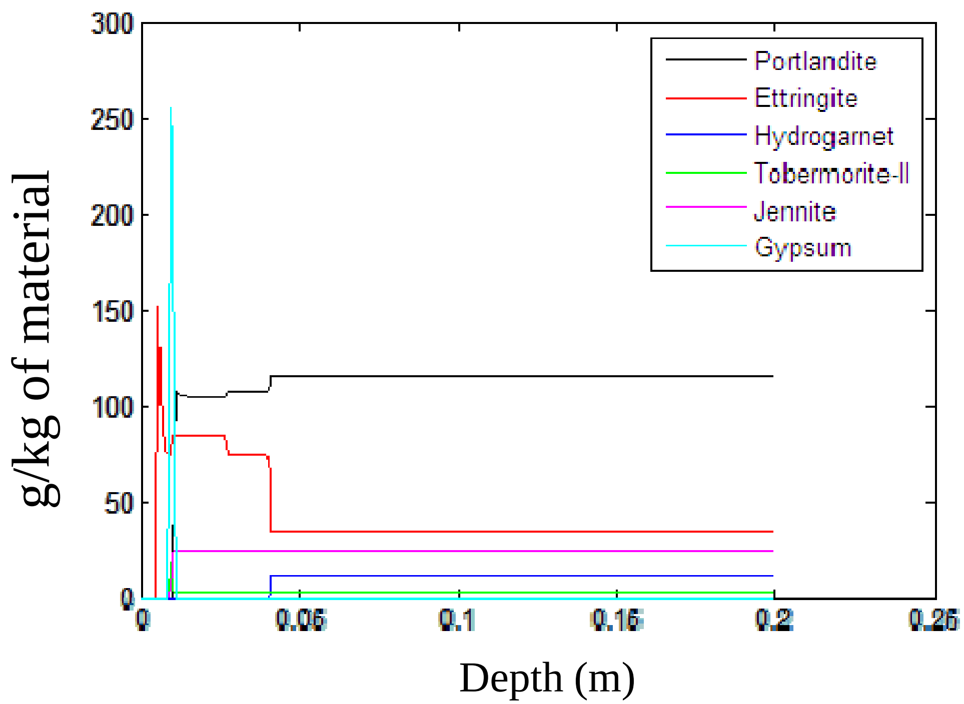


- Calcium leaching prominent near the boundary
- Gypsum formation front prominent as a large sulfur peak
- Damage rate progression nonlinear process
- Approximate rate of progression: 0.0013 m/year

Example of Deterministic Durability Analysis



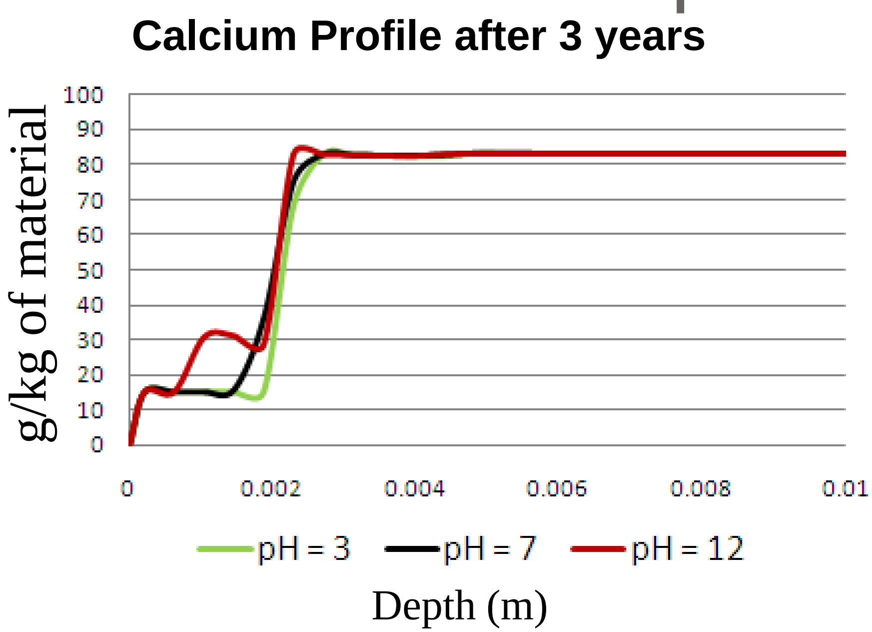


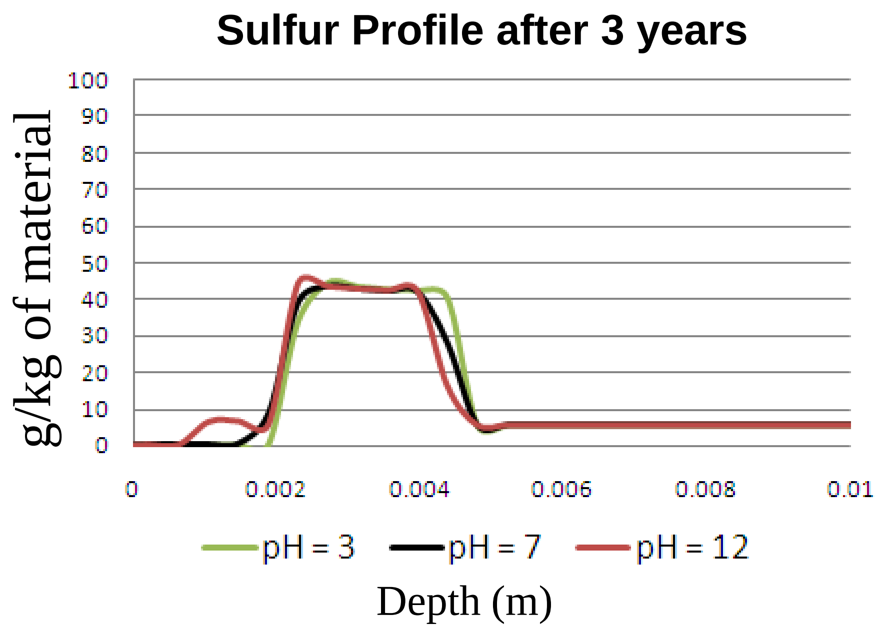


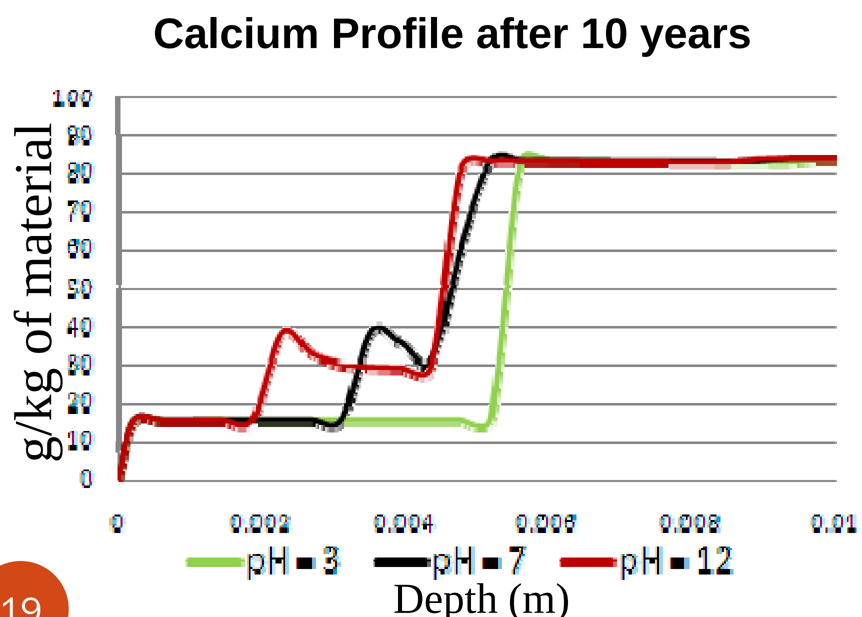
Examples of Sensitivity Analysis

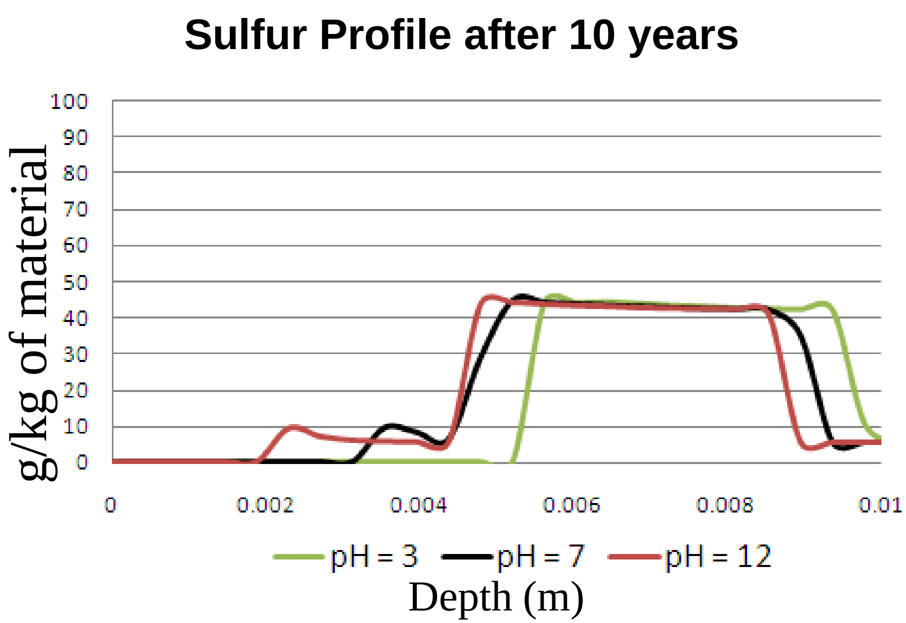
- Factors considered
  - External solution – pH
  - Structure – initial porosity & tortuosity, cement type
- Simulation details
  - 25 mm x 25 mm x 285 mm US type I sample, all faces exposed
  - 350 mmol/L Na_2SO_4 external solution
  - External solution pH : 7
  - 7 day renewal of solution
  - Liquid to solid volume ratio : 10
  - Porosity : 0.3
  - Fraction of available porosity : 0.5
  - Tortuosity : 36
  - Cement : water : sand (mass ratio) = 1 : 0.5 : 3

External Solution pH

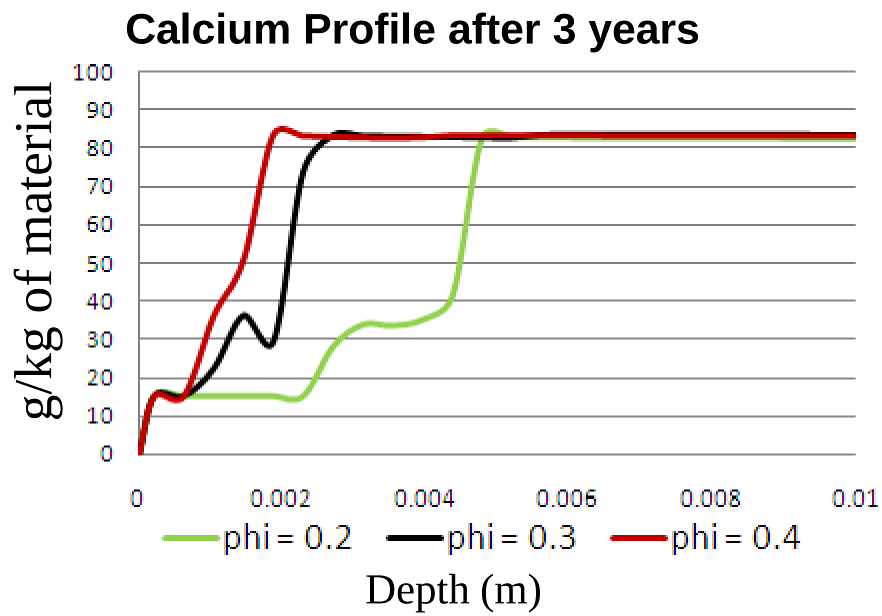


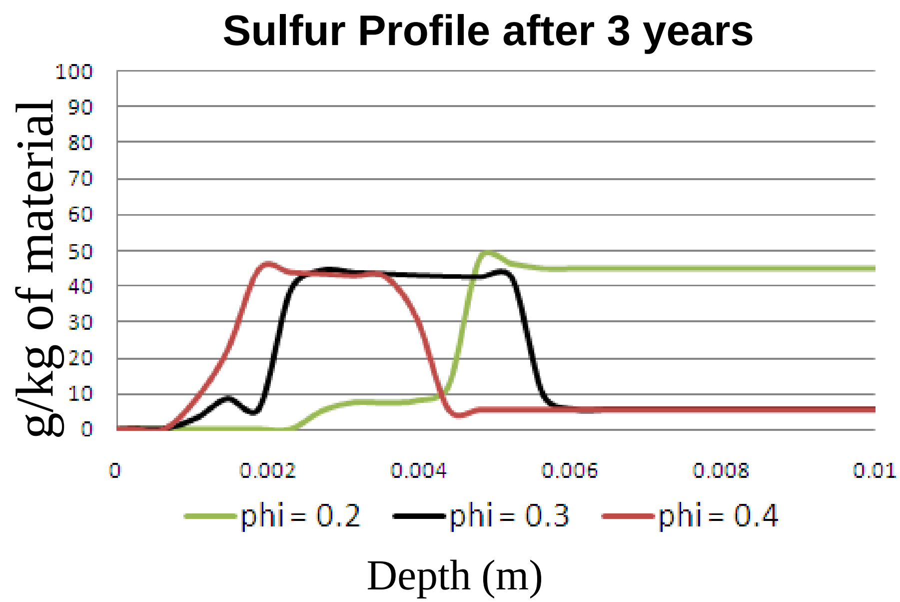


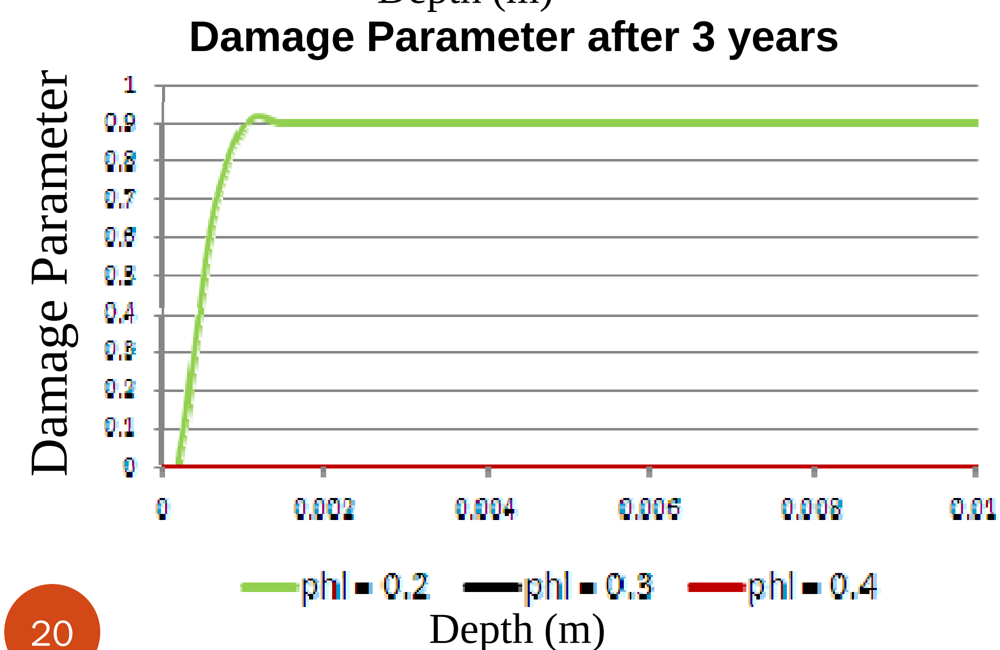




Initial Porosity

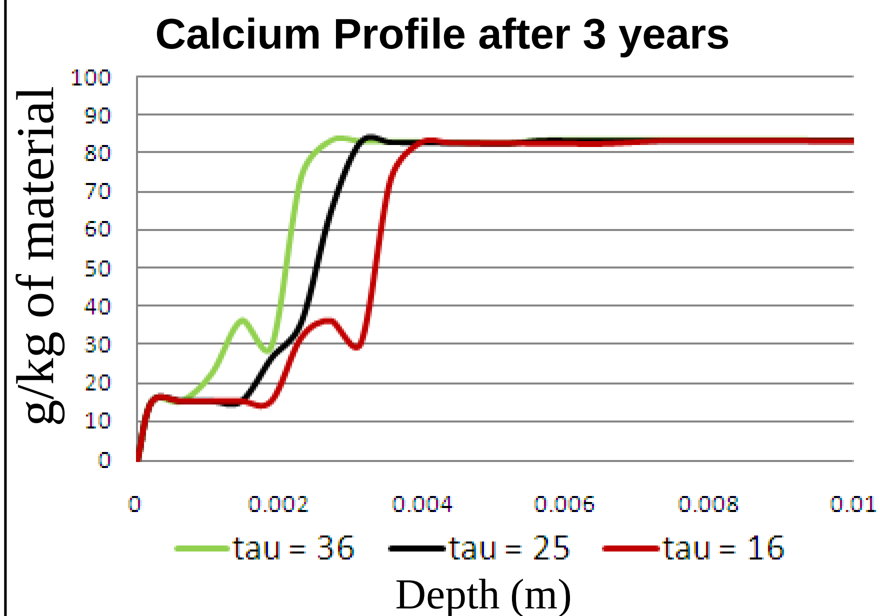


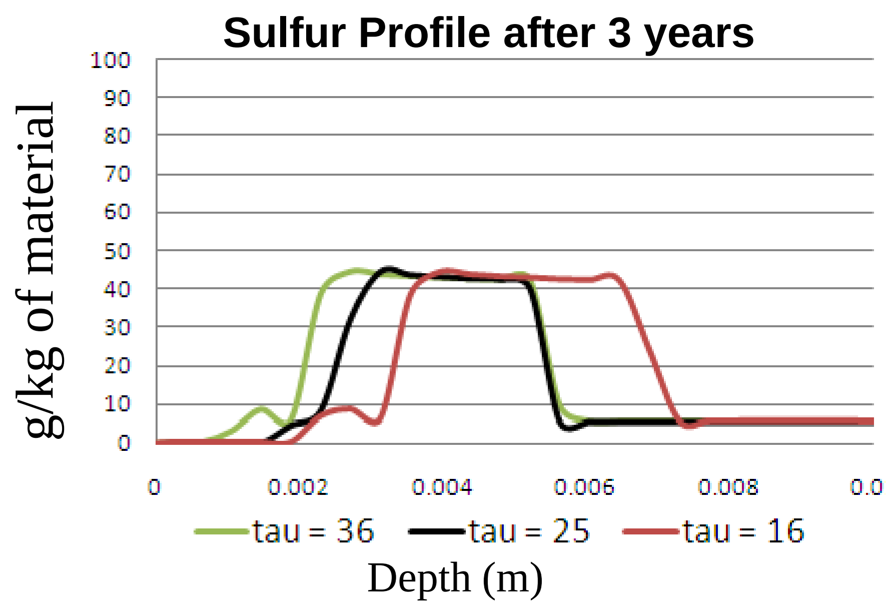




Damage increases if porosity decreases (all other factors remaining constant)

Initial Tortuosity

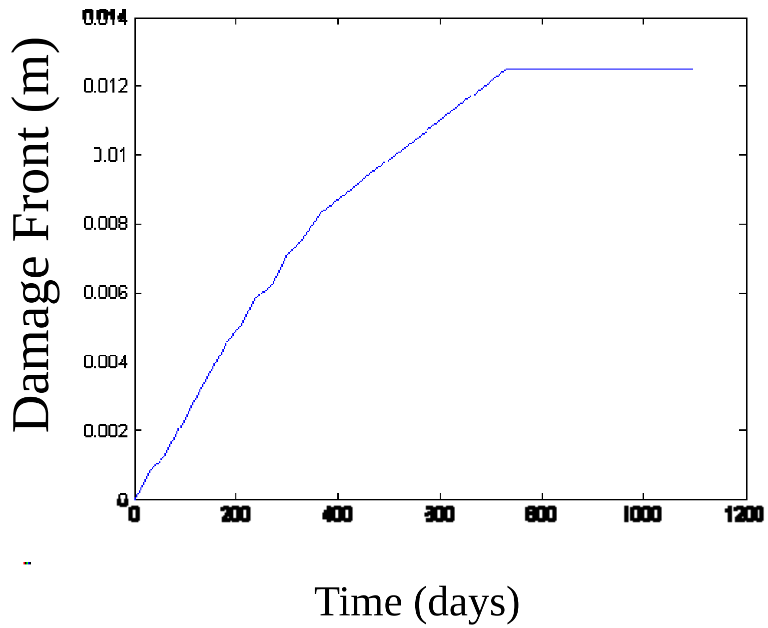




Tortuosity affects the rate at which mineralogical features will change as it affects the diffusion of the ions. If the path is more tortuous, ions will take more time to move from one point in the structure to another.

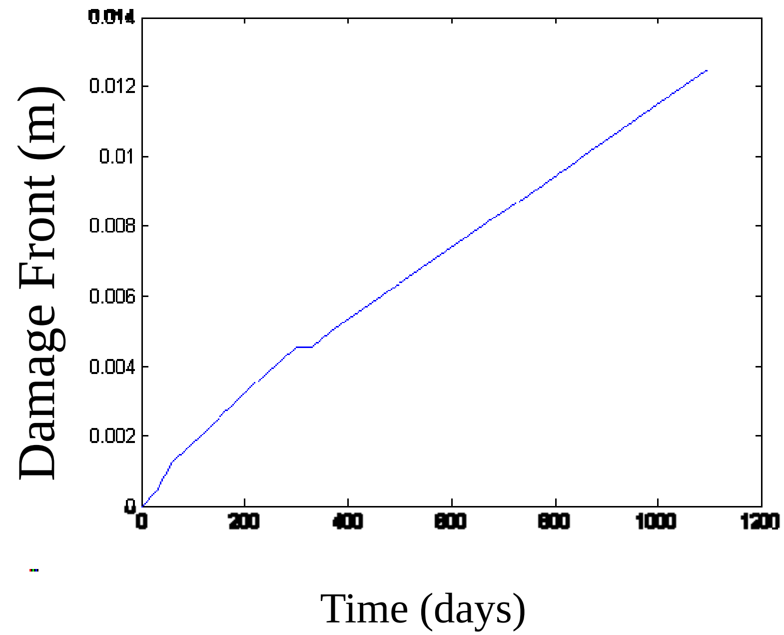
Cement Type

Cement Type N1



Hydrogarnet = 0.0424 moles/m^3

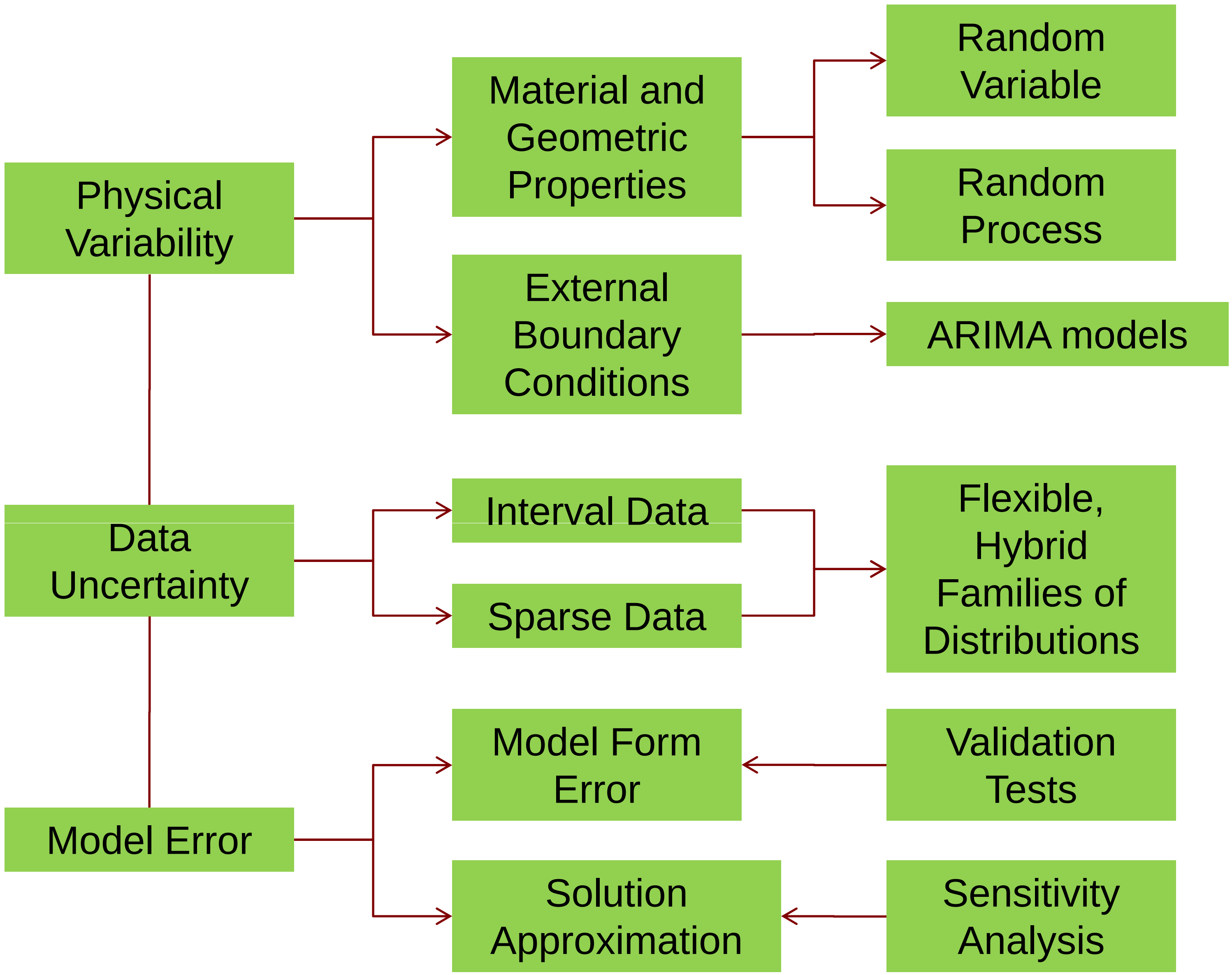
Cement Type D1



Hydrogarnet = 0.0116 moles/m^3

- **Fraction of available porosity is assumed to be 0.2 to evaluate the rate of damage progression**
- **Damage increases with increase in Calcium Aluminate content**

Sources of Uncertainty



Uncertainty Quantification

Method 1

Repeat N_2 times

Generate samples for parameters having Data Uncertainty

↓

Repeat N_1 times

Generate samples for parameters having Physical Variability

↓

Perform simulation

↓

Generate sample for Model Error

↓

Durability Assessment

↓

Distribution of Durability

↓

Confidence Bounds on Durability Curve

Uncertainty Quantification

Method 2

Repeat N times

Generate samples for parameters having Data Uncertainty

↓

Generate samples for parameters having Physical Variability

↓

Perform simulation

↓

Generate sample for Model Error

↓

Durability Assessment

↓

Distribution of Durability

Model Error Quantification

Sources

- Input data measurement (D1) , e.g., initial porosity
- Output data measurement (D2), e.g., experimental verification data
- Discretization – time and space (T and S) : Richardson extrapolation
- Uncertainty quantification method (U) : Sampling method and truncation of response surface
- Model form (M) : Need to quantify using experimental observations

Model form error

$$\varepsilon_{\mathrm{obs}} = y_{\mathrm{pred}} - y_{\mathrm{obs}}$$

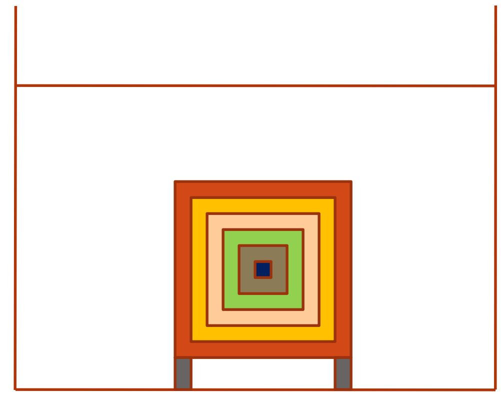
$$= f(\varepsilon_{\mathrm{D}_1}, \varepsilon_{\mathrm{T}}, \varepsilon_{\mathrm{S}}, \varepsilon_{\mathrm{U}}) + \varepsilon_{\mathrm{M}} - \varepsilon_{\mathrm{D}_2}$$

$$\Rightarrow \varepsilon_{\mathrm{M}} = \varepsilon_{\mathrm{obs}} + \varepsilon_{D_2} - f(\varepsilon_{D_1}, \varepsilon_T, \varepsilon_S, \varepsilon_U)$$

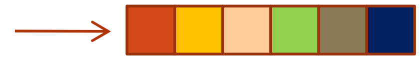
Example of Durability Analysis

Problem Description

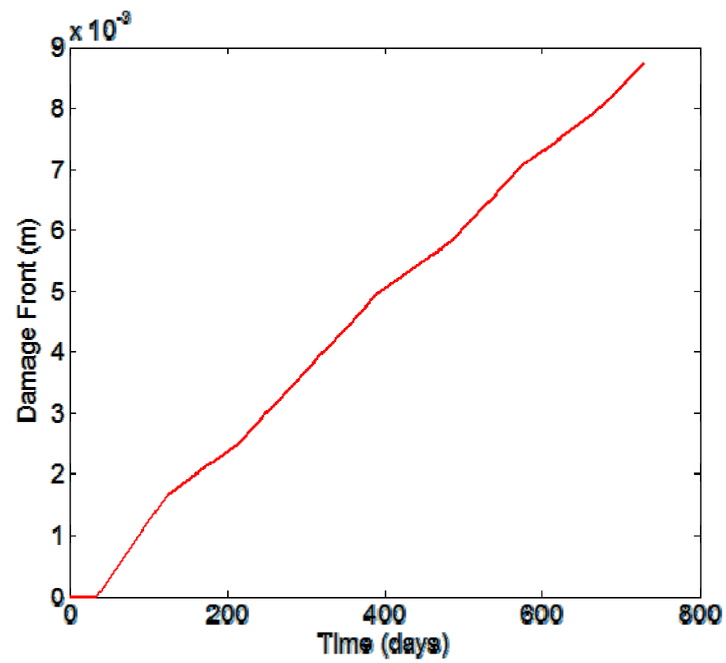
- US type I cement mortar sample (25mm x 25mm x 285mm) with cement, water and sand mass ratio 1:0.5:3
- Na_2SO_4 solution to sample volume ratio 10
- Simulation performed for 2 years
- Uncertainty quantification
    - Physical variability
        - Initial porosity, tortuosity, pH and concentration of external solution and renewal rate of the solution
    - Data uncertainty
        - Fraction of porosity available for solid product deposition, peak stress and Young's modulus



Actual problem



1D Idealization



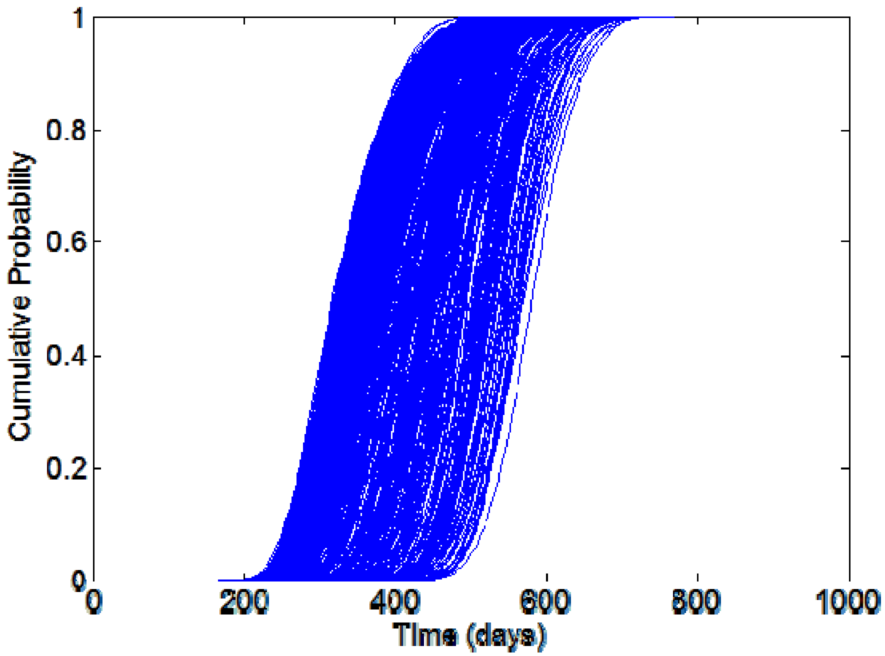
Example of Durability Analysis

Statistical description of the parameters

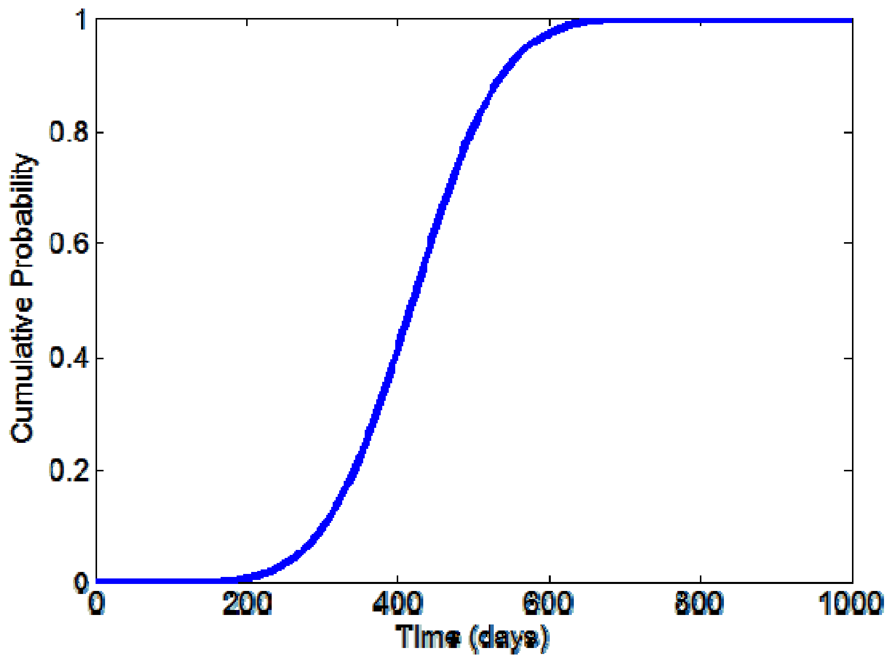
Input Type	Distribution
Initial porosity	N(0.3, 0.03)
Initial tortuosity	N(36, 3.6)
pH of external solution	N(7, 1.4)
Solution concentration (moles/L)	N(0.35, 0.07)
Renewal rate of solution (days)	U(5, 15)
Fraction of porosity available	U(LB, UB) LB~U(0.05, 0.15) UB~U(0.25, 0.35)
Peak stress (MPa)	N(f_t, 0.5) f_t~N(3, 0.3)
Initial Young's Modulus (GPa)	N(E_0, 5) E_0~N(20, 2)

Assessment of Time to Failure

Method 1



Method 2



- **Failure Criterion : 50% of the structure reaches maximum damage level**

Conclusions

- A coupled reactive transport and damage mechanics model has been developed for assessment of degradation of cementitious materials under external sulfate attack
- Sensitivity analysis was carried out to identify influential parameters
  - Long term effect of pH of external solution
  - Damage more if porosity less
  - Effect of tortuosity on rate of damage
  - Damage increases if calcium aluminate content increases
- Durability analysis approach demonstrated considering various sources of uncertainty

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