PROBABILISTIC DURABILITY ANALYSIS OF CEMENTITIOUS MATERIALS UNDER COMBINED SULFATE ATTACK AND CALCIUM LEACHING – 10149

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ABSTRACT

A deterministic mechanistic model coupled with probabilistic methodology is presented in this paper to evaluate durability of cementitious materials under the coupled effects of calcium leaching and external sulfate attack. The deterministic degradation modeling methodology includes (i) diffusion of ions from the external solution (e.g., sulfate salts) and leaching out of ions from the structure (e.g., calcium), (ii) chemical reactions which are assumed to occur under local equilibrium conditions, and (iii) damage accumulation using a continuum damage mechanics approach. The objective of the proposed methodology is to compute the probability of reaching a degradation measure as a function of time. The probabilistic framework accounts for various sources of uncertainty – physical variability due to inherent randomness of physical processes and model parameters, data uncertainty due to sparse or interval data, and model uncertainty due to assumptions and approximations in modeling a physical process. Various approaches for statistical representation of the uncertainties are investigated and incorporated in the durability assessment framework. The methodology for assessing the durability of the structure is implemented using nested and single-loop Monte Carlo Simulation techniques.

INTRODUCTION

Low activity nuclear wastes are mixed with cementitious materials and placed in underground reinforced concrete vaults. The waste materials contain various salts of sulfate, carbonate, chloride, nitrite etc. Sulfate attack from sulfate contained in the waste form is identified to be one of the potentially important degradation mechanisms for the reinforced concrete containment structures [1]. When a cementitious structure is exposed to an aggressive chemical solution, calcium leaches out of the structure. Thus in this work, sulfate ingression and calcium leaching are considered to be coupled degradation phenomena.

The assessment period for the vaults is generally 10,000 years and is therefore dependent on the long-term durability of the containment structures. It is not feasible to perform experiments to evaluate performance of the structures at this time scale. Thus a mechanistic model is needed for this purpose including all the essential steps of the degradation process. When sulfate ions

diffuse through a structure, they react with the cement hydration products to form expansive products. This induces strain leading to cracking and eventual failure. The numerical model used in this work incorporates the three stages of the degradation process – diffusion of ions into and out of the structure, chemical reactions and damage accumulation due to cracking.

If long term structural response is of interest, it is important to consider the uncertainty due to variability of the system parameters and the fluctuations in the initial and boundary conditions over time. Several service life assessment models available in the literature include variability of the parameters [2-5], but assess resistance to degradation using empirical relations. In addition to physical variability of the parameters, data uncertainty due to sparse or interval data, and model uncertainty due to various assumptions and approximations, introduce additional uncertainty in the model predictions. A probabilistic framework is presented in this paper to assess the durability of structures under the combined effects of sulfate attack and calcium leaching, incorporating various sources of uncertainty.

NUMERICAL MODELING FRAMEWORK

The numerical model used in this paper incorporates three essential stages of degradation of cement-based structures under the combined effects of sulfate ingression and calcium leaching – diffusion of ions, chemical reactions and damage accumulation. Brief descriptions of the stages are given in the following subsections. Detailed descriptions of the following approaches are given in [6].

Diffusion of ions

Assuming the structure to be saturated, porous and under isothermal condition, the governing equation for diffusion of an ion is expressed as

$$\frac{\partial(\varphi c_i)}{\partial t} = \mathrm{div}\left(\frac{D_i^0 \varphi}{\tau}(\mathrm{grad}(c_i) + c_i \mathrm{grad}(\ln \gamma_i))\right) \quad (1)$$

where c_i is the concentration of the i^{th} ion, D_i^0 is the free solution diffusivity of the ion, φ is the porosity, τ is the tortuosity and γ_i is the chemical activity coefficient of the ion. The modified Davies equation [7] is used to calculate the chemical activity of the ions, which produces better results for highly concentrated ionic solutions such as concrete pore solutions than other formulations of activity coefficient [8], and is given as

$$\ln \gamma_i = -\frac{A z_i^2 \sqrt{I}}{1 + \alpha_i B \sqrt{I}} + \frac{(0.2 - 4.17e-5I) A z_i^2 I}{\sqrt{1000}} \quad (2)$$

where z_i is the valence of the ion, I is the ionic strength of the solution, and A and B are temperature dependent parameters.

Chemical reactions

Some of the main hydration products which are generally present in a matured cement-based structure are calcium silicate hydrate (CSH), calcium hydroxide or Portlandite (CH), ettringite ($C_6A\bar{S}_3H_{32}$), calcium monosulfoaluminate ($C_4A\bar{S}H_{12}$), hydrogarnet (C_3AH_6), etc. When sulfate ions diffuse through a structure, a series of reactions take place. If the diffusing species is sodium sulfate, it reacts with Portlandite to form gypsum and with calcium monosulfate and tricalcium aluminate to form ettringite. The gypsum can then react with any of the calcium aluminate phases, e.g. calcium monosulfate, tricalcium aluminate, hydrogarnet, tetracalcium aluminate hydrate, etc., to form ettringite. Initially, calcium in the pore solution is supplied by Portlandite. When Portlandite is depleted from the system, calcium silicate hydrate dissociates to form calcium hydroxide and silica gel. The main reactions are given in Eqs. (3) – (9).

$$Na_2SO_4 + CH + 2H \rightarrow C\bar{S}H_2 + 2NaOH \quad (3)$$

$$C_4A\bar{S}H_{12} + 2C\bar{S}H_2 + 16H \rightarrow C_6A\bar{S}_3H_{32} \quad (4)$$

$$3C_4A\bar{S}H_{12} + 3Na_2SO_4 \rightarrow 6NaOH + 2Al(OH)_3 + 21H + 2C_6A\bar{S}H_{32} \quad (5)$$

$$C_3A + 3C\bar{S}H_2 + 26H \rightarrow C_6A\bar{S}_3H_{32} \quad (6)$$

$$C_3A + 3Na_2SO_4 + 3CH + 32H \rightarrow 6NaOH + C_6A\bar{S}H_{32} \quad (7)$$

$$C_4AH_{13} + 3C\bar{S}H_2 + 14H \rightarrow C_6A\bar{S}_3H_{32} + CH \quad (8)$$

$$C_3AH_6 + 3C\bar{S}H_2 + 20H \rightarrow C_6A\bar{S}_3H_{32} \quad (9)$$

In addition to the above mentioned reactions, formation and dissolution of several other solid phases also occur simultaneously to maintain the equilibrium state of the pore solution. A geochemical speciation code, ORCHESTRA [9], is used in this work to calculate the equilibrium phases of the solids. The change in volume due to the dissolution and precipitation of solids is expressed as

$$\Delta V_s = \sum_{m=1}^{M} \left(V_m - V_m^{init}\right) \quad (10)$$

where M is the number of solid phases, and V_m^{init} and V_m are the initial and current volume of the m^{th} solid. The volume of the solid is calculated by multiplying number of moles of each mineral obtained from the equilibrium calculations and molar volume of the respective mineral phase. If the final volume of solids is more than the initial volume, the additional volume can only be accommodated in the pore space. Thus the decrease in porosity is expressed as

$$\varphi = \varphi_0 - \Delta V_s \tag{11}$$

where φ and φ_0 are the current and the initial porosities respectively. Similarly, if the final solid volume is less than the initial solid volume, porosity increases which can also be calculated from Eqs. (10) and (11). The change in diffusivity due to the change in porosity is calculated using an empirical equation given as [10]

$$H_D(\varphi) = \frac{e^{\frac{4.3\varphi}{V_p}}}{e^{\frac{4.3\varphi_0}{V_p}}} \tag{12}$$

where V_p is the volume of the paste. Eq. (12) is a correction factor which is multiplied with the diffusivity ($\frac{D_i^0 \varphi}{\tau}$) in Eq. (1) and is used as the changed diffusivity for the next time step.

Damage accumulation

As mentioned in the previous subsection, if the final solid volume is more than the initial solid volume, the additional solid is accommodated in the pore space. The solid grows and eventually exerts pressure on pore wall of the surrounding cement matrix. Cracks start to form when the stress induced in the cement matrix exceeds the strength of the material. The solids deposited in the pores and the pores themselves are morphologically different. Thus the solids do not have to fill up the total pore volume in order to start exerting pressure. Thus it is assumed that a fraction of the pore space (b) is available for solid product deposition before strain can develop. This is a model parameter which needs to be calibrated using experimental results. The net solid volume which contributes to the strain development is calculated as

$$\overline{\Delta V_s} = \Delta V_s - b\varphi V \tag{13}$$

where V is the volume of the representative volume element. The volumetric strain is calculated as

$$\bar{\varepsilon} = \frac{\overline{\Delta V_s}}{V} \tag{14}$$

if $\overline{\Delta V_s} > 0$. Otherwise it is zero. The uniaxial strain is calculated as

$$\varepsilon = \frac{\bar{\varepsilon}}{3} \tag{15}$$

assuming the structure to be homogeneous and isotropic. A continuum damage mechanics-based approach is used in this paper to relate the strain to the cracked state of the structure using experimental stress-strain curve of the material. The tensile stress-strain relation of cementitious materials exhibits three sequential regions – linear ascending, nonlinear ascending and nonlinear descending regions. There is no damage accumulation in the linear ascending region. In the nonlinear ascending region, a crack density parameter C_d which reflects accumulated damage can be expressed as

$$C_d = k(1 - \frac{\varepsilon^{th}}{\varepsilon})^m \text{ for } \varepsilon \geq \varepsilon^{th} \tag{16}$$

where ε is the strain, ε^{th} is the threshold strain at which micro-cracks start forming and k and m are parameters that need to be calibrated from the experimental stress-strain diagram. The damage parameter ω can be conceptually defined as the ratio of damaged strength to the undamaged strength of the material. Assuming that the damage parameter is not affected by the Poisson's ratio of the damaged material, ω can be expressed as [11, 12]

$$\omega \approx (16/9)C_d \tag{17}$$

The post-peak response of the material is modeled by using the relations [13] given as

$$\frac{\sigma}{f_t'} = \sqrt{\frac{\tan\left(\frac{\pi\omega_0}{2}\right)}{\tan\left(\frac{\pi\omega}{2}\right)}} \tag{18}$$

$$\frac{w}{w_0} = \frac{\sigma}{f_t'}\frac{\log[\sec(\frac{\pi\omega}{2})]}{\log[\sec(\frac{\pi\omega_0}{2})]} - 1 \tag{19}$$

where σ is the stress, f_t' is the maximum tensile stress, ω_0 is the damage parameter at peak stress, w and w_0 are the current post-peak deformation and the deformation at the peak stress respectively. The damage parameter starts from 0 at the threshold strain (ε^{th}) and reaches 1 at failure. The maximum allowable value of damage parameter is assumed to be 0.9 in the numerical simulations presented in this paper to allow for additional uncertainties and adequate margin in safety.

Change in diffusivity

The effect of development of strain on the formation of cracks is discussed in the previous subsection. The presence of cracks enhances diffusivity which leads to more diffusion of ions from outside and more leaching out of ions from inside of the structure. When the cracks are dilute in concentration, the changed diffusivity is expressed as [14, 15]

$$D = H_D(\varphi)\frac{D_i^0\varphi}{\tau}\left(1+\frac{32}{9}C_d\right) \tag{20}$$

When the microcracks coalesce and form macrocracks, the diffusivity is calculated using the following expression [14, 15]

$$D = H_D(\varphi)\frac{D_i^0\varphi}{\tau}\left[\left(1+\frac{32}{9}C_d\right)+\frac{(C_d-C_{dc})^2}{(C_{dsc}-C_d)}\right] \tag{21}$$

where C_{dc} is the conduction percolation threshold below which concentration of cracks is sparse and C_{dsc} is the rigidity percolation threshold at which the cluster of cracks transects the volume. The values for the thresholds are obtained from the literature [14, 16, 17].

PROBABILISTIC DURABILITY ANALYSIS

The numerical model described in the previous section is used in this work to simulate degradation of cement-based materials under the combined effects of calcium leaching and sulfate ingression. The model requires several input and model parameters for performing simulations. These parameters can be obtained from the experiments or from the literature. In either case, the values of the parameters will have some uncertainties associated to them leading to uncertainty in the model response. Various methods are available in the literature for quantifying the uncertainties in the parameters and propagating it through the model which leads to quantification of uncertainty in the model prediction. Finally, the durability of a structure subject to a specific set of performance criteria is expressed as a statistical distribution.

Uncertainty quantification

There are mainly three sources of uncertainty – (i) physical variability due to the inherent randomness of the variables, (ii) data uncertainty due to sparse or interval data, and (iii) model uncertainty due to assumptions and approximations used during the modeling process. The approaches for quantification of uncertainty in the parameters are described in this subsection.

Physical variability – This comes from the inherent randomness of the parameters e.g. material properties and external boundary conditions. For a homogeneous structure modeled at the resolution of macro-scale, initial material and geometrical properties can be modeled as random variables. For example, bulk density of the material, mortar-water ratio, total open porosity,

external sulfate solution concentration etc. are modeled as a normal random variable by Rigo *et al.*, 2005 [18]. But some parameters may vary not only from sample to sample (as is the case for random variables), but also in spatial or time domain. In these cases, they can be modeled as *random fields or processes* [19, 20]. Some of the well known methods for simulating random fields/processes are spectral representation (SR) [21], Karhunen-Loeve expansion (KLE) [22-24], polynomial chaos expansion (PCE) [20, 23-25], etc. Some boundary conditions generally exhibit a recurring pattern over shorter periods and also a trend over longer periods. These can be numerically represented by a seasonal model [26] using an autoregressive integrated moving average method. In this paper, the physical variations in the parameters are incorporated by defining them as random variables with probability density functions (PDFs).

Data uncertainty – In a numerical simulation, data uncertainty comes from complete or partial lack of knowledge of some input parameters. If the available data set is small or if only ranges of values are available, statistical distributions can be constructed by using an empirical distribution function or a flexible family of distributions such as the Johnson family of distributions [27, 28]. In either case, the parameters are expressed as statistical distributions, whose parameters are also expressed as statistical distributions. In this paper, model parameters having data uncertainty are simulated using statistical distributions with random parameters.

Model uncertainty – This can come from various approximations and assumptions made during the modeling process such as incomplete knowledge of the physics of the phenomenon, and analysis approximations. Verification, validation, calibration, and error quantification are different steps to handle model uncertainty. Recently, a method was developed to quantify model error by combining errors due to model form, numerical discretization, uncertainty analysis method and input and output measurements [20]. In this method, experimental data are needed to evaluate the model form error. Model uncertainty is not included in this work due to the lack of experimental data and to keep the formulation simple.

Uncertainty propagation

The input parameters of the numerical model can be simulated using the methods described in the previous subsection. Finally, this problem can be formulated as a time-dependent reliability analysis to assess the evolution of the probability of reaching a specified level of degradation with time [2, 3, 5, 29-31] by Monte Carlo Simulation (MCS) [32], First Order Reliability Method [32], etc. MCS is the most commonly used method in the literature which can be time consuming. Various efficient sampling techniques can be used to minimize the computer time or storage requirement, e.g., Latin hypercube sampling, importance sampling, etc.

Two approaches are explored to quantify the uncertainty in the durability assessment framework. The first method incorporates a nested Monte Carlo simulation as shown in Figure 1, in order to estimate the confidence bounds on the durability curve, i.e., the probability of

reaching a particular degradation measure as a function of time. As shown in the figure, N_2 samples for parameters having data uncertainty are generated. For each of N_2samples, N_1 samples for parameters having physical variability are generated and simulations are performed. The model errors (if available) can be added to the simulation results at this stage. Thus a total of $N_1 \times N_2$ simulations are performed. A durability curve can be constructed using each of N_1 model responses from the inner loop Monte Carlo simulations and comparing the result with a particular performance requirement, e.g., the. maximum allowable stress or strain in the structure. Thus, confidence bounds on the durability curve are calculated by using N_2 durability curves obtained from the outer loop Monte Carlo simulations. This method is computationally expensive for a large finite element multiphysics time dependent problem. A surrogate model, e.g., Gaussian process model, can be used in such cases to reduce the computational time.

The second method incorporates the aforementioned uncertainties in a single loop Monte Carlo simulation as shown in Figure 2 by consolidating the nested simulations into one loop. In this case, N samples are generated for parameters having physical variability and data uncertainty. Simulations are performed for each set of samples generated and probability of reaching a particular degradation measure as a function of time is calculated. Thus a single durability curve is obtained as a result of the simulations in this case.

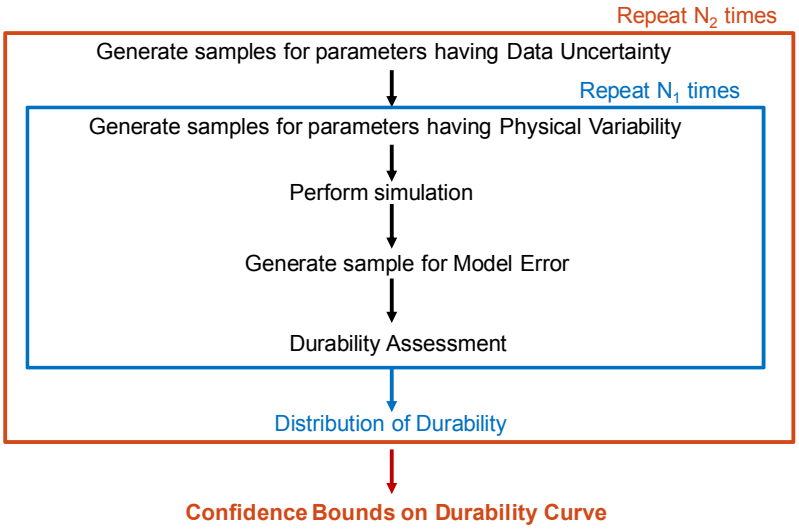

Figure 1: Probabilistic durability assessment using nested Monte Carlo simulation.

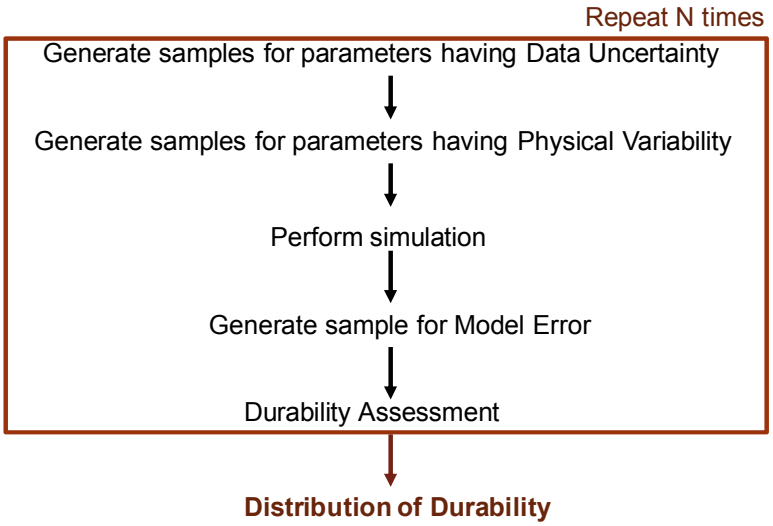

Figure 2: Probabilistic durability assessment using single loop Monte Carlo simulation.

Demonstration of probabilistic durability analysis

A simulation test case is set up by immersing a U.S. Type I cement mortar sample of size $25\ \text{mm} \times 25\ \text{mm} \times 285\ \text{mm}$ in a tank of Na_2SO_4 solution. The liquid volume to solid volume ratio is 10. The cement, water and sand mass ratio is $1:0.5:3$. A one-dimensional idealization of the problem is simulated for 2 years for each set of random variables generated for the Monte Carlo simulation. The descriptions of the parameters are given in Table 1. $N(\mu, \sigma)$ represents a normal distribution with mean μ and standard deviation σ. $U(LB, UB)$ represents a uniform distribution with LB lower bound and UB upper bound.

Table 1: Statistical descriptions of the parameters.

Input Type	Distribution
Physical variability	
Initial porosity	$N(0.3, 0.03)$
Initial tortuosity	$N(36, 3.6)$
pH of the external solution	$N(7, 1.4)$
Solution concentration (moles/L)	$N(0.35, 0.07)$
Renewal rate of solution (day)	$U(5, 15)$
Data uncertainty	
Fraction of porosity available (b)	$U(LB, UB)$

	$LB \sim U(0.05, 0.15)$
	$UB \sim U(U(0.25, 0.35)$
Peak stress (MPa)	$N(f_c, 0.5)$
	$f_c \sim N(3, 0.3)$
Initial Young's modulus (GPa)	$N(E_0, 5)$
	$E_0 \sim N(20, 2)$

The durability of the structure is assessed by assuming a particular performance requirement as a failure criterion. The structure is assumed to have failed if 50% of the thickness of the structure reaches the maximum damage level. An element in the numerical model reaches the maximum damage level when the damage parameter associated with that element reaches the maximum value which is assumed to be 0.9 as mentioned in the previous section. As mentioned earlier, the nested Monte Carlo simulation is computationally expensive. Therefore, a Gaussian process surrogate model has been built for assessing time to failure of the structure, trained using 50 numerical simulations of the full multiphysics model to reduce the computational time. Then 500 samples are generated for parameters having data uncertainty and physical variability and the nested Monte Carlo simulation is performed. It is important to acknowledge the fact that additional uncertainty is introduced in the simulation due to the use of the surrogate model which is not included in this paper to keep the formulation simple. Figure 3 shows the ensemble of the durability curves. The 50 numerical simulations using the full multiphysics model are then used in a single loop Monte Carlo simulation to generate a durability curve as shown in Figure 4.

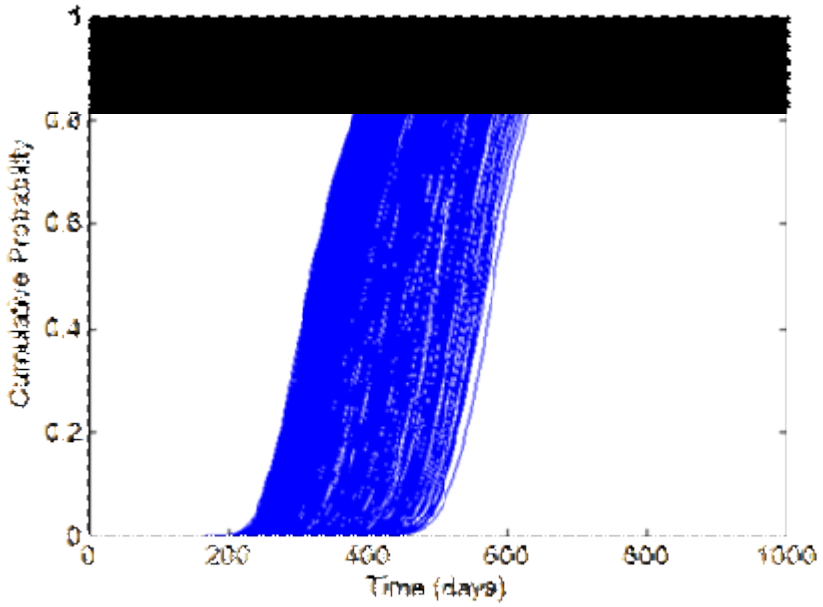

Figure 3: Cumulative probability of time to failure as a function of time using nested Monte Carlo simulation technique that provides the full envelope of probability.

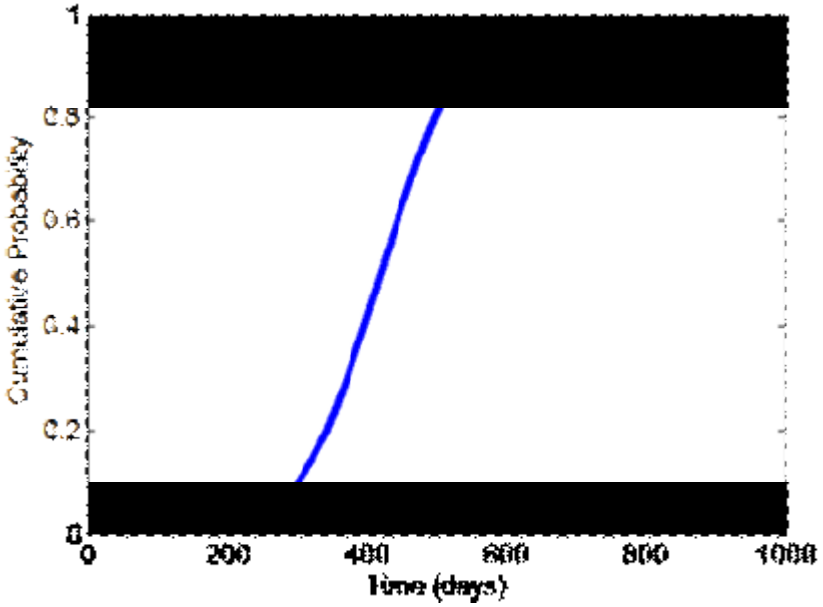

Figure 4: Cumulative probability of time to failure as a function of time using single loop Monte Carlo simulation technique.

The durability curve in Figure 4 is the unconditional cumulative distribution function, whereas the curves in Figure 3 are conditioned on the values of the distribution parameters. Figure 3

explicitly shows the contribution of data uncertainty, whereas Figure 4 integrates all three sources of uncertainty into one cumulative distribution function curve.

CONCLUSION

A numerical model for degradation assessment of cementitious materials under combined sulfate attack and calcium leaching is developed in this paper by combining a diffusion model and a chemical reaction model with a continuum damage mechanics based model. Several sources of uncertainty in the degradation assessment process (physical variability, data uncertainty and model uncertainty) are discussed. The methods for quantifying the uncertainty in the model parameters and propagating them through the model are discussed using nested and single loop Monte Carlo simulations. Applicability of the probabilistic durability analysis framework is demonstrated using an example case.

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