Concrete Degradation Modeling in the Evaluation of Entombment as a Decommission Option

by

K.A. Snyder
Building and Fire Research Laboratory
National Institute of Standards and Technology
Gaithersburg, MD 20899 USA

and

J. Philip
Nuclear Regulatory Commission
Rockville, MD 20852 USA

Reprinted from Spectrum 2002: International Conference on Nuclear and Hazardous Waste Management. Proceedings. Reno, NV, August 4-8, 2002, CD-ROM, 2002.

Concrete Degradation Modeling in the Evaluation of Entombment as a Decommissioning Option

K.A. Snyder[a] and J. Philip[b]

[a] *National Institute of Standards and Technology, Building Materials Division, MS-8621, Gaithersburg, MD 20899*
kenneth.snyder@nist.gov

[b] *Radiation Protection, Environmental Risk & Waste Management Branch*
Division of Systems Analysis and Regulatory Effectiveness
Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission (USNRC)
Rockville, MD 20852
jxp@nrc.gov

Abstract – *For entombment to be a viable option for the decommissioning of nuclear structures, the effectiveness of available engineered barriers needs to be assured. Barrier performance should be estimated with the aid of computer models that can accurately predict the response of the barrier to foreseeable physical and chemical conditions. For concrete barriers, virtually all degradation mechanisms are controlled by the transport of water and ionic species within the pore space. These, in turn, are controlled by the appropriate transport coefficients. For sound concrete, the transport coefficients are sufficiently small enough that isolation is expected. It is the presence of cracks within the concrete that compromises the barrier by increasing the transport coefficients dramatically. Therefore, additional efforts to characterize the performance of concrete barriers must focus on quantifying the existing cracks and flaws within the concrete. This characterization would include sampling (when possible), nondestructive techniques, and computer modeling. All of these are incorporated into a coherent monitoring plan.*

I. INTRODUCTION

Entombment can be considered as an option for decommissioning reactors and other nuclear structures so that the licenses for facilities on which these structures reside may be terminated. Prior to the entombment process, the nuclear facility permanently ceases operations and spent fuel is removed from it. After preliminary decommissioning activities are completed, radioactively contaminated components to be left on site would remain in a structure (e.g., a reactor containment building). The radioactive materials are then entombed with engineered barrier systems that would include filling the structure with cement, absorbent grouts, or infills. The entombed structure may be surrounded with sorbent backfills and then capped with reinforced concrete to mitigate transport of radionuclides to the environment and resist inadvertent intruder incursions. Isolation or containment of the radioactive materials necessitates that the engineered systems in the entombment perform adequately for the necessary duration of isolation to mitigate the leaching and migration of radionuclides to the environment and ensure that the relevant decommissioning dose criteria are met.

Effective performance predition of the entombed structure requires concrete degradation modeling. The parameters of importance for the modeling would include the porosity, the formation factor, the water-to-cement ratio, and the permeability of the concrete. The chemical assessment would require knowing both the quantity and the spatial distribution of the ionic species inside the concrete and immediately exterior to it. The greatest factor affecting the transport properties of the concrete is the existence of cracks within the structure. The program 4SIGHT [1] was developed as a tool for estimating the service life of underground concrete structures. Enhancements were made after validation tests of the code's transport model.[2] Additionally, a cracking model and a Monte Carlo calculation based on parameter uncertainty to address the inherent issue of variability were incorporated in the code.[3] A description of the features of the 4SIGHT code and its use in the effectiveness and performance evaluation of an entombed concrete structure is presented.

II. COMPUTER MODEL

The 4SIGHT computer program [1] was originally developed to predict the performance of underground concrete vaults. Previously, models had been developed for predicting performance of concrete subjected to a specific degradation process, but no model existed for the simultaneous consideration of a number of degradation processes, along with the coupled effect that one degradation mechanism (e.g., leaching) could have on another (e.g., chloride diffusion and subsequent corrosion of the steel reinforcement).

The 4SIGHT program succeeded in modeling these coupled effects by modeling the transport of ionic species through the pore space. From the stoichiometry of possible reactions between the ionic species and available salts and minerals, the pore space would change through either dissolution or precipitation. Using computer models of cement paste hydration, sensible relations were established between changes in porosity and corresponding changes in the transport coefficients. Through this coupling between reaction and transport, 4SIGHT is able to simulate the synergistic effect of multiple degradation mechanisms. In the field of concrete materials performance prediction, the program is one of the first of its kind.

II.A. Transport

The transport portion of 4SIGHT is based on an electro-diffusion equation [4] that treats the Brownian motion and the electrostatic interactions separately. The flux $\mathbf{j}_i$ of the i-th diffusing species can be related to gradients in both the amount-of-substance concentration c_i of that species and the electrostatic potential ψ:

$$\mathbf{j}_i = -\frac{D_i^\circ}{F}\left(1 + \frac{\partial \ln \gamma_i}{\partial \ln c_i}\right)\nabla c_i - z_i u_i c_i \nabla \psi \tag{1}$$

The quantities D°, F, and γ_i are the dilute limit self-diffusion coefficient of the species, the material formation factor, and the species activity coefficient, respectively. The quantities z_i and u_i are the species valence and species conventional electrochemical mobility, respectively. The dynamical behavior of the system is determined from mass conservation and the porosity ϕ:

$$\frac{\partial \phi c_i}{\partial t} = -\nabla \cdot \mathbf{j}_i \tag{2}$$

In the absence of an external electrical field, the complete system of equations requires there to be zero total current:

$$\sum_i z_i \mathbf{j}_i = 0 \tag{3}$$

Although Eqns. 1,2, and 3 apply to systems containing an arbitrary number of ionic species, this approach is favorable for use with porous materials [5] in that only two material parameters, the formation factor and porosity, are required to characterize diffusive transport. The equations have been used to characterize diffusion in nonreacting porous materials [6] containing salt concentrations near 0.1 mol/L. Because cement paste pore solution may contain hydroxyl concentrations in excess of 1 mol/L, current work involves validating the equations at these concentrations. To achieve this, calculations of the diffusion coefficient of binary salts at concentrations in excess of 2 mol/L are compared to published data. [7]

II.B. Transport-Reaction Feedback

The advantage of the 4SIGHT approach is the feedback between transport and reaction. The pore solution is maintained in equilibrium with the solid salts within the pore space through dissolution and precipitation. The result is a change in both the porosity and the connectivity of the pore space. These changes are reflected in both the material porosity and the formation factor. The change in the porosity can be calculated directly from the quantity of salt dissolved or precipitated. Estimating the change in the formation factor is more difficult.

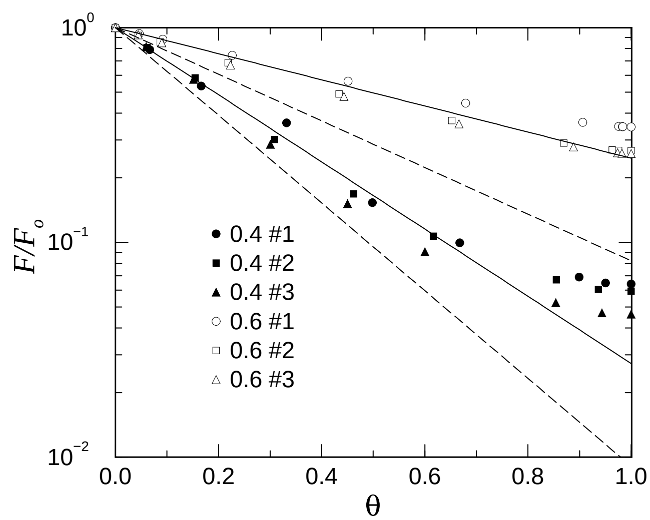

Figure 1: Relative change in the formation factor F as a function of the fraction θ of calcium hydroxide that has been leached.

The relationship between changes in salt content and changes in the formation factor were studied using a microstructural model for hydrated cement paste.[8] The relationship is based on a study of effect of calcium hydroxide leaching on the formation factor F. The results are expressed as fraction of the available calcium hydroxide that has been leached θ, where $0 \leq \theta \leq 1$.

From the data, a relationship was found between the relative change in the formation factor F/F_o and the fraction θ of calcium hydroxide leached:

$$\frac{F}{F_o} = \mathrm{e}^{-A\theta} \tag{4}$$

The coefficient A is a function of the water-to-cement mass ratio $\frac{w}{c}$:

$$A = 8 - 11\frac{w}{c} \tag{5}$$

The performance of these relations is shown in Fig. 1 for three different cements at two $\frac{w}{c}$ ratios. The solid curves in the figure are the estimates for 0.4 and 0.6 $\frac{w}{c}$ ratios. The dashed curves are the estimates for 0.3 and 0.5 $\frac{w}{c}$ ratio systems.

III. PERFORMANCE ANALYSIS

The initial prediction of concrete performance will be based on as much information as is available. Whenever possible, samples will be taken. Unfortunately, the nature of these structures will probably preclude extensive sampling for analysis. Under these circumstances, one must make sound engineering estimates. To facilitate this, one can use tools such as the NIST microstructural model [9] to make predictions of both the physical transport parameters and the relevant cement chemistry.

III.A. Material Properties

The initial condition assessment will attempt to characterize both the physical and chemical condition of the facility. The important physical parameters will include the appropriate transport parameters (formation factor, permeability, and porosity) and also the spatial characterization of cracks within the structure. The chemical properties are mainly the spatial distribution of ionic species inside the concrete and the concentration of external ionic species. In addition, microstructural analysis may be required in order to predict future reactions that are possible.

III.B. Cracks

While there is a body of research on characterizing various transport parameters in porous materials, characterizing the spatial distribution of cracks in a massive concrete structure will be a challenging hurdle. Analysis indicates that the single most important parameter used to characterize the influence a crack will have on transport is the depth to which the crack penetrates the concrete. The influences of the crack width and the spacing between cracks are relatively small by comparison to penetration depth. Therefore, nondestructive evaluation techniques must be investigated for their applicability to this problem.

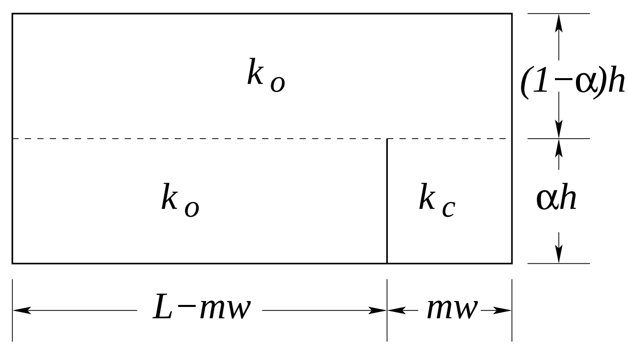

Figure 2: Crack model schematic for a concrete slab with span L, depth h, and containing m cracks, each of width w. k_o is the permeability of the uncracked concrete, and k_c is the permeability of each crack.

The influence of crack depth on the transport properties of a slab with span L and depth h can be characterized analytically using the schematic shown in Fig. 2. Let there be a number m cracks, each with width w, within the span, each penetrating into the slab a proportion α of the total depth. The permeability of the uncracked concrete is k_o. The permeability of each crack k_c is based on the assumption that the crack walls are parallel and smooth [3]:

$$k_c = \frac{w^2}{12} \tag{6}$$

The total flow through the entire slab can be estimated from the composite properties of the slab. The composite permeability k_p of the slab to the depth αh is a weighted sum of the two permeabilities:

$$k_p = \left(1 - \frac{mw}{L}\right) k_o + \frac{mw}{L} k_c \tag{7}$$

The bulk permeability k_b of the entire slab is estimated by analogy to electrical conductors in series:

$$k_b = \frac{1-\alpha}{k_o} + \frac{\alpha}{k_p} \tag{8}$$

This relationship simplifies to reveal asymptotic behavior with respect to α:

$$k_b = \frac{k_o}{1-(1-k_o/k_p)\,\alpha} \tag{9}$$

A sensitivity analysis is simplified by expressing the bulk permeability k_b differently:

$$k_b = k_o\left[\frac{1+\beta}{1+(1-\alpha)\beta}\right] \tag{10}$$

The intermediate quantity β is a function of the crack properties:

$$\beta = \frac{mw}{L}\left(\frac{k_c}{k_o}-1\right) \tag{11}$$

Given that the permeability k_o of concrete is in the range 10^{-16} m^2 to 10^{-18} m^2, and that cracks in concrete have widths on the order of 10^{-4} m, the parameter β can be simplified slightly:

$$\beta = \frac{mw^3}{12k_oL} \tag{12}$$

It should be remembered that, for cracks in concrete, the value of β is far greater than 1.

Changes in the bulk permeability Δk_b can be expressed as a Taylor expansion:

$$\Delta k_b = \frac{dk_b}{dm}\,\Delta m + \frac{dk_b}{dw}\,\Delta w + \frac{dk_b}{d\alpha}\,\Delta\alpha + \cdots \tag{13}$$

Substitution for the differentials gives the following relationship:

$$\begin{aligned}\frac{\Delta k_b}{k_b} &= \left(\frac{k_b}{k_o}\right)\frac{\beta}{1+\beta} \\ &\quad\times\left[\frac{\alpha}{1+\beta}\,\frac{\Delta m}{m} + \frac{3\alpha}{1+\beta}\,\frac{\Delta w}{w} + \Delta\alpha\right]\end{aligned} \tag{14}$$

The effects of changes in the relative number of cracks m and the crack widths w are reduced by a factor of β ($\beta \gg 1$). The α term, however, lacks β in the denominator. Therefore, relative changes in the crack depth can be dramatic, as compared to relative changes in either the crack width or number of cracks.

IV. PROBABILISTIC CALCULATION

Although the performance prediction is fundamentally deterministic, uncertainty is incorporated through parameter uncertainty. By incorporating probabilistic models for the input parameters, a Monte Carlo calculation of the performance for an ensemble of input parameters yields a probabilistic result. This approach has been applied to both the formation of cracks [3] and to the overall performance of the concrete.[2]

The input parameters are characterized by any one of four probability density functions: delta function, [10] uniform distribution, normal distribution, and lognormal distribution.[11] The user specifies the relevant parameters for each distribution, and the total number of iterations to perform. The result is a distribution of responses from the calculation.

V. MONITORING

A plan for assuring entombment's effectiveness will likely require a combination of initial condition assessment, performance prediction, and monitoring to verify the model prediction. Since no 300-year data exist on modern cement-based composites (cement production in the United States changed dramatically in the 1970's with emphasis on early age strength in order to reduce construction cost), the most viable means of assuring isolation is through monitoring. The initial condition assessment and performance prediction will establish what parameters require monitoring and how often monitoring must be performed.

Since the performance prediction will be probabilistic, one can place uncertainty limits on the expected service life. At monitoring intervals, the results can then be incorporated into the model for refinement of the expected time to failure. In this way, uncertainty in the prediction model may be reduced and monitoring intervals can be better refined.

VI. SUMMARY

A combination of condition assessment, performance prediction, and monitoring can be implimented in a meaningful way to achieve an effective path to entombment. The condition assessment techniques will include both sampling and computer modeling in order to characterize the intact concrete. Nondestructive evaluation techniques will be required to characterize the depth of crack penetration, which is the quantity that has the greatest impact on transport. The performance prediction will incorporate the current knowledge of transport and relevant degradation reactions. The combination of assessment and model prediction will be incorporated into a monitoring strategy that uses both physical observation and estimated future predictions. The period between monitoring will be governed by both model and measurement uncertainty, and the results can be incorporated into revised monitoring plans.

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