Project Number: DE-FG03-99SF21891
Project Title: Monitoring the Durability Performance of Concrete in Nuclear Waste Containment
Principal Investigator: Franz-Josef Ulm, MIT
Date: 06/30/00

Technical Progress Report N^{o}4

RECEIVED
JUL 14 2000
OSTI

Abstract

This technical progress report covers the three months period from April 2000 through June 2000. Research during this period was devoted to tasks 1.1, 2 and 3.1.

Task 1.1 Model of Calcium Leaching in Cracked Materials

The analysis of the effect of cracks on the acceleration of the calcium leaching process of cement-based materials has been pursued. During the last period (Technical Progress Report No 3), we have introduced a modeling accounting for the high diffusivity of fractures in comparison with the weak solid material diffusivity. It has been shown through dimensional and asymptotic analysis that small fractures do not significantly accelerate the material aging process. This important result for the overall structural aging kinetics of containment structure has been developed in a paper submitted to the international journal 'Transport in Porous Media' (see section Published Papers at the end of this report).

If it is now obvious that a pure diffusive mass transport in a cracked material is not a significant source of calcium leaching acceleration, less appears to be known on the material chemical degradation due to an arrival of 'pure' water through the material fractures. When penetrating the material, the pure water may effectively induce an important leaching since the calcium present in the fracture can be evacuated at 'high' velocity by the water flow. This mass transport phenomenon of calcium transported by the water flux is referred to as a convective (or advective) mass transport. This phenomenon has been studied in the same manner as for the pure diffusive mass transport process, and the dimensional and asymptotic (i.e. large time) analysis shows that the degraded depth along the fracture is on the form:

$$y_d(t) = C_1 \frac{Vb}{\phi} \sqrt{\frac{t}{D_m}} \tag{1}$$

where b is the half fracture opening, V is the average fluid velocity in the fracture, D_m is the effective diffusion coefficient in the matrix, ϕ is the material porosity and t is the time. Equation (1) indicates that the degradation process evolves as a function of the square root of time as it is the case for a one dimensional leaching process of a solid material. However,

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

equation (1) also points out that a high value of the fluid velocity in the fracture V may induce an acceleration of the material chemical degradation. In order to investigate this risk for containment structures, the previous modeling of calcium leaching (see TPR 2 and 3) has been enhanced to account for convective mass transport in fractures. The developments realized are briefly described in Task 2 and some model-based simulations are discussed.

Task 1.1 is now almost completed, and the during the next steps of task 1. the focus will be on part 1.2 devoted to coupled chemo-mechanical modeling.

Task 2: Computational Mechanics and Analysis

The modeling accounting for calcium convection induced by a fluid flow in a fracture was implemented in the Finite Element program CESAR-LCPC, following the former developments of calcium leaching (TPR 2) and diffusion mass transport in fracture (TPR 3). Developments were validated with analytical solutions described in the literature. When considering high fluid velocity with respect to the diffusion process, the Finite Element approximation used for the convective term can induce numerical dispersion and overshoot. To overcome these problems, the following simulations analyzing the effect of a fluid flow in a fracture on the material degradation were performed with small grid and time step.

Developments were tested on the 2D-degradation of a rectangular cement paste of width 10 cm and length 50 cm. A straight crack with a 0.04 cm aperture is situated along the left side of the material. The material is initially undegraded, and the degradation process starts at the bottom of the rectangular geometry where the aggressive boundary condition is prescribed. The fracture may induce an acceleration of the degradation process due to either a faster diffusion of calcium toward the boundary condition or a fresh arrival of pure water in the fracture. In what follows, we briefly discuss the results of three model-based simulations with pure diffusion (Figure 1), convection for a fluid velocity of 5 cm/d (Figure 2) and 10 cm/d (Figure 3).

Figure 1 represents the solid calcium concentration in the material at 440 years for a pure diffusive mass transport in the fracture (i.e. no convection). In accordance with TPR 3, figure 1 shows that the fracture does not enhance the overall kinetics of material degradation. Considering now a convective mass transport in the fracture, figures 2 and 3 show the solid calcium concentration in the material at the same time as figure 1 for fracture fluid velocities respectively equal to 5 and 10 cm/d. Contrarily to the pure diffusive case (i.e. figure 1), figure 2 and 3 point out that a fluid flow in the fracture ensures a more important renewal of the fracture solution, and then a significant degradation of the material around the fracture in comparison with the material degradation at the bottom of the material. Analysis of the effect of the fluid velocity on the material chemical degradation is still under revue and a paper is in preparation on that topic.

As a conclusion, it has been shown that a pure diffusion process in cracked cement-based material does not significantly increase the overall structural aging kinetics of containment structure. On the other hand, cracked materials submitted to environmental conditions that

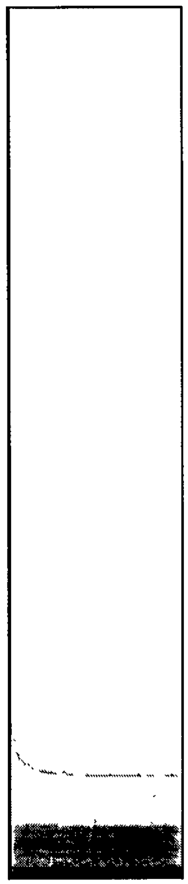

Figure 1: Solid calcium concentration at 400 years with a pure diffusive mass transport in the fracture.

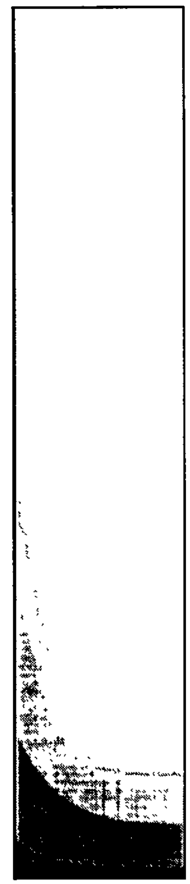

Figure 2: Solid calcium concentration at 400 years with fluid velocity of 5 cm/d in the fracture.

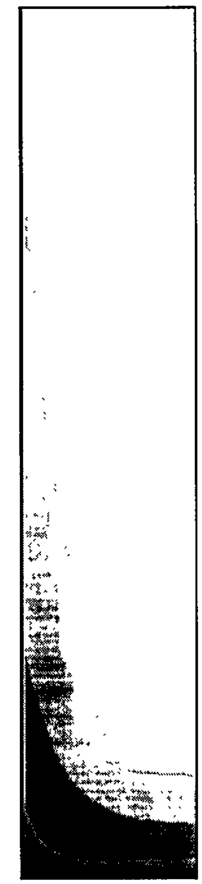

Figure 3: Solid calcium concentration at 400 years with fluid velocity of 10 cm/d in the fracture.

may induce an arrival of fresh water in the material fractures with significant fluid velocity appears to be a more important source of chemical degradation for the structure.

Task 3.1: Material testing

Continuing the material testing, we started calcium leaching and triaxial testing of mortar.

The utilized mortar is composed of the same type of cement as in the previous phase of the project (testing of pure cement paste). A fine Nevada 120 sand is added at a mass ratio sand/cement of 2. The sample size remains unchanged from the cement paste experiments (i.e. 11.5 mm in diameter and 23.5 mm length). The relation between sample size and grain size will allow to draw conclusions about the behavior of real concrete structures.

In order to leach the mortar, the same technique as for the cement paste was applied (For a detailed description see TPR 2 and 3). The mortar was put into a 6M Ammonium Nitrate solution and kept immersed for more than 45 days to achieve complete leaching. The visually observed propagation of the dissolution front was the same as for the cement paste at 1.5 $\mathrm{mm}/\sqrt{\mathrm{day}}$. This result was expected and has been reported before in the literature for pure water calcium leaching.

First results of the triaxial test that started recently show again a pressure dependent material behavior. Yet, due to the added granular material, the explanation of the observed material response will be more challenging.

A second area work for task 3.1 was the design and fabrication of material to perform triaxial test that lead to failure on the tension meridian. For this to happen the vertical stress must be smaller at the failure point than the confinement stress. As the confinement stress is applied hydrostatically in our test configuration a way of unloading the vertical axis had to be found. The fabrication of the necessary parts is completed now and the corresponding tests will start soon.

Project Communication

Published Papers

In the period covered by this report, results of the performed research were presented by the P.I. and his coworkers on the ASCE EM2000 conference in Austin, TX, and on the 11th NIST/ACBM Computer Modeling Workshop in Gaithersburg, MD. A paper entitled "Triaxial Behavior of Calcium Depleted Cementitious Materials" was published as part of the EM2000 proceedings. In addition, two scientific papers were submitted for review and publication: The paper entitled "Mechanical Properties of Calcium Leached Cement Paste" by F. H. Heukamp, F.-J. Ulm, and J. T. Germaine was submitted to "Cement and Concrete Research". The paper entitled "Coupled Diffusion-Dissolution Around a Fracture Channel: The Solute Congestion Phenomenon" by M. Mainguy, and F.-J. Ulm was submitted to "Transport in Porous Media".

Press coverage

The "Monitoring the durability performance of concrete in nuclear waste containment" - project was extensively covered in the press during the report period. In April a one page article about the project appeared in "Tech Talk" the weekly MIT newspaper. Moreover, the MIT "research digest", a paper issued by the MIT news office presenting current research at MIT reported on the project. In addition, printed articles about the project were published in "Business Week" and the "Norwich Bulletin" (Norwich, CT). A radio interview about the project was given by the principal investigator to "Deutschlandfunk" a German based world wide transmitting radio station. The web site *http://www.office.com*, a web portal featuring spezialised bussiness and scientific information, devoted two pages to the project under *http://www.office.com/global/0,2724,56-18045|1,FF.html.*

Web Page Development

Under the url *http://cist.mit.edu* a section on calcium leaching has been developed featuring press coverage of the DOE sponsored research project and first results. An animation is available showing the calcium dissolution front propagation in a container based on results from task 2.

Milestone plan: The initial milestone plan remains valid; no changes were necessary.

Task listing:
No task has been finished yet. Planned completion dates remain unchanged

Cost performance: