Project Title: Decontamination of Radionuclides From Concrete During and After Thermal Treatment

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Principal Investigators: Brian P. Spalding, Oak Ridge National Laboratory[1], PO Box 2008, Oak Ridge, TN 37831-6036, 865-574-7265, spaldingbp@ornl.gov and
Zdenek P. Bazant, Walter P. Murphy Professor of Civil Engineering and Materials Science, Northwestern University, Evanston, IL 60208, 847-491-4025, z-bazant@nwu.edu

Graduate Research Assistant: Goangseup Zi, Dept. of Civil Engineering, Northwestern Univ., Evanston, IL 60208, 847-491-3351, g-zi@nwu.edu

RESEARCH OBJECTIVE

This project will determine the effect of heating concrete on its engineering properties and the potential to aid in removal of common DOE radionuclide contaminants including ^{137}Cs, ^{90}Sr, ^{60}Co, and U. In the chemical properties subtask, effects on direct volatilization of radionuclides during heating, as well as their subsequent extractability behavior, will be established over a range of temperatures up to the melting point of Portland cement. The main objective of the mechanics and thermodynamics subtask of this investigation is to develop a powerful computational capability to evaluate the effects of various regimes of rapid heating of a contaminated concrete wall or slab.

RESEARCH PROGRESS AND IMPLICATIONS

This report summarizes project progress through June 2000, I.e., 20 months into this planned 36-month project interval. In the chemical properties subtask, direct volatilization of radionuclides during heating of hydrated Portland cement, as well as their behavior during subsequent extractions, were established over a range of temperatures up to the melting point of Portland cement. Techniques for spiking Portland cement with shorter-lived radioisotopes (^{134}Cs, ^{85}Sr, and ^{57}Co) and U were developed and gamma-ray spectroscopy was used to nondestructively measure their activity before and after heating. An extraction procedure (employing five sequential extractions each with water, followed by 0.1*N* $CaCl_2$, followed by 0.2*N* HCl) for fully-hydrated but pulverized Portland cement was adapted to contrast radionuclide behaviors after thermal treatments. Complete volatility of ^{134}Cs was observed at temperatures of 1200°C or greater. Thus, decontamination of ^{137}Cs from cement would be feasible by direct heating. However, no volatility of other radionuclides was observed so that their potential for facilitated decontamination depends on their extraction behavior following thermal perturbation.

The sequential extraction of the four radionuclides has been completed for pulverized cement samples heated from ambient to 1400°C at 100°C intervals. Both ^{134}Cs and ^{85}Sr were readily removed from cement during initial water-based extractions for final temperatures <800°C. In contrast, the highly alkaline nature of Portland cement, with its large content of $Ca(OH)_2$, resulted in the negligible extractability of both ^{57}Co and U into water or calcium chloride where extract pHs were >10. The release of both these radionuclides was delayed until

the dilute hydrochloric acid extracts decreased the pH below 4. Significant patterns in the extraction behaviors of all radionuclides with temperature were observed in ranges where significant mineral alterations are known to occur. When surface-contaminated cohesive cement plugs were employed during heating and subsequent extraction, the volatile and extractable behavior of both ^{57}Co and ^{134}Cs were found to be very similar to their behavior in pulverized cement. Cement plugs heated above 1000°C, readily disintegrated during subsequent extractions indicative of their loss of cohesive strength after heating above this temperature. After aging periods varying between 20 and 175 days, the behavior of radionuclides in unheated pulverized cement during subsequent extraction sequences was similar.

The work in the mechanics and thermodynamics subtask concentrated on effective and realistic computational simulation of the mechanical and thermodynamic aspects of the simulation of concrete spallation induced by rapid heating, both by conductive heating from the surface and by microwave heading within the bulk.

Microwave heating offers an attractive possibility of inducing concrete spallation and egress of pore water. The development of the computational model was based on the assumption of normal incidence of TEM (Transverse Electro-Magnetic) wave. The heat source is the dissipated energy (given by Poynting vector in the electro-magnetic field theory). A simple expression of heat generation by microwave heating has been developed by an averaging process in time and space domains over the wave period. The resulting expression is similar to the classical Lambert's law (which is used in microwave heating studies in food engineering), except that there is an extra term for the steel reinforcement inside the body of concrete.

A 'finite volume' computer code has been written to simulate the heat and mass transfer during the thermal process. The coupled heat and mass transfer model of concrete developed by Bazant and Thonguthai has been used. An important feature of the finite volume method is that the conservation equations for each finite volume are satisfied exactly. However, the very fine meshes needed for adequate resolution in multidimensional simulation have been found to pose extreme demands for computer time, in the case of the finite volume method. The finite volume method shows good performance in the case of conductive heating from the surface. Especially, a sharp drying front and an exponentially decaying temperature profile are reproduced well. But the method does not perform well in the case of microwave heating. The problem is caused by a jump in permeability at 100 degrees C, and the corresponding discontinuity in the field is not captured well. Currently, work is proceeding on a mixed finite element scheme that incorporates the main advantages of the finite volume method, i.e., the exact enforcement of mass balance.

The results so far show that in conductive heating from the surface, spalling is produced mainly by thermal stresses, since the pore pressure induced does not exceed about 10 atm., which by itself is not sufficient to cause a break up of concrete. For microwave heating, by contrast, the simulations show a that the maximum pore pressures generated can be about 10-times larger (about 100 atm), which suffices by itself to cause spalling.

PLANNED ACTIVITIES

In the chemical subtask, the behavior of the four radionuclides, after thermal treatment of cement, will be followed using additional extractants composed of common chelating agents including EDTA, DTPA, NTA, EDDHA, oxalic acid, and citric acid as well as other specific chemical agents including acetic acid, carbonic acid, sodium chloride, and hydrochloric acid. The potential influence of concrete aggregate types, including both siliceous and carbonaceous rock, on the volatility and extraction behavior of these radionuclides will also be examined.

Concomitant measurements of cement compressive strength will be obtained to correlate with the observed chemical changes mediated by thermal treatment.

In the mechanics and thermodynamics subtask, the main focus is now on the simulation of the spalling fracture in an objective (fracture energy based) manner. The crack band model, previously developed by Bazant, is being used for this purpose. The nonlinear triaxial behavior of concrete is simulated by microplane model M4, which is being extended to fracture so as to allow the use of finite elements of varying sizes. It is expected that complete simulations of the concrete layer to various regimes of heating will become available soon.

INFORMATION ACCESS

Complete details of the poster, presented at the EMSP National Workshop in Atlanta in April, are available through the EMSP workshop web site (http://emsp.osti.gov/em52/NWS2000_Posters/id64896.pdf) under project number 64896. Detailed information on radionuclide volatilization during heating and subsequent chemical extraction of cement, as well as radionuclide and chemical modeling of soil, can be obtained at http://www.ornl.gov/LANGMUIR/bpsintro.htm. A research report has been submitted for publication in the chemical literature summarizing the volatility of radionuclides during heating and their subsequent extraction behavior from the thermally-altered Portland cement.

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