The Expanded Capabilities of the Cementitious Barriers Partnership Software Toolbox Version 2.0 – 14331

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ABSTRACT

The Cementitious Barriers Partnership (CBP) Project is a multi-disciplinary, multi-institutional collaboration supported by the U.S. Department of Energy (US DOE) Office of Tank Waste Management. The CBP program has developed a set of integrated tools (based on state-of-the-art models and leaching test methods) that help improve understanding and predictions of the long-term structural, hydraulic and chemical performance of cementitious barriers used in nuclear applications. The CBP Software Toolbox – "Version 1.0" was released early in FY2013 and was used to support DOE-EM performance assessments in evaluating various degradation mechanisms that included sulfate attack, carbonation and constituent leaching. The sulfate attack analysis predicted the extent and damage that sulfate ingress will have on concrete vaults over extended time (i.e., > 1000 years) and the carbonation analysis provided concrete degradation predictions from rebar corrosion. The new release "Version 2.0" includes upgraded carbonation software and a new software module to evaluate degradation due to chloride attack. Also included in the newer version are a dual regime module allowing evaluation of contaminant release in two regimes – both fractured and un-fractured. The integrated software package has also been upgraded with new plotting capabilities and many other features that increase the "user-friendliness" of the package. Experimental work has been generated to provide data to calibrate the models to improve the credibility of the analysis and reduce the uncertainty. Tools selected for and developed under this program have been used to evaluate and predict the behavior of cementitious barriers used in near-surface engineered waste disposal systems for periods of performance up to or longer than 100 years for operating facilities and longer than 1000 years for waste disposal. The CBP Software Toolbox is and will continue to produce tangible benefits to the working DOE Performance Assessment (PA) community.

INTRODUCTION

The Cementitious Barriers Partnership (CBP) (http://cementbarriers.org/) is a multi-disciplinary, multi-institutional collaboration supported by the United States Department of Energy (US DOE) Office of Tank Waste Management. The objective of the CBP is to develop a set of tools to improve understanding and prediction of the long-term structural, hydraulic, and chemical performance of cementitious barriers used in nuclear applications. The participating institutions are Savannah River National Laboratory, Vanderbilt University and Consortium for Risk Evaluation with Stakeholder Participation (CRESP) III, Hans van der Sloot Consultancy, Nuclear Research and Consultancy Group (NRG), U.S. National Institute of Standards and Technology

(NIST), SIMCO Technologies Inc., U.S. DOE, and U.S. NRC.

CBP research and development efforts are organized around a set of reference cases [1] that provide the following functions: (i) a common set of system configurations to illustrate the methods and tools developed by the CBP, (ii) a common basis for evaluating methodology for uncertainty characterization, (iii) a common set of cases to develop a complete set of parameter and changes in parameters as a function of time and changing conditions, and (iv) a basis for experiments and model validation, and (v) a basis for improving conceptual models and reducing model uncertainties. These reference cases include the following two reference disposal units and a reference storage unit: (i) a cementitious low activity waste form in a reinforced concrete disposal vault, (ii) a concrete vault containing a steel high-level waste tank filled with grout (closed high-level waste tank), and (iii) a spent nuclear fuel basin during operation. Each case provides a different set of desired performance characteristics and interfaces between materials and with the environment. Experimental and model simulation efforts that support Version 1 and 2 of the CBP Toolbox have focused on external sulfate attack, chloride attack, carbonation, dual regime flow and primary constituent leaching phenomena [2-11].

Based on these components, the CBP has developed a suite of software for simulating reactive transport in cementitious materials and certain degradation phenomena named the “CBP Software Toolbox” [8,9,12,13]. The primary software components are LeachXS™ / ORCHESTRA, STADIUM®, and a GoldSim interface for probabilistic analysis of selected degradation scenarios. The current version supports analysis of external sulfate attack (including damage mechanics), carbonation, and primary constituent leaching. The LeachXS™ component embodies an extensive material property measurements database with emphasis on cementitious materials used in DOE facilities, such as Saltstone (DOE Savannah River site) and Cast Stone (DOE Hanford site), tank closure grouts, and barrier concretes. The US EPA Leaching Environmental Assessment Framework (LEAF) (http://vanderbilt.edu/leaching/) test methods are also applicable to soils remediation and evaluation of alternative waste forms and treatment process effectiveness.

In FY2013-FY2014, the CBP demonstrated continued tangible progress toward fulfilling the objective of developing a set of software tools to improve understanding and prediction of the long-term structural, hydraulic and chemical performance of cementitious barriers used in nuclear applications. In November 2012, the CBP released “Version 1.0” of the CBP Software Toolbox, a suite of software for simulating reactive transport in cementitious materials and important degradation phenomena. In addition, the CBP completed development of new software for the “Version 2.0” Toolbox to be released in early FY2014 and demonstrated use of the Version 1.0 Toolbox on DOE applications.

The current primary software components in both Versions 1.0 and 2.0 are LeachXS/ ORCHESTRA (LXO), STADIUM, and a GoldSim interface for probabilistic analysis of selected degradation scenarios as shown in Figure 1. The CBP Software Toolbox Version 1.0 supports analysis of external sulfate attack (including damage mechanics), carbonation, and primary constituent leaching. Version 2.0 includes the additional analysis of chloride attack and dual regime flow and contaminant migration in fractured and non-fractured cementitious material as shown in Figure 2.0 [14,15].

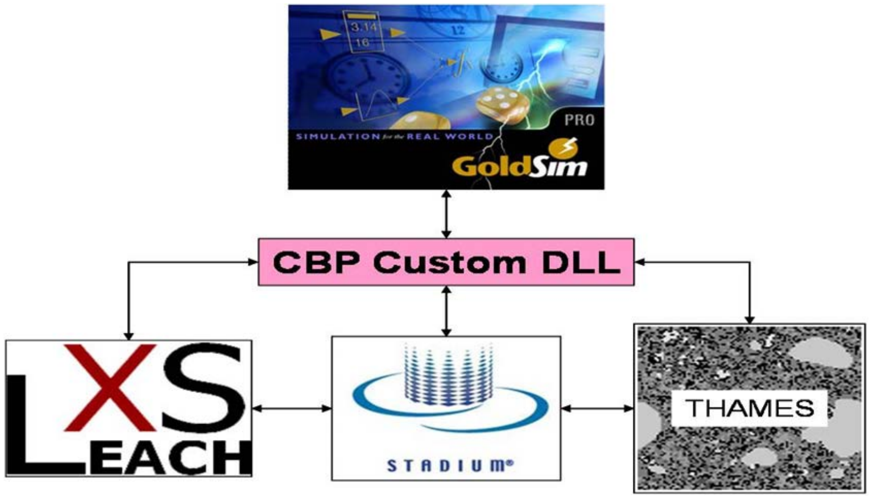

Fig. 1. CBP Software Toolbox Software

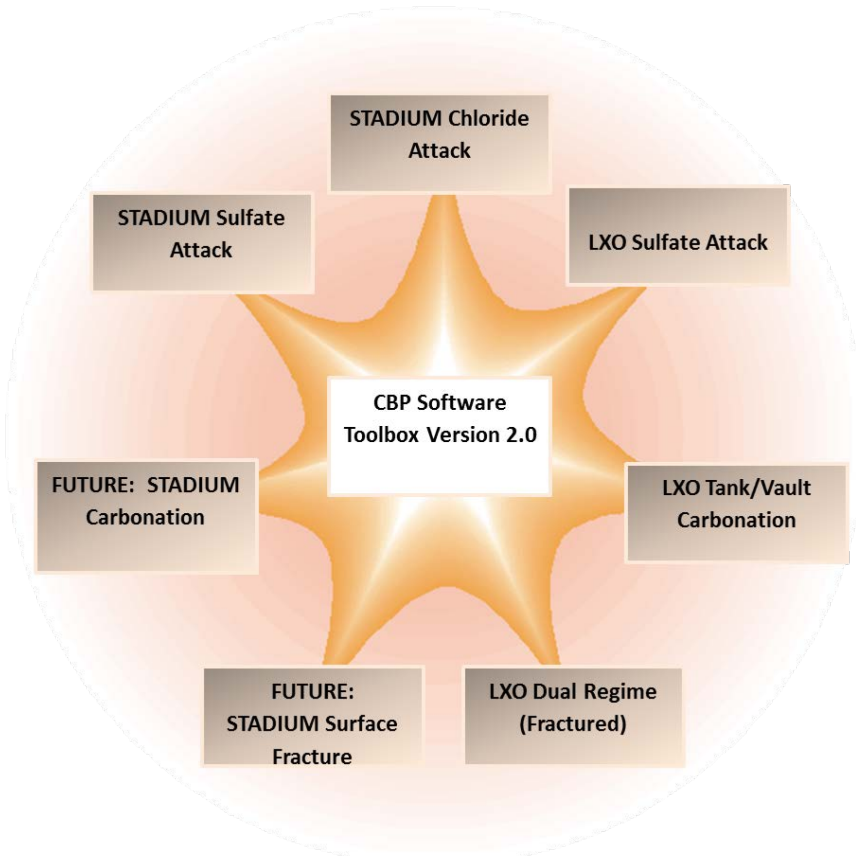

Fig. 2. CBP Software Toolbox Software Modules in Version 2.0

CBP SOFTWARE TOOLBOX DEMONSTRATIONS

The CBP Software Toolbox has produced tangible benefits to the DOE Performance Assessment (PA) community. In FY2013, the CBP Software Toolbox software components were used to produce two major evaluations: one in support of the Saltstone Disposal Facility (SDF) at Savannah River Site (SRS) and the other for a representative Hanford High Level Waste (HLW) Tank [14,16]. The results of the SDF analysis on sulfate attack and carbonation degradation were used directly to support a DOE Performance Assessment Special Analysis. These key analyses are presented in this section, along with, CBP Toolbox enhancements and the developmental efforts of the several new software tools.

CBP Software Toolbox Demonstration on the Saltstone Disposal Facility

The CBP Software Toolbox Version 1.0 was utilized on DOE applications by evaluating the performance of the DOE Savannah River Site Saltstone Disposal Facility under the most probable degradation mechanisms and consequent release of constituents. The results from this demonstration are being utilized in the modification of the current SDF Performance Assessment (PA) via a DOE Special Analysis.

The SDF disposes of low-level radioactive salt solution originating from liquid waste storage tanks at the site using cementitious materials. Cementitious materials play a prominent role in the design and long-term performance of the SDF. The saltstone grout exhibits low permeability and diffusivity, and thus represents a physical barrier to waste release. The waste form is also reducing, which creates a chemical barrier to waste release for certain key radionuclides, notably Tc-99. Similarly, the concrete shell of an SDF disposal unit (SDU) represents an additional physical and chemical barrier to radionuclide release to the environment. Together the waste form and the SDU compose a robust containment structure at the time of facility closure. However, the physical and chemical state of cementitious materials will evolve over time through a variety of phenomena, leading to degraded barrier performance over timescales of thousands of years.

Previous studies of cementitious material degradation in the context of low-level waste disposal have identified sulfate attack, carbonation influenced steel corrosion, and decalcification (primary constituent leaching) as the primary chemical degradation phenomena of most relevance to SRS exposure conditions. In this study, using the CBP Software Toolbox, degradation time scales for each of these three degradation phenomena were estimated for the SDF. The combined effects of multiple phenomena were then considered to determine the most limiting degradation time scale for each cementitious material (i.e., vault and waste form). The CBP Software Toolbox was used to provide estimates of the degradation times using the numerical simulation codes in the Toolbox (http://cementbarriers.org).

CBP Analysis of a USDOE High-Level Waste Tank Closure Scenario using an Upgraded Carbonation Module

The CBP is focused on reducing uncertainties in current methodologies for assessing cementitious barrier performance and increasing the consistency and transparency in the assessment process. One important set of US Department of Energy challenges is assessing the integrity and closure of the high-level waste tanks that currently store millions of gallons of highly radioactive wastes. Many of these tanks are decades past their design lives, have leaked or been overfilled, and must be emptied and closed to satisfy regulatory agreements. Carbonation-induced corrosion has been identified as a primary degradation and possible failure mechanism for the HLW tanks prior to closure. After closure, the synergistic impact of carbonation and oxidation may be to increase the release and transport of contaminants of concern.

In Version 2.0 of the Toolbox, the CBP has upgraded the LeachXS™/ ORCHESTRA carbonation module originally in Version 1.0 and used it to evaluate a representative HLW tank closure scenario including the potential impacts of carbonation on waste tanks prior to and post closure (see Figure 3.0). CBP software tools, including LeachXS™/ORCHESTRA, are being used to simulate waste tank carbonation, major constituent leaching and contaminant releases to evaluate the source term and near-field conditions. The performance of the closed tanks over centuries, if not millennia, must be assessed to evaluate the potential release of residual radionuclides to the environment. Simulations included sensitivity analyses for uncracked concrete to varying input parameters to evaluate sensitivity of the resulting predicted carbonation results. Input parameters included composition, soil-gas CO2 concentration, concrete saturation, porosity, CO2 effective diffusivity, mineral set, and thermodynamic parameters. The results indicated that carbonation to pH 9 (from depassivation of embedded steel) requires at least on the order of 700 years and possibly up to 7000 years based on a probabilistic analysis on the modeling.

This CBP analysis of a HLW Tank provides an efficient method for assessing effectiveness of current closure grouts and designing of future grouts. Future work will ensure longer simulation times and consideration of gas exchange reactions (e.g. CO2 and O2) to provide useful additional information.

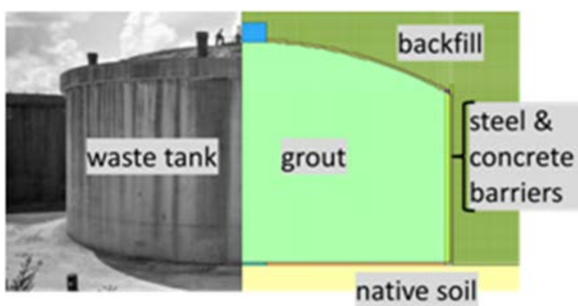

Fig. 3. CBP Analysis of a Representative USDOE High-Level Waste Tank

CBP SOFTWARE TOOLBOX DEMONSTRATIONS

CBP Dual Regime Module Development

In FY2013, a reactive transport model was developed as part of the CBP to simulate the release of radionuclides from grout that is both in-tact and fractured and to estimate the evolution of pH. In tank closure, as well as, any cementitious closure facility where grout is used to stabilize radionuclide species, the grout can be assumed to have varying extents of cracking. The partially or completely degraded grouted tank is idealized as a “dual regime” system as shown in Figure 4 comprised of two regimes:

1. A mobile region with cracks and macropores, and
2. An immobile/ stagnant region comprising of the solid matrix with micropores.

The transport profiles of the species are calculated by incorporating advection of species through the mobile region, diffusion of species through the immobile/stagnant region, and exchange of species between the mobile and immobile regions. A geochemical speciation code in conjunction with the pH dependent test data for a grout material is used to obtain a mineral set that best describes the trends in the test data of the major species (see Figure 5). The dual regime reactive transport model predictions are compared with the release data from an up-flow column percolation test. The coupled model is then used to assess effects of crack state of the structure, rate and composition of the infiltrating water on the pH evolution at the grout-waste interface. The coupled reactive transport model developed in this work can be used as part of the performance assessment process for evaluating potential risks from leaching of a cracked tank containing elements of human health and environmental concern.

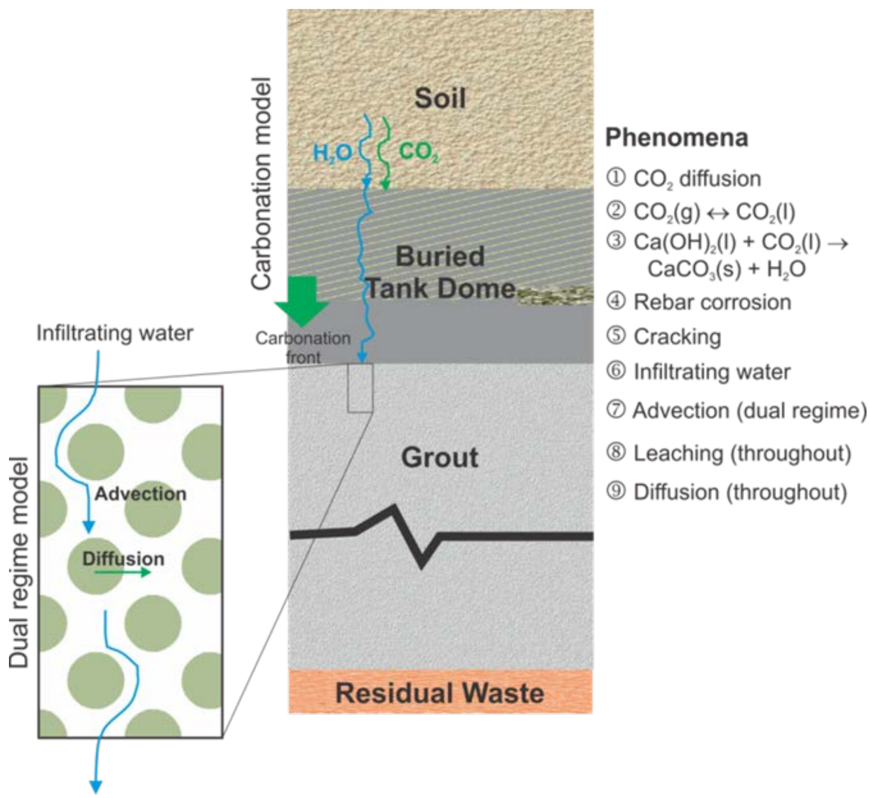

Fig. 4. Dual-Regime System

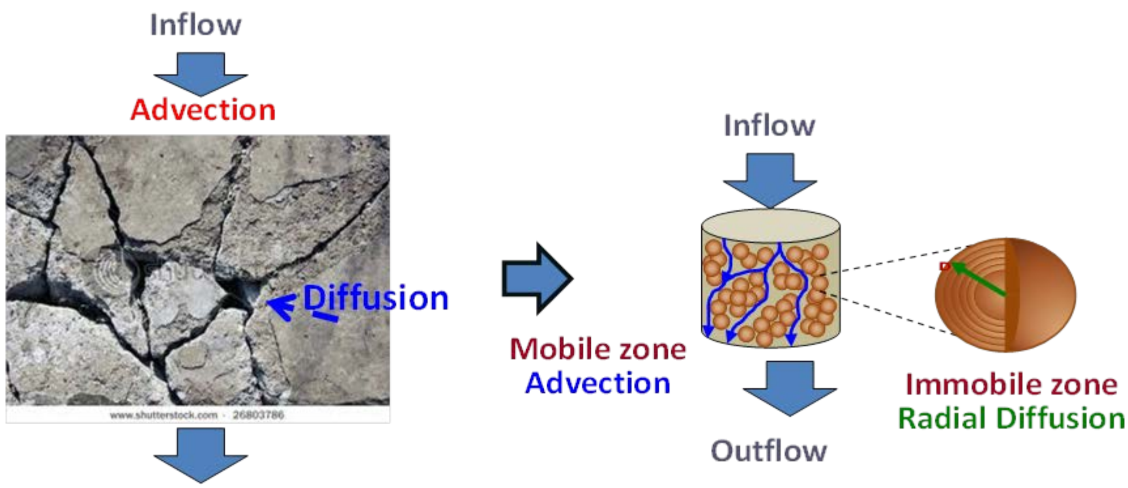

Fig. 5. Model Framework of Fractured and Solid Matrix in a Dual Regime System

CBP Chloride Attack Module

The CBP Software Toolbox, Version 2.0, includes a STADIUM chloride attack module to predict chloride ingress and corrosion initiation predictions. STADIUM uses time-step finite element analysis to simulate the progress of harmful ions such as chloride and hydroxide through concrete, by considering the chemical and physical properties of the concrete being analyzed. The STADIUM® Chloride Attack module software simulates the physical and chemical changes in concrete as it reacts with its environment, taking into account concrete variables such as chemical composition, permeability, ion diffusion coefficients, moisture transport coefficients, tortuosity, and many other factors, as well as exposure conditions such as ambient humidity, temperature, and chemical aggression. This added software module provides a method to accurately predict the degradation of cementitious materials exposed to chloride and other harmful ions.

CBP SOFTWARE TOOLBOX ENHANCEMENTS

CBP Software Toolbox Version 2.0 Code Integration Enhancements

In FY2013, enhancements were made to the CBP Software Toolbox Version 1.0, specifically the code integration functionality aspects of the Toolbox (Ref.17). The most significant enhancements were:

1) Improved graphical display of model results – The CBP Toolbox has been modified with improved methods for displaying results including two and three-dimensional plotting capabilities as shown in Figure 6.
2) Improved error analysis and reporting – In previous years, the CBP developed a custom Dynamic Link Library (DLL) to enable data communication between the Toolbox software components, allowing comparative analysis. Recently, error messages have been enhanced to more clearly explain problems encountered during execution of the DLL instruction files. The additional information will allow the User to quickly

identify the problem and take corrective actions.

3) Increase in the default maximum model mesh size from 301 to 501 nodes – Modifications to increase the mesh size provides a finer resolution necessary to operate the multi-layers within the STADIUM model. The finer resolution will improve the distinctions between the layers.
4) The ability to designate the LeachXS™/ORCHESTRA simulation time frames through the GoldSim interface – The original version of the CBP Software Toolbox did not allow the User to change the degradation simulation time of the cementitious barrier. Now the Goldsim User Interface (GUI) has been modified for LXO so that the User can determine the time frames desired or dictated by the performance assessment.

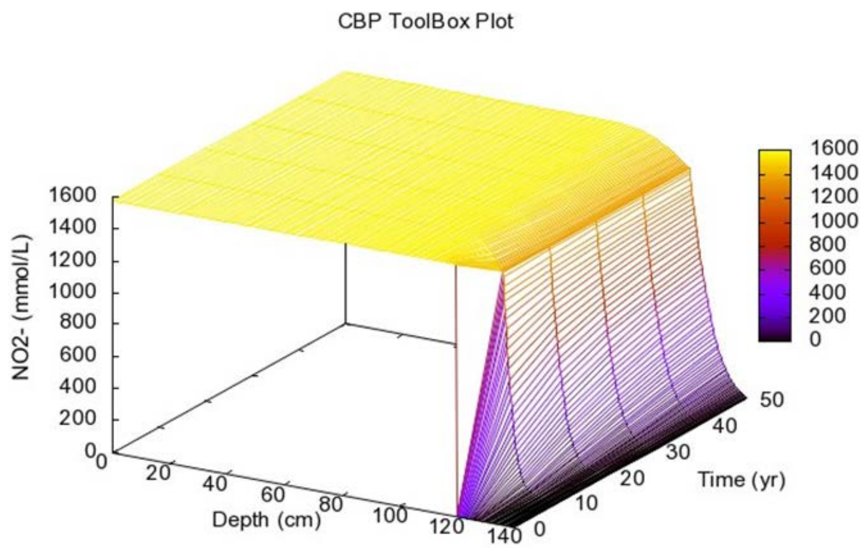

Fig. 6. Three-dimensional surface plot of nitrate concentration from a 2-layer model

CONCLUSIONS

The CBP Software Toolbox has been used to provide important technical insights to the DOE PA process regarding sulfate attack on the DOE Saltstone Disposal Facility and carbonation on a typical high-level waste tank. Current development efforts in the areas of carbonation, transport in fractured and intact media, and chloride attack have resulted in the second release of the Toolbox. Future development efforts on gas and liquid phase oxidation of ground blast furnace slag and Tc-99 mobility, damage mechanics, and flow and transport in fractured cementitious materials will enable further tangible contributions to DOE PAs and future upgrades of the CBP Toolbox.

Since the 2009 CBP review of DOE PAs, the influence of carbonation on the corrosion rate of steel embedded within cementitious materials has been identified by the CBP as another critical need for improved predictive capabilities. Embedded steel occurs in the common form of reinforcing bar (rebar) material, and as the primary tank liner in the context of DOE liquid radioactive waste tank closures. Damage to cementitious barriers and waste forms is considered a primary degradation phenomenon in many DOE applications. In response, development efforts to add carbonation modules to the CBP Software Toolbox began in 2012 and the initial Toolbox release includes one based on LeachXS™/ORCHESTRA. Refinement of that module was realized in the Version 2.0 release and addition of a STADIUM®-based module is anticipated in FY2014.

Recognizing that physical damage to cementitious materials typically occurs in the form of cracking, ongoing CBP development efforts are also focused on predicting damage through fracture mechanics considerations, determining the hydraulic and transport properties of fractured materials, and implementing corresponding Toolbox simulation capabilities. Version 2.0 includes software that differentiates transport through a dual regime of fractured and intact cementitious material. Future versions of the Toolbox anticipated in FY2014 will include software that calculates the unsaturated hydraulic properties of fractured and in-tact media.

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