

Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Characterization of Unsaturated Hydraulic Conductivity in Fractured Media Using the Multistep Outflow Method – 15461

Greg Flach *, Kenneth Dixon *, Ralph Nichols *

* Savannah River National Laboratory

ABSTRACT

The multistep outflow method is routinely used to characterize the unsaturated hydraulic conductivity of soils. The technique involves placing a soil sample in a pressure plate apparatus, subjecting the sample to multiple gas pressures in discrete steps through time, and measuring the transient volume of pore fluid extracted. Unsaturated hydraulic property values are estimated through inverse modeling of the experimental conditions. In this study the multistep outflow concept was applied to micro-cracked cementitious materials to assess the efficacy of the technique for these materials. The cementitious materials tested were salt-waste simulant grout samples artificially damaged through oven-drying.

Compared to typical soils, fractured media exhibit higher saturated conductivity and lower air-entry pressure, and the volume of fluid extractable from fractures is much lower than soil porosity. To accommodate these material differences the standard test apparatus was modified to incorporate a higher conductivity ceramic pressure plate, a high-precision digital balance for logging outflow mass, a low volume (diameter) effluent line, multiple inline high-precision gas regulators, and a high-precision low-range pressure gauge. Testing to date indicates that the modified apparatus can provide a viable means to measure the unsaturated hydraulic properties of micro-fractured cementitious materials. However the accuracy/uniqueness of inverse modeling is limited by the inherent characteristics of fractured media: high saturated conductivity, low air-entry pressure, and strong non-linearities. Hydraulic property results in the form of van Genuchten / Mualem curves are presented for three fractured grout specimens.

DOE Performance Assessments often involve cementitious barriers and/or waste forms that are predicted or assumed to degrade over time due to various mechanisms such as carbonation-influenced reinforcing steel corrosion, external sulfate attack, differential settlement, and seismic activity. Physical degradation typically takes the form of small-scale cracking / fracturing, and the affected materials reside in unsaturated hydrogeologic zones. In these cases, unsaturated hydraulic properties are needed for fractured cementitious materials to simulate moisture movement and contaminant transport within and around the facility. The outflow extraction method, as implemented in the present study, provides a suitable method for estimating these material properties.

INTRODUCTION

DOE Performance Assessments (PAs) and augmenting Special Analyses (SAs) often involve analysis of cementitious waste forms and barriers that are known or postulated to physically degrade over long time periods, typically extending thousands to tens of thousands of years into the future (e.g. [1], [2], [3], [4]). Degradation mechanisms may include differential settlement, seismic events, freeze-thaw cycling, steel corrosion influenced by carbonation and/or chloride ingress, alkali-silica reaction (ASR), and sulfate attack (e.g. [5], [6], [7]). The physical damage resulting from local-scale degradation mechanisms is commonly assumed to take the form of small-scale cracking. Unsaturated hydraulic properties must be defined for fractured cementitious materials in order to simulate moisture movement and contaminant transport within and around the facility.

Fractured media flow can be represented by a number of modeling concepts. Recent Savannah River Site (SRS) PAs and SAs conceptualize fractured cementitious materials as homogeneous continua composed of blended (or superimposed) fractions of intact matrix and fractures ([8], [9], [10], [11]). Local hydrologic equilibrium is assumed between matrix and fractures, such that a single pair of water retention and relative permeability curves describes the behavior of the composite material. This construct has been identified as the *Composite Porosity (Equivalent Continuum)* or *Equivalent Matrix and Fracture Continuum* modeling approach (Figure 2-2 of [12]). The concept is most appropriate for smaller scale fractures and steady-state flow conditions, such that local equilibrium occurs between fractures and matrix.

The composite porosity concept leads to a two-tier or double-hump relative permeability (and hydraulic conductivity) curve as a function of capillary pressure/head. The shape of the composite curve is a natural result of blending individual characteristic curves (describing matrix and fracture regions) that have distinctly different saturated conductivities and air-entry pressures. For further information and blending equations see Section 2.1 of Reference [9] for example. The characteristic curves for the fracture region may be estimated from a mechanistic model of capillary water retention and film flow for an idealized fracture geometry (e.g. [13], [14]). Alternatively, Reference [15] states that “theoretical and experimental work suggests that relative permeability and capillary pressure behavior of fractures is similar to that of highly permeable media with intergranular porosity ... Accordingly, we used the customary van Genuchten correlations, with parameters chosen as for coarse sands.” [16], [17], [18]. Thus high permeability granular materials are reasonable surrogates for natural fractures with rough surfaces and/or infilled with granular material. The van Genuchten [19] / Mualem [20] functional forms are commonly adopted to define the individual characteristic curves, but that is not required. Figure 1 illustrates the two-tier or double-hump nature of composite porosity hydraulic curves using two material sets and multiple blending fractions (labeled “Degradation fraction”) as specific examples taken from a recent SRS Saltstone Special Analysis ([9], [2]). Increasing levels of damage are represented by an increasing blending fraction.

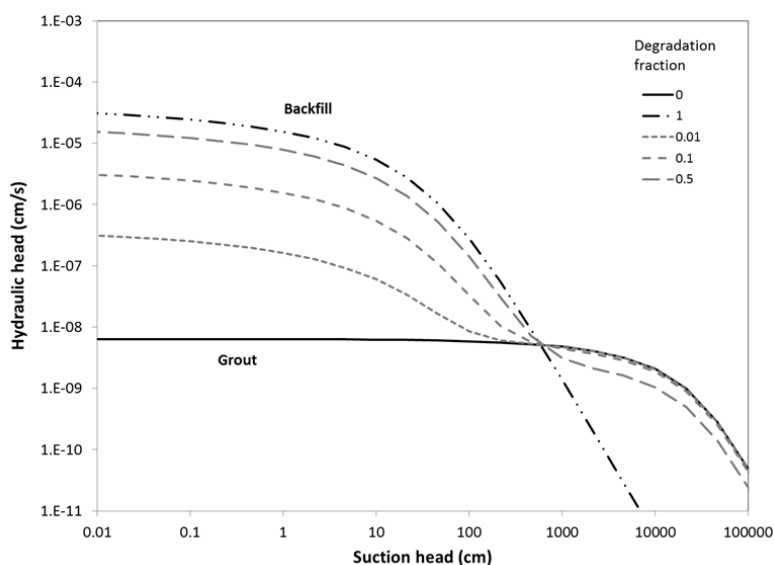


Fig. 1. Two-tiered unsaturated hydraulic conductivity variations resulting from the *Composite Porosity* concept; grout and backfill soil end-members example.

While theoretical functions based on idealized configurations or high-permeability surrogate materials produce the desired qualitative behavior of fractures, experimental datasets that might be used to quantitatively validate these or develop new functions are difficult to obtain [21] and “virtually nonexistent” ([14], [22]). Reference [23] recently identified only References [16], [24] and [25] as experimental data reported in the literature. Reference [16] constitutes a numerical experiment and the latter studies involve two-phase air-water flow between nominally flat plates.

Recognizing the need for an experimental technique to measure the unsaturated hydraulic properties of unsaturated fractured media, SRNL researchers [26] recently applied the multistep outflow method concept ([27], [28], [29]) to fractured cementitious materials. The multistep outflow method involves monitoring the transient flow of water extracted through a high air-entry pressure porous plate in response to step increases in gas pressure. Water retention and relative permeability curve (e.g. van Genuchten) parameters are determined through inverse fitting of a Richards equation flow simulation model to the transient outflow data. HYDRUS-1D [30] is commonly chosen as the inverse modeling software. The technique has become well established for characterizing soils, although parameter uniqueness and uncertainty are issues inherent with inverse modeling and obtaining accurate measurements is a persistent challenge.

Compared to typical soils, fractured media exhibit higher saturated conductivity and lower air-entry pressure, and the volume of fluid extractable from fractures is much lower than pore volumes. To accommodate these material differences, Reference [26] modified a multistep outflow method apparatus suitable for soils testing by incorporating a higher conductivity ceramic pressure plate, a high-precision digital balance for logging outflow mass, a low volume (diameter) effluent line, multiple inline high-precision gas regulators, and a high-precision low range pressure gauge. Testing [26] with SRS Saltstone grout samples suggests that the modified apparatus/method can provide a viable means to measure the unsaturated hydraulic properties of micro-fractured cementitious materials. However the accuracy and uniqueness of inverse modeling results are limited by inherent characteristics of fractured media: high saturated conductivity, low air-entry pressure, and strong non-linearities. Furthermore, the test method has not been rigorously validated against a known standard.

In this study, additional testing on micro-cracked saltstone samples is reported, and the previous experimental data are further analyzed along with the present data using multiple inverse model fitting approaches. More complete information is provided in Reference [31].

EXPERIMENTAL APPARATUS AND PROCEDURE

Reference [26] fully describes the experimental apparatus and procedure used to test fractured grout samples, and note some of the complications in using the method to investigate the hydraulic properties of fractured media. A brief description of the apparatus and test method are included herein for completeness.

The outflow extraction system (Figure 2) consists of the outflow extractor, a peristaltic pump, and a high precision balance to monitor effluent from the extractor. The outflow extractor contains a porous ceramic that provides the interface between the applied pressure inside the extractor and ambient atmospheric pressure outside the extractor. The sample rests inside the extractor on the surface of the porous ceramic plate. For the purposes of this testing, a high flow porous ceramic plate with an air entry pressure of 1000 cm H₂O was selected. The saturated hydraulic conductivity of the plate is reported by the manufacturer (Soil Moisture, Inc.) to be 8.6E-06 cm/s. Prior to the start of each test, the plate was saturated with de-aired water. This was accomplished by pressurizing a known mass of de-aired water in the chamber. This allowed the de-aired water to flow through the plate until the chamber was empty thereby saturating the plate. Care

was taken not to exceed the air entry pressure of the plate during this process. Outflow was monitored on the digital balance to determine when the bulk of the water had been pushed through the plate.



Fig. 2. Experimental apparatus for outflow extraction method.

The outflow extractor consists of a single inlet port for pressure application. The outlet side of the extractor consists of two ports, one which is used to drain effluent from the sample and a secondary port that is used to remove air from beneath the ceramic plate prior to testing. Using an arrangement of valves, the two outlet ports, and a peristaltic pump, de-aired water was circulated beneath the ceramic plate to remove entrapped air. Once this process was completed, a valve was used to close the secondary outlet port. Small diameter tubing was connected to the primary outlet port. The end of this tubing was maintained at a height equal to the bottom of the sample. Outflow experiments were carried out by pressurizing the extractor using compressed air. Several pressure increments were used in each transient test ranging from about 5 to 100 cm H₂O. These pressures were maintained using a series of high precision multistage gas regulators. Effluent from each sample was monitored using a digital balance and logged directly to a computer every five seconds. Ultimately, the balance served as a drop counter and the mass of each drop of water was converted to volume using a density of 1.00 g/cm³. Cumulative outflow was converted to cumulative flux by dividing the volume of effluent by the total area of the sample.

FRACTURED CEMENTITIOUS MATERIAL TESTING

Three fractured grout samples, identified as TR430, TR436 and TR437, were tested. Micro-fracturing in the specimens was induced by oven drying. The three specimens exhibit different fracture networks. TR430 has a macro-fracture among smaller cracks. TR436 also has a macro-fracture from being broken in half through mechanical loading. The TR437 sample was not broken, but nonetheless has one notably larger fracture among many smaller micro-cracks. Figure 3 is an example radiography image showing the extent of cracking in the TR437 specimen at 25% depth from the top. The saturated hydraulic conductivities of TR436 and TR437 were measured independent of outflow extraction testing with a flexible wall permeameter (ASTM D5084-03); however, the saturated conductivity of TR430 was not measured with this technique.

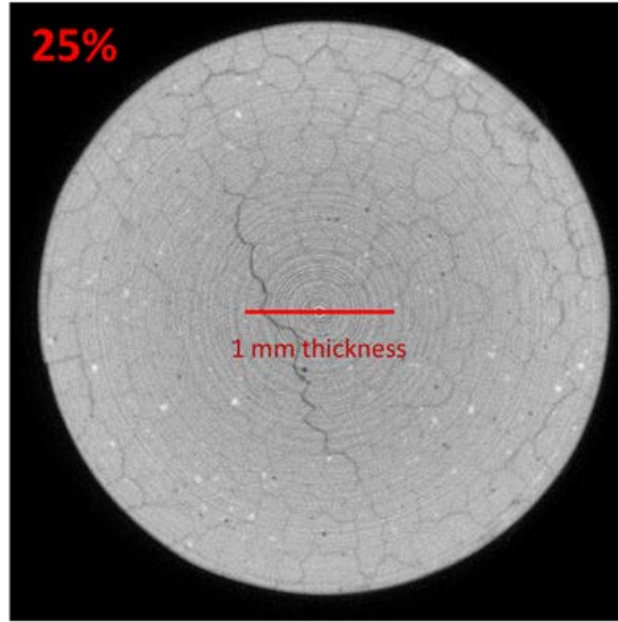


Fig. 3. Radiography image showing fracture network in TR437 at 25% depth from the top surface.

Five outflow extraction experiments were conducted, one each for TR436 and TR437 and three for TR430 ([26], [31]). Figure 4 presents measured cumulative outflow data for TR437. Also shown are simulated cumulative outflow curves based on inverse modeling with HYDRUS-1D for various combinations of fitted van Genuchten [19] / Mualem [20] parameters, defined as follows.

Effective saturation S_e is defined in terms of water content θ , or equivalently saturation S , as

$$S_e \equiv \frac{\theta - \theta_r}{\theta_s - \theta_r} = \frac{S - S_r}{1 - S_r} \quad (\text{Eq. 1})$$

where the subscripts refer to saturated (s) and residual (r) conditions. The van Genuchten functional form for effective saturation is [19]

$$S_e(\psi) = \frac{1}{[1 + (\alpha\psi)^n]^m} \quad (\text{Eq. 2})$$

where ψ is suction or tension head. The Mualem assumption is [20]

$$m = 1 - \frac{1}{n} \quad (\text{Eq. 3})$$

The functional form for relative permeability is

$$k_r(\psi) = S_e^{1/2} \left[1 - \left(1 - S_e^{1/m} \right)^m \right]^2 \quad (\text{Eq. 4})$$

Finally, unsaturated hydraulic conductivity $K(\psi)$ is the product of saturated hydraulic conductivity K_s and relative permeability

$$K(\psi) = K_s k_r(\psi) \quad (\text{Eq. 5})$$

The parameter α controls the air-entry pressure and n controls the slope of conductivity versus tension head in the unsaturated region.

In this study θ_r is assumed to be zero for fractured media, leaving θ_s , α , n and K_s as potential fitting parameters (with consideration of Equation (3)). Figure 5 summarizes the inverse modeling results for four selections of fitted parameters. When K_s is not fitted, its value is set to that measured from flexible-wall permeameter testing for TR436 and TR437. Because K_s was not measured for TR430, this specimen was not subjected to inverse modeling with K_s fixed and thus TR430 curves are absent from Figure 5(a) and (c). For TR436 and TR437 the fitted K_s value is roughly an order of magnitude lower than the independently measured value. However, the estimated values of α and n are similar regardless of whether K_s is fitted. Fitting θ_s , α and n produced unreasonably high values for saturated water content θ_s (fracture volume). Fitting all four parameters produced more parameter variability within and between cases, indicating lower uniqueness and higher uncertainty. The more reliable results are considered to be those shown in Figure 5(a) and (b).

Figure 5 also compares the three fractured grout specimens to two sand-clay soils commonly used in Savannah River PAs and SAs to represent the natural environment and as surrogates for degraded cementitious materials. Discounting Figure 5(c), representing inverse fitting results with excessively large θ_s , and Figure 5(d), representing less reliable parameter estimates and curves, nominal hydraulic conductivity for the fractured cementitious materials decreases with a steeper slope than nominal K for soils (Figure 5(a), (b)). This behavior apparently reflects easier dewatering of fractures compared to intergranular porosity for soils with a fine-grained fraction, as one would expect.

The estimated hydraulic curves a) generally conform to expectations, b) are similar for the three TR430 tests indicating a level of experimental reproducibility, and c) are similar for TR430, TR436 and TR437, indicating a level of consistency across all three specimens which have similar fracture networks. These observations suggest that the outflow extraction method, as adapted herein, is a viable technique for characterizing the unsaturated hydraulic conductivity of micro-fractured cementitious materials. This conclusion is tentative considering the lack of rigorous method validation using a standard specimen with known properties. Furthermore, while the technique appears to be viable for micro-fractured media, which can be expected to behave similar to porous materials at some level, fractured media with larger apertures would probably not be suitable for testing because of extremely low air-entry pressure and high saturated permeability.

Assuming that lower hydraulic conductivity is conservative with respect to facility performance, Figure 5 suggests that soils may serve as conservative surrogates for damaged cementitious materials, provided their saturated conductivities are sufficiently large for the given fracture network.

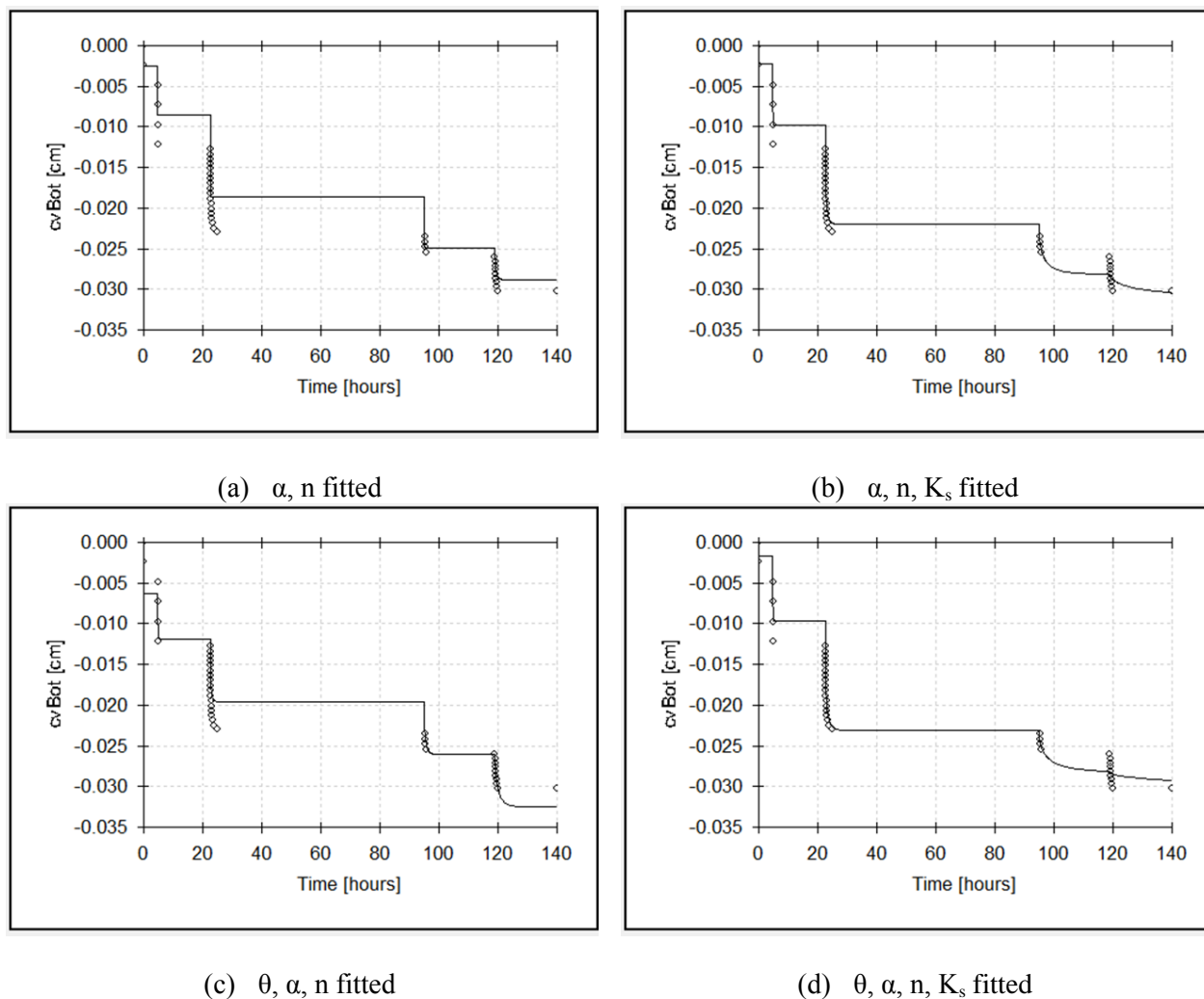


Fig. 4. Measured and simulated cumulative flux for TR437.

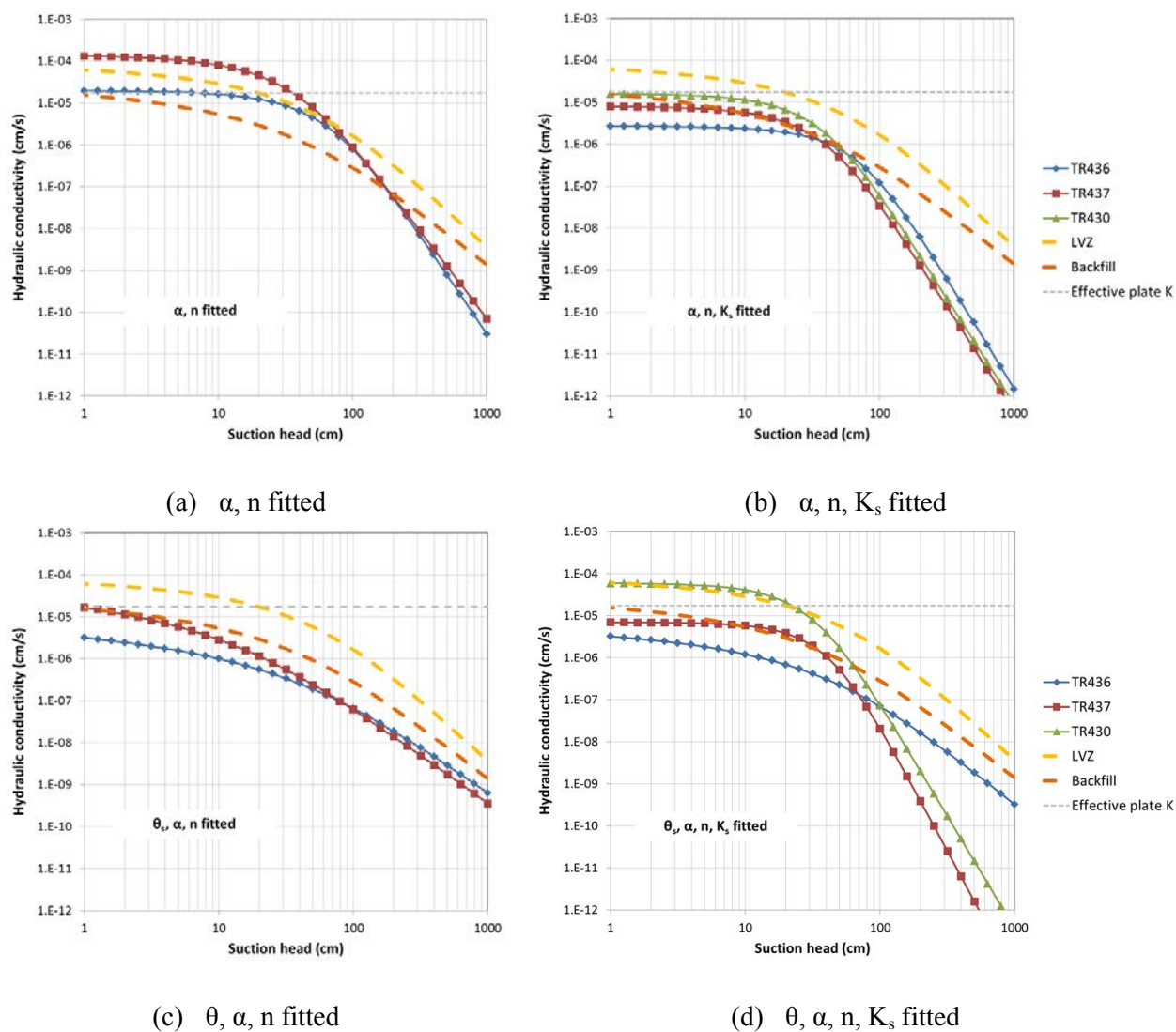


Fig. 5. Unsaturated hydraulic conductivity estimates for fractured grout samples compared to reference Savannah River Site soils.

CONCLUSIONS

An *outflow extraction method* apparatus and procedure, adapted for use with fractured media, were used to measure the unsaturated hydraulic conductivity of three micro-fractured grout specimens through inverse modeling with HYDRUS-1D. The estimated hydraulic curves generally conform to expectations and appear to be reproducible based on repeat testing and similar results being achieved for similar fracture networks. These observations suggest that the outflow extraction method is a viable technique for characterizing the unsaturated hydraulic conductivity of micro-fractured cementitious materials. However, rigorous method validation has not been done to confirm this tentative conclusion. With respect to PA applications, comparisons of the unsaturated conductivity for the micro-fractured grout samples suggests that soils may serve as conservative surrogates for damaged cementitious materials, assuming that higher conductivity at higher tension heads is conservative for facility performance.

REFERENCES

1. SRR Closure & Waste Disposal Authority. *Performance Assessment for the Saltstone Disposal Facility at the Savannah River Site*. SRR-CWDA-2009-00017, Rev. 0. October 2009.
2. Savannah River Remediation LLC. *FY2013 Special Analysis for the Saltstone Disposal Facility at the Savannah River Site*. SRR-CWDA-2013-00062, Rev. 1. June 2013.
3. Savannah River Remediation LLC. *FY2014 Special Analysis for the Saltstone Disposal Facility at the Savannah River Site*. SRR-CWDA-2014-00006, Rev. 1. June 2014.
4. Savannah River Remediation LLC. *Performance Assessment for the H-Area Tank Farm at the Savannah River Site*. SRR-CWDA-2010-00128, Rev. 1. November 2012.
5. Cementitious Barriers Partnership. *Review of Mechanistic Understanding and Modeling and Uncertainty Analysis Methods For Predicting Cementitious Barrier Performance*. CBP-TR-2009-002, Rev.0. November 2009.
6. Langton, C. A. *Chemical Degradation Assessment for the H-Area Tank Farm Concrete Tanks and Fill Grouts*. SRNL-STI-2010-00035, Rev. 0. January 2010.
7. Flach, G. P. and F. G. Smith III. *Degradation Of Cementitious Materials Associated with Saltstone Disposal Units*. SRNL-STI-2013-00118, Rev. 2. September 2014.
8. Jordan, J. M., G. P. Flach and M. L. Westbrook. *PORFLOW Modeling Supporting the H-Tank Farm Performance Assessment*. SRNL-STI-2012-00465, Rev. 0. August 2012.
9. Jordan, J. M. and G. P. Flach. *PORFLOW Modeling Supporting the FY13 Saltstone Special Analysis*. SRNL-STI-2013-00280, Rev. 0. May 2013.
10. Flach, G. P. and G. A. Taylor. *PORFLOW Modeling Supporting the FY14 Saltstone Special Analysis*. SRNL-STI-2014-00083, Rev. 1. April 2014a.
11. Flach, G. P. and G. A. Taylor. *Sensitivity Analysis for Saltstone Disposal Unit Column Degradation Analyses*. SRNL-STI-2014-00505, Rev. 0. October 2014b.
12. Altman, S. J., B. W. Arnold, R. W. Barnard, G. E. Barr, C. K. Ho, S. A. McKenna, and R. R. Eaton. *Flow Calculations for Yucca Mountain Groundwater Travel Time (GWTT-95)*. Sandia Report SAND96-0819. September 1996.
13. Or, D. and M. Tuller. *Flow in unsaturated fractured porous media: Hydraulic conductivity of rough surfaces*. Water Resources Research vol. 36 no. 5. p. 1165-1177, May 2000.

14. Tuller, M. and D. Or. *Unsaturated Hydraulic Conductivity of Structured Porous Media: A Review of Liquid Configuration-Based Models*. Vadose Zone Journal vol. 1. p. 14–37. August 2002.
15. Pruess, K. *On water seepage and fast preferential flow in heterogeneous, unsaturated rock fractures*. Journal of Contaminant Hydrology vol. 30. p. 333–362. 1998.
16. Pruess, K. and Y. W. Tsang. *On Two-Phase Relative Permeability and Capillary Pressure of Rough-Walled Rock Fractures*. Water Resources Research vol. 26 no. 9. p. 1915–1926. 1990.
17. Firoozabadi, A. and J. Hauge. *Capillary pressure in fractured porous media*. J. Pet. Technol. p. 784–791. June 1990.
18. Persoff, P. and K. Pruess. *Two-phase flow visualization and relative permeability measurement in natural rough-walled rock fractures*. Water Resour. Res. vol. 31 no. 5. p. 1175–1186. 1995.
19. van Genuchten, M. T. *A closed-form equation for predicting the hydraulic conductivity of unsaturated soils*. Soil Science Society America Journal vol. 44 no. 5 pp. 892–898. 1980.
20. Mualem, Y. *A new model for predicting the hydraulic conductivity of unsaturated porous media*. Water Resour. Res. vol. 12 no. 3 p. 513–522. doi:10.1029/WR012i003p00513. 1976.
21. Liu, H.-H. and G. S. Bodvarsson. *Constitutive Relations for Unsaturated Flow in a Fracture Network*. Journal of Hydrology vol. 252. p. 116–125. 2001.
22. Monachesi, L. B. and L. Guarracino. *A Fractal Model for Predicting Water and Air Permeabilities of Unsaturated Fractured Rocks*. Transport in Porous Media vol. 90. p. 779–789. doi 10.1007/s11242-011-9815-9. 2011.
23. Li, Y., Y.-F. Chen and C.-B. Zhou. *Hydraulic Properties of Partially Saturated Rock Fractures Subjected to Mechanical Loading*. Engineering Geology vol. 179. p. 24–31. 2014.
24. Chen, C. *Liquid-Gas Relative Permeabilities in Fractures: Effects of Flow Structures, Phase Transformation and Surface Roughness*. Stanford University. 2005.
25. Chen, C.-Y. and R. N. Horne. *Two-Phase Flow in Rough-Walled Fractures: Experiments and a Flow Structure Model*. Water Resources Research vol. 42. W03430, doi:10.1029/2004WR003837. 2006.
26. Dixon, K. L. and R. L. Nichols. *Method Development for Determining the Hydraulic Conductivity of Fractured Porous Media*. SRNL-STI-2013-00522, Rev. 0. September 2013.
27. Kool, J. B. and J. C. Parker. *Analysis of the Inverse Problem for Transient Unsaturated*

28. Eching, S. O. and J. W. Hopmans. *Optimization of Hydraulic Functions from Transient Outflow and Soil Water Pressure Data*. Soil Science Society of America Journal vol. 57 no. 5 p. 1167-1175. 1993.
29. Hopmans, J. W., J. Šimůnek, N. Romano and W. Durner. *Inverse Modeling of Transient Water Flow, In: Methods of Soil Analysis, Part 1, Physical Methods, Chapter 3.6.2*. Eds. J. H. Dane and G. C. Topp. Third edition. SSSA. Madison, WI. p. 963-1008. 2002.
30. Šimůnek, J., M. Th. van Genuchten, and M. Sejna. *The Hydrus-1D Software Package for Simulating the One-Dimensional Movement of Water, Heat, and Multiple Solutes in Variably-Saturated Media, Version 4.14*. Department of Environmental Sciences, University of California-Riverside, Riverside, CA, 92521. 2008.
31. Flach, G. P., K. L. Dixon, F. G. Smith III and R. L. Nichols. *Characterization of Unsaturated Hydraulic Conductivity in Fractured Media Using the Multistep Outflow Method*. SRNL-STI-2014-00618, Rev. 0. 2015.