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Dynamic Leaching Method for Intact Saltstone Samples – 18495

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ABSTRACT

At the Savannah River Site (SRS) low activity salt solution is stabilized via encapsulation within a grout termed saltstone. Saltstone is emplaced into large (multi-million liter) concrete storage containers referred to as Saltstone Disposal Units (SDUs). Technetium-99 (Tc-99) and iodine-129 (I-129) are long-lived radionuclides contained in the low activity salt waste and subsequently incorporated into the grout waste form: both radionuclides are contributors to the potential long-term radiation exposure of the public and environment surrounding the SDUs. With respect to Tc-99, in the reducing, high pH environment within the grout, Tc-99 is expected to be relatively immobile since it exists in a reduced Tc(IV) oxidation state in the form of sparingly soluble hydrated oxides ($TcO_2.xH_2O$) or sulfides (TcS_x). However, in the presence of O_2 (associated with the future infiltration of air or oxygenated ground waters into the saltstone monolith) it is possible for redox-sensitive Tc(IV) to transition into highly soluble (and mobile) Tc(VII) species, such as pertechnetate (TcO_4^-), which are more readily transported to the surrounding environment. In contrast, I-129 is highly soluble and its leaching behavior is not anticipated to be significantly impacted by potential changes in redox environment or pH within saltstone.

Traditional approaches to quantifying the leaching behavior of Tc-99 and I-29 from cementitious matrices have involved partitioning experiments using size-reduced (crushed/ground) samples, and determination of the radionuclide fraction immobilized by the cementitious solids. Such experiments create artificially high solid-liquid contact areas that likely result in higher Tc-99 and I-129 leachate concentrations than would be expected for intact, monolithic samples. In the current study a new technique, termed the Dynamic Leaching Method (DLM), is being used to initially investigate the Tc-99 leaching behavior from monolithic saltstone samples; the system will be used at a later date to evaluate the leaching characteristics of I-129. The data derived using this technique is intended to inform the SRS Saltstone Disposal Facility (SDF) Performance Assessment (PA) which models the long-term transport of radionuclides from the SDUs to the environment. The DLM utilizes a flexible-wall permeameter to achieve saturated leaching under an elevated hydraulic gradient to simulate and accelerate the future transport of groundwater through saltstone. Leachates are regularly analyzed with respect to radionuclide concentrations, key constituent concentrations (e.g., Al, Ca, Mg, Na, Si, CO_3^{2-}, NO_2^-, NO_3^-, SO_4^{2-}), pH, and reduction potential (E_h) in order to develop an understanding of how chemical changes associated with the pore solution influence grout weathering and radionuclide leaching behavior. This paper provides a brief update with respect to data derived from preliminary Tc-99 leaching evaluations as the DLM was being developed, but the paper is predominantly focused on new sample data in which the DLM technique, pre- and post-DLM saltstone sample characterization, and leachate analysis protocols have been optimized.

INTRODUCTION

The Savannah River Site (SRS) is a U.S. Department of Energy (DOE) facility currently storing 133 million liters (35 million gallons) of Cold War legacy, high-level waste (HLW) with a combined activity of 9.73E+12 megabecquerels (MBq) (263 million Curies). This waste is stored in 43 active underground tanks and will ultimately be dispositioned via vitrification. However, the treatment of HLW at SRS will generate approximately 380 million liters (100 million gallons) of low activity salt solution containing less than 0.1% of the total SRS radionuclide inventory. The salt solution is dispositioned at the Saltstone Disposal Facility (SDF) via combination with a mixture of ground-granulated blast furnace slag

(GGBFS), Class F fly ash (FA), and Type II ordinary Portland cement (OPC) at the Saltstone Production Facility (SPF) to form a flowable grout referred to as saltstone. Once mixed the radioactive grout is transferred to concrete vaults, termed Saltstone Disposal Units (SDUs) (Fig. 1), where it subsequently cures and encapsulates the waste.

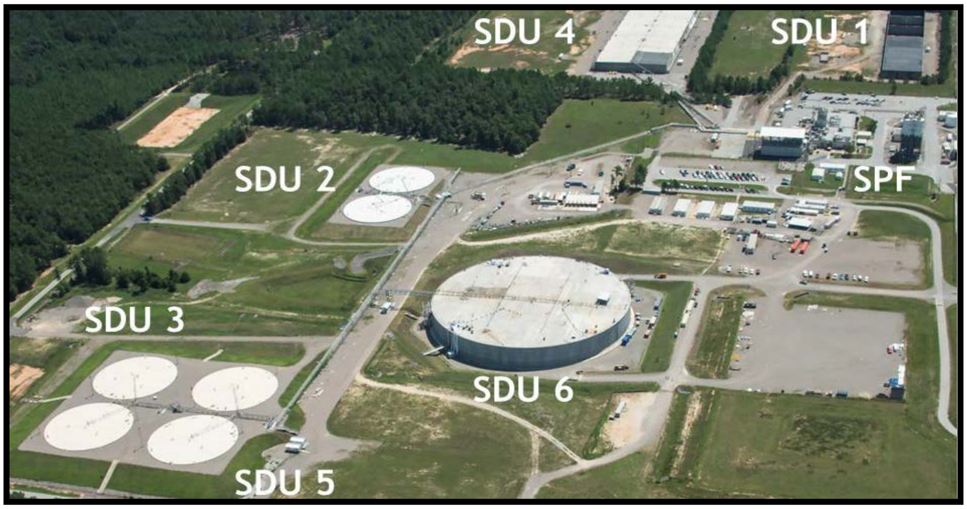

Fig. 1. SRS SDF: low activity salt solution is mixed with cementitious materials in the SPF and transferred to the SDUs for permanent disposal.

A Performance Assessment (PA) is conducted for the SDF to model the long-term (hundreds to thousands of years after SDF closure) fate and transport of the grout-encapsulated contaminants, and to determine the potential for future interactions with the public and the environment [1]. A predominant mechanism controlling the release of contaminants from the SDF is the adsorption characteristics of the saltstone which are element dependent and differ as the reduction potential (E_h) and pH of the saltstone environment changes over time. However, in some instances, contaminant mobility may be controlled by the limited solubility of a particular species. This is indeed the case for Tc-99, a redox sensitive contaminant that under reducing conditions exists in its reduced Tc(IV) oxidation state in the form of sparingly soluble hydrated oxides ($TcO_2.xH_2O$) or sulfides (TcS_x). It is believed that ferrous iron (Fe^{2+}) and sulfide (S^{2-}), both present in the GGBFS component of saltstone, serve as Tc reductants. However, in the presence of O_2 (associated with the future infiltration of air or oxygenated ground waters into the saltstone monolith) it is possible for redox-sensitive Tc(IV) to transition into the highly soluble (and mobile) Tc(VII) species, pertechnetate (TcO_4^-), after which Tc-99 mobility will be controlled predominantly by adsorption. The SDF PA generally utilizes empirically derived data (from simulant samples) as inputs for contaminant transport simulation. However, this data has traditionally been derived from exposing size-reduced (crushed/ground) saltstone materials to simulated SRS groundwater and determining solid-liquid partitioning coefficients (K_d) that describe the distribution of contaminant between the liquid and the saltstone material. Such experiments create artificially high solid-liquid contact areas that likely result in higher Tc-99 leachate concentrations than would be expected for intact, monolithic samples. Alternate techniques evaluating intact, monolithic samples have been developed at Vanderbilt University as part of the Leaching Environment Assessment Framework (LEAF) (http://www.vanderbilt.edu/leaching/). In particular, United States Environmental Protection Agency (USEPA) Method 1315, *Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using a Semi-Dynamic Tank Leaching Procedure* [2], has been previously utilized to evaluate Tc-99 leaching characteristics from saltstone, and is considered a vast improvement over previous tests using size-reduced materials that focus on equilibrium partitioning rather than the rate of contaminant release from intact monoliths under physically realistic conditions. EPA Method 1315 is considered a semi-dynamic method in which a sample is submerged in a leachate solution and the leachate is periodically removed and analyzed to determine the concentrations of relevant contaminants; this data is subsequently used to calculate the "apparent" diffusion rate of the contaminants from the sample, which includes for multiple processes in addition to diffusion, including contaminant dissolution or desorption within the monolith. In the current study a new technique, termed the Dynamic Leaching Method (DLM), is being used to investigate the Tc-99 leaching behavior of intact, monolithic saltstone samples. The DLM facilitates the continuous transport of leachate through the interior structure of an intact monolith via application of a hydraulic gradient across the sample. The method is described in more detail in the subsequent section. Preliminary data with respect to the evaluation of Tc-99 leaching from saltstone using DLM was presented in Ref. 3. Fig. 2 provides an update of that data for a sample that, at the time of writing, had been subjected to 30 months of testing.

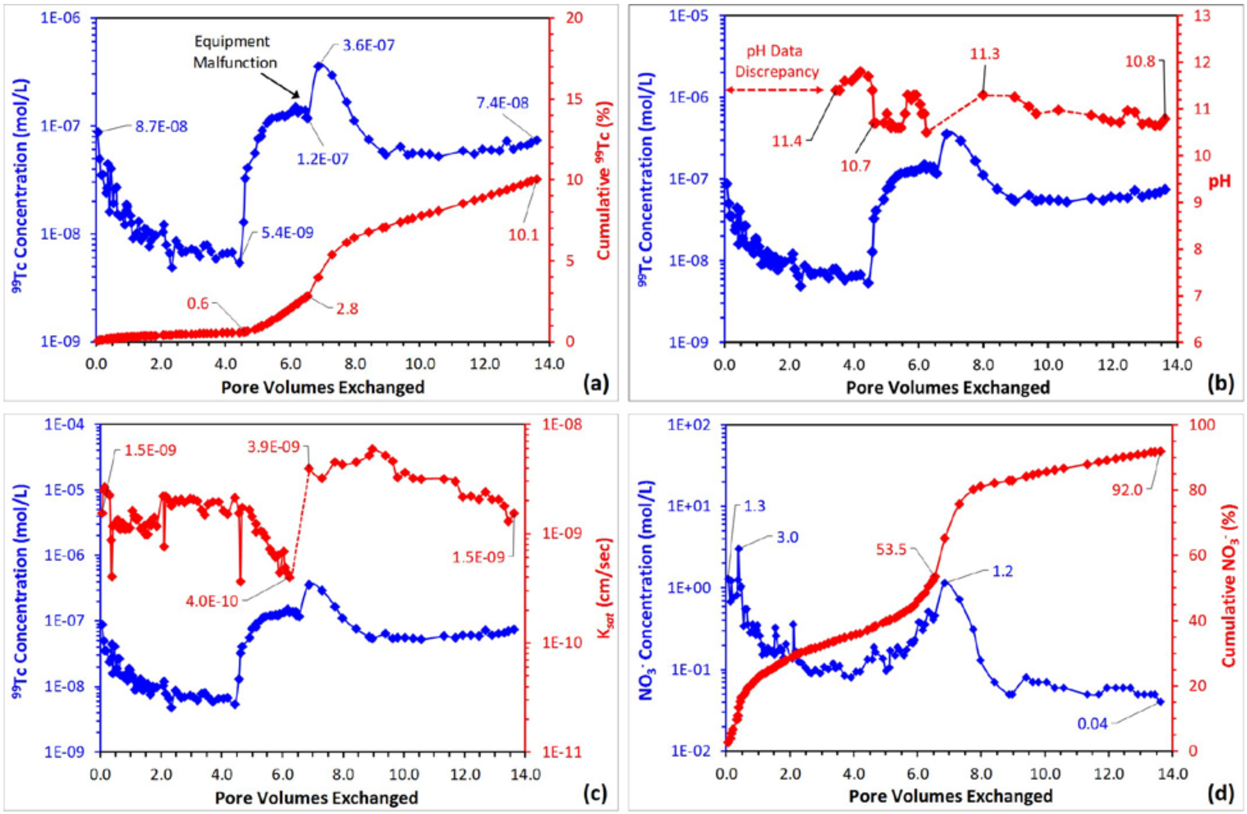

Fig. 2. Relationships between (a) Tc-99 concentration, (b) pH, (c) K_{sat}, (d) NO_3^- concentration and pore volumes exchanged.

Discussion of the data in Fig. 2 will not constitute a key part of this paper since important datasets including leachate pH and E_h are incomplete due to complications with accurate measurements of these properties and with preventing air exposure to the leachates prior to measurement. As such it is impossible to correlate rapid order-of-magnitude changes in Tc-99 concentrations, Fig. 2(a), to changes in the chemical leaching environment. In addition, leachates from the earlier testing were not analyzed for other key constituents of interest (e.g., Al, Ca, Mg, Na, Si, CO_3^{2-}, NO_2^-, NO_2^-, SO_4^{2-}), that should enable the development of an understanding of how chemical changes associated with the grout pore solution influence radionuclide leaching behavior. Furthermore, half way through testing an equipment malfunction resulted in a loss of the confining pressure and potential exposure of the sample to air. The timing of the malfunction is indicated in Fig. 2(a) and resulted in a marked increase in Tc-99 leachate concentration, and an order of magnitude increase in saturated hydraulic conductivity (K_{sat}). As such, the primary focus of this paper will be discussion of data derived from an enhanced DLM system with improved protocols for analyzing leachate constituents. In addition, to supplement the data interpretation, saltstone samples will be analyzed to provide pre- and post-DLM properties such as minerology, elemental composition, density, porosity, and residual reduction capacity. Fig. 3 depicts the sample and leachate analysis proposed to enhance the understanding of how contaminant leaching behavior is influenced by changes in the saltstone chemical and physical properties. Additional information regarding the proposed analysis techniques and rationale is provided in Table I.

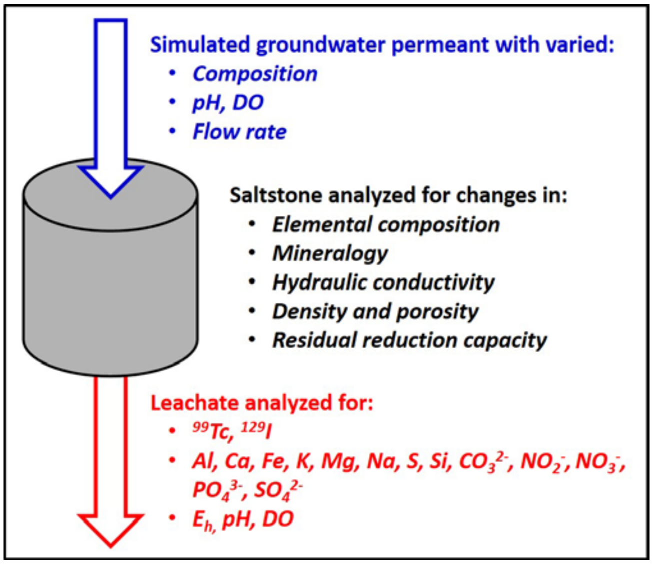

Fig. 3. DLM sample and leachate analysis to support enhanced understanding of contaminant leaching characteristics (DO = dissolved oxygen).

The scope and purpose defined in Table I are not new concepts. Previous work has been conducted in an effort to elucidate information regarding chemical and physical changes within the cementitious matrix that could be correlated with the materials physical integrity and varied contaminant leaching behavior [4, 5]. However, both studies utilized size-reduced (crushed) saltstone that was subsequently packed into columns through which groundwater simulants were passed. An advantage of these experiments is the higher K_{sat} of the packed material which makes it possible to pass large volumes of groundwater simulant through the sample in a short period of time. For comparison, during the 30-month DLM testing (Ref. 3 and Fig. 2) approximately 14 pore volumes of groundwater were passed through the sample at an approximate flow rate of 0.5 mL/day. In contrast, the flow rate used in Ref. 4 was 95 mL/day. At this juncture, it is not known if lower flow rate experiments with intact saltstone samples will elicit data that is more representative of field conditions, and a direct comparison of equivalent saltstone samples utilizing the column and DLM techniques is perhaps warranted. However, the potential issues of artificially high liquid-solid contact areas associated with K_d measurements and unintended exposure to air (in the case of redox sensitive samples) may also apply to size-reduced sample flow-through experiments. In addition, the intact monolith is composed of an intricate and tortuous network of micropores which can potentially be obstructed by solids that precipitate as the pore solution chemistry evolves. Indeed, such mechanisms may account for the reduction in K_{sat} indicated in Fig. 2(c). It is important to note, however, that both experimental techniques utilize accelerated transport through the material in comparison to the time scales (100s to 1000s of years) predicted for actual saltstone monoliths.

TABLE I: Proposed analysis of DLM saltstone samples and leachates.

Component or Property	Required For				Technique	Purpose
	Permeant	Bulk Monolith		Leachate		
		Before Testing	After Testing			
Al	Simulant known	✓	✓	✓	XRF, ICP-MS	• Leachate – determine components that are being leached from saltstone as a function of pore volume exchange • Saltstone Sample – confirmation of components lost during leaching (e.g., loss of Ca and Si likely attributed to CH, CSH dissolution) • Mass balance: pre-DLM sample constituents || post-DLM samples constituents + ∑leachate constituents
Ca		✓	✓	✓	XRF, ICP-MS	
Fe		✓	✓	✓	XRF, UV/Vis, ICP-MS	
K		✓	✓	✓	XRF, ICP-MS	
Mg		✓	✓	✓	XRF, ICP-MS	
Na		✓	✓	✓	XRF, ICP-MS	
S		✓	✓	✓	UV/Vis	
Si		✓	✓	✓	XRF, ICP-MS	
CO_3^{2-}		✓	✓	✓	Alk Titration	
NO_2^-		✓	✓	✓	UV/Vis	
NO_3^-		✓	✓	✓	UV/Vis	
PO_4^{3-}		✓	✓	✓	UV/Vis	
SO_4^{2-}		✓	✓	✓	UV/Vis	
^{129}I		Spiked amounts known	✓	✓	LSC	• Primary purpose of testing is to determine the leaching behavior of ^{99}Tc and ^{129}I from an intact saltstone sample • Leaching data as a function of pore volumes exchanged will inform the PA
^{99}Tc			✓	✓	LSC, ICP-MS	
pH	✓	×	×	✓	pH Probe	• pH and E_h effect component solubility • May provide rationale for observed changes in ^{99}Tc conc. in leachate • DO content in leachate will inform PA assumption that excess reduction capacity in saltstone consumes DO
E_h	✓	×	×	✓	E_h Probe	
DO	✓	×	×	✓	DO Probe	
Mineralogy	×	✓	✓	×	XRD	• If CH and C(A)SH are dissolving the mineral phases and proportions will change: this can be determined by XRD
Reduction Capacity	×	✓	✓	×	Glasser Ce(IV) titration	• If reductants (e.g., S^{2-}, Fe^{2+}) are leached from the saltstone sample the reduction capacity would be expected to decrease • Provides input to PA regarding consumption of reduction capacity
Density	×	✓	✓	×	Archimedes (ASTM)	• If minerals in the saltstone are dissolving and being leached then the density and porosity of the sample might be expected to change
Porosity	×	✓	✓	×		
K_{sat}	×	✓	✓	×	Permeameter (ASTM)	• K_{sat} will be continually measured during the DLM tests • K_{sat} variations may impact PA model

EXPERIMENTAL

Sample Preparation

For this study, a Tc-99 spiked saltstone grout with a GGBFS-FA-OPC mass % ratio of 45-45-10 was prepared by mixing with simulant salt solution (Table II) at a water-to-solids ratio of 0.6. Samples are poured into 5-cm diameter by 10-cm height molds and allowed to cure for at least 6 months.

TABLE II: Salt solution used in preparation of simulant saltstone.

Material	Molarity (M)
Sodium Hydroxide, NaOH	1.594
Sodium Nitrate, $NaNO_3$	3.159
Sodium Nitrite, $NaNO_2$	0.368
Sodium Carbonate, Na_2CO_3	0.176
Sodium Sulfate, Na_2SO_4	0.059
Aluminum Nitrate, $Al(NO_3)_4.9H_2O$	0.054
Sodium Phosphate, $Na_3PO_4.12H_2O$	0.012

DLM Technique

The DLM technique is based on American Society for Testing and Materials (ASTM) D5084, *Standard Test Method for Measurement of Hydraulic Conductivity of Saturated Porous Materials Using a Flexible Wall Permeameter* [6], which is typically used for K_{sat} measurement by applying a hydraulic gradient across the sample while ensuring internal sample flow. A photo of the updated DLM set-up is provided in Fig. 4. Samples tested on the DLM are typically 2.5 cm height by 5 cm diameter. Diffuser plates and end pieces (for permeant delivery and leachate removal) are placed at the top and bottom of the sample which is subsequently fitted with a rubber, confinement membrane. The entire sample configuration is immersed in water inside the confinement vessel. A confining pressure of approximately 140 kPa (20 psi) is applied to the sample to compress the rubber membrane against the exterior sample surface in order to prevent from leachate flowing around rather than through the saltstone monolith.

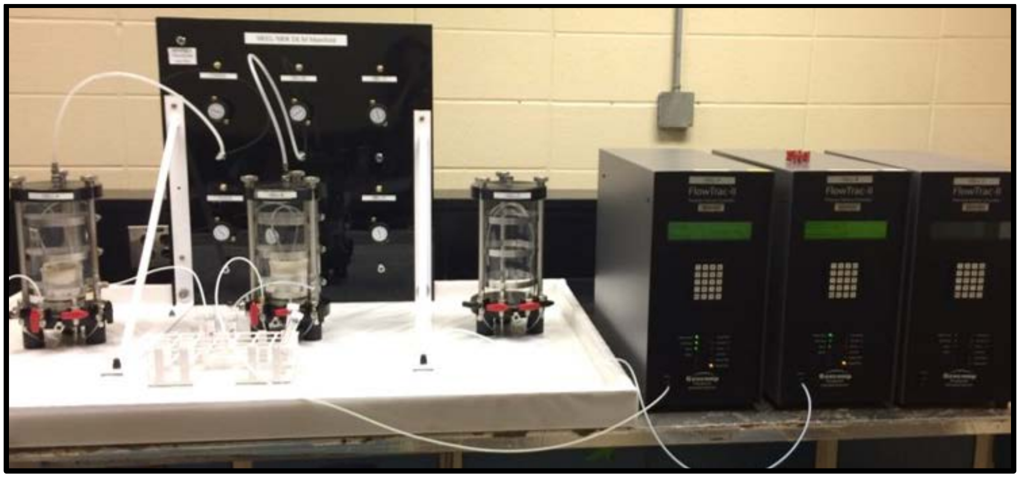

Fig. 4. Photo of revised DLM system.

Figs. 5 A and B compare the original and revised DLM systems. In comparison to the previous work [3], the following changes have been made to the equipment. Firstly, the permeant flow through the sample is achieved via a mechanical pump (Geocomp FlowTrac II, Acton, MA) which enables a constant flow rate to be set; the pump maintains the desired flow rate while recording changes in the back pressure limiting flow. In the previous DLM system the pressure applied to the permeant was constant and the flow rate varied based on changes to the sample K_{sat}. These pumps were incorporated into the system in an attempt to determine if flow rates could be increased and held constant, thereby reducing test times and providing control over pore solution residence times within the sample. However, it should be noted that there appear to be some limitations of the set-up in terms of realistically achievable flows, and ultimately flow rates may be restricted to approximately 0.5 mL/day (no matter which system is utilized) in order to ensure that the flow of permeant is through rather than around the sample.

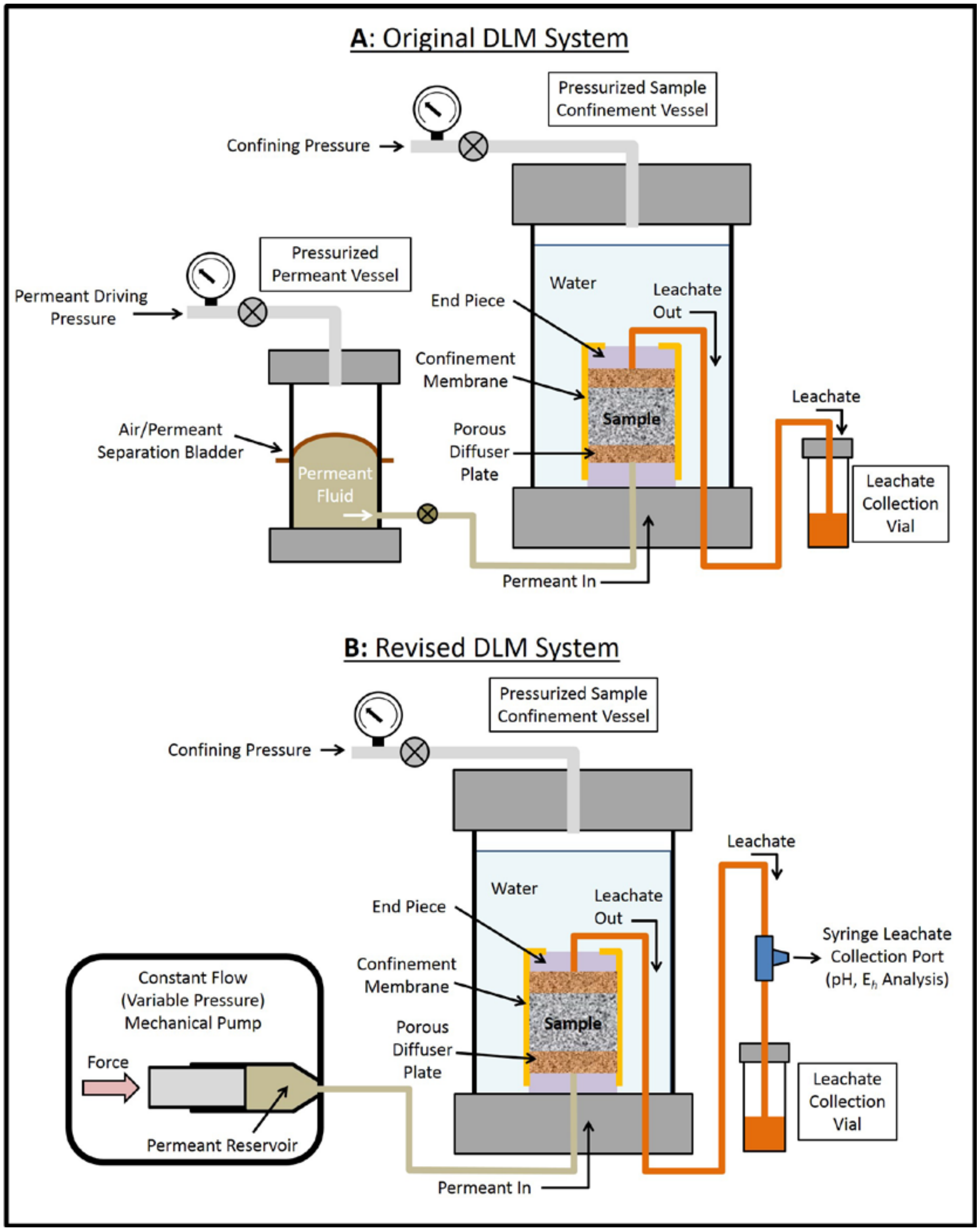

Fig. 5. (A) Original DLM system; (B) Revised DLM system.

Another significant change to the system relates to retrieval of samples for pH/E_h analysis. In previous testing unintended exposure of the leachate to air during sample collection resulted in lower than expected pH and higher than expected E_h. The new system depicted in Fig. 6 allows removal of leachate from the outlet flow through a sealed septa access port that limits exposure of the leachate to the atmosphere prior to measurement.

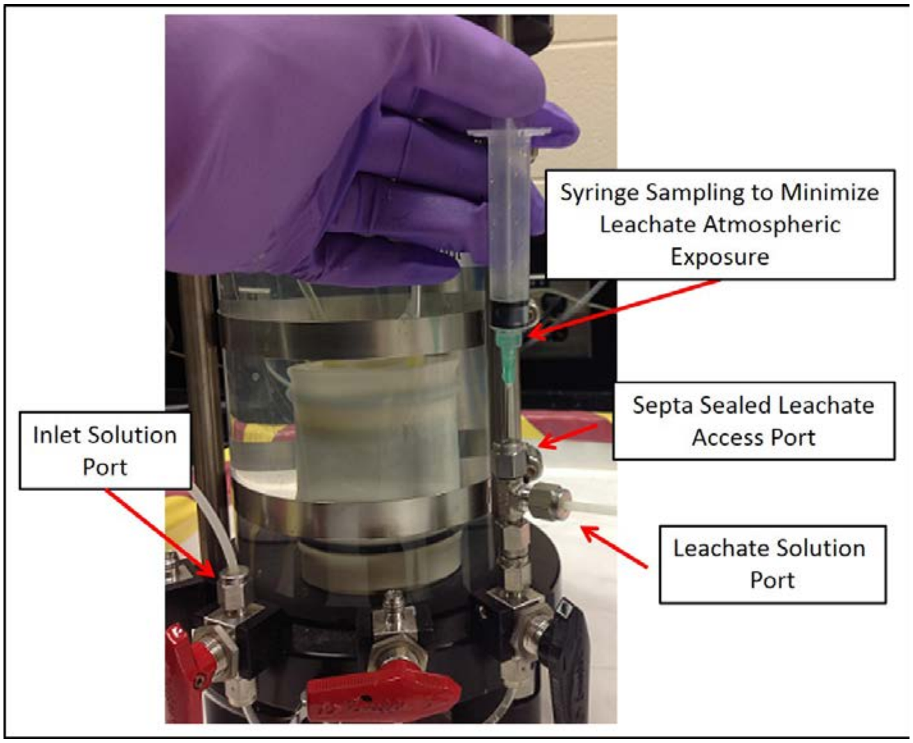

Fig. 6. Alternate system for sampling leachate for ph-E_h analysis.

The permeant used to simulate groundwater transport through the saltstone samples was an SRS-simulated groundwater solution as defined in Table III.

TABLE III: SRS-simulated groundwater.

Constituent/Parameter	**Concentration** (mg L^{-1})
Na	1.39
K	0.21
Ca	1.00
Mg	0.66
Cl	5.51
SO_4	0.73
pH	5.0

Sample Analysis

As stated previously the intent of this work is to assemble chemical and physical information about the sample and leachate before, during, and after DLM testing in an effort to better understand the relationship between the chemical and physical changes of the grout sample and their effects on contaminant leaching. For pre-DLM sample testing an adjacent piece of sample to the sectioned DLM sample will be used as shown in Fig. 7.

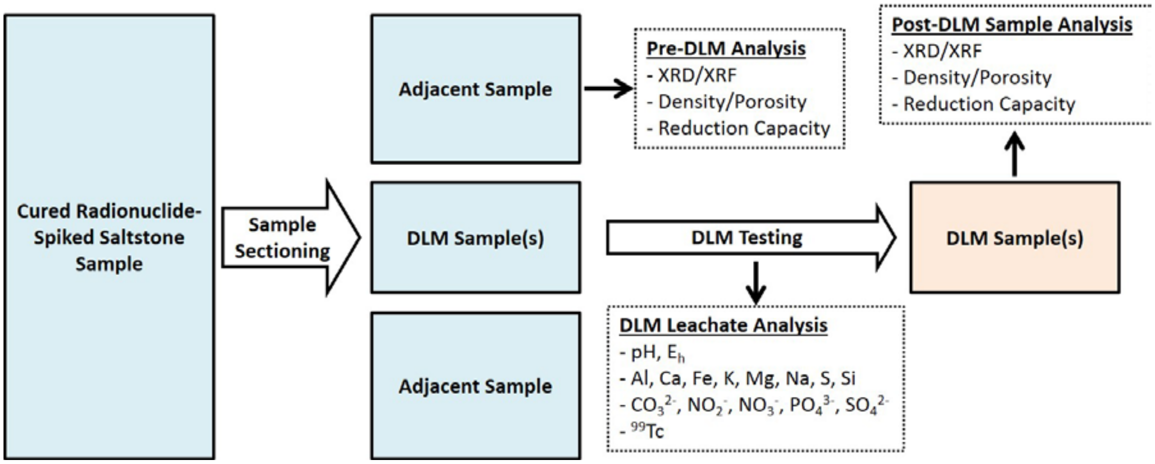

Fig. 7. DLM sample sectioning and subsequent analysis.

To date powder X-ray diffraction (XRD), density/porosity, and reduction capacity have been performed only on a pre-DLM sample cured for 6 months. XRD analysis was performed on a D2 Phaser x-ray diffractometer (Bruker AXS, Madison, WI). Sample density/porosity was measured according to ASTM C-642, *Standard Test Method for Density, Absorption, and Voids in Hardened Concrete* [7]. Reduction capacity was measured for the individual dry feeds (GGBFS, FA, and OPC) and a saltstone sample via the Angus and Glasser Method [8].

Leachate Analysis

The concentrations of Na, K, Mg, Ca, Al, P, Fe, Mn, and Si in the DLM leachates were determined by inductively coupled plasma–mass spectrometry (ICP-MS) using a NexION 300 (Perkin Elmer, Inc.) according to the QA/QC protocols outlined in EPA Method 6020B [9]. The leachate alkalinity (equiv. mg $CaCO_3$/L), NO_3^-, NO_2^-, SO_4^{2-}, and S^{2-} were determined using standardized water test kits and a Orion AquaMate 8000 UV/Vis Spectrophotometer. Technetium-99 concentrations in the effluent were determined by liquid scintillation counting (LSC).

RESULTS

The pre-DLM sample density and porosity was measured at 1.71 g/cm^3 and 52% which are generally consistent with previous data [10]. The reduction capacity of the individual dry feed components and cured saltstone are provided in Table IV. Whilst the reduction capacity of the GGBFS and OPC are equivalent to measurements from a previous study [11] both the FA and the saltstone sample for this study indicate lower values. This is quite possibly simply related to batch-to-batch variations in dry feed materials received from the supplier. Pre-DLM XRD for a sample cured for 6 months is provided in Fig. 8. The predominant phases are nitratine ($NaNO_3$ – a major salt waste component), quartz (SiO_2), calcite ($CaCO_3$), mullite ($3Al_2O_3.2SiO_2$), and gypsum ($CaSO_4.2H_2O$). These phases are consistent with those detected by XRD in previous studies on saltstone phase determination [12, 13].

TABLE IV: Reduction capacity of dry feed components and saltstone sample cured for 6 months.

Materials	**Reduction Capacity (μeq g^{-1})**	
	This Study	**Ref. 11**
GGBFS	925	819
FA	Negligible	299
OPC	135	198
Saltstone	464	607 - 849

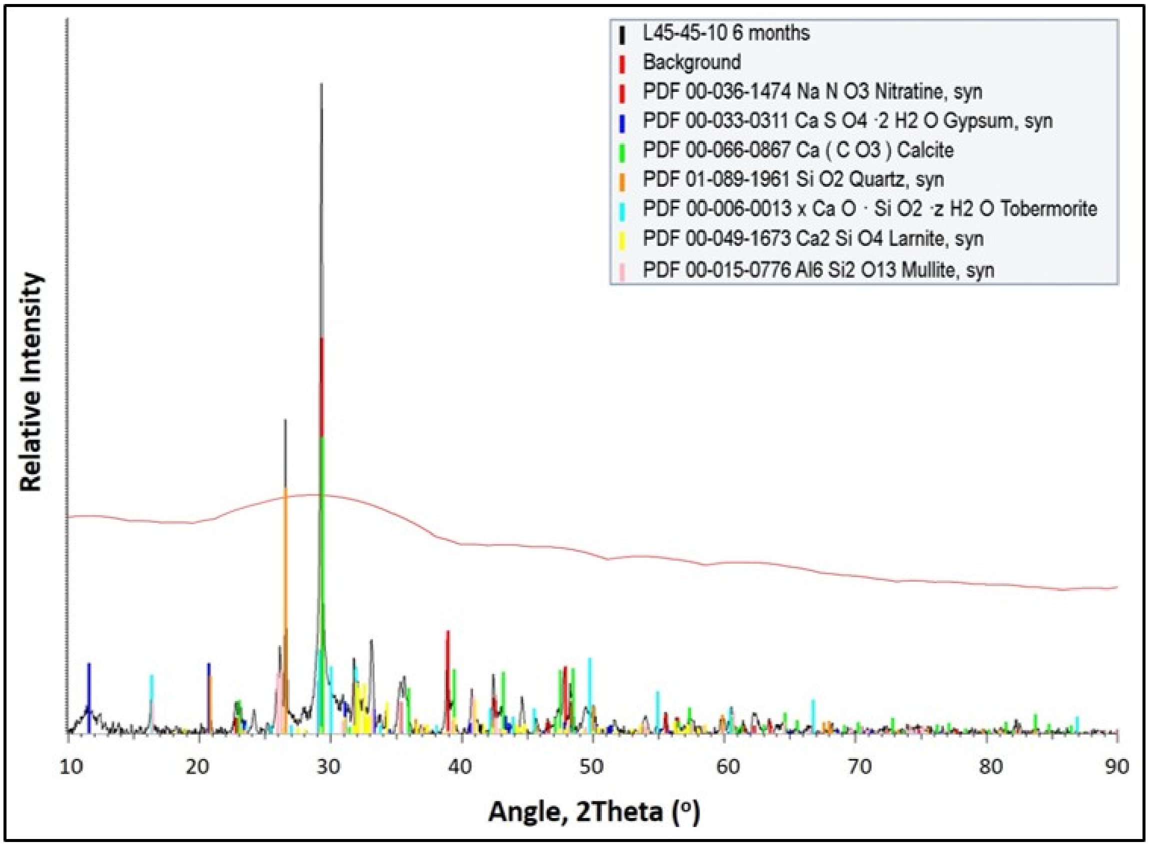

Fig. 8. XRD pattern for pre-DLM sample cured for 6 months.

Fig. 9 indicates leachate concentrations for an array of constituents. It should be noted that at this juncture in the study the data is limited to a single sample with less than a pore volume of permeant passed through the monolith. As such it would be premature to attempt to define data trends and correlate them with variations in leachate Tc-99 concentration. However, a number of data points are noteworthy. Firstly, as expected sodium (Na) and nitrate (NO_3^-), the two primary constituents of the simulant salt solution used to prepare the grout, indicate the highest concentrations in the range of 1-3 mol/L. Secondly, the relative proportions of calcium (Ca) and silicon (Si) are of note. With respect to the leaching of conventional (non-alkali-activated) cementitious materials, it is anticipated that the transport of water through the material results initially in the dissolution of portlandite [$Ca(OH)_2$] followed by the dissolution of the calcium silicate hydrate (C-S-H) gel [14]. For the six leachates analyzed the Ca/Si molar ratio ranges 0.02 to 0.05 which implies the dissolution of silica (SiO_2) phases. A similarly low Ca/Si ratio in the leachate was also observed in Ref. 5.

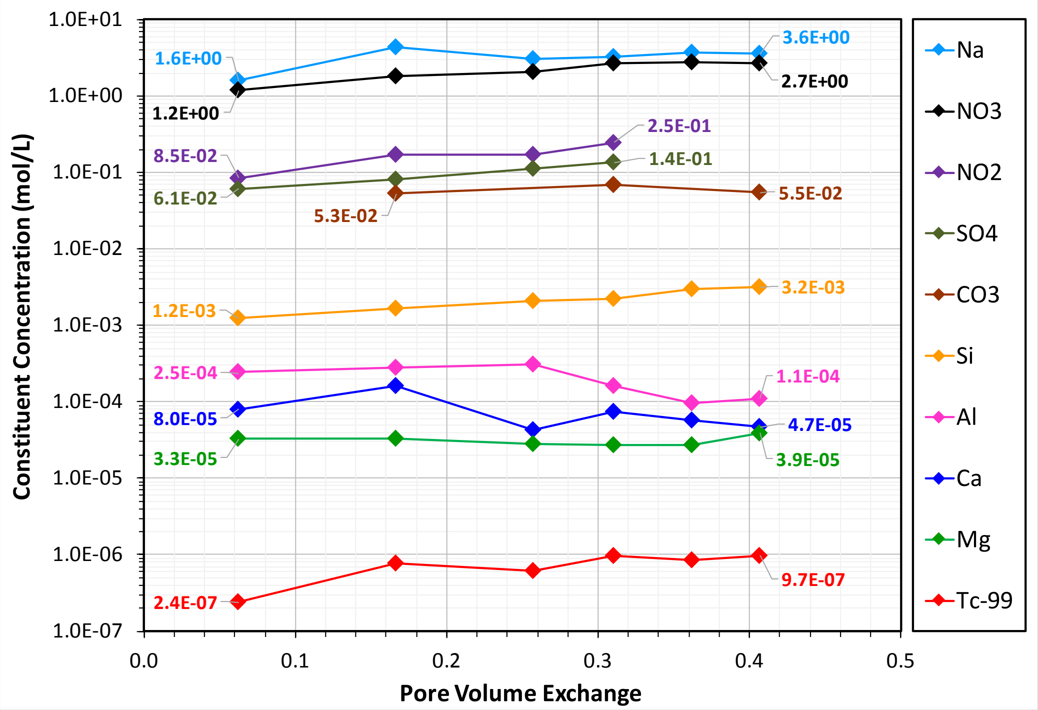

Fig. 9. DLM leachate concentration data.

Tc-99 Concentration, pH, and E_h

The Tc-99 concentration is plotted with pH in Fig. 10. For the six leachates analyzed the Tc-99 concentration is initially 2.4E-07 mol/L and subsequently increases to 9.7E-07 mol/L. Over this pore volume range the pH drops from 12.8 to 11.8.

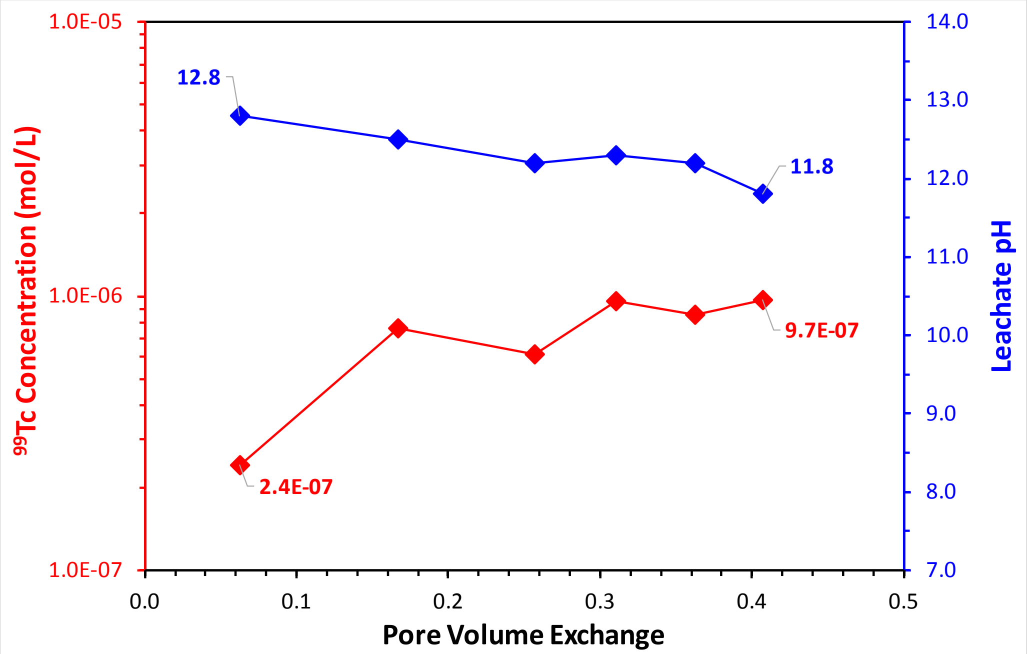

Fig. 10. Leachate pH and Tc-99 concentration.

Fig. 11 indicates shows the Fig. 10 data plotted on the pH-solubility diagram for Tc compounds. The data from the current study (◇) matches the predicted solubility of $TcO_2.xH_2O$. Fig. 11 also provides pH data (◇) for leachates acquired from the original sample tested per Ref. 3 and indicated in Fig. 2. Again, these data points predominantly fall around the theoretical (pH-dependent) solubility limit of $TcO_2.xH_2O$ and suggest that after 30 months of analysis and the transport of 14 pore volumes through the material, the Tc remains in a reduced state.

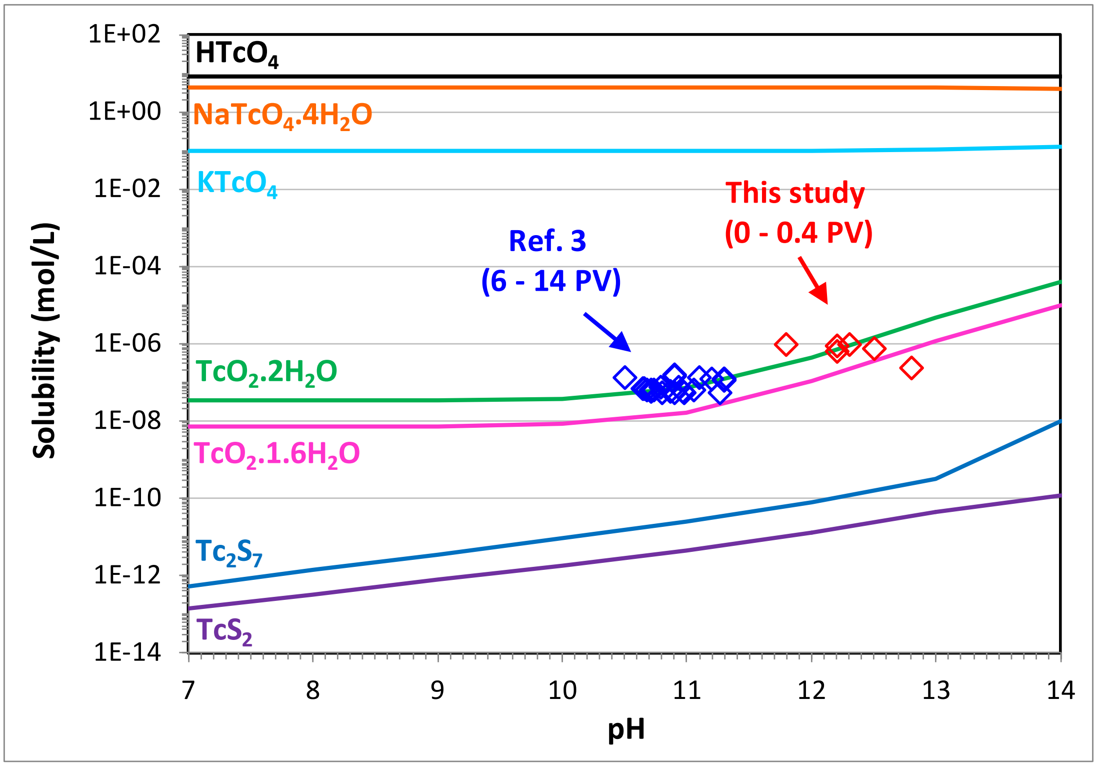

Fig. 11. pH-solubility diagram for Tc compounds.

This figure was generated using the geochemical computer code PHREEQC-2 (USGS-99-4259) and the Lawrence Livermore National Laboratory (LLNL) database (thermos.com.V8.R6.230)

Fig. 12 indicates E_h-pH data for the leachate samples from this study and the previous study [3]. The theoretical redox boundary that separates oxidized Tc(IV) and reduced Tc(VII) is drawn from data provided in Ref. 15. Either side of the boundary represents the pH-E_h values associated with the thermodynamically-favored oxidation states of Tc(IV) or Tc(VII). This initial data suggests that the sample environment is consistently in the region for which the oxidized Tc(VII) would be the thermodynamically-favored phase. The data (predominantly E_h) is also in contrast to data defined in Refs. 4 and 5 in which E_h values of approximately -400 mV were measured in the initial stages of testing. However, the leachate concentration of Tc-99 suggests the presence of reduced $TcO_2.xH_2O$ phases. One question that arises is whether it can be assumed that the leachate E_h equates to the E_h of the pore solution throughout the sample? At this juncture, the authors do not have an explanation for the measured data but again, even with respect to the longer-term sample (Ref. 3) which has a positive E_h, the Tc-99 concentrations continue to support the presence of reduced Tc(IV) phases.

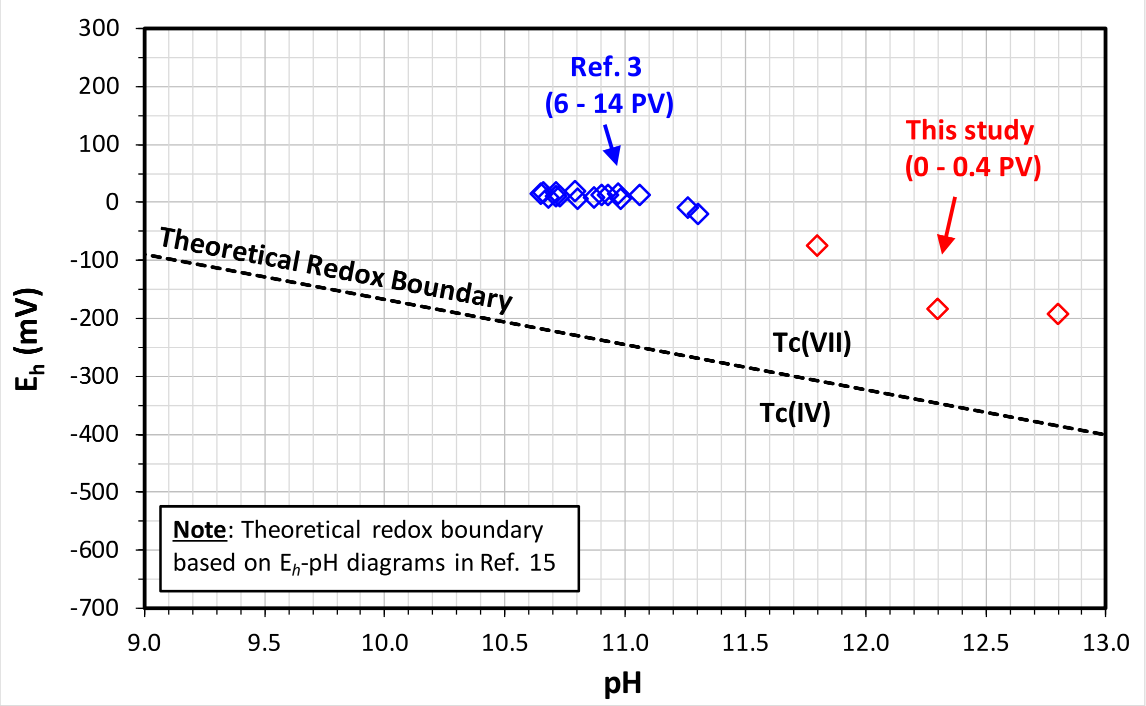

Fig. 12. E_h-pH data for DLM leachates.

CONCLUSION

The DLM technique has been developed to evaluate the leaching characteristics of radionuclides, such as Tc-99 and I-129, from a cured, intact, cementitious monolith. The method enables continuous flow of permeant through the monolith, and subsequent (periodic) analysis of the leachate to determine the pH, Eh, and concentrations of Tc-99 and other key constituents associated with the pore solution and/or dissolution of the cementitious matrix as multiple pore volumes of permeant are transported through the sample. The intent of this work is to correlate Tc-99 release with changes in the chemical (and physical) characteristics of the sample; potential correlations are expected to better inform the SDF PA. The work defined in this paper is predominantly associated with recent tests utilizing an improved DLM system with the ability to better control flow rate and to sample leachate while minimizing atmospheric exposure. The data set presented is preliminary and includes: (1) pre-DLM sample analysis (XRD, density/porosity, and reduction capacity) that will be compared to equivalent analysis conducted on the post-DLM sample, and (2) a suite of leachate chemical data associated with six leachate samples obtained during the transport of approximately 0.4 pore volumes through the monolith. The pre-DLM sample data is in general agreement with previous saltstone analysis. With respect to the leachate data it is too early to identify trends and/or correlations between the Tc-99 leaching characteristics and the leachate chemistry. At this point, and as expected, it does appear that the concentration of the Tc-99 in the leachate is predominantly controlled by the pH-dependent solubility of reduced $TcO_2.xH_2O$ phases.

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