Laboratory contaminant migration assessment of soil cement mixtures

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ABSTRACT
A literature review of contaminant transport through cement-based solidified/stabilized wastes related to advection and diffusion of various contaminants is presented. Common testing methods are reviewed and typical values for hydraulic conductivities and diffusion coefficients are reported. Based on a synthesis of this information, an approach to testing methodology and apparatus is presented for a pure diffusion test on cement-based S/S treated soil.

RÉSUMÉ
Ce document présente une revision de la littérature scientifique sur la mobilité des contaminants solidifiés et/ou stabilisés (S/S) au ciment en fonction des concepts d'advection et de diffusion. Les méthodes d'essais courantes y sont réexaminées et des valeurs typiques sont fournies pour la conductivité hydraulique ainsi que les coefficients de diffusion. En se basant sur la synthèse de ces informations, une approche sur la méthode d'essai est présentée pour un test de diffusion fait sur un sol solidifié et/ou stabilisé au ciment.

1 INTRODUCTION

Solidification/stabilization (S/S) is a widely used treatment method for disposal and treatment of radioactive and hazardous wastes as well as contaminated soil (the subject of this paper). Binders such as lime, fly ash, and bitumen are sometimes used in this process, however cement binders are prevalent in the technology. Cement-based S/S involves the immobilization of potentially harmful components of the waste form (often contaminated soil) through chemical and physical interactions with a cementitious binder (Shi and Spence, 2004).

In cement-based S/S treatment, contaminated soils are subjected to two separate phenomena which are often lumped together or used interchangeably; solidification and stabilization. The mechanisms responsible for immobilization of the contaminants are different for each phenomenon. The term "solidification" refers to changes in the physical properties of the contaminated soil (Batchelor, 2006; Bone *et al.*, 2004). This typically includes an increase in compressive strength and a change in pore structure that leads to a lower permeability, a lower diffusivity, and to the physical encapsulation of a contaminated soil material into a solid product (the soil-cement matrix). The term "stabilization" refers to changes in contaminant mobility, solubility, and/or toxicity (Bone *et al*., 2004; Shi and Spence, 2004). Stabilization involves the chemical reaction of contaminated soil with reagents to produce more chemically stable and less hazardous waste forms (e.g. by forming insoluble metal hydroxides) (Paria and Yuet, 2006; Mulligan *et al*., 2001).

Traditionally, in the design of cement-based S/S treatment, there has been limited focus on the diffusive properties of the treated waste. However, in other low-hydraulic conductivity barriers, it has been shown that diffusion plays a significant role in contaminant migration (Rowe *et al*., 2004). In particular, if the hydraulic conductivity of the material is less than approximately 10^{-9} m/s the only measurable contaminant migration may be via diffusion. As discussed by Rowe *et al.* (2004), well designed barrier systems with low hydraulic conductivities and low diffusivities are capable of protecting the environment in perpetuity. Stegemann and Côté (1991) report that for cement-based S/S materials the infiltration of water is expected to be negligible and the rate of a contaminant release is expected to be governed by diffusion when hydraulic conductivities are less than 10^{-9} m/s. This paper provides a brief literature review on contaminant transport through cement-based S/S treated soils and on the current practices used to measure relevant contaminant transport properties. The second part of this paper focuses on the development of a diffusion apparatus and a testing methodology for cement-based S/S treated soils.

2 LITERATURE REVIEW

Understanding the transport properties of the contaminant of interest, the properties of the transport medium, and their interaction with one another is essential when estimating the potential long term effects of a contaminant on the environment. The goal of cement-based S/S treatment is ultimately to minimize the effect of contaminants on the environment. This goal is achieved by converting hazardous wastes into more environmentally desirable waste forms. The focus in design of cement-based S/S systems is typically on (Shi and Spence, 2004):

1. compatibility between cement and waste materials
2. chemical fixation of contaminants
3. "leachability" of contaminants from treated waste
4. durability of the treated waste or contaminated materials
5. cost effectiveness of mix designs

Items 1 and 4 have been studied in detail for a variety of wastes (e.g. Bricka and Jones, 1993; Al'Tabbaa and King, 1998) and can usually be optimized with an appropriate mix design. Item 5 is typically evaluated at the design level on a case-by-case basis and can also be optimized with an appropriate mix design. Item 2 has received a lot of attention in the literature (e.g. Paria and Yuet, 2006; Malviya and Chaudhary, 2006; Chen *et al.*, 2008) particularly with respect to heavy metals and other inorganic contaminants. Item 3, "leachability" is a term that is often used with cement-based S/S materials but can relate to the "stabilization" term and/or the "solidification" term in "S/S". For many inorganic contaminants, "stabilization" is the mechanism responsible for the treatment. However, the "leachability" of contaminants, which are not entirely "stabilized", may show reduced "leachability" because the contaminant has been entrapped in the pore structure soil-cement matrix. Solidification is hypothesized to be the mechanism responsible for the success of cement-based S/S to treat mid to high molecular weight organic compounds. Unfortunately, "leachability" is often assessed using conventional leaching tests which do not evaluate the potential of the cement-based S/S treatment to perform the desired task with respect to solidification. Conventional leaching tests also often provide little beyond a "pass or fail" criterion and more importantly do not provide information which could assist in a pre-design selection stage. Some additional details on these leaching tests are presented in section 2.1.

When the solidification mechanism is responsible for immobilization of contaminants, it is the diffusivity of the contaminant from the low hydraulic conductivity matrix which may control the "leachability". Diffusion is a subject area that is relatively well understood and is used to design barrier systems for municipal solid waste landfills. It is logical to assume that a diffusion testing apparatus and a diffusion testing procedures can be adopted to evaluate cement-based S/S materials. A diffusion testing apparatus would provide information not only relevant from a proof of treatment standpoint but also to provide information to designers regarding the long term performance of cement-based S/S systems.

2.1 Diffusive Transport

Diffusion is defined as the movement of contaminant from a location of high concentration to an area of low concentration due to Brownian motion. The mass flux (f) transported by diffusion may be expressed by Equation 1 (Rowe *et al.,* 2004).

$$f = -n_e D_e \frac{dc}{dz} \qquad [1]$$

where: f is the mass flux [$ML^{-2}T^{-1}$], n_e is the effective porosity, D_e is the effective diffusion coefficient [L^2T^{-1}] and dc/dz is the concentration gradient (the change in concentration divided by the distance between the two locations; [ML^{-4}]). The effective diffusion coefficient may be related to the free solution diffusion coefficient, Do [L^2T^{-1}] by the tortuosity factor, τ, as shown in Equation 2 (Rowe *et al.*, 2004). Tortuosity is a complex factor that considers; increased length of flow path, changes in fluidity, and electrostatic interactions due to the presence of solid particles (Rowe *et al.*, 2004). The free solution diffusion coefficient represents the maximum rate of diffusion a species can experience in pure water at infinite dilution (Rowe *et al.*, 2004).

$$D_e = \tau D_o \qquad [2]$$

In efforts to obtain information on the "leachability" and diffusivity of cement-based S/S treated wastes, numerous leaching tests have been employed. Many of the existing leaching tests are similar to one another but contain variations in controlled conditions such as: contact time, liquid/soil ratio, pH, and leachant composition (Garrabrants and Kosson, 2005). The two most common leaching tests are the Toxicity Characteristic Leaching Procedure (TCLP) (USEPA, 2004a) and the Synthetic Precipitate Leaching Procedure (SPLP) (USEPA, 2004b). In many countries, the TCLP is sometimes as the only test to characterize the treated waste and/or the suitability of treatment. The TCLP has been criticized as it may not accurately represent field conditions and may lead to inaccurate predictions of the long term leaching properties of the waste (Spence and Shi, 2005; Perera *et al.*, 2005; Butcher *et al.*, 1996). In both the TCLP and SPLP the sample is size reduced (crushed) which accelerates leaching and minimizes or eliminates the solidification aspect of treatment (changes in pore size, diffusivity, etc.).

There are few standard approaches to measuring the diffusive properties of cement-based S/S treated soils. Semi-dynamic tank leaching tests such as ANSI/ANS 16.1 (American Nuclear Society, 2005) and NEN 7345 (Netherlands Normalisation Institute, 1995) may be used to determine information relating to the kinetics of leaching (i.e. a diffusion coefficient) (Perera *et al.*, 2005; Moore *et al.*, 2005). Semi-dynamic leaching tests are performed on intact samples of cement-based S/S products (unlike the more common TCLP and SPLP). Typically an intact sample is placed in contact with demineralised water and the concentration of the contaminant in the leachant is measured as the leachant is replaced at specific time intervals. This process allows for a solution to Fick's second law of diffusion, which in turn allows for determination of a diffusion coefficient (Perera *et al.*, 2005). Results of the ANSI/ANS-16.1 leaching test are often reported as a leachability index (LI), which is calculated as shown in Equation 3.

$$LI = -\frac{(\Sigma \log_{10} D_e)}{N} \qquad [3]$$

where: LI is the leachability index, D_e is the diffusion coefficient for each leach interval (cm^2/s), and N is the number of leach intervals.

The leachability index typically varies from values of about 5 (very mobile) to 15 (very immobile) (Kundu and Gupta, 2007). Kundu and Gupta (2007) report that a treated waste with a leachability index of 8 or greater may be disposed of in a landfill while treated wastes with a leachability index of less than 8 are not appropriate for landfill disposal. The calculated D_e often changes with time during dynamic leaching tests which is in indication that diffusion is not always the primary mechanism measured in this test (Andrés *et al.*, 1993). Sometimes the results are governed by other mechanisms such as dissolution or surface wash off (Malviya and Chaudhary, 2006).

Diffusion through soils has been studied extensively. Examples of steady state diffusion tests may be found in literature such as Dutt and Low (1962), Kemper and van Schaik (1966), Lai and Mortland (1962), and Gillham *et al.* (1984). These diffusion tests can be separated into two different categories, transient and steady state diffusion tests. Further details on these two types of tests may be found in Rowe *et al.* (2004). Transient diffusion tests, which involve the development of a concentration versus distance profile to be measured chemically or radioactively, may be performed in relatively short test times. At the conclusion of a transient diffusion test, the experimental results may be plotted and the retarded diffusion coefficient (D_R) may be determined by fitting calculated data to the results. In the case of a conservative and non-reactive species (with no sorption) such as chloride, tritium, or deuterium, the calculated diffusion coefficient, D_R, is equal to the effective diffusion coefficient, D_e. D_R, is different from D_e in that D_R includes not only the effects of diffusion but also of sorption. The two may be related through Equation 4 for the case of linear sorption (Rowe *et al.* 2004):

$$D_R = \frac{D_e}{1 + \left(\frac{\rho_d}{\theta}\right) K_d} \qquad [4]$$

where: ρ_d is the dry density of the soil [ML^{-3}], θ is the volumetric water content, and K_d is the distribution coefficient [L^3M^{-1}] which reflects the degree of sorption. steady state diffusion test yields only one property while transient diffusion tests have the advantage of yielding values for both D_e and K_d.

A summary of diffusion coefficients on various soil types (mainly clays) for a variety of contaminants is presented in Rowe *et al.* (2004). Studies by Barone *et al.* (1989) similarly investigated the diffusion of chloride through intact rock. In this case, however, distilled water was placed in contact with samples of shale having a high initial concentration of chloride in their pore water and Cl^- was diffused out of the sample.

There has been a significant amount of research regarding chloride diffusion through concretes due to the potential for chloride to cause corrosion of reinforcing steel (e.g. Vedalakshmi *et al.*, 2008; Kayali and Zhu, 2005). This is often measured using tests such as ASTM C1556-04 which results in a solution to Fick's second law of diffusion and thus the parameter D_R (which is referred to as an apparent diffusion coefficient, D_a, by the test procedure).

Tiruta-Barna *et al.* (2006) studied the leaching behaviour of both inorganic (Na^+, K^+, Ca^{2+}) and low level polycyclic aromatic hydrocarbon (PAH) organic pollutants (i.e. naphthalene and phenanthrene) through cement-based solidified/stabilized soils. As part of this study, a model was developed to describe the relevant physiochemical and mass transfer properties of typical contaminants. Dynamic leaching tests were performed and a curve fitting procedure was used to determine the relevant (retarded) diffusion coefficients (D_R) using the measured and theoretical elute concentrations. The results are presented below in Table 1. A range of values is presented for the diffusion of naphthalene as there were multiple tests performed with varying concentrations of naphthalene and ratios of methanol and water as co-solvents.

Table 1. Retarded diffusion coefficients for various compounds (Tiruta-Barna *et al.*, 2006).

Contaminant	D_R (m^2/s)
Na^+	3.0×10^{-12}
K^+	5×10^{-11}
Cl^-	5×10^{-11}
Ca^{2+}	5×10^{-12}
Naphthalene	2×10^{-11} to 4×10^{-12}
Phenanthrene	3×10^{-12}

Malviya and Chaudhary (2006) also performed dynamic leaching tests (NEN 7345) on heavy metal contaminated samples and reported (retarded) diffusion coefficients for a number of heavy metals and ions of interest. The average result of 10 tests (over a range of waste/binder and water/solid ratios) for each constituent presented by Malviya and Chaudhary (2006) are summarized in Table 2.

Table 2. Retarded diffusion coefficients for various inorganic compounds (Malviya and Chaudhary, 2006).

Contaminant	D_R (m^2/s)
Na^+	$9.74 \cdot 10^{-12}$
K^+	$2.37 \cdot 10^{-11}$
Cl^-	$4.68 \cdot 10^{-11}$
SO_4^{2-}	$3.32 \cdot 10^{-12}$
Ca^{2+}	$1.25 \cdot 10^{-13}$
Pb^{2+}	$1.09 \cdot 10^{-12}$
Zn^{2+}	$8.20 \cdot 10^{-15}$
Fe^{2+}	$4.30 \cdot 10^{-12}$
Mn^{2+}	$2.89 \cdot 10^{-12}$
Cu^{2+}	$9.70 \cdot 10^{-13}$

The diffusion coefficients presented by Malviya and Chaudhary (2006) and Tiruta-Barna *et al.* (2006) are presented as "effective diffusivities" as defined by Godbee and Joy (1974). This particular definition of "effective diffusivity" is derived from Fick's second law and is not equivalent to the effective diffusion coefficient discussed throughout this paper or presented by authors such as Rowe *et al*., (2004). The Fickian model does not separately account for diffusion and sorption parameters. The diffusion coefficient obtained from similar test is often referred to as the retarded diffusion coefficient (D_R) by some authors (Rowe *et al*., 2004) and is referred to similarly throughout this document.

Tits *et al*. (2003) performed tritium diffusion testing on cement pastes. The results were fit to two different models, one assuming no sorption and one including linear sorption. It was concluded that there was a strong indication of linear sorption, theorized to be caused by diffusion into dead-end pores.

2.2 Advective Transport

Advective transport is the movement of contaminant with the flux of water. The mass flux (mass of contaminant per unit area per unit time) transported by advection alone may be expressed by Equation 5 (Rowe *et al*., 2004).

$$f = n_e v c \tag{5}$$

where: f is the mass flux [$ML^{-2}T^{-1}$], n_e is the effective porosity of the soil, c is the concentration of contaminant [ML^{-3}], and v is the groundwater velocity [LT^{-1}] which is governed by Darcy's Law (Equation 6).

$$v = \frac{ki}{n_e} \tag{6}$$

where: k is the hydraulic conductivity [LT^{-1}], n_e is the effective porosity, and i is the hydraulic gradient.

From a cement-based S/S mix design perspective, advective transport is limited by minimizing the hydraulic conductivity of the material. Mature cement mortars have shown experimental hydraulic conductivities of 10^{-12} to 10^{-13} m/s. Cements blended with secondary materials such as blast furnace slag or fly ash have been shown to typically exhibit even smaller values (Garrabrants and Kosson, 2005). The hydraulic conductivity of cement-based S/S materials is also normally quite low. Perera *et al*. (2005) reported a range of hydraulic conductivities ranging from a minimum of 4×10^{-18} m/s to a maximum of 4×10^{-6} m/s. The target hydraulic conductivity is often evaluated on a case-by-case basis but can often be specified to reach as low as 10^{-9} m/s for in-ground S/S treatment (Perera *et al*., 2005; Shi and Spence, 2004). This value is similar to those typically used for other contaminant barriers such as clay liners and cut-off walls.

Due to the low hydraulic conductivity of cement-based S/S materials, falling head test methods such as ASTM D5084-00 and BS 1377: Parts 5(5) and 6(6) are often used (Perera *et al*., 2005). Stegemann and Côté (1991) performed falling head conductivity tests (in a method similar to ASTM D5084) on cement-based S/S treated wastes. This study determined that there was generally poor reproducibility of hydraulic conductivity results and that the differences between individual specimens of the same cement-based S/S product were the largest source of variability. Cullinane and Channell (1996) also experienced a high variability in permeability testing of individual specimens of the same cement-based S/S product.

Wang (1997) discussed the effect of cracks on permeabilities of concrete samples. It was determined that the presence of microcracks (< 50μm in diameter) seem to have a negligible effect on hydraulic conductivity. However, when cracks of larger diameter exist, there is an effect on the hydraulic conductivity as water is able to travel through the network of cracks increasing the hydraulic conductivity of the material and ultimately increasing the rate of contaminant transport.

3 EXPERIMENTAL DESIGN

Based on the literature review presented in section 2.1, an attempt was made to develop a testing apparatus and methodology for examining diffusion coefficients for soil-cement mixtures. The procedures presented below allow for a physical characterization of the sample in terms of its porosity, tortuosity, and diffusion coefficient. Details of the sample preparation and diffusion testing are presented in sections 3.1 and 3.2, respectively.

3.1 Sample Preparation

A "synthetic" soil was prepared by mixing 80% silica sand with 20% kaolin in a 20L plastic bucket. Water was added to result in a 13% moisture content. Mixing was performed using a combination of a large scoop, a tamping rod, and a drill with an attached paint mixer. A cement grout was also mixed until homogeneous using the drill with attached paint mixer and a water to cement ratio of 2:1. To obtain a range of different pore distributions, and to study the effect of varying cement contents, grout was mixed into the synthetic soil in five different quantities such that the cement content was 5, 10, 15, 20, and 25% of the dry unit weight of the soil.

After mixing, the specimens were promptly molded. Samples for diffusion testing were cast in molds cut from 70mm diameter Shelby tubes into lengths of approximately 150mm. A base for these molds was created using a sheet of plastic, cut to fit over the end, and taped firmly in place. As suggested by Stegemann and Côté (1990), a layer of mixture was placed in the mold and tamped 32 times (when required). Two layers were used for the 50mm cubic samples and three for the larger 70mmm cylindrical samples. Strokes were distributed evenly over the cross-section of the mold and pressure just sufficient to ensure uniform filling of the mold was applied. Tamping was not performed for the 20 and 25% cement mixtures due to their low viscosity. When placing the final layer, the mold was then filled so that the mixture extended slightly over the top. The top of the sample was then cut off flush with a wet trowel.

To prevent the sample from sticking to the molds, the 50mm cubic molds were greased before casting and a thin plastic insert was cut to fit inside the cylindrical molds. Immediately after casting, all specimens were placed in separate, tightly sealed plastic bags. Specimens were removed from the molds and placed again in tightly sealed plastic bags after 14 days.

3.2 Diffusion Testing

Before a diffusion testing apparatus could be developed, modeling was necessary to estimate the effect of several parameters (i.e. source reservoir volume, time of test, concentration of source). The computer program POLLUTE (Rowe and Booker, 1999) was used.

A porosity of 0.20 was assumed in initial modeling as this is an approximate value reported in the literature for cement-based S/S treated wastes (De Windt and Baddredine, 2007; Balzamo *et al.*, 1996). Tritiated water was chosen as the tracer as it has no charge, is non-reactive (Rowe *et al.*, 2004) as it has shown negligible interaction with soil particles and cement hydrates (Delagrave *et al.*, 1998). A free diffusion coefficient of $2.44 \cdot 10^{11}$ m^2/s was used for tritiated water (Willingham *et al.*, 2004; Philips and Brown, 1968, GIllham *et al.*, 1984; Klitzsche *et al.*, 1976). Tortuosity was estimated through potassium diffusion results presented by Malviya and Chaudhary (2006) and the free diffusion coefficient of potassium (Rowe *et al.*, 2004). Based on this information, and applying Equation 2, a diffusion coefficient of $1.9 \cdot 10^{-10}$ m^2/s was used for modeling diffusion of tritiated water.

A diffusion cell was constructed out of PVC and its dimensions are as shown in Figure 1. The 150mm length of the sample and duration of the test were chosen such that the sample can be divided into six appropriately sized sections at the conclusion of the test. It was desired that each section have sufficient reactivity such that, after extraction, it could be counted accurately with a liquid scintillation counter. At shorter durations there is less penetration of chloride and a full profile may not be measureable. The sampling rates and source reservoir size were chosen so that the diffusion of tritium into the sample would measurably affect the concentration in the reservoir. The additional information on contaminant transport provided by the change in source reservoir concentration will yield a better fit for contaminant transport parameters.

Diffusion testing is performed on samples approximately 150mm in length and 69mm in diameter. Samples are typically allowed to hydrate for approximately 3 months prior to testing. The samples are first saturated in the flexible wall permeameter using the method described above for hydraulic conductivity samples. The samples are then removed from the hydraulic conductivity cell and the outside cylindrical and bottom surfaces are dried before two coats of epoxy are applied. The sample is then liberally coated in vacuum grease and placed in the diffusion cell. A small amount of silicone vacuum grease is applied to where the top of the sample meets the diffusion cell as shown in Figure 1.

The reservoir of the diffusion cell is filled with distilled water and additional water is added as required to keep the water level constant. Tritiated water is then placed in the source reservoir of the diffusion cell. During the two month duration of the test, weekly 10µL samples are taken from the source reservoir and replaced with an equal amount of distilled water. Samples are placed in scintillation vials with scintillation fluid, wrapped in tinfoil, and stored until liquid scintillation counting. Liquid scintillation counting is performed by the Department of Pharmacology at Dalhousie University. Receipt, storage, transport, and disposal of all tritiated water and contaminated materials were treated in accordance with radiation safety procedures (Dalhousie and Canadian Nuclear Safety Commission regulations).

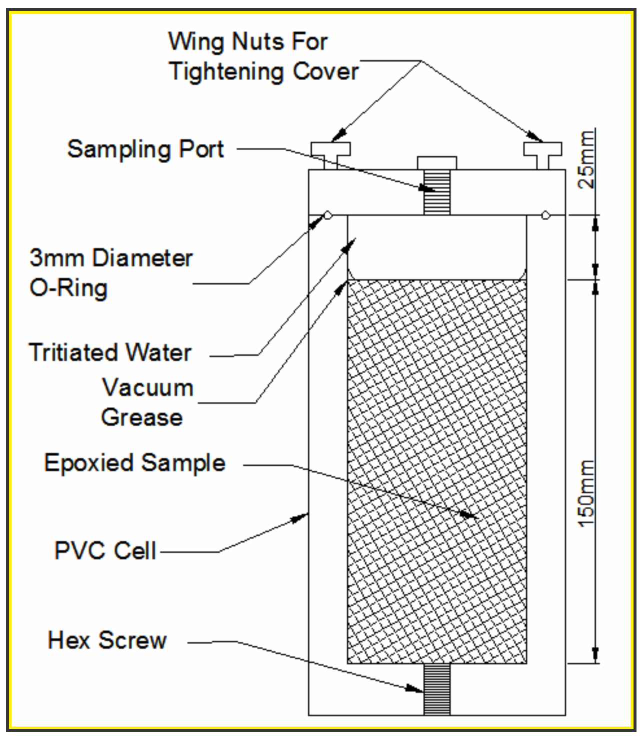

Figure 1. Diffusion test apparatus developed for this study.

The cement creates a basic environment which may potentially cause tritiated water to chemoluminesce and interfere with liquid scintillation counting results. To prevent this chemiluminescence, a small amount (~0.05mL) of weak acid (trichloroacetic acid) is added to all scintillation vials to control the pH.

Upon completion of the test, the sample is sectioned into 6 pieces of approximately equal sizes (25mm). The samples are then crushed and placed in a centrifuge tube with 100mL of distilled water. Each sample is centrifuged to allow solids to settle out of the mixture. The liquid is decanted and a 3mL sample of it placed with 2mL of scintillation fluid and trichloroacetic acid in a scintillation vial. The vial is wrapped in tinfoil and stored until liquid scintillation counting.

4.0 CONCLUSIONS

A literature review of contaminant transport through cement-based S/S treated soils has been presented in this paper. It was shown that there is a lack of consistent testing methodologies in the literature for obtaining contaminant transport properties for cement-based S/S material. It was also shown that dynamic leaching tests often combine transport mechanisms such as sorption and diffusion when reporting diffusion test results which can limit the application of test results to the particular contaminant and soil cement mixture. A procedure and apparatus for diffusion testing has been presented in the paper with the aid of a 1D contaminant transport modelling program. Future work will examine the influence of cement content and water content on diffusion properties of the soil-cement mixtures.

ACKNOWLEDGEMENTS

The authors would also like to acknowledge the Cement Association of Canada, the Portland Cement Association, and the Natural Sciences and Engineering Research Council of Canada for funding of the project. Jonathan Blay of Department of Pharmacology at Dalhousie University is also acknowledged for his assistance in this project.

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