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MONITORING SUBSURFACE BARRIER INTEGRITY USING PERFLUOROCARBON TRACERS

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ABSTRACT: Subsurface barriers are an extremely promising remediation option to many waste-management problems. It is recognized that monitoring of the barrier is necessary to provide confidence in the ability of the barrier to contain the pollutants. However, the large size and deep placement of subsurface barriers make detection of leaks a challenging task. Therefore, typical geophysical methods are not suitable for the monitoring of an emplaced barrier's integrity. Perfluorocarbon tracers (PFTs) have been tested as a means of barrier verification at the Hanford geotechnical test facility, where a soil/cement barrier was emplaced around a buried drum. PFTs were injected beneath the drum for three days in the center of the barrier 3 m below grade. The concentration of PFTs in seven external and two internal monitoring wells has been measured as a function of time over a 17-day period. The data have been analyzed through numerical modeling to determine barrier integrity and PFT diffusion rates through the barrier. This paper discusses the experimental design, test results, data analysis, and modeling of PFT transport in the subsurface system.

INTRODUCTION

Subsurface barriers are an extremely promising remediation option to many waste-management problems. Potential uses of subsurface barriers include surrounding and containing buried waste; as secondary confinement for underground storage tanks; to direct or contain subsurface contaminant plumes; and to restrict remediation methods, such as vacuum extraction, to a limited area. Subsurface barriers are being considered for use at many of the Department of Energy sites including: Sandia National Laboratory, Idaho National Engineering Laboratory, Oak Ridge National Laboratory, Hanford, Fernald, and Rocky Flats. Barriers are also considered an important remediation option by the U.S. Environmental Protection Agency (Siskind and Heiser 1993).

The ability to verify barrier integrity through monitoring will be required to gain public acceptance of subsurface barriers as either a primary or secondary means of confinement of wastes. To effectively contain the wastes, the barriers should be continuous and have few or no breaches. Currently, no placement technology can guarantee the completeness of the engineered barrier. A breach may be formed by many processes including discontinuous grout application; joint formation between grout panels; cracking during curing; localized "tears" due to differential settling; wet/dry cycling; and, over time, degradation of the grout due to chemical attack.

The large size and deep placement of subsurface barriers makes the demonstration of barrier integrity a challenging task. This becomes magnified if the permissible leakage from the site is low. Several geophysical techniques exist for the determination of the barrier's physical properties. These include the four major types of well-logging techniques (nuclear, electrical, acoustic, and thermal) as well as tracer technologies. A detailed review of the applicability of all of these techniques can be found in Heiser (1994). The major finding of the review are summarized next.

Nuclear logging techniques, neutron and gamma logging, are only accurate over a small spatial range and would require a prohibitive number of bore holes (spacing every meter). Even at this spacing, these techniques are able to detect bulk properties but are not able to accurately detect fractures.

Electrical and electromagnetic logging techniques such as electrical resistance tomography, radio-imaging method, and ground-penetrating radar are able to determine bulk properties at depth, but their spatial resolution is not high enough to detect discrete fractures. Electrical cross-bore-hole tomography (Daily and Rameirez 1993) and bore-hole induction logs (Boyd et al. 1994) have been used to map affected areas for remediation and to determine grout penetration in soils, but these studies have been limited to defining general locations and could not be used to infer barrier integrity. The radio-imaging method has been used at Sandia National Laboratory on their Chemical Waste Landfill to detect soil units of 0.5 m in size (Borns et al. 1993). Such resolution is still not accurate enough for barrier continuity verification.

Acoustic logging shows some promise in being able to locate small breaches. However, due to multiple reflections of the acoustic waves, interpretation of the output is often difficult. Seismic tomography, a form of acoustic logging, has been tested at Sandia National Laboratories Mixed Waste Landfill and was able to detect general grout locations; however, the technique was considered inadequate for verifying barrier continuity. In a study of high-resolution seismic imaging of fractures in rock, it was concluded that minor structures such as cracks were not detectable at the frequencies used in the study (Majer et al. 1991).

Thermal logging measures temperature variation within a region. As such, it may be useful to follow the curing of cementitious or thermosetting grouts and therefore define the region of grout injection. However, it will not yield data on barrier continuity.

Tracer techniques involve emplacement or injection of a substance that will migrate to a collection well. Based on the rate of arrival at the monitoring well and the transport properties of the tracer and materials in the subsurface system, estimates of barrier integrity can be obtained. For subsurface soil systems, the tracer can be radioactive or nonradioactive liquids or gases. Radioactive tracers can be incorporated into the barrier grout and the radiation field can be monitored to ascertain the location of the grout. In this case the migrating substance is the radiation particle.

For barrier integrity studies in the unsaturated zone, gas-phase tracers are needed. Liquid-phase tracers will not have high enough mobility to be useful for determining barrier integrity on a time scale of weeks. Gas-phase tracers will provide direct information for volatile compounds and will give evidence of a breach long before liquid solute contamination

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would escape from the barrier. Theoretically, gas-phase tracers show promise to be able to detect fractures on the order of a few centimeters in size. This needs to be demonstrated on field-scale applications.

Gas-phase tracers include perfluorocarbon tracers (PFTs) and sulfur hexaflourides. Both have been applied for leak detection in subsurface systems. PFTs have been used to detect leaks in buried natural-gas pipelines, the rate of dioxin movement into a commercial building from surrounding contaminated soil, the rate of leaking dielectric fluid from subsurface electrical cables, the rate of leaking gasoline from underground storage tanks, and the rate of radon ingress into residential basements (D'Ottavio and Deitz 1987, Horn et al. 1991).

In this study PFTs were chosen as the tracer. PFTs have the following advantages and characteristics as compared to other tracers (Deitz 1986).

- Negligible background concentrations; consequently, only small quantities are needed.
- PFTs are nontoxic, nonreactive, nonflammable, environmentally safe (do not contain chlorine), and commercially available.
- PFT technology is the most sensitive of all nonradioactive tracer technologies. Concentrations in the range of 10 parts per quadrillion of air can be easily measured.
- PFT technology can simultaneously deploy, sample, and analyze up to six PFTs with the same instrumentation. This results in lower costs and flexibility in experimental design, testing, and data interpretation.
- PFT concentrations can be analyzed in a few minutes in the field or in the laboratory using gas chromatography.

The ability to use multiple tracers at a single site can help to improve the spatial resolution of the breach. Theoretically, the combination of monitoring data with numerical modeling of the movement of the PFTs can be used to locate hole size down to a few centimeters in size. Testing of the resolution that can be obtained in the field remains to be done.

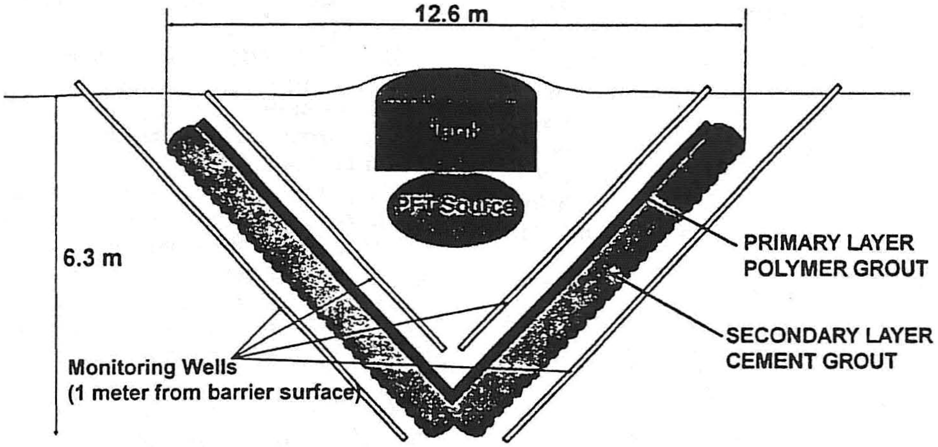
The focus of this paper is to describe the barrier verification tests conducted using PFTs at the Hanford geotechnical test facility and the analysis of the data from the test. The objective of the test was to demonstrate the proof-of-concept that PFT technology can be used to determine if small breaches form in the barrier and for estimating the effectiveness of the barrier in preventing migration of the gas tracer to the monitoring wells. The next section describes the test facility and the experiment. The experimental results and the analysis of the data follows. Based on the findings of this study, conclusions are offered and suggestions for future work are presented.

EXPERIMENTAL SYSTEM

Testing was performed at the Hanford geotechnical test facility that has been in existence since 1982 (Phillips and Fischer 1982). It was developed to obtain information on low-level waste subsurface burial subsidence control alternatives. Over the years, this facility has been used in numerous subsurface testing programs. In this study, a low-permeability barrier was emplaced around and beneath a simulated waste drum without disturbing the waste drum. The containment structure is a multibarrier comprised of a cementitious grout lined with a polymer grout. The system design called for the two grouts to be emplaced in a close-coupled fashion such that the polymer barrier is bonded to the cementitious barrier.

The demonstration of barrier integrity using tracers was conducted in two parts. In the first phase, PFTs were injected into the system beneath the center of the simulated waste tank approximately 3 m below grade prior to emplacement of the polymer grout. This permits evaluation of the cement grout as a barrier to release and allows estimation of transport parameters in the cement grout. After completing the tests and analyzing the data, the polymer grout was injected and the test repeated. This paper discusses the results of the first phase of testing. Analysis of the data from the second phase is continuing and will be reported at a later time. In the remainder of the paper, all discussion will refer to the soil/cement barrier system prior to injection of the polymer grout.

The physical system under study is displayed in Fig. 1. The approximate dimensions of the system are 6.3 m deep and 12.6 m in diameter. The cementitious barrier wall was constructed by injecting two parallel rows of grout at an angle of $\pi/4$ rad (45°) to the ground surface. The top view of the system is displayed in Fig. 2. In this study, the barrier was covered by 60 cm of soil. The use of a sloping barrier wall forms an inverted cone. The second row of the barrier is used to increase the thickness of the grout barrier and help ensure that large-scale breaches in the barrier do not occur. The average thickness of the cementitious grout barrier is 1 m (this is the thickness in the plane parallel to the barrier). Eight monitoring wells are uniformly spaced parallel to the barrier wall at a distance of approximately 1 m from the wall. Two internal monitoring wells were also placed to permit measurement of



Tank dimensions, 3 m diameter by 2.4 m height
Concrete barrier, 1.1 m thick
Polymer barrier, 0.3m thick
All monitoring wells 1 m from barrier surface

FIG. 1. Schematic Drawing of Side View of Subsurface Barrier System

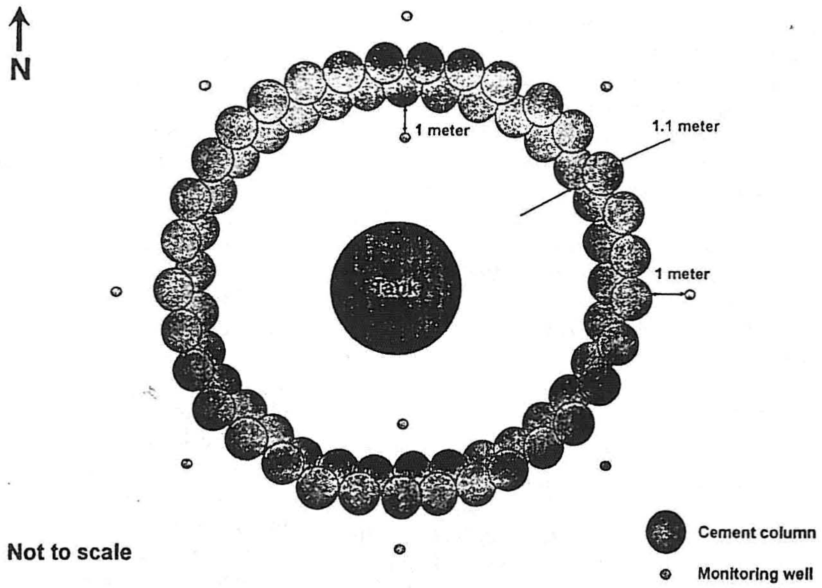


FIG. 2. Schematic Drawing of Top View of Subsurface Barrier System

the concentrations inside the barrier. The monitoring point of the internal wells are located approximately 1 m from the source. The tank represents a simulated waste form and has dimensions of 3 m in diameter and 2.4 m in height.

To support the PFT testing procedure, modeling of the PFT gas movement has been conducted for the subsurface system with the soil/cement barrier. The modeling was used to estimate the rate at which the tracer will travel through the barrier and reach the monitoring wells, to demonstrate the effects of small holes (1–10 cm) on concentration at the well, and the effects of the waste tank on movement of the gas. This modeling information was used to assist in the determination of injection rates for PFTs into the subsurface system, the frequency and duration of sampling, and the time to flush the tracers from the system prior to testing the soil/cement/polymer barrier.

CONCEPTUAL MODEL

The problem involves transport of an injected tracer through an engineered barrier (soil/cement) to a monitoring well. To model this requires knowledge of the rate of tracer injection, location of injection, geometry of the system, location of the monitoring wells, and transport properties of the PFT through the soil and soil/cement barrier.

The diffusion equation with a time-dependent external source is used to solve for the movement of the tracer from the injection location throughout the modeled domain. In cylindrical coordinates the equation is (Sullivan and Suen 1989)

$$\frac{\partial C(r, \theta, z, t)}{\partial t} = \nabla \cdot D(r, \theta, z, t)\nabla \cdot C(r, \theta, z, t) + M_s(r, \theta, z, t) \tag{1}$$

where $C(r, \theta, z, t)$ = tracer concentration (mass/length^3); $D(r, \theta, z, t)$ = diffusion coefficient (length^2/time); and $M_s(r, \theta, z, t)$ = rate of tracer injection into the system (mass/length^3/time).

In this application, the source term, M_s, is provided by injection of PFT into the center of the inverted cone directly beneath the waste tank. Therefore, $M_s(0, z, t)$ describes the source used in these experiments. In addition, to simplify the analysis cylindrical symmetry is assumed. This is a valid assumption provided the barrier is intact. If the barrier is not intact and has a breach, it may not be possible to assume cylindrical symmetry. Therefore, to simulate this physical system described in the previous section (Fig. 1), a two-dimensional cylindrical slice through the center of the cone was taken. The tank in Fig. 1 was represented as a no-flow boundary. For simplicity and because of the lack of site-specific data, it is assumed that there are two distinct materials in the subsurface system, the soil and the soil/cement barrier. The soil inside the barrier is assumed to be identical to that outside the barrier in terms of its transport properties (e.g., diffusion coefficient).

Simulations with a completely intact barrier were performed as a baseline. Then the effects of having small imperfections (caused by imperfect grouting) were studied. In this analysis, the imperfections are represented as a hole through the entire wall. Due to the assumption of cylindrical symmetry, the hole is assumed to be uniform around the circumference of the barrier. While this assumption should not be realized in practice, it is adequate for preliminary studies of the effects of a hole in the azimuthal plane of the imperfection. The range of hole sizes that were modeled was between 1 and 10 cm.

The dominant transport process for air in soil systems is believed to be diffusion (Hillel 1983). Advection resulting from barometric pressure changes can facilitate the release of the tracers to the atmosphere and was considered in the initial phase of the modeling. The advection due to barometric pressure changes is expected to vary cyclically. These pressure changes will lead to times when the flow is into the soil and times when the flow is directed out of the soil. The results indicated that for likely values of the cyclical advection velocity, diffusion would be the dominant transport process as expected and advection could be neglected (Sullivan et al. 1996). Substantial differences between the predicted concentrations of the diffusion only and diffusion with the cyclical advection case occurred only in the top meter of the soil.

The PFT perfluoromethylcyclohexane (PMCH) was used in these tests. The measured diffusion coefficient for the tracer PMCH in air is 5×10^{-2} cm^2/s. Measured values in the soil system at the Hanford geotechnical test facility are not available. To account for tortuosity effects in the soil, the diffusion coefficient of the PMCH in the soil has been estimated as 10^{-2} cm^2/s for the base case. This value is similar to that for radon gas in dry soils (Nielson and Rogers 1982). The test site is arid; therefore, dry soils are expected. As the moisture content of the soils increase, the diffusion coefficient decreases. This effect is generally minor until saturation of the soils is approached. At saturation, the diffusion coefficient will be that of the tracer in water, which is generally four or five orders of magnitude lower than in air. Estimates of radon diffusion coefficients as a function of degree of saturation are available (Nielson and Rogers 1982). The same diffusion coefficient is used for the soil inside and outside of the barrier.

The diffusion coefficient through the soil/cement was selected as 10^{-4} cm^2/s. The range of diffusion coefficients for radon gas through residential concretes is 10^{-4}–5×10^{-3} cm^2/s (Rogers et al. 1984). A value from the low end of the range was selected in an attempt to provide a lower estimate of release to the monitoring wells and to ensure that sampling would be able to detect the PFTs. It is expected that diffusion coefficients in the soil/cement barrier will be toward the high end of the residential concrete data.

COMPUTATIONAL MODEL

The subsurface barrier system is modeled in cylindrical geometry using a two-dimensional finite-element transport code, BLT (Sullivan and Suen 1989). This problem has two size scales. The first scale is that of the system itself. The height from the bottom of the subsurface barrier to the ground surface is 6.3 m. The radius of the barrier was also approximately 6.3 m and a total distance of 12.8 m was simulated in the direction parallel to the ground surface. The second scale is that of the size of the potential breach that is on the order of a few centimeters. It would require nearly 1,000,000 computational points to model the entire system on the scale of 1 cm. This is not computationally feasible. To account for the two scale sizes, variable mesh spacing was used. A fine mesh (order of 1 cm) was used in the region of the hypothetical breach. The mesh was increased in size as the distance away from the hypothetical breach increased. The slanting soil/cement barrier was modeled through definition of the finite elements used to represent the barrier to also slant at a $\pi/4$ rad (45°) angle. These two details led to a complicated finite-element mesh with 3,000 computational points (Fig. 3). Removing the assumption of azimuthal symmetry would require a three-dimensional simulation of the transport of the contaminant. This would require at least an order of magnitude more detail to simulate the spatial resolution appropriately.

Initially, the system is tracer free with the initial condition of zero concentration at all locations. The boundary condition assumed zero flux at the centerline due to the assumed symmetry. The specified-concentration boundary conditions where *C* is specified as zero were used at the top boundary and right-hand boundary defined in Fig. 3. The right-hand boundary is located at a large enough distance such that the tracer does not reach the boundary during the simulation period of 0.1 years. The top boundary was selected to have zero concentration to represent PFT concentrations in the atmosphere that are assumed to be zero. Numerical analysis performed with flow out of the ground surface and into the atmosphere indicated that the zero concentration boundary condition is an excellent approximation due to the higher transport rates in the atmosphere (higher diffusion and advection rates). The bottom boundary in Fig. 3 is represented as a zero mass-flux boundary. This will cause a slight overestimation of the concentrations in the modeled domain.

In the base case, the source was treated as a point source being injected at the centerline at an elevation 97 cm above the bottom of the facility. Because the injection of tracer during the experiment was through a 1/4-in. o.d. copper tube, the point-source approximation is an adequate representation of the experiment. This location is 40 cm above the top of the soil/cement layer in Fig. 3. Two injection scenarios were modeled: a 3.7-day pulse injection and continuous injection over the entire simulation period of 36.5 days. The air-injection rate was assumed to be 30 cm^3/min at a unit tracer concentration. The mathematical representation of the system as defined in (1) exhibits a linear response to the injection concentration. This linearity property was used to normalize all of the simulation concentrations to the injection concentration.

PFT tracers are nonreactive in soil systems and can be detected at levels of one part in 10^{15}. Typically, injection concentrations are on the order of 1 ppm. Therefore, the detection limit will be approximately 10^{-9} of the incoming concentration. One objective of the modeling work was to define the time at which the PFTs will first be detected at the monitoring wells and the time evolution of concentration at the monitoring wells. For the purposes of defining the experimental protocol (source strength and duration of injection), the minimum detection limit for the PFT was multiplied by a safety factor of 100. This provides a goal for the concentration to reach a value of one part in 10^{13} in the monitoring wells. Assuming an injection concentration of 1 ppm, the projected concentrations

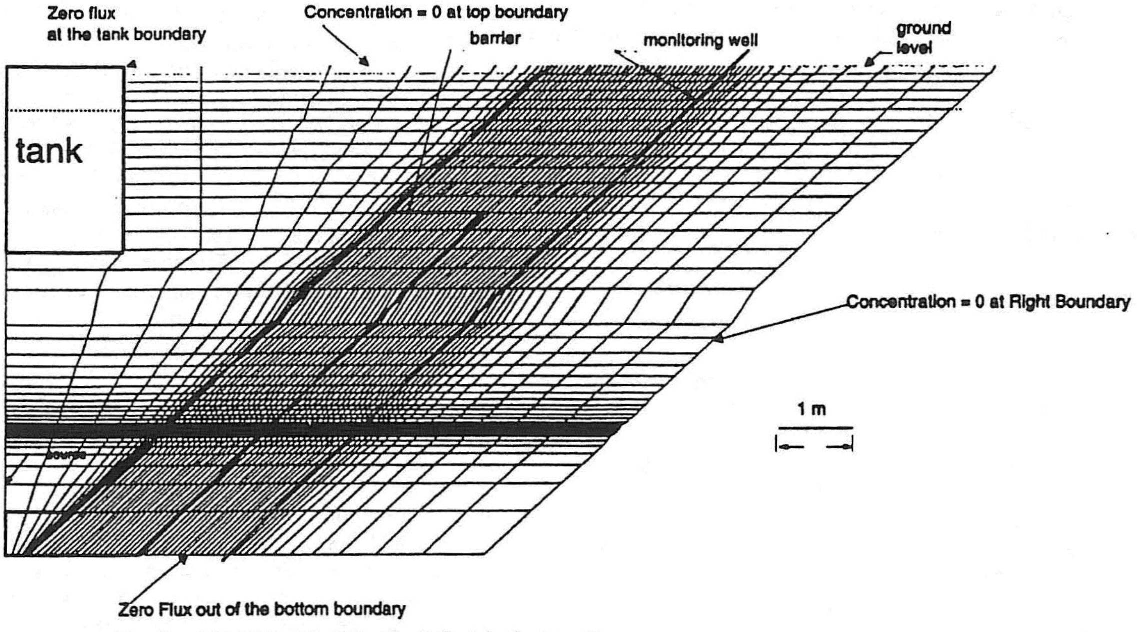


FIG. 3. Finite-Element Grid Used to Simulate Flow Through Subsurface Barrier System

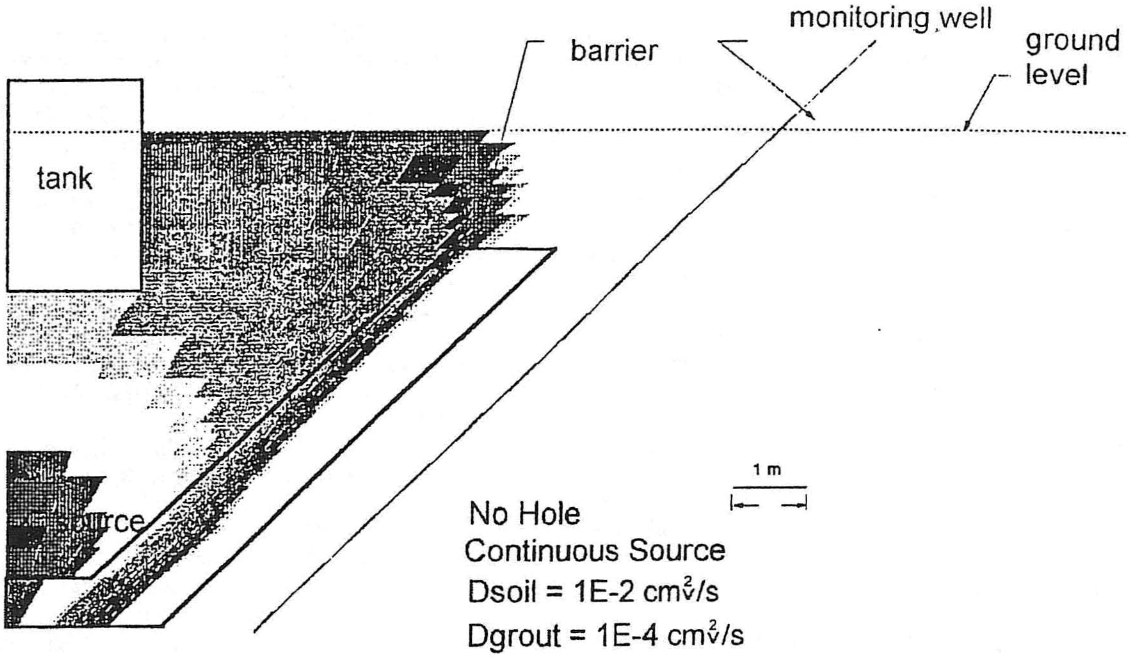


FIG. 4. Concentrations at 14.6 Days for Intact Barrier with Soil Diffusion Coefficient of 2×10^{-2} cm²/s and Barrier Diffusion Coefficient of 2×10^{-3} cm²/s

at the well normalized to the source concentration must exceed 10^{-7}.

Due to the choice of values of the diffusion coefficients from the low end of the expected range for the soil and soil/grout, it is expected that the concentrations estimated based on the base-case model parameters will be much less than the actual concentrations. In effect, the model predictions will overpredict the time it takes to reach the design-goal concentration of one part in 10^{13}.

MODEL RESULTS FOR DESIGNING EXPERIMENT

The computer code BLT (Sullivan and Suen 1989) was used to solve the preceding equations for the tracer plume due to injection of the tracer. A variety of cases with a wide range of parameters was considered to assist in gaining an understanding of the system behavior. The objective of these simulations was to estimate the time evolution of tracer concentration at the monitoring well.

The time evolution of the tracer plume was followed for 36.5 days. In the base case, a diffusion coefficient of 10^{-2} cm²/s for all soil regions in the modeled domain and 10^{-4} cm²/s in the grout wall. In the base case, it is assumed that the subsurface barrier wall is intact and no substantial breach occurs. In the simulation tracer was injected for the entire simulation period. The results of the simulation at 14.6 days after the start of the experiment are presented in Fig. 4. The contour plot color key is presented in Fig. 5. All projected concentrations are normalized to the injection concentration.

In Fig. 4, it is seen that for the base-case parameters, the simulated soil/cement wall provides an effective barrier to migration of the PFTs. Concentrations at the well 14.6 days after the beginning of tracer injection are more than eight orders of magnitude less than the injection concentration. Inspection of the output files indicates that the projected baseline concentrations are nine orders of magnitude less than the injection concentration at this time. Concentrations at the monitoring well exceeded the minimum design-basis value of 10^{-7} after 30 days.

To determine the effect of the barrier diffusion coefficient on release, the base case was modified by increasing the barrier diffusion coefficient by an order of magnitude to 10^{-3} cm²/s. This value is expected to be more representative of the soil/cement barrier because it is in the middle of the range of measured radon diffusion coefficients through residential concretes (Rogers et al. 1984). With a soil/cement diffusion coefficient of 10^{-3} cm²/s, a review of the output files indicated that the predicted concentrations at the monitoring well at 14.6 days reached a maximum normalized concentration of 7×10^{-6} and averaged more than 10^{-6}. This is much greater than the design-basis concentration.

■	>=5E-2
■	1E-2 - 5E-2
▨	5E-3 - 1E-2
□	1E-3 - 5E-3
░	5E-4 - 1E-3
░	1E-4 - 5E-4
▨	5E-5 - 1E-4
▨	1E-5 - 5E-5
▨	5E-6 - 1E-5
▨	1E-6 - 5E-6
■	5E-7 - 1E-6
■	1E-7 - 5E-7
░	5E-8 - 1E-7
□	1E-8 - 5E-8
□	<1E-8

FIG. 5. Contour Plot Color Key (Normalized Concentration)

To determine the effect of a small breach in the barrier a 5-cm gap was simulated as having the same diffusion coefficient as the soil, 10^{-2} cm²/s. The hypothetical gap in the barrier was located at an elevation of 1.8 m from the bottom of the modeled domain, 0.8 m higher than the source. The total distance from the source to the edge of the hypothetical gap in the barrier is 1.8 m. The results of this simulation at 14.6 days after the start of the injection (Fig. 5), indicate that the breach has a pronounced effect on the tracer plume. Streaming through this 5-cm breach is clearly evident. The peak normalized concentration at the well is 4×10^{-6}. Average concentrations along the lower section of the well are above 10^{-6}, a three-order-of-magnitude increase over the projected concentration for the intact wall. In fact, the projected concentrations at the monitoring well for the 5-cm hole simulation was of the same order of magnitude as the case with the barrier diffusion coefficient increased an order of magnitude over the base-case value.

In all three cases, the tracer plume within the region bounded by the subsurface barrier is almost identical. Average concentrations in this region are approximately 10^{-3}, four orders of magnitude larger than at the monitoring well location for the case with a barrier breach. This indicates that only a small fraction of the tracer reaches the monitoring wells under the conditions simulated.

The gap size varied from 1 to 10 cm and the results were similar. Even a 1-cm gap would permit the concentration of PFT tracer that reaches the monitoring well to exceed the base case (an intact barrier) value by 2–3 orders of magnitude in the early stages (i.e., before diffusion through the barrier becomes an important source at the wells). The large increase in predicted release due to a small breach indicates that resolution of breaches on the order of a centimeter in size should be possible. However, this needs to be demonstrated on field-scale problems. The assumption of radial symmetry most likely will not be valid if a breach occurs. Most likely any breach would have been localized in space. In this case, a three-dimensional representation of the subsurface system would be appropriate.

In the field experiment, a breach would be indicated if the measured concentrations in the eight external monitoring wells differed by a several orders of magnitude. Relatively uniform concentrations at the monitoring wells indicates that a breach has not occurred.

COMPARISON OF EXPERIMENTAL RESULTS WITH MODEL PREDICTIONS

To test the concept of monitoring barrier performance with PFTs, tracers were continually injected for three days into the area contained by the soil/cement barrier (Fig. 1), just beneath the empty tank at the center of the region bounded by the cone. Air spiked with the tracer PMCH was injected through a 1/4-in.-o.d. copper tube at a rate of 0.2–0.25 cm^3/s at a PMCH concentration of 373 ppm. Seven monitoring wells were located parallel and approximately 1 m outside of the barrier. The wells are designated by compass direction, i.e., N for north, NE for northeast, etc. The eighth well at the location designated as west malfunctioned. Measurements for PFTs were taken from each well for 18 days after the start of injection. Exterior monitoring wells were slotted over their length. Samples were taken by inserting a sampling port at the end of a steel rod. The rod was inserted to the appropriate sampling depth and a sample was taken passively. Sample sizes ranged from 1–60 mL.

PFT concentrations within the region bounded by the barrier were measured during the three-day injection period. Interior wells were slotted over the length of the well. Therefore, measurements are representative of the average over the well volume. The data showed a net drift toward one side of the barrier. The interior monitoring well designated as N, for north, had measured concentrations approximately one-order-of-magnitude greater than the interior well designated S, for south. If diffusion was the only transport mechanism, the concentrations at these two wells, which are equidistant from the source, would be equal. Therefore, advection is occurring. The cause of this net drift is not known; however, it has been postulated that it is due to the injection flux (0.2–0.25 cm^3/s). This and other possible explanations are under investigation. The fact that there is a drift indicates that the exterior concentrations on wells near the north side should exceed those on the south side by an order of magnitude provided the barrier is intact (i.e., no breach).

The time evolution of measured concentration normalized to the injection concentration at the seven exterior monitoring wells is displayed in Fig. 6. Each of the monitoring wells shows similar behavior over time. The spread in the measured concentrations is approximately one order of magnitude and this is consistent with the internal well-monitoring data; i.e., highest concentrations are measured on the north side of the facility. The drop in concentration between the inner and outer monitoring wells (i.e., across the barrier) was approximately four orders of magnitude at the end of the injection period of three days. There was no evidence of a substantial breach in any region as the drop in concentration across the barrier was consistent at all monitoring wells. The actual measured PFT

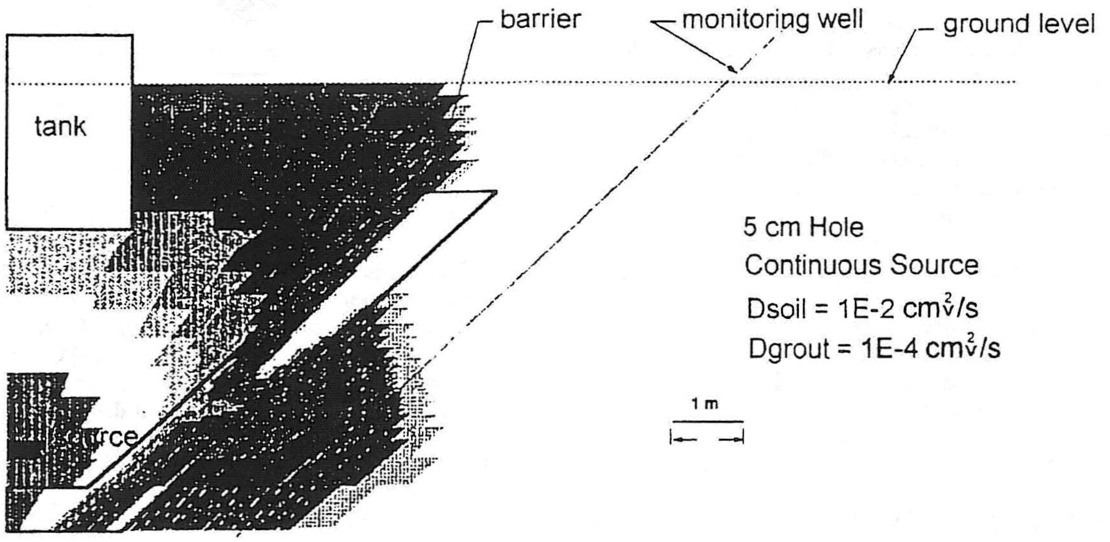


FIG. 6. Concentrations at 14.6 Days for Barrier with 5-cm Breach and Soil Diffusion Coefficient of 2×10^{-2} cm^2/s and Barrier Diffusion Coefficient of 2×10^{-3} cm^2/s

concentrations are in the range of 100 parts per trillion (normalized concentration of 3×10^{-7}) to 100 parts per quadrillion (normalized concentration of 3×10^{-9}).

To estimate diffusion coefficients in the soil and the barrier, prospective model evaluations were performed. The computational model is similar to the one described to examine the influence of a breach in the barrier (Fig. 2), with the exception that the dimensions were changed to match the as-built dimensions exactly and the source location was changed to reflect the experimental conditions. The major change in input involved increasing the barrier thickness to 1.15 m and adjusting the location of the source to directly under the simulated waste tank. To facilitate comparison of the measured values obtained from the slotted wells, average concentrations were estimated by taking the numerical average of concentrations along the region of the monitoring well.

The base-case diffusion coefficient values, $D_{soil} = 10^{-2}$ cm^2/s and $D_{wall} = 10^{-4}$ cm^2/s, provided concentration estimates (on

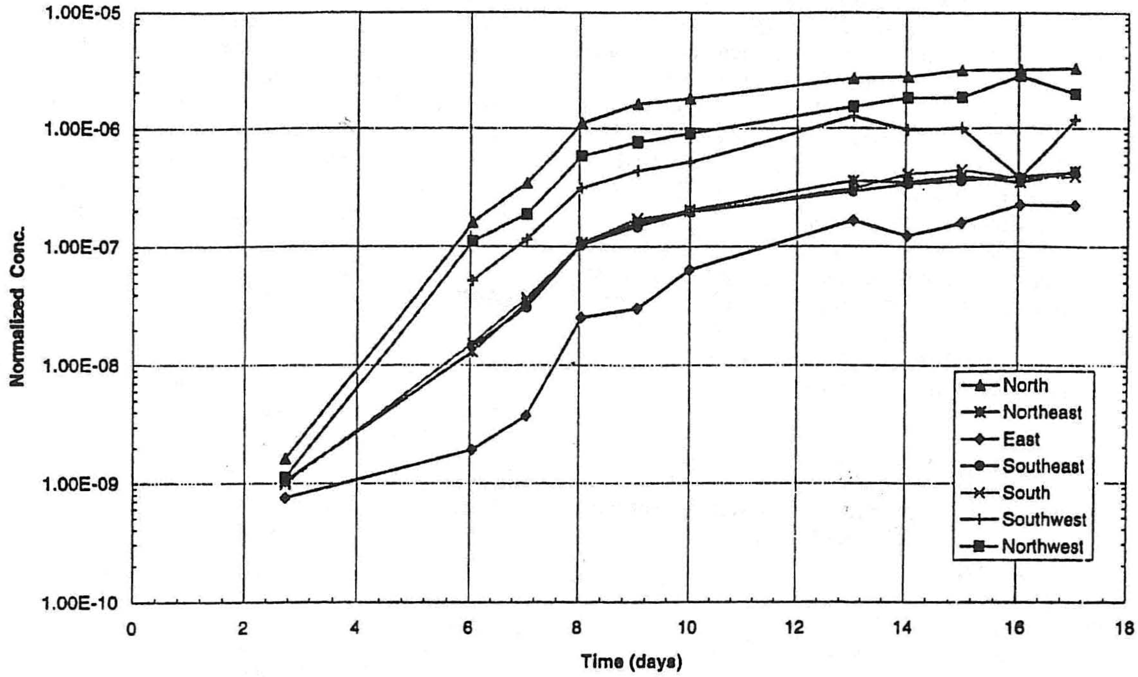


FIG. 7. Time History of PFT Concentration in Seven Exterior Monitoring Wells

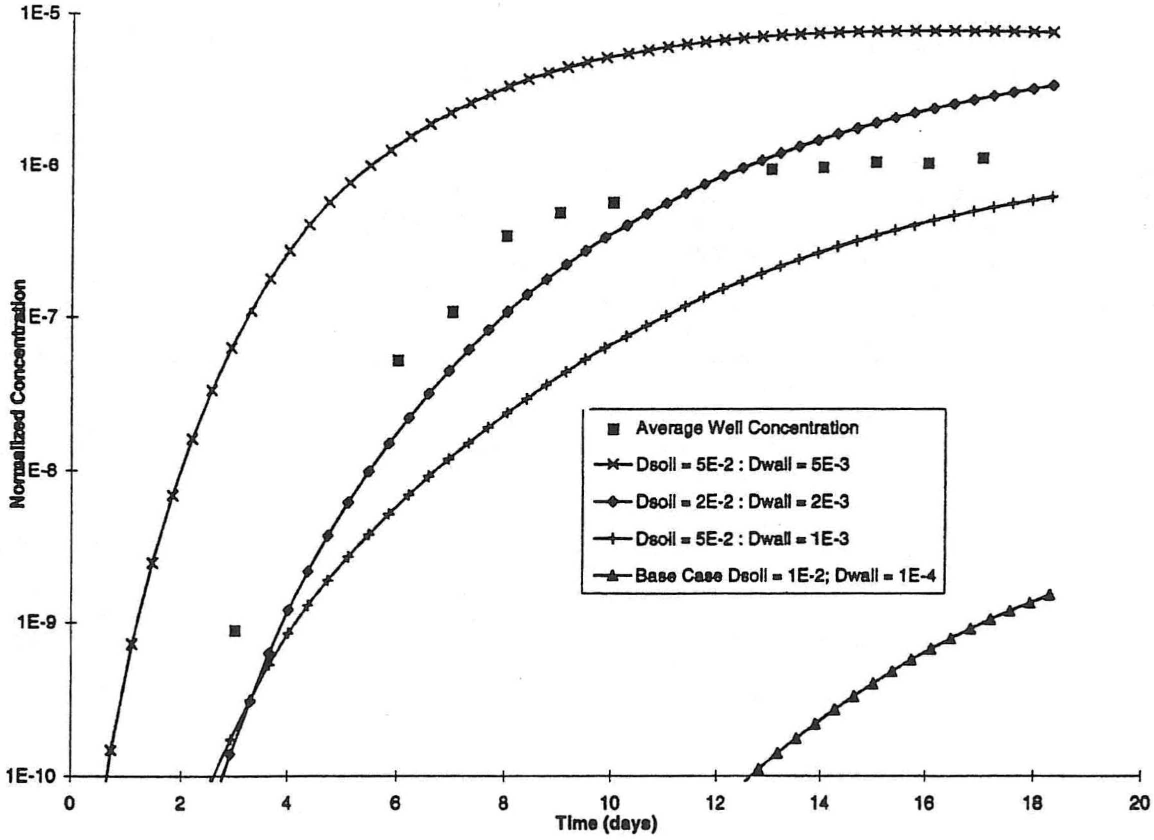


FIG. 8. Comparison of Time History of Average Monitoring Well Concentrations with Model Results

the order of 10^{-9} after 18 days) that were far lower than the measured value (on the order of 10^{-7}). This was expected because the base-case values were chosen with the intent of underpredicting the amount that would reach the wells to ensure that detection would be possible.

A range of different values of the diffusion coefficients was simulated. The results have been compared to the measured average value of the seven monitoring wells and are displayed in Fig. 7. The predicted results of the base-case simulation are also included on Fig. 8 and the predictions for these parameters are much less than the measured data, as expected. From the evaluations with different diffusion coefficients, the soil diffusion coefficient for the PFT has been determined to lie between 1 and 5×10^{-2} cm^2/s under the test conditions. The diffusion coefficient for the soil/cement barrier has been determined to lie between 1 and 5×10^{-3} cm^2/s. The best fit was obtained using a soil diffusion coefficient of 2×10^{-2} cm^2/s and a barrier diffusion coefficient of 2×10^{-3} cm^2/s. Attempts to improve the fit by regression analysis or other statistical techniques have not been undertaken at this time. At this time, it is felt that due to the net drift exhibited during injection, diffusion was not the only process leading to transport during the injection phase. Therefore, fine tuning the estimate would have little meaning within the limits of the model.

ADDITIONAL EXPERIMENTS

After completion of the PFT testing for the soil/cement barrier, a polymer grout liner was injected on the interior of the barrier to further reduce the potential for transport. PFT injection tests were repeated and the data are currently undergoing evaluation. In this experiment, the sampling points were located every 0.65 m (2 ft) along the length of the monitoring wells.

Upon completion of the PFT injection tests for the close-coupled barrier system, the soil surrounding the barrier was excavated. Visual inspections of the close-coupled barrier confirmed that a breach did not occur in either the soil/cement or polymer components of the barrier.

CONCLUSIONS

The proof-of-concept that PFTs could be used as a method for monitoring barrier performance has been demonstrated. A field-scale experiment was conducted, the data collected and analyzed. The results support the feasibility of detecting tracers outside of the barrier on the time frame of a few weeks.

Modeling the transport of PFT tracers in a subsurface system consisting of soil and a soil/cement barrier has been conducted. For the base case, a two-order-of-magnitude difference in the PFT diffusion coefficient in the soil and barrier, small holes (on the order of a few cm) simulated as having the same transport properties as the soil, should be easily detectable. In an actual system, the diffusion properties may lie between the intact barrier and the soil. This will make resolution of the spatial locations more difficult. As the difference in diffusion coefficients of the soil and barrier decreases, the ability to detect small holes also decreases.

Site-specific data on transport parameters were not available. Therefore, the model evaluations were compared to the experimental data and used to estimate the diffusion coefficient for the PFT through the soil and barrier. The best fit to the data indicates that the soil diffusion coefficient is approximately 2×10^{-2} cm^2/s and the barrier diffusion coefficient of 2×10^{-3} cm^2/s. These values are in the range of expected values based on diffusion coefficients of other gases through soil and cementitious systems.

The fact that the barrier emplacement was successful in that no large-scale breaches were formed prevented field-scale demonstration of the accuracy of PFTs in defining a breach. Model evaluations indicate the feasibility of locating breaches down to a few centimeters in size. However, experimental verification of this concept is needed. It is recommended that tests be performed on subsurface barriers with preformed breaches of known location, size, and geometry. In addition, work should be done for partial breach failure (e.g., a region with half the thickness of the barrier to simulate improper grouting). These types of tests are needed to permit demonstration of the resolution that can be obtained by PFTs and build confidence in the ability to understand, monitor, and predict the behavior of subsurface barriers.

APPENDIX. REFERENCES

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