

Scale-up Testing—Foam as A Remedial Ammendment Carrier - 11029

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ABSTRACT

This paper describes results from intermediate-scale, two-dimensional testing of foam injection into sedimentary materials collected from the U.S. Department of Energy (DOE) Hanford Site area. The testing was performed to evaluate the transport of new, more robust, foam generation formulas developed by PNNL on field applicability. The mechanisms of transport, foam stability, delivery pressure, foam migration and the ability of foam to deliver remedial amendments for stabilization of uranium contamination were evaluated. Testing was accomplished in a test bed that is designed to focus on two-dimensional flow, in the form of two, thin rectangular boxes. Each of the boxes holds approximately 135 liters (255 kilograms) of sediment. A total of six sets of tests have been conducted, the last two of which will be described here. During the fifth and sixth tests, foam, generated by means of mechanical blending, was injected into the central screened segment of the left side of each box while air was extracted from multiple screened segments along the right side of each box. During both of the tests the boxes were loaded to contain a rectangular zone of fine material and a rectangular zone of coarse material. Portions of the sediment were augmented with uranium-rich calcite to produce known concentrations of uranium. The foam generating formulas used in both tests contained sodium phosphate and sodium tripolyphosphate as a remedial amendment. Subsequent to each of the two tests, the test beds were disassembled, and samples of the sediments were taken and analyzed for a number of parameters, depending on the specific test. The data indicated that uranium contamination may be successfully immobilized and that the directional movement of the injected foam can be controlled.

INTRODUCTION

The production of nuclear fuels and weapons has also resulted in the production of numerous forms of legacy waste within the U.S. Department of Energy (DOE) Complex. To make progress toward remedial methods for waste cleanup, while minimizing risk and cost and maximizing success, the DOE Environmental Management (EM) organization has identified action areas to advance the state of knowledge. One example of such an action area site is at the BC Cribs located on the DOE Hanford site. Pacific Northwest National Laboratory (PNNL) is leading a multi-agency consortium investigating foam delivery as a remediation tool for the deep vadose zone contamination at the Hanford site.

Some of the most challenging legacy waste remediation problems are those associated with the deep vadose zone sites. These areas pose unique problems for remediation, as the site specific geology, geochemistry, depth of contamination and the variety of contaminants present substantial uncertainties. Conventional remediation technologies and surface remedies have limited applicability and efficacy in controlling water flux and contaminant behavior and are limited in applicability due to the depth and heterogeneous nature of the vadose zone. Alternatively, chemical fixation technologies can be used as a means of in situ immobilization of metals and radionuclides. The objective of this initiative is to transform foam technology, initially developed within the oil and gas industry, into a viable method for delivering remedial amendments to vadose zone environments, to immobilize technetium-99 and uranium. PNNL has focused on the development of foam producing chemistries and bench-scale evaluations of this application. In collaboration with this PNNL effort, MSE is tasked with completing the scale-up testing for the foam injection technology. Six sets of scale-up tests have been performed in test boxes designed to focus on two-dimensional flow. The tests were used to evaluate foam stability, delivery pressure, injection rate, effect of foam adsorption to sediments, influence of formation heterogeneities,

post-injection moisture distribution, and the ability of foam to deliver remedial amendments for stabilization of uranium or technetium contamination.

INTERMEDIATE-SCALE TESTING

The intermediate-scale laboratory testing and data analysis was completed at the MSE Test Facility in Butte, Montana. Testing was performed to evaluate the field applicability of the process as related to foam stability, delivery pressure, foam migration and the ability of foam to deliver remedial amendments while maintaining the performance of those amendments for stabilization of uranium contamination.

Test Objectives

The overall objective of the project is to develop a foam delivery technology to be used for the distribution of remedial amendments to deep vadose zone sediments for the in situ immobilization of metal and radionuclide contaminants in those sediments. To that end, the scale-up testing was designed to:

- investigate the ability of a unique foam formulation generated by mechanical methods to propagate through a meso-scale sediment volume;
- investigate the pressure gradient required to propagate the foam through the sediment volume;
- investigate the ability of changes in extraction volume and position to alter the movement of foam through the sediment; and
- test the immobilization of uranium by the remedial amendment carried by foam within the sedimentary volume.

Injection Testing Approach

The scale-up testing was performed in a test bed that was comprised of two, thin-rectangular boxes designed to focus on 2-D flow. Each box had dimensions of 100 centimeters (cm) in length by 90 cm high by 15 cm wide and held approximately 135 liters (L) [255 kilograms (kg)] of sediment. The boxes were formed of steel sheeting with a front face of clear acrylic and a removable lid to allow for loading and sampling of the contained sediment. Foam was injected into the central section of each box through a segment of polyvinyl chloride (PVC) slotted well casing, and air was extracted from the boxes through a similar system. During the fifth test, air was extracted from three spigots aligned along the right side of each test box. During the sixth test, air was extracted from the aforementioned spigots along the right side of each box and from an additional extraction spigot located near the right end of the box lid. A frontal view schematic of the test bed used for the testing is shown in Fig. 1.

Each box was accompanied by a foam generating system consisting of a pressurized air system, a solution pump, a mechanical mixer in the form of a speed controlled blender and a water-based cooling system as shown in Fig. 2. Foam was produced by mixing the foam generating formula (including the remedial amendment) with air. That mixture was injected into the blender carafe where the rotating blades of the blender produced dense foam with individual bubble diameters that are less than 0.1 millimeter. The resulting foam was then injected into the test boxes using the pressure from the air system as the force of injection.

Each test box also included five sample spigots, nine pressure transducers, seven time-domain reflectometry (TDR) probes and a PC for data collection and retrieval. These devices were incorporated into the back wall of each of the test boxes so they did not interfere with visual observations being made from the front of each box. The instruments were used for determining the real-time pressure gradient and moisture distribution within each box during the testing sequences. The sample spigots were used to acquire samples of the foam as it traveled through the sediment. Fig. 3 illustrates the instrumentation within the test boxes while Figure 4 is a schematic showing the location of the instruments.

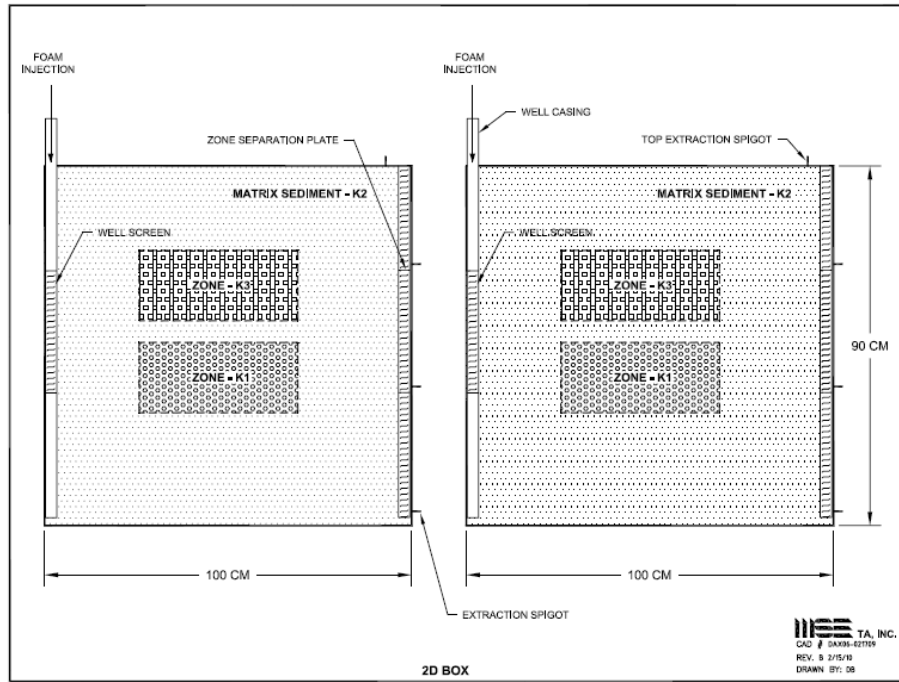


Fig 1. Schematic frontal view of test bed.



Fig 2. Mechanical blender used for foam generation.

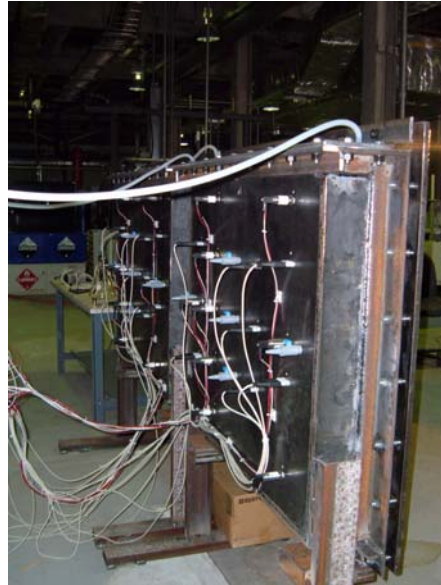


Fig. 3. Back of the test boxes showing the instrument locations.

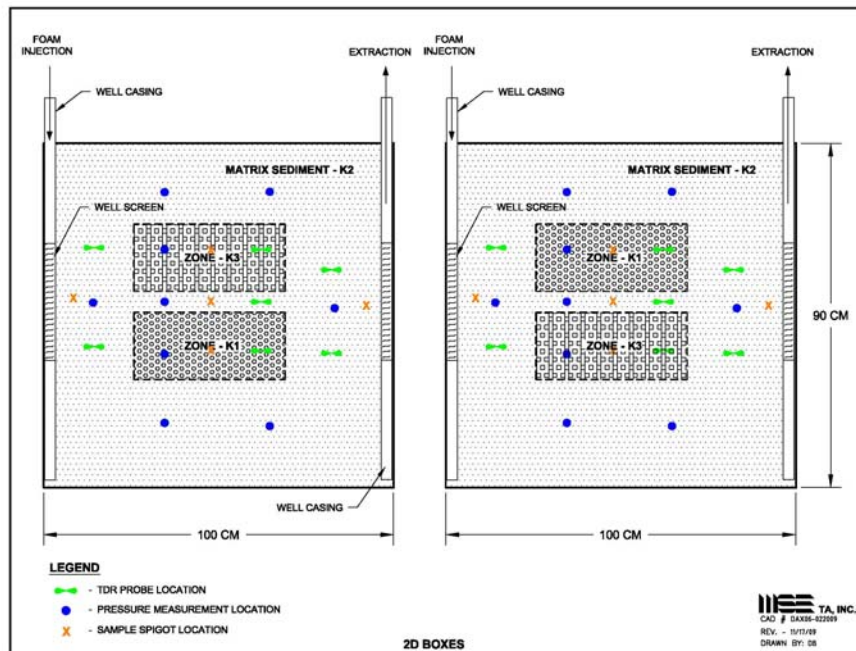


Fig 4. Schematic showing instrument locations in test bed.

Three sediments from the Hanford area were used for the tests. The first of these sediments (designated as K1) was quite coarse with particles ranging from 8 millimeters (mm) to less than 0.075 mm. The hydraulic conductivity of a packed sample of this sediment was 1.15×10^{-1} cm/second (s). The initial moisture concentration of the K1 sediment was approximately 3.5% by volume. The second sediment (designated K2) was also quite coarse with particles again ranging from 8 mm to less than 0.075 mm.

The hydraulic conductivity of a packed sample of this sediment was 4.69×10^{-2} cm/s. The initial moisture concentration of the K2 sediment was approximately 5% by volume. The third sediment (designated K3) was coarse sand with particles ranging from 2 mm to less than 0.075 mm. The hydraulic conductivity of a packed sample of this sediment was 2.93×10^{-3} cm/s. The initial moisture concentration of the K3 sediment was approximately 9% by volume. The grain-size distribution of the three sediments is shown in Fig. 6.

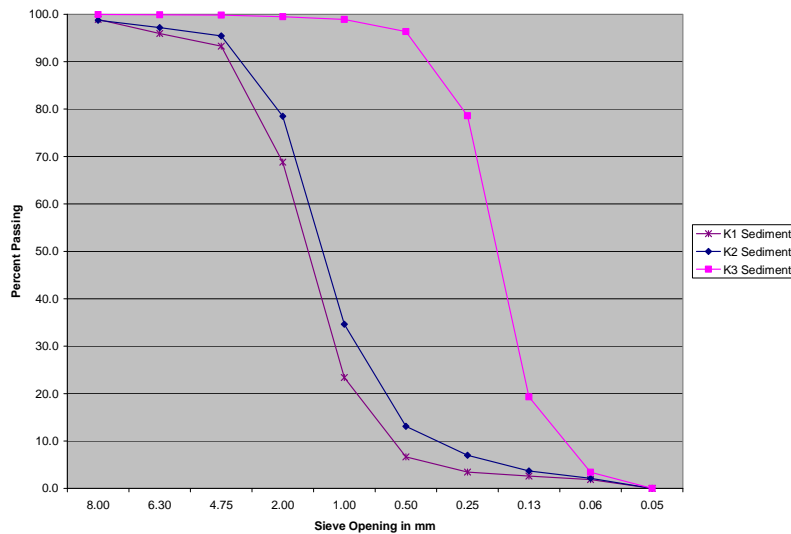


Fig. 5. Grain size distribution of test sediments.

Two sets of tests were conducted using both boxes for each test. Descriptions of those test sets are listed below.

- Test 5: Both of the boxes were loaded so as to produce two rectangular heterogeneous zones (one zone with K1 sediment and one with K3 sediment) surrounded by K2 sediment (see Figure 1). In addition, the central portion of the fine-grained sediment zone was augmented with uranium-rich calcite to produce an area containing approximately 300 mg/Kg total uranium. Foam was produced using PNNL formula # 172. The injection rate for both boxes was 60 milliliters per minute (mL/min). Air was extracted from each box at the same rate as foam was injected. The total phosphate concentration for the first box was 5000 ppm while the total phosphate concentration for the second box was 3000 ppm. The phosphate concentration for both boxes, was developed with a 75:25 ratio of orthophosphate to polyphosphate.
- Test 6: Both of the boxes were loaded in the same manner as in Test 5. In addition, the upper-right portion of the fine-grained sediment zone was augmented with uranium-rich calcite to produce an area containing approximately 300 mg/Kg total uranium. Additionally, the upper-central portion of the coarse-grained zone was augmented with uranium-rich calcite to produce an area containing approximately 300 mg/Kg total uranium, in the left box and 600 mg/Kg in the right box. Foam was produced using PNNL formula # 173. The injection rate for both boxes was 70 milliliters per minute (mL/min). The phosphate concentration for the first box was 5000 ppm while the phosphate concentration for the second box was 3000 ppm. The phosphate concentration for both boxes, was developed with a 90:10 ratio of orthophosphate to polyphosphate. Air was extracted from all four of the extraction spigots of the first box from the beginning of the test. Air was extracted from the second box from only the three side spigots until the foam front reached a position approximately one half of the horizontal distance across the box. At that point air was extracted from all four of the

extraction spigots of the second box. Overall, air was extracted from each box at the same rate as foam was injected.

Subsequent to each of the tests, the test boxes were disassembled, and samples of the sediments were collected from a grid-like pattern through each box. These samples were then analyzed for a number of parameters including percent moisture, total uranium and leachable uranium. In addition, a number of parameters including the foam quality and stability were tested via sampling throughout the duration of each test, and a number of operating parameters that included the injection pressure, the fluid and air injection rates, the extraction rate, and the internal pressure within each box were monitored throughout the duration of each test. Finally, the propagation of the foam wetting front was tracked through the sediment within the box during each test.

TEST RESULTS

The initial concept of the two sets of injection tests was to determine if the new foam formulation could propagate across the entire length of the box, if the foam movement could be manipulated using a horizontal extraction well and if at least 3 pore volumes of the foam solution could be moved across the box.

Test 5

As it was injected, the foam moved into the sediment and formed arcuate wetting fronts that expanded with time of injection. When the edges of the wetting front reached the upper and lower edges of the box the shape of the wetting front was irregular, but distinctly vertical. Throughout the test, the movement of foam bubbles could be discerned within the coarse-grained portions of the sedimentary mass behind the wetting front. In addition, as the foam fronts contacted the fine-grained zones, the fine-grained sediment appeared to extract moisture into those zones by a process of wicking, this was inferred since actual movement of the bubbles could not be observed within the fine-grained zones. After approximately 7 hours of injection the wetting front reached the extraction end of both injection boxes. Within thirty minutes of the wetting front reaching the extraction end of the box, liquid was being extracted from the extraction ports. The injection was terminated at that point due to the potential for exceeding the functional pressure of the carafes.

Subsequent to the completion of Test 5, samples of the sediment from both of the test boxes were analyzed for contained moisture. Fig. 6 shows the results of these analyses.

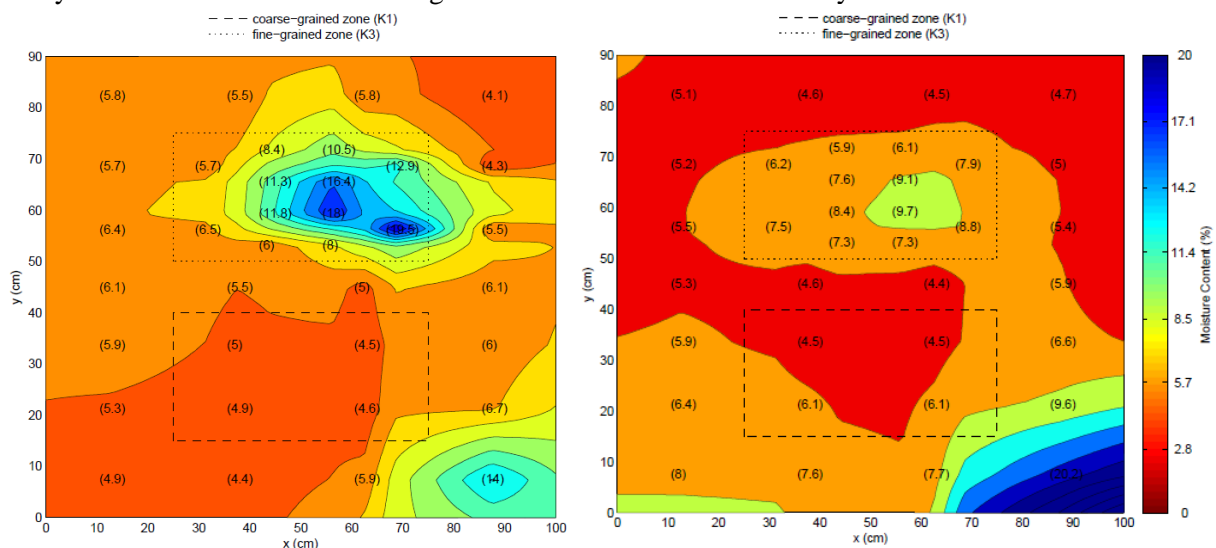


Fig. 6. Test 5 post-test gravimetric moisture percentages.

During Test 5, the pressure probes inserted into the back of both test boxes were operational. The data accumulated from those probes are shown in Fig. 7. A number of the early changes in pressure are due to operational changes being made to the test box while in operation. The probes denoted a general increase in internal pressure in the boxes as the test proceeded with similar increases in both boxes.

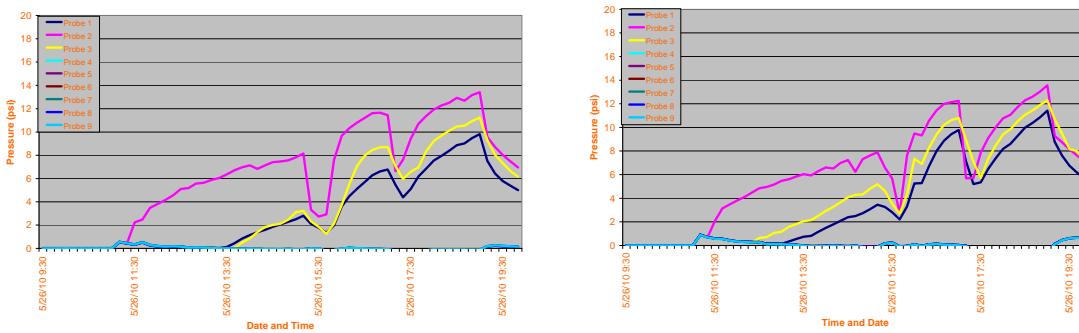


Fig. 7. Test 5 internal pressures

Time-domain reflectometry data was collected from the probes associated with both boxes used during test number 5. The results of that data collection are shown in Fig. 8. As can be seen from that figure, the TDR probes responded to changes in the volumetric water content of the surrounding sediments. Several of these changes in water content can be attributed to the arrival times of the foam wetting front at the positions of the probes.

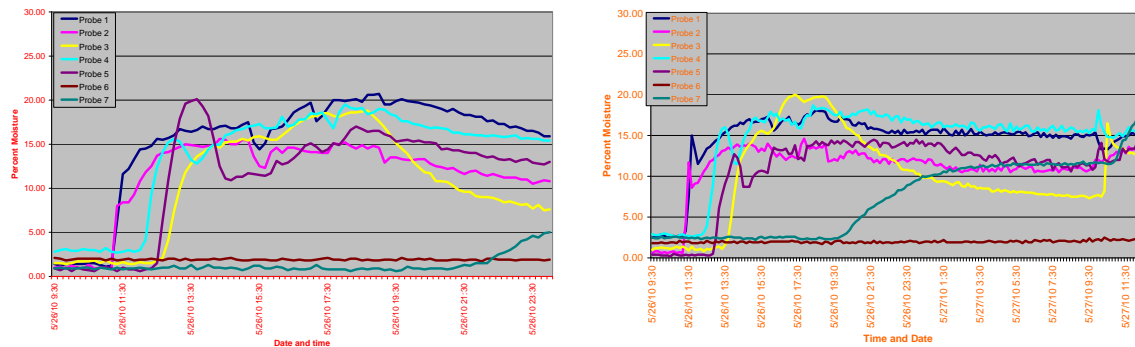


Fig. 8. Test 5 TDR data

TDR Probe number 3 was located in the fine-grained heterogeneous zone of both boxes. The information collected by that probe denotes a rapid increase in moisture content followed by a static moisture period, followed finally by a slow decrease in measured moisture. This pattern within the TDR data could be caused by the aforementioned wetting and drying sequence of the fine-grained sediments. This observation and hypothesis is the subject of further investigation.

The post-test moisture pattern within the sediments and observations made during the test revealed that a small section of the sediment in the lower right corner of each box contained more moisture than the remainder of the lower section of sediment. This moist section of sediment was decreased in size during the later stages of the test and likely would have disappeared had the test been allowed to run for a longer period of time. The fine-grained sediments contained more moisture than the coarse-grained material with

higher moisture concentrations in the fine-grained sediments of the left hand box than in those of the right hand box.

The total and leachable concentrations of uranium in the post-test sediment samples were measured to determine the effectiveness of the remedial amendment augmented foam to immobilize in-situ the uranium contamination. The results of these analyses are shown in Figures 9 and 10 respectively.

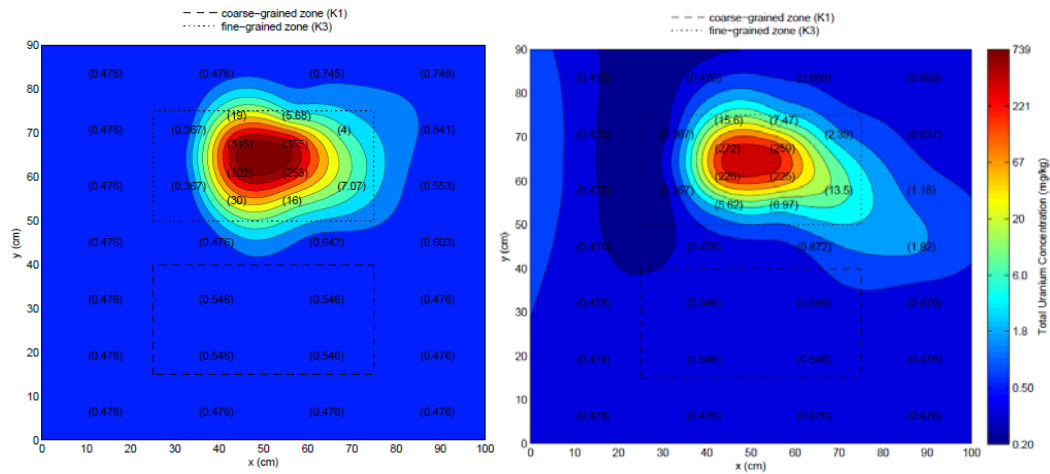


Fig. 9. Test 5 total uranium concentration

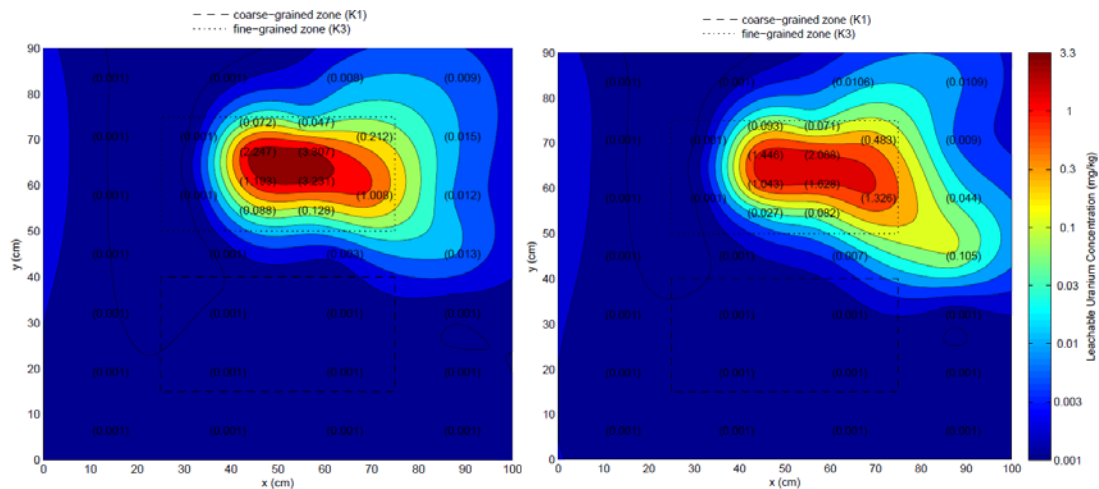


Fig. 10. Test 5 leachable uranium concentration

Test 6

The injection of foam into the two test boxes for Test 6 was conducted one-box-at-a-time. The injection into the right box was conducted first followed by the left box. During the injection into the right box, the extraction port that penetrated the lid of the box was used for extraction from the beginning of the injection. During the injection into the left box this same extraction port was only used for extraction after the foam wetting front had moved approximately one-half of the horizontal distance into the box.

Do to the variances in the extraction of air from the boxes, the movement of foam in the sediments of the two boxes also differed. The initial direction of foam movement within the sediments of the right box

used for Test 6 was both upward toward the top of the box and outward into the sediments. Overall, the direction of foam movement was toward the extraction port that penetrated the lid of the box. After approximately 1 hour of injection a flow path was established across the upper third of the box. This process continued for the first two hours of injection when foam began to be extracted from the upper extraction port on the right side of the box. No foam or liquid was extracted from the extraction port on the lid of the box. After approximately two hours the extraction port on the lid of the box was closed. The foam flow path was not affected by this action and foam began to accumulate in the upper right corner of the box. Foam from this accumulation began to move downward along the right edge of the box, toward the lower extraction wells, leaving a dry area of sediment in the right-central portion of the box. The injection was stopped after a period of approximately seven hours due to an issue with the electrical system supplying power to the test. Throughout this portion of Test 6, the movement of foam bubbles could be easily observed within the coarse-grained sediments both across the top of the box and near the center of the box. In addition, as the foam front contacted the fine-grained zone, the fine-grained sediment appeared to extract moisture into those zones possibly by a process of wicking, as movement of bubbles was not observed within the fine-grained zones.

The initial direction of foam movement within the sediments of the left box used for Test 6 was very similar to that observed during Test 5. As it was injected, the foam moved into the sediment and formed an arcuate wetting front that expanded with time of injection. When the edges of the wetting front reached the upper and lower edges of the box the shape of the wetting front was irregular, but distinctly vertical. Throughout the test the movement of foam bubbles could be discerned within the coarse-grained portions of the sedimentary mass behind the wetting front. As was previously observed, when the foam front contacted the fine-grained zone, the fine-grained sediment increased in moisture content. Five hours after injection was initiated the foam front had reached a point approximately one-half of the horizontal distance across the box, at which point the horizontal extraction port located in the lid of the box was activated. When the extraction port in the lid of the box was opened the movement of the foam front changed and rather than remaining vertical, the upper quarter of the front began to extend. This process continued until foam began to be extracted from both the lid port and the upper port on the side of the box approximately eight hours after the initiation of injection. As the injection continued, the extended portion of the wetting front expanded until all three of the extraction ports along the right edge of the box were emitting foam some eleven hours after the initiation of injection. Foam continued to be extracted from the three ports along the right edge of the box through the remainder of the injection period, while foam was extracted from the lid port for only the three hour period from eight to ten hours after injection was initiated. The injection was stopped after an injection period of approximately 23 hours at which point between 2 and 2.5 pore volumes of foam had been injected.

Subsequent to the completion of Test 6, samples of the sediment from both of the test boxes were again analyzed for contained moisture. Fig. 9 shows the results of these analyses.

Throughout the duration of Test 6, the color of the fine-grained heterogeneous zones varied. These zones initially had a homogenous, light tan color. As the test continued, those areas of the fine-grained zones nearest the flowing foam became darker in appearance, potentially due to the previously described wicking process. Later in the test, those portions of the fine-grained zones that initially darkened began to lighten, possibly as the contained moisture decreased. This drying of the fine-grained sediment may have been due to the movement of foam and gas used to form the foam through the test boxes and is the subject of further investigation.

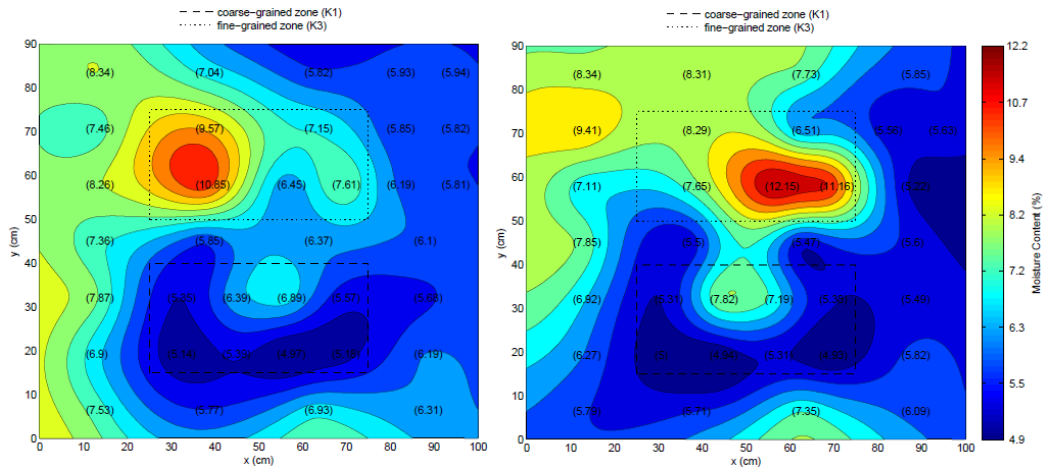


Fig. 11. Test 6 post-test moisture percentages.

TDR data was collected from the probes associated with both boxes of Test 6. The results of that data collection are shown in Fig. 12. As can be seen from that figure, the TDR probes responded to changes in the volumetric water content of the surrounding sediments. Several of these changes in water content can be attributed to the arrival times of the foam wetting front at the positions of the probes.

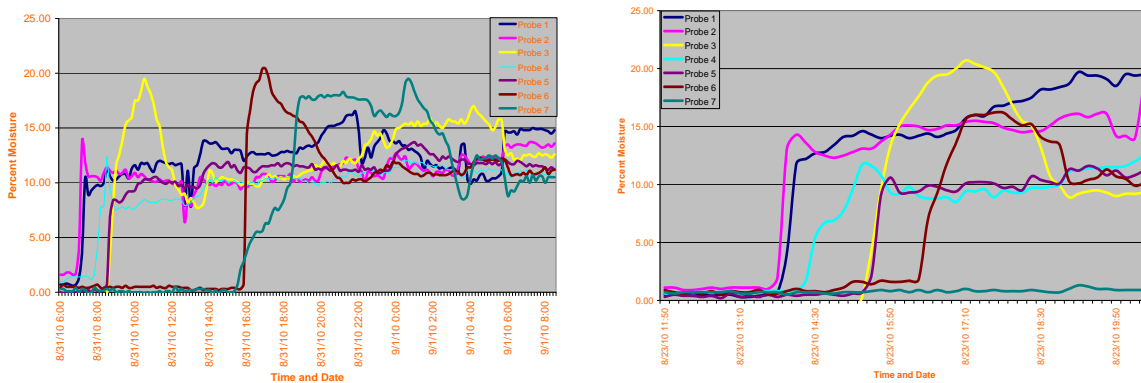


Fig. 12. Percent moisture by TDR - Test 6.

During Test 6, the pressure probes inserted into the back of both test boxes were operational. The data accumulated from those probes is shown in Fig. 13. Both of the data plots denote an increase in internal pressure in the boxes as the test proceeded.

The total and leachable concentrations of uranium in the post-test sediment samples were measured to determine the effectiveness of the remedial amendment augmented foam to immobilize in-situ the uranium contamination. The results of these analyses are shown in Figures 14 and 15 respectively.

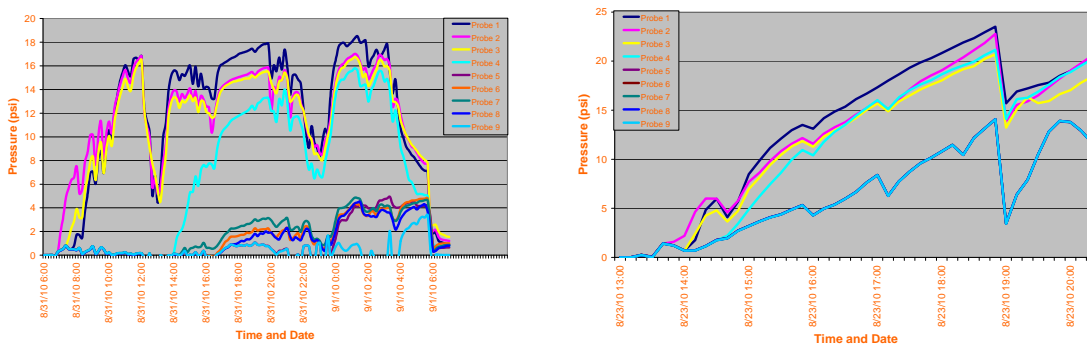


Fig. 13. Test 6 Internal pressures.

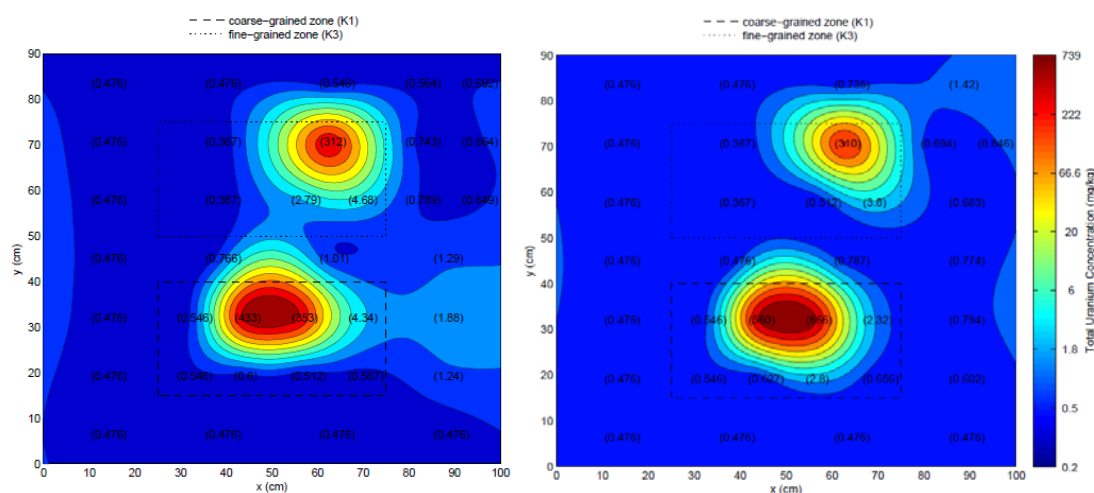


Fig. 14. Total uranium concentration.

CONCLUSIONS

The movement of foam through the sediments was similar for both of the tests. In each test, the foam wetting front produced an arcuate form that moved into the sediment until the edges of the arc intersected the edges of the sediment mass. At that time, the shape of the wetting front came to resemble an irregular vertical line. Subsequent foam movements extended the vertical line toward the right side of the box. The movement of foam bubbles could be easily observed within the coarse-grained sediments behind the wetting front; however, no bubble movement could be discerned within the fine-grained sediment. As the foam wetting front came into contact with the heterogeneous zones, the fine-grained zone was wetted by the foam in a pattern paralleling the edges of the zone. No excess wetting could be discerned in the coarse-grained zone. As the test progressed, the wetness of the fine-grained sediments decreased but remained above that of the coarse-grained sediments. As previously noted, detailed mechanisms controlling foam migration and associated sediment wetting-drying cycles are the subject of further investigation.

The addition of an extraction port near the upper-right corner of the test boxes used during the sixth test showed that the path of foam movement through the sediment can be significantly altered by the extraction well scheme.

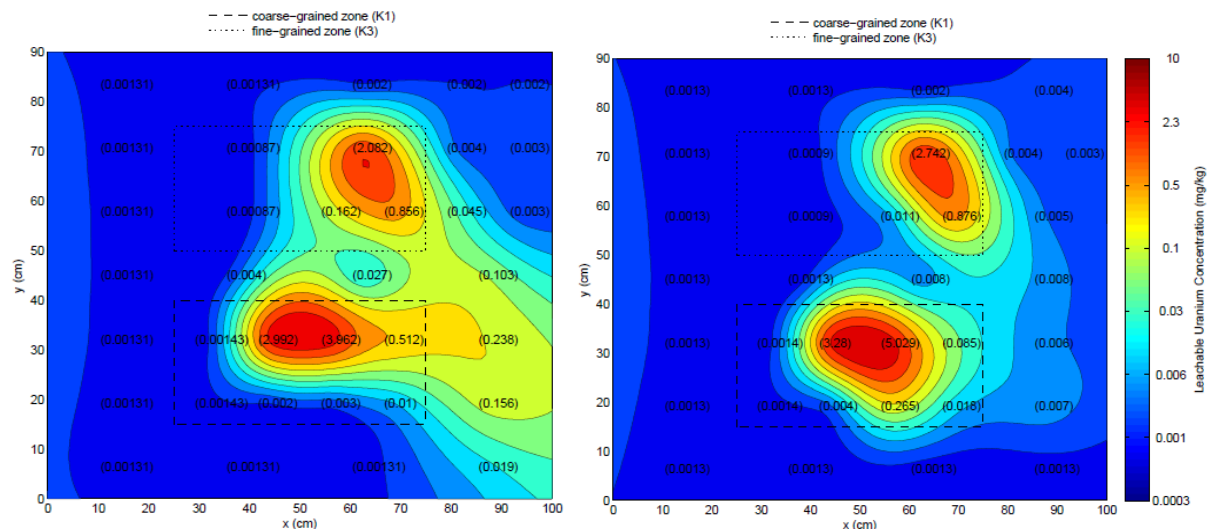


Fig. 15. Leachable uranium concentration.

The foam front in each test reached the extraction end of the respective box. Upon the foam front reaching the right side of each test box, a small amount of fluid resulting from pore water and foam breakdown was extracted from the right side of the test boxes. In the sixth test significant quantities of foam were extracted subsequent to the initial extraction of fluid

Ultimately, the experiments provided considerable useful information regarding the ability to manipulate and direct foam movement through the experimental system. In the final test multiple pore volumes of foam were injected and extracted from the box.

Data generated from the sediments analyzed subsequent to the injections showed a moderate increase in the moisture content of the sediments with the largest increase in the fine-grained sediment zones. The total and leachable concentrations of uranium in the sediment samples indicated that bulk of the uranium in each test box had remained within or very near to the emplaced zone suggesting potential in-situ immobilization. These tests have thus confirmed the feasibility of using foam carried mixtures of orthophosphate and tripolyphosphate as a remedial amendment for in-situ immobilization of uranium.

RECOMMENDATIONS

It is recommended that new foam generating systems with field applicability be developed for testing, both at bench and intermediate scale. These studies should focus on determining the efficacy of producing and injecting functional quantities of viable foam as they are required to maintain the process of injection. The intermediate-scale version of this system should be used to increase the length of time that additional intermediate-scale tests are conducted to determine the effects of the increase on the movement of uranium out of the contaminated zones (both fine- and coarse-grained) into uncontaminated sediments.

ACKNOWLEDGEMENTS

Initially this project was conducted for the DOE Environmental Management Consolidated Business Center from funding provided by the U.S. Department of Energy (DOE) Office of Environmental Management (EM) Office of Technology Innovation and Development (EM-30), Soil and Groundwater Remediation Technology Development Program (EM-32) through the Western Environmental Technology Office under DOE Contract Number DE-AC09-96EW96405. Follow-on work was conducted for Battelle Memorial Institute, Pacific Northwest Division under Contract Number 124946.