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The Cementitious Barriers Partnership Experimental Programs and Software Advancing DOE’s Waste Disposal/Tank Closure Efforts – 15436

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ABSTRACT

The U.S. Department of Energy Environmental Management (DOE-EM) Office of Tank Waste Management sponsored Cementitious Barriers Partnership (CBP) is chartered with providing the technical basis for implementing cement-based waste forms and radioactive waste containment structures for long-term disposal. DOE needs in this area include the following to support progress in final treatment and disposal of legacy waste and closure of High-Level Waste (HLW) tanks in the DOE complex:

- Long-term performance predictions and
- Flow sheet development and flow sheet enhancements
- Conceptual designs for new disposal facilities

The DOE-EM Cementitious Barriers Partnership is producing software and experimental programs resulting in new methods and data needed for end-users involved with environmental cleanup and waste disposal. Both the modeling tools and the experimental data have already benefited the DOE sites in the areas of performance assessments by increasing confidence backed up with modeling support, leaching methods, and transport properties developed for actual DOE materials. In 2014, the CBP Partnership released the CBP Software Toolbox – “Version 2.0” which provides concrete degradation models for: 1) sulfate attack, 2) carbonation, 3) chloride initiated rebar corrosion, and includes constituent leaching. These models are applicable and can be used by both DOE and the Nuclear Regulatory Commission (NRC) for service life and long-term performance evaluations and predictions of nuclear and radioactive waste containment structures across the DOE complex (Figure 1) including:

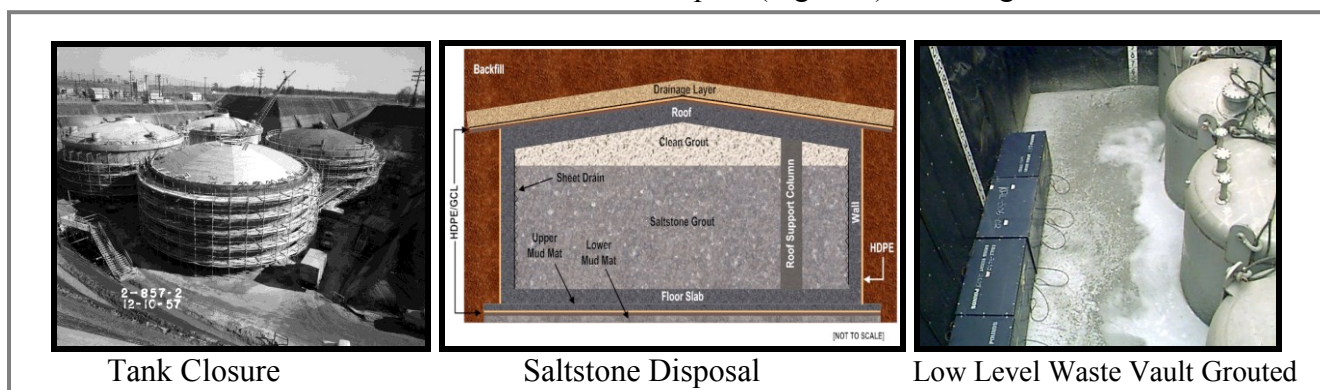


Figure 1: Various DOE Cementitious Barriers in Waste Management

- Future SRS Saltstone and HLW tank performance assessments and special analyses
- Hanford site HLW tank closure projects and other projects in which cementitious barriers are required
- The Advanced Simulation Capability for Environmental Management (ASCEM) project which requires source terms from cementitious containment structures as input to their flow simulations
- Regulatory reviews of DOE performance assessments
- Nuclear Regulatory Commission reviews of commercial nuclear power plant (NPP) structures which are part of the overall US Energy Security program to extend the service life of NPPs

In addition, the CBP experimental programs have had a significant impact on the DOE complex by providing specific data unique to DOE sodium salt wastes at Hanford and SRS which are not readily available in the literature. Two recent experimental programs on cementitious phase characterization and on technetium (Tc) mobility have provided significant conclusions as summarized below:

- Recent mineralogy characterization discussed in this paper illustrates that sodium salt waste form matrices are somewhat similar to but not the same as those found in blended cement matrices which to date have been used in long-term thermodynamic modeling and contaminant sequestration as a first approximation. Utilizing the CBP generated data in long-term performance predictions provides for a more defensible technical basis in performance evaluations.
- In addition, recent experimental studies related to technetium mobility indicate that conventional leaching protocols may not be conservative for direct disposal of Tc-containing waste forms in vadose zone environments. These results have the potential to influence the current Hanford supplemental waste treatment flow sheet and disposal conceptual design.

INTRODUCTION

The Cementitious Barriers Partnership, sponsored by DOE-EM Office of Tank Waste Management, is a collaborative partnership between Savannah River National Laboratory (SRNL), Energy Research Centre (ECN), Vanderbilt University, and SIMCO Technologies, Inc. established under a Cooperative Research and Development Agreement (CRADA). The National Institute of Standards and Technology (NIST) and the Nuclear Regulatory Commission (NRC) are also providing support to the CBP CRADA through an Interagency Agreement (IAA) and MOU, respectively. The CBP is funded through both DOE-EM Office of Tank Waste Management and the Consortium for Risk Evaluation with Stakeholder Participation (CRESP). The CBP is chartered with providing the technical basis and simulation tools for implementing cement-based waste forms and radioactive waste containment structures. Recently, in 2014, the CBP released its new Software Toolbox - Version 2.0, a software package providing new concrete degradation models that assist in lifetime predictions for cementitious structures. The

experimentally based software is used to predict degradation depths and damage due to sulfate attack and carbonation for actual DOE cementitious waste structures as shown in Figure 2.

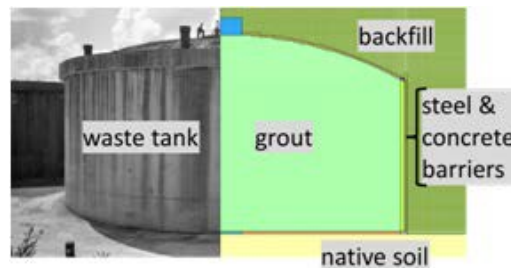


Figure 2: Cementitious Barriers – Tank Integrity and Closure Model Constructs

The CBP modeling and experimental products have already had a beneficial impact to DOE disposal and closure efforts, with some of the highlights discussed in this paper. The CBP software tools and experimental programs have already been used to support DOE Performance Assessments (PAs) such as the Saltstone Disposal Facility at the Savannah River Site and have the potential to impact the design of new facilities. The CBP also hosted workshops across the DOE-Complex in 2014 to familiarize the end-users to the existing benefits of the software and experimental programs and to hear first-hand of needed areas of uncertainty that CBP could respond to in future development work.

DISCUSSION

CBP Software Tools and Impact

The CBP Software Toolbox is a suite of software for simulating reactive transport in cementitious materials and certain degradation phenomena. The primary software components are LeachXS/ORCHESTRA (LXO), STADIUM, and a GoldSim interface for probabilistic analysis of selected degradation scenarios. The current version released in 2014, “Version 2.0”, supports analysis of external sulfate attack, carbonation degradation analysis, chloride attack and dual regime modeling for fractured and in-tact cementitious materials. The LeachXS component embodies an extensive material property measurements database with emphasis on cementitious materials used in DOE facilities, such as Saltstone (Savannah River) and Cast Stone (Hanford), tank closure grouts, and barrier concretes. The integrated software is shown in Figures 3 and 4 with the various software components and modules, respectively, which simulate various degradation and leaching mechanisms in DOE-specific cementitious materials.

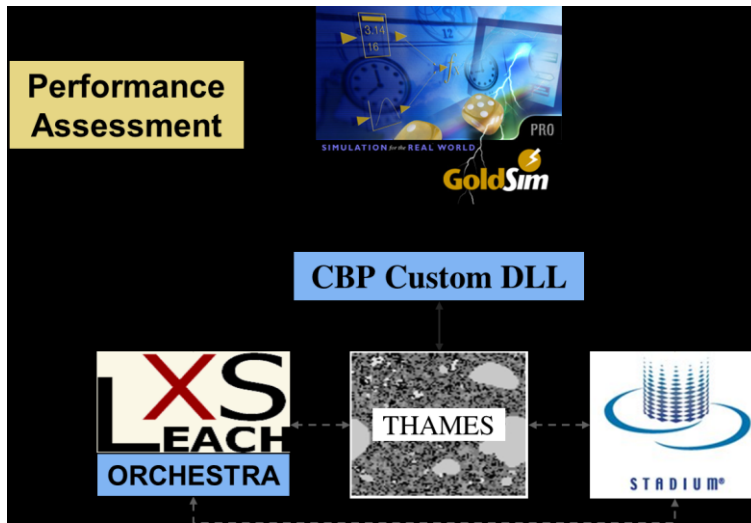


Figure 3: CBP Partner Software Components

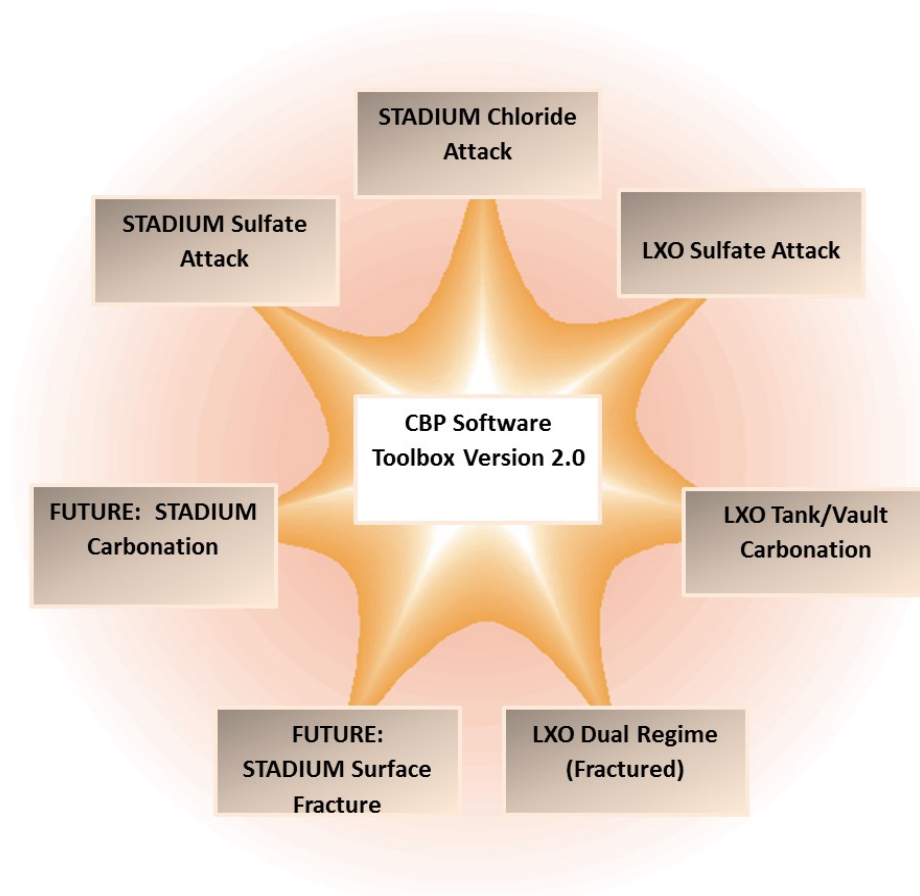


Figure 4: CBP Software Simulating Cementitious Degradation & Leaching Mechanisms

The integrated software package has also been upgraded with new plotting capabilities and many other features that increase the “user-friendliness” of the package. Experimental work has been generated to provide data to calibrate the models to improve the credibility of the analysis and reduce the uncertainty. Tools selected for and developed under this program have been used to evaluate and predict the behavior of cementitious barriers used in near-surface engineered waste disposal systems for periods of performance up to or longer than 100 years for operating facilities and longer than 1000 years for waste disposal. The CBP Software Toolbox is and will continue to produce tangible benefits to the working DOE performance assessment community.

The CBP is currently working on experimental programs and developing simulation tools to model other degradation mechanisms that include damage mechanics which estimates damage based upon sulfate ingress. Currently, the performance assessments must assume damage based upon the ingress but these tools will aid on the degree of damage which will reduce unrealistic conservatism in the PA. Other degradation mechanisms of cementitious materials that the CBP is developing include alkali silica reactions and acidic soil attack. Experimental programs are continuing in determining hydraulic properties of fractured cementitious materials, redox sensitive (i.e., Tc) oxidation rates and surface fracture studies. For all these mechanisms, experimental programs are designed and software constructed from the experimental results. Figure 5 shows the timeline of CBP development plans

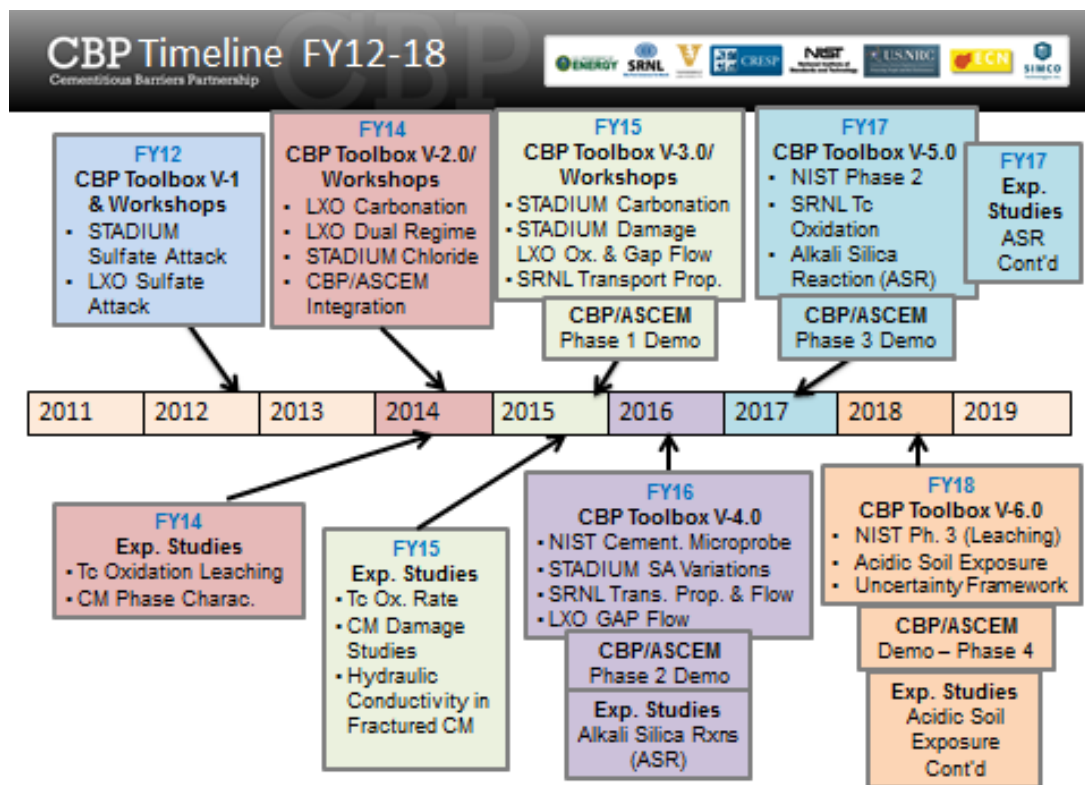


Figure 5: CBP Experimental and Modeling Integrated Timeline

CBP- Hosted Workshop To Showcase CBP Software and to Address DOE Emerging Issues

SRNL hosted a webinar on the new CBP Software Toolbox Version 2.0 at the request of DOE-Savannah River (SR) and Hanford participants in July 2014, and the CBP has hosted workshops at Hanford, NIST, and SRNL in late summer through fall in 2014. In these workshops, new capabilities of the CBP Software Toolbox were presented, including user-interface improvements, an enhanced carbonation module, and the addition of a dual regime transport module for fractured and aggregate systems. Specific applications and simulation needs of DOE-SR and other participants were discussed at the workshops so that future CBP experimental programs and software tools could be designed to address specific current needs of the DOE Tank Waste Management Programs. Software additions to solve DOE emerging issues with cementitious barriers identified in recent CBP workshops are shown in Table 1 along with the CBP plan in addressing these issues.

Table 1: DOE Emerging Issues and How the CBP is Addressing These Issues

DOE Emerging Issues <i>(Identified in 2014 CBP Workshops)</i>	How CBP is Addressing DOE Emergent Issues
Bulk Chemistry Variations & Impact to Cementitious Phases (Impact to Modeling Results)	SRNL FY14 Experimental Program provided initial work on CM phases. Future work will increase understanding.
Need Damage Model & Impact to Hydraulic Conductivity	SIMCO STADIUM Damage Module Planned Release (<i>FY15</i>)
Need CBP Software Tools on Additional Degradation Mechanisms (Oxidation, Alkali-Silica Reactions (ASR))	CBP Program on Alkali-Silica Reactions and impact to DOE cementitious barriers (<i>FY16</i>)
Need DOE Compliant QA Documentation of CBP Software	CBP Program to Develop QA Guidelines that comply with DOE Sites (<i>FY15</i>)
Need Realistic Predictions of Damaged CM Properties	SIMCO STADIUM Damage Module Release/ NIST THAMES Software to simulate time-evolution of CM Prop. (<i>FY15-17</i>)
Need Protection of Tc in Waste Form from Oxidation	SRNL FY15 Experimental Program Method development to measure oxidation rate
Need CBP Sulfate Attack Software to simulate High Sulfate Conc. in Waste.	SIMCO STADIUM Software Demo. Case with 50,000 ppm sulfate concentration (<i>FY15</i>)
Is Liner Needed in New SRS Disposal Units for Cost Savings?	SIMCO STADIUM Software Demo. Case with and without liner (<i>FY15</i>)
CBP Software needs to reflect various material layers of disposal units	SIMCO STADIUM Software Demo. Case to include multiple sub-layers of concrete (<i>FY15</i>)
Need to simulate less saturated conditions at Hanford soils vs 100% saturated	SIMCO STADIUM Software Demo. Case to conduct simulations w/ varying saturations
Need Time-Evolution of Cementitious Materials (CM) and Transport Properties	NIST THAMES Software Designed to simulate Changes in CM/ Properties

In response to DOE needs that were brought up in the meetings, the CBP partners are planning enhancements to the existing software modules to provide continued support for DOE site operational and post-closure scenarios. Examples of some of these enhancements include equipping the STADIUM external sulfate attack modules to include composite barriers and various degrees of saturation based on exposure conditions that vary between arid and tropical environments. The CBP will be developing the future program to include the near- and long-term development work to support these user needs listed in Table 1.

Degradation Studies of DOE Cementitious Barriers Associated with Salt-Based Waste Forms

The saltstone facilities at the DOE Savannah River Site (SRS) stabilize and dispose of low-level radioactive salt solution originating from liquid waste storage tanks at the site. The Saltstone Production Facility (SPF) receives treated salt solution and mixes the aqueous waste with dry cement, blast furnace slag, and fly ash to form a grout slurry which is mechanically pumped into concrete disposal cells that compose the Saltstone Disposal Facility (SDF). The solidified grout is termed “saltstone”.

Cementitious materials play a prominent role in the design and long-term performance of the SDF. The saltstone grout exhibits low permeability and diffusivity, and thus represents a physical barrier to waste release. The waste form is also reducing, which creates a chemical barrier to waste release for certain key radionuclides, notably Tc-99. Similarly, the concrete shell of a saltstone disposal unit (SDU) represents an additional physical and chemical barrier to radionuclide release to the environment. Together the waste form and the SDU concrete wall compose a robust containment structure at the time of facility closure. However, the physical and chemical state of cementitious materials will evolve over time through a variety of phenomena, leading to degraded barrier performance over Performance Assessment (PA) timescales of thousands to tens of thousands of years. Previous studies of cementitious material degradation in the context of low-level waste disposal have identified sulfate attack, carbonation influenced steel corrosion, and decalcification (primary constituent leaching) as the primary chemical degradation phenomena of most relevance to SRS exposure conditions.

In this study, degradation time scales for each of these three degradation phenomena were estimated for saltstone and concrete associated with each SDU type. The combined effects of multiple phenomena were then considered to determine the most limiting degradation time scale for each cementitious material. Degradation times were estimated using a combination of analytic solutions from literature and numerical simulation codes provided through the CBP Software Toolbox.

The modeling study found that degradation of these concrete barriers generally occurs from combined sulfate attack and corrosion of embedded steel following carbonation. Saltstone is projected to degrade very slowly by decalcification, with complete degradation occurring in excess of 200,000 years for any SDU type.

CBP/ASCEM Integration

An initial ASCEM / CBP demonstration is planned that will focus on a representative high-level waste tank closure scenario relevant to the DOE-EM complex. CBP modeling tools will be used to model the source term and near-field conditions and releases, and ASCEM software will be used to model far-field conditions to a downstream point of interest. The purpose of the joint analysis is to demonstrate specialized CBP capabilities for simulating reactive transport in cementitious materials, and ASCEM reactive transport capabilities (cement/waste form leachate impacted far-field soil), and guide ASCEM and CBP development of interfaces to external codes and information.

As illustrated in Figure 6, closure of a representative liquid waste tank includes grouting the residual waste and the empty spaces in the tank. The CBP near-field model domain will encompass the reducing cementitious grout in the tank, the steel tank liner, the concrete tank wall or shell, and a sufficient amount of adjoining soil (approximately 1 m thickness) to adequately capture significant leachate interactions between the concrete wall and soil.

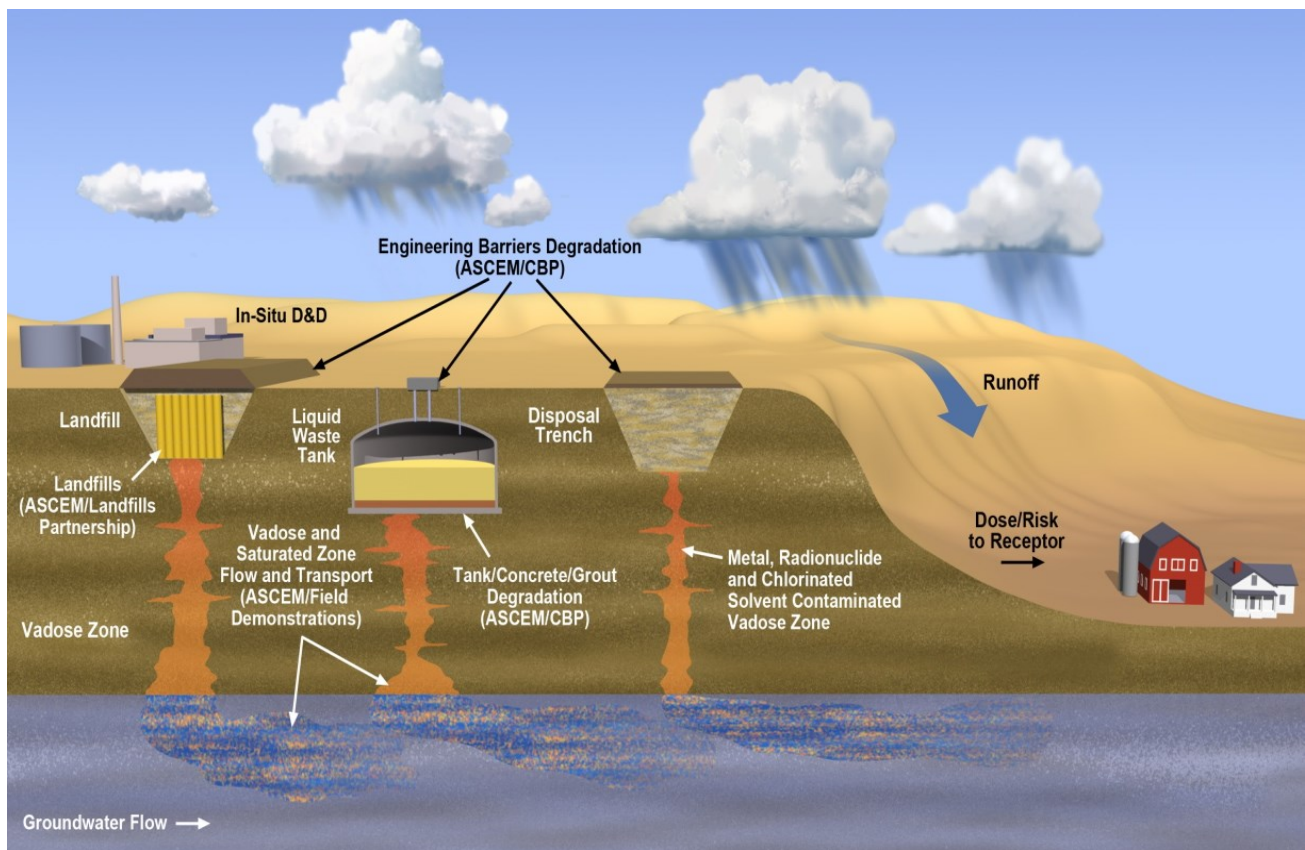


Figure 6: CBP / ASCEM Integrated Software Approaches

CBP Experimental Programs and Impact

The CBP provides great value to the DOE complex through the development of the experimentally-based simulation tools that model pertinent degradation mechanisms much needed for technical support of the DOE facility PAs. The experimental programs, in addition to providing data for calibration and validation of the CBP software tools, provide support directly to future DOE facility flowsheets and design. For example, the CBP has recently issued the results of experimental studies related to Tc mobility suggesting that conventional leaching protocols may not be conservative for direct disposal of Tc containing waste forms in vadose zone environments. Applying understanding through experimental efforts to simulation tools and facility design is a necessary need in the DOE complex.

Tc Oxidation and Mobility Studies in DOE Salt Waste Forms

SRNL completed an experimental program on oxidation of DOE cementitious materials, specifically, slag-based salt waste forms that are representative of both SRS and future Hanford waste. Both the SRS and Hanford waste streams contain soluble redox sensitive technetium (Tc) which requires stabilization to meet disposal requirements and poses great challenges to performance predictions of cementitious barriers used in stabilizing Tc due its high solubility and mobility. SRNL developed a new method known as depth-discrete sampling and leaching method that provides a way to estimate the contaminant oxidation rate, and in turn, provides much needed data for modeling Tc releases to the environment.

In the oxidation studies, slag-based salt waste form samples were exposed to both Hanford sediments and deionized (DI) water. Soluble Tc was leached from all of the depth-discrete subsamples which strongly suggests that oxygen was present in the entire length of both samples. The results also reveal that the oxidation rate of the sample exposed to soil was faster than that of the sample exposed to water. This conclusion is consistent with the more rapid transport of ions through a gas phase as compared to a liquid phase. This conclusion has the potential to challenge current leaching protocols of redox sensitive contaminants in water.

The SRNL technetium oxidation and mobility studies may have an impact on performance predictions and challenge the current conceptual models for release at the DOE sites. During a series of grout working group meetings, SRNL subject matter experts discussed the experimental data and current conceptual model, lines of inquiry for diagnosing the observed discrepancies, and alternative conceptual models. Additional data are required to fully understand and quantify the rate of oxygen ingress and oxidation of Tc. However, these scoping studies have provided insights to the multiple mechanisms affecting the solubility and leachability of Tc and other redox sensitive contaminants.

Characterization of Mineral Phases in Salt-Based Waste Forms

In this CBP SRNL experimental program, x-ray diffraction results for a series of DOE sodium salt waste forms and cementitious binders were measured. Characterization of the matrix mineralogy is important for predicting contaminant leaching and evolution of the materials as a function of time and changing conditions. The objective of this study was to provide initial phase characterization for the cementitious salt waste form. This information can be used to: 1) generate a base line for the evolution of the waste form as a function of time and conditions, 2) design new binders based on mineralogy of the binder, 3) understand and predict anion and cation leaching behavior of contaminants of concern, and 4) predict performance of the waste forms for which phase solubility and thermodynamic data are available. Characterization of the mineralogy is also important for understanding the buffering effects that the waste form has on infiltrating water / leachates.

Hydraulic Conductivity Method Development in Fractured Cementitious Materials

Another SRNL CBP experimental program included the method development of measuring important transport properties in fractured cementitious materials. Specifically, the scope was to develop a method to measure one of the most important thermodynamic properties in modeling, the hydraulic conductivity of fractured material. DOE Performance Assessments often involve cementitious barriers and/or waste forms that are predicted or assumed to degrade over time due to various mechanisms such as carbonation-influenced reinforcing steel corrosion, external sulfate attack, differential settlement, and seismic activity. Physical degradation typically takes the form of small-scale cracking / fracturing, and the affected materials reside in unsaturated hydrogeologic zones. In these cases, unsaturated hydraulic properties are needed for fractured cementitious materials to simulate moisture movement and contaminant transport within and around the facility. The outflow extraction method has been developed to provide a suitable method for estimating these material properties.

Exposure of DOE Barriers to Aggressive Solutions Experimental Study

The SIMCO Technologies, Inc. (SIMCO) experimental work aimed at supporting the assessment of long-term durability of concrete barriers containing sulfate-bearing wasteform, present at both SRS and Hanford. The high sulfate content of the wasteform pore solution is the main cause of concern for the durability of concrete barriers. The purpose of the study was to improve understanding of the complex concrete/wasteform reactive transport problem, in particular the role of pH in sulfate attack. The samples were immersed in sulfate contact solutions and analyzed to measure the impact of the aggressive environment on the material.

This experimental program characterization study was performed on three hardened ASTM Type I cement pastes mixtures exposed to 1) a neutral-pH and 2) a basic-pH solutions, both containing a high level of sulfate. After three months of exposure, various techniques were used to quantify the penetration of sulfate in the paste samples and the damage sustained as a result of sulfate exposure:

- Layer-by-layer analysis of sulfate content through acid dissolution,
- Microprobe analysis,
- Mercury intrusion porosimetry (MIP),
- X-ray diffraction (XRD).

The data collected indicated that in the sulfate (Na_2SO_4) solution, damage occurs to the pastes. Sulfate profiles, either from layer-by-layer acid dissolution analysis or by microprobe, confirm the penetration of sulfate in the material. Limited XRD data show that in the damaged portion next to the surface, ettringite and gypsum was formed. Alterations to the microstructure were confirmed by MIP measurements. Close to the surface, where the paste is most damaged, some of the finer pores were filled, as indicated by a reduction of the pore volume in the 10nm-100nm pore range. However, for pores in the 20nm–2 μm pore range, pore volume increased. This newly created volume can be associated with microcracks, likely created by the formation of ettringite and gypsum. These observations are valid for all three paste mixtures. The rate of sulfate ingress and degradation was directly related to the mix characteristics: higher water-to-cement ratio showed higher rates of degradation.

In the case of the high pH sulfate solution ($\text{Na}_2\text{SO}_4 + \text{NaOH}$), no signs of damage was observed on any of the paste mixtures. Contrary to the previous case, the deleterious mineral phases associated with sulfate exposure did not form in the high pH environment. A possible explanation for this is the absence of gypsum formation at high pH.

Although these results need further confirmation, they indicate that the high sulfate content found in the wasteform pore solution will not necessarily lead to severe damage to concrete typical of that used in SRS Saltstone Disposal Units 1/4. Good quality mixtures could thus prove durable over the long term and act as an effective barrier to prevent radionuclides from reaching the environment. Additional experiments with contact solutions that mimic more closely wasteform pore solution and the impact to the new SDU units concrete are needed in future studies.

Transport Properties of Damaged DOE Cementitious Materials

The objective of the CBP project is to develop a set of tools to improve understanding and prediction of the long-term structural, hydraulic, and chemical performance of cementitious barriers used in low level waste storage applications. One key concern for the long-term durability of concrete is the degradation of the cementitious matrix, which occurs as a result of aggressive chemical species entering the material or leaching out in the environment, depending on the exposure conditions. As degradation of the matrix and alteration of the microstructure progresses, properties of the materials like porosity, diffusivity and permeability are affected, which in turn affect degradation rate. The prediction of modifications to the different properties as a function of damage level sustained by the material is needed to properly model the long-term performance of engineered barriers.

The objective of this SIMCO experimental study was to provide data to relate damage level in cementitious materials to changes in transport properties, which can eventually be used to support model development. In order to get results within a reasonable timeframe and to induce as much as possible uniform damage level in materials, concrete samples were exposed to freezing and thawing (F/T) cycles. The methodology consisted in exposing samples to F/T cycles and monitoring damage level with ultrasonic pulse velocity measurements. Upon reaching pre-selected damage levels, samples were tested to evaluate changes in transport properties. The data collected in this study can later be used and compared to similar data collected on samples damaged by extended exposure to aggressive solutions in order to establish a relationship between the alteration sustained by hydrated paste and relevant transport properties.

The methodology consisted in damaging samples using freezing and thawing cycles and monitoring the damage level on the basis of ultrasonic pulse velocity measurements. Upon reaching selected damage levels, samples were tested to evaluate their porosity and tortuosity. Experimental results showed that diffusion coefficient is more sensitive than porosity to internal damage. Mobility of an ionic species is not only related to pore volume, but also to pore connectivity and tortuosity.

These results have a significant impact on modeling efforts. Models relating porosity to tortuosity and permeability are unlikely to provide the correct basis for predicting long-term durability of concrete sustaining internal pressures and microcrack formation. Other avenues like the modeling of internal crystallization pressure need to be explored.

CONCLUSIONS

The CBP partnership provides competence, continuity, and credibility to the DOE need to use engineered barriers in near surface disposal facilities and to advance progress in final treatment and disposal of legacy waste and closure of HLW tanks in the DOE complex. Therefore, the CBP will continue to focus efforts in:

- Oxidation of redox sensitive contaminants, specifically, technetium (Note: Tc is of significant concern because it is long-lived and highly mobile in the environment.)
- Alkali silica reaction degradation predictions for DOE cementitious containment structures
- Alkali silica module applied to Nuclear Power Plant concrete (required for service life prediction)
- Validation of all models and higher levels of quality assurance
- Multiple mechanism model (i.e., synergistic impact of dual and triple mechanisms impacting the degradation of structures)
- Analysis (experiments/modeling) of additional mechanisms that impact DOE cementitious containment(e.g., acidic soil exposure)
- Analysis (experiments/modeling) of the change in microstructure and transport properties as the cementitious material degrades

The CBP Software Toolbox has been used to provide important technical insights to the DOE PA process regarding sulfate attack on the DOE Saltstone Disposal Facility and carbonation on a typical high-level waste tank. Current development efforts in the areas of carbonation, transport in fractured and intact media, and chloride attack have resulted in the second release of the Toolbox. Recognizing that physical damage to cementitious materials typically occurs in the form of cracking, ongoing CBP development efforts are also focused on predicting damage through fracture mechanics considerations, determining the hydraulic and transport properties of fractured materials, and implementing corresponding Toolbox simulation capabilities.

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