

Application of the New US EPA Leaching Environmental Assessment Framework (LEAF) to DOE Environmental Management Challenges – 14383

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ABSTRACT

An integrated Leaching Environmental Assessment Framework (LEAF) is being developed and implemented in the United States. LEAF provides a basis for evaluating remediation, beneficial use and disposal options for a wide range of materials including residuals from energy production and industrial processes, contaminated soils and low activity wasteforms. LEAF evaluates leaching behavior of materials using a tiered approach that considers pH, liquid-to-solid ratio (L/S), and waste forms across a range of field conditions. Implementation of LEAF includes development and validation of leaching test methods, data management tools, and scenario-based assessment in the context of prior information and using percolation and diffusion mass transfer models. The LEAF leaching test methods have undergone required inter-laboratory precision and repeatability testing and are now included in SW-846, a compendium of EPA methods to evaluate the physical and chemical properties of wastes and secondary materials.

INTRODUCTION

Leaching, the release of constituents to contacting water, is a primary pathway for potential impact to human health and the environment during solid and hazardous waste disposal and beneficial use secondary materials. Leaching test methods and assessment procedures are necessary to determine whether or not specific disposal or use options for a particular material will be sufficiently protective to avoid adverse impacts through constituent release to soils, surface water or groundwater. In the United States, beneficial use of secondary materials is regulated by individual states without national regulatory requirements. In contrast, waste management (i.e., disposal) is regulated at the national level through the Resource Conservation and Recovery Act (RCRA). Under RCRA, leachability is evaluated based on a plausible mismanagement scenario, defined as co-disposal with municipal solid waste. As a result, the primary leaching test generally used in the United States is the Toxicity Characteristic Leaching Procedure (TCLP), which was designed to simulate waste co-disposal with municipal solid waste. However, field leaching conditions for many waste disposal practices and contaminated sites requiring evaluation, and almost all beneficial uses, have limited or no relationship to the leaching conditions in a municipal waste landfill. Cleanup at former defense nuclear facilities is regulated through multiple federal and state authorities, including CERCLA, RCRA and the Atomic Energy Act. For many cases relevant to waste management and cleanup at former defense nuclear facilities, the relevant waste form is monolithic, and therefore the monolithic leaching procedure ANS 16.1 has been used to assess leachability based on criteria for a resulting leaching index. However, the leaching index approach also assumes constant diffusivity of constituents of potential concern (COPCs), including radionuclides, and without regard to the field liquid contacting conditions.

The development of an alternative leaching characterization and assessment approach enables improved decision making for cleanup, beneficial use and disposal decisions for a wide range of materials and management scenarios. Given the range of potential cleanup, beneficial use and

disposal scenarios, considering both the nature of the materials along with the geographic and engineering contexts in which the materials may be managed, it is impractical to have individual tests that are designed to simulate all the likely scenarios. Kosson et al., [1] proposed an alternative approach to leaching assessment that is based on laboratory measurement of intrinsic leaching characteristics of materials using a limited set of leaching tests and using the testing results in conjunction with scenario-based mass transfer models to estimate potential leaching under a range of management scenarios. This approach, referred to as the leaching environmental assessment framework (LEAF), is being further developed under the direction of USEPA as a basis for more robust leaching assessment for a range of possible applications, including evaluation of beneficial uses of secondary materials, waste treatment and disposal practices, soil remediation, and life-cycle assessments. This effort is also being carried out in coordination with closely related initiatives within the European Union. Currently, LEAF is being used by USEPA to evaluate potential leaching from coal combustion residuals (e.g., fly ash, scrubber residues) resulting from wider use of multi-pollutant controls at U.S. coal-fired power plants and for use in concrete [2,3,4,5,6], by DOE to evaluate effectiveness of low activity stabilized wasteforms [7].

LEACHING ENVIRONMENTAL ASSESSMENT FRAMEWORK (LEAF)

LEAF is an integrated framework for evaluating leaching behavior of materials using a tiered approach that considers pH, liquid-to-solid ratio (L/S), and waste form across a range of field conditions [1]. Implementation of LEAF includes leaching test methods, data management, assessment in the context of prior information and using scenario-based mass transfer models [8,9,10,11,12,13], and statistical quality control [14]. The LEAF leaching test methods that have been adapted for inclusion into SW-846 are:

- Method 1313 - liquid-solid partitioning as a function of eluate pH using a parallel batch extraction test,
- Method 1314 - liquid-solid partitioning as a function of liquid-solid ratio using a parallel batch test,
- Method 1315 - liquid-solid partitioning as a function of liquid-solid ratio using an up-flow column test, and
- Method 1316 - mass transfer in monolithic or compacted granular materials using a semi-dynamic tank leach test.

The LEAF test methods are currently available at the EPA SW-846 website.¹ Microsoft Excel[®] templates are available for use along with the test methods to facilitate leaching test method calculations and data entry (www.vanderbilt.edu/leaching). Leach XS™ and LeachXS Lite™ (see below) also available for data viewing and analysis.

Method 1313 and Method 1316 are parallel batch procedures intended to characterize the liquid-solid partitioning at conditions approaching equilibrium as a function of final extract pH and liquid-to-solid ratio (L/S), respectively. The test parameters and values specified in these methods have been described in a background information document on the LEAF leaching methods [15].

Method 1313 is an equilibrium-based leaching test provides aqueous extracts representing the liquid-solid partitioning (LSP) curve of constituents as a function of eluate pH. This procedure consists of nine parallel batch extractions at targeted pH values and one extraction at the

¹ http://epa.gov/wastes/hazard/testmethods/sw846/new_meth.htm

natural pH² of the material. The solid material may require particle-size reduction by crushing in order to facilitate the approach to solid-liquid equilibrium within a reasonable extraction timeframe. Dilute acid or base in deionized water is added to each extraction according to a pre-test titration in order to achieve final extract pH values at specified target values ranging between 2 and 13 at an L/S of 10 mL/g-dry. The extraction contact time ranges from 24 to 72 hours based on the grain size of the “as tested” material (i.e., the material after any particle size reduction or air drying required to improve the handling of the “as received” material). The pH and conductivity of the final extract solution are recorded and vacuum- or pressure-assisted filtration is used to separate the liquid and solid phases prior to chemical analysis of the eluate. Eluate concentrations for constituents of interest are plotted as a function of eluate pH allowing for comparison to quality control and assessment limits. Eluate concentrations also may be interpolated to the target pH points to provide a uniform basis for comparison of results as the recorded eluate pH is likely to differ slightly from target values within specified pH tolerances.

Method 1316 is an equilibrium-based leaching test intended to provide eluates over a range of L/S values from 10 to 0.5 mL/g-dry using five parallel batch extractions in DI water. No acid or base is added to the extractions such that the results can indicate changes in eluate pH with L/S. As in Method 1313, particle size reduction of the solid material may be required in order to facilitate the approach to solid-liquid equilibrium. The contact time for the extractions ranges from 24 to 72 hours based on grain size in a similar manner as in Method 1313. The pH and conductivity of the final extract solution are recorded. Solid and liquid phases are separated by vacuum- or pressure-assisted filtration and prepared for chemical analysis. This method provides data on the changes in equilibrium chemistry (i.e., ionic strength, constituent concentrations) as the L/S value approaches that found within the solid phase pore solution.

Method 1314 is a percolation column test designed to evaluate the release of constituents from solid materials as a function of cumulative liquid-to-solid ratio (L/S). During Method 1314, fractions of a continuous elution of water through a packed bed of granular material are collected at nine specified L/S values. The concentrations of constituents in the collected eluates³ are used to derive the cumulative mass release from the column. Both eluate concentration and cumulative release are reported as a function of L/S and may be used as test outputs for assessment purposes.

Method 1315 is a semi-dynamic tank leaching procedure used to determine the rate of mass transport from monolithic materials (e.g., concrete materials, bricks, tiles) and compacted granular materials (e.g., soils, sediments, fly ash) as a function of time. Test samples are leached in a series of nine deionized water tanks for specified interval durations. Although the direct method result is eluate concentrations, the test outputs for Method 1315 include the mean interval flux during each test interval and the cumulative mass released through the end of a leaching interval as derived from the eluate concentration and other test information.

Fig. 1 provides an overview of the types of data generated and applications in assessment for use and disposal scenarios. Leaching test eluate concentrations provide a basis for comparison of materials and initial screening of potential risk, while use of leaching test results to

² The natural pH (also referred to as “own pH”) is the final eluate pH response of a deionized water extraction of a solid material (i.e., no acid or base added) conducted at an L/S 10 mL/g-dry.

³ This paper follows the LEAF terminology where “eluate” is used to refer to the solution resulting from a leaching test and “leachate” is used to refer to the solution collected from or measured in the field. The distinction is made within LEAF to minimize confusion when comparing laboratory leaching test data to field leaching data.

parameterize scenario-based mass transfer models that include appropriate consideration of leaching chemistry, water flux and attenuation processes provides a source term for detailed impact assessment. The primary mass transfer models used for scenario assessment are (i) diffusion-controlled coupled release assuming local equilibrium between solid and porewater within monolithic materials, and (ii) percolation considering a two-regime system (e.g., dual porosity or percolation coupled with radial diffusion from aggregate or monolithic materials) and also assuming local equilibrium within each region and location in the material through which water is percolating.

Results from a detailed impact assessment can be used to back-calculate threshold values from leaching tests to use for subsequent decisions and on-going quality control as secondary materials are produced for beneficial use applications. The minimum dilution and attenuation factor (DAF) can be calculated that is necessary to reduce the maximum leaching concentration of all constituents of potential concern to less than the drinking water standard or other performance criteria at the point of compliance between the anticipated field pH domain for the management scenario (e.g., pH 5.4-12.4). These results suggest an approach to defining the performance requirements for management scenarios. For example, a management scenario for a specific material (e.g., one of the wastefoms or contaminated soils tested) may need to result in a DAF of greater than X (e.g., 600) to reduce anticipated source term leaching concentrations of a constituent (e.g., chromium) to less than the maximum contaminant level (MCL; commonly referred to as the drinking water standard).

Clearly, there is a considerable range in the leachable concentrations of constituents from materials of different types and from different sources, as well as a considerable range of DAFs resulting from different potential management scenarios and locations. This suggests a need for developing DAF ranges for specific scenarios and evaluation in combination with material specific leaching test results. The effective dilution and attenuation achieved by the specific scenario can be further apportioned to the design of the engineered system (e.g., material compaction, blending with other materials, hydraulic controls) and the natural system (e.g., annual infiltration, attenuation during transport in the vadose zone or groundwater). Conservative regional values for DAFs also can be based on statistical (Monte Carlo) evaluation. Overall, a robust, flexible and practical evaluation system that distinguishes between environmentally acceptable and unacceptable management for specific materials is needed to facilitate safe beneficial use and ensure protection of water resources.

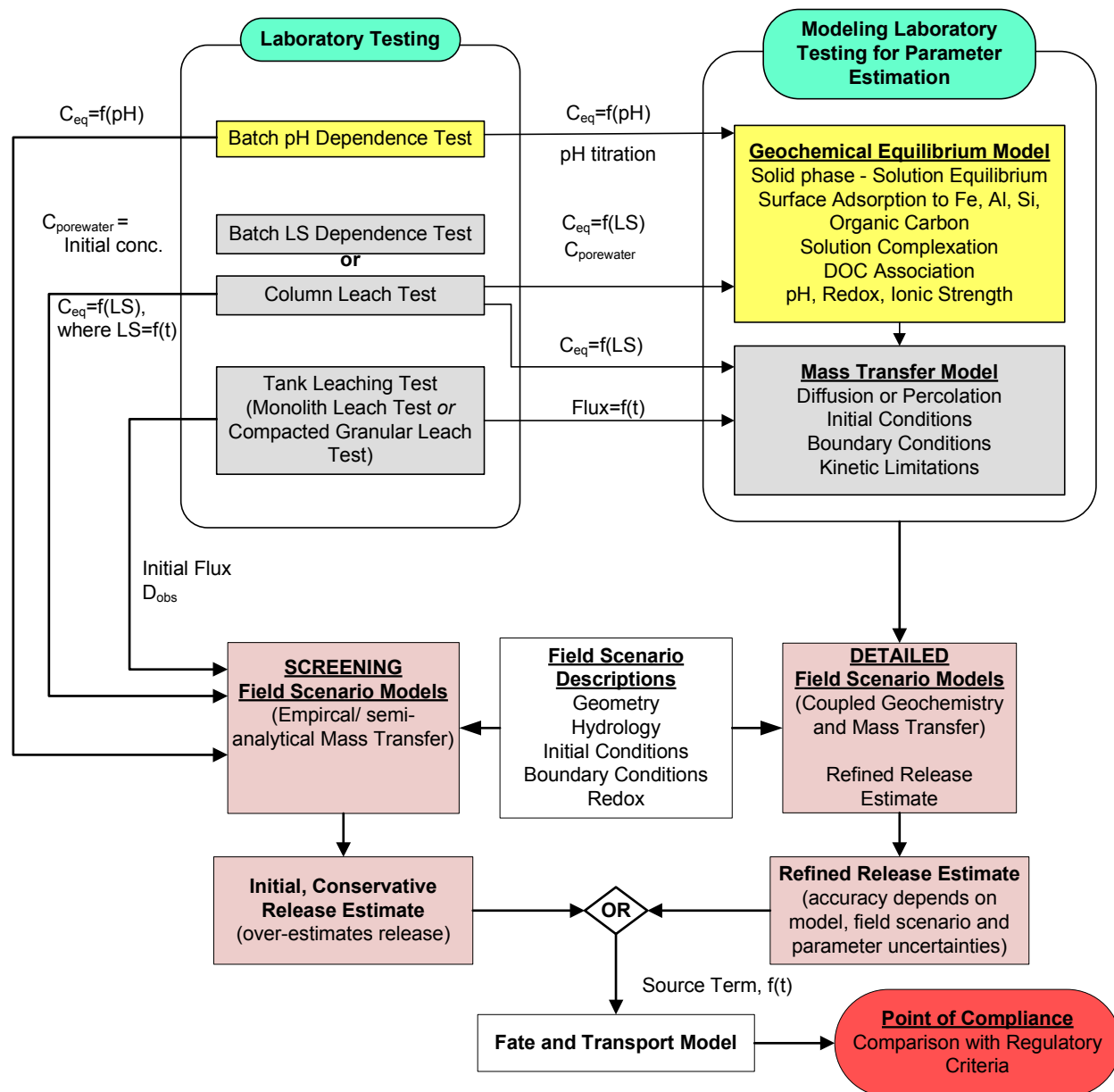


Fig. 1. Information Flow for Using Laboratory Leaching Test Results for Assessing Use and Disposal Scenarios Based on either Empirical Results or Geochemical Speciation with Reactive Transport Modeling.

INTER-LABORATORY TEST METHODS VALIDATION

An inter-laboratory round-robin testing program to validate Methods 1313, 1314, 1315 and 1316 was completed in 2012 [16,17], and the methods completed review and were posted to EPA SW-846 Methods web site in 2013.

Both Methods 1313 and 1316 involve preparation of fine-grained subsamples of test materials by sieving and crushing (as appropriate to yield subsamples with 85 wt% less than specified grain size), parallel batch extractions of the material using common laboratory equipment, and

filtration to prepare eluate solutions for chemical analysis. Due to the procedural similarities of these parallel batch extraction methods, validation of both Method 1313 and Method 1316 was carried out in parallel. The validation study followed the two-phase approach of the EPA *Guidance for Methods Development and Methods Validation for the RCRA Program* [18]. Three study materials – a coal combustion fly ash, a solidified/stabilized waste analog, and a contaminated field soil – were chosen to challenge the methods to a wide variety of waste types. In Phase I, participating laboratories were solicited to demonstrate proficiency in performing each method using a single study material. Those laboratories passing with demonstrated proficiency were asked to perform each method for two additional study materials. Statistical analysis to provide performance information was completed for at least 10 constituents on each of the three study materials. For each study material, each participating laboratory submitted three test replicates for chemical analysis while one laboratory, with sufficient experience to serve as a reference laboratory, provided six test replicates. All chemical analyses were conducted at a single laboratory in order to minimize variability associated with differing analytical methods and quality control criteria.

For Method 1313 and Method 1316, the results for each material were processed using \log_{10} transformation and linear interpolation/extrapolation of measured extract concentrations to yield a set of data used in statistical analysis. The reference laboratory data was used to develop intervals for 95% prediction limits about the mean which were compared to 95% robust confidence limits about the median of all the means from each laboratory. Variance components were calculated using a one-way analysis of variance (ANOVA) in order to determine within-lab, between-lab and overall standard deviations. Repeatability and reproducibility statistics were derived from the within-lab and overall standard deviation values, respectively. The results showed that median Method 1313 repeatability for eight common analytes in three materials ranged from 5 to 20% while reproducibility was slightly higher at 20 to 50%. For the same analytes and materials, Method 1316 repeatability was between 6 and 15% while reproducibility was found to be 8 to 30%. These results are as good as or better than precision results for European single-batch leaching tests.

For the percolation column test, Method 1314, the study materials included contaminated smelter site soil and brass foundry sand. For validation of the Method 1315 mass transfer test, both monolithic and granular materials were tested through use of a cementitious solidified waste analog and a contaminated smelter site soil. The number of laboratories participating in the Method 1314 validation was limited to seven due to the costs associated with providing columns and pumps to the participating laboratories, ensuring consistent equipment for all participating laboratories. For Method 1315, ten laboratories were asked to participate in the validation study. All ten laboratories provided analytical samples for the first study material (material code SWA); however, only eight laboratories provided analytical samples for the second study materials due to a combination of time and support commitments. The concentrations of ten selected analytes from each test and study material were analyzed by inductively coupled plasma-optical emission spectroscopy and the results were used to determine method precision based on a statistical approach adapted to this validation study.

The mean precision for Method 1314 cumulative mass release for ten analytes in each of two study materials was 6% repeatability, expressed as a repeatability relative standard deviation (RSD_r) and 16% reproducibility, expressed as reproducibility relative standard deviation (RSD_R). Method precision for Method 1314 eluate concentrations collected directly from the participating laboratories was approximately twice that of the method precision for cumulative release at 12% repeatability and 24% reproducibility. For Method 1315, method precision was based on mean interval flux and cumulative release rather than eluate concentrations since these eluate

concentrations are not recommended under LEAF for use in environmental assessment. The Method 1315 precision for mean interval flux was 12% repeatability and 30% reproducibility while the precision for cumulative release was 8% repeatability and 21% reproducibility. For both Method 1314 and Method 1315, the distribution of reproducibility estimates for individual analytes was considerably better than the reproducibility distribution reported for the current EPA regulatory leaching method, the Toxicity Characteristic Leaching Procedure (SW-846 Method 1311).

TESTING AND DATA MANAGEMENT, EVALUATION AND REPORTING

LeachXS Lite is a LEAF tool that allows the user to evaluate and characterize the leaching of constituents in materials under various conditions based on comparisons derived from leaching test results. LeachXS Lite is a simplified version of full software package LeachXS™ [19], which facilitates data comparison with field results and a range of scenarios based using either empirical relationships or mass transfer and geochemical speciation models to provide a source term for constituent leaching. Licenses for LeachXS Lite are available free of charge and LeachXS Lite can be downloaded from www.Vanderbilt.edu/leaching; user registration is required. Development of LeachXS Lite is on-going with additional functionality and data intended to be added with subsequent versions over the next several months. Updated versions will also be available for downloading from the above cited website. LeachXS and LeachXS Lite have been developed jointly by The Energy Research Centre of The Netherlands (Petten, The Netherlands), Vanderbilt University (Nashville, TN, USA) and DHI (Hørsholm, Denmark).

LeachXS Lite provides facilitated data management and leaching data comparison:

- Direct import of leaching data from Excel data templates (Methods 1313-1316),
- Comparison of leaching from different materials or leaching tests, allowing inferences about leaching mechanisms and material characteristics and behavior under different conditions,
- Comparison of leaching to reference values (e.g., method detection limits, applicable pH domain, water quality indicators; either preloaded or user defined) for individual constituents,
- Comparison of leaching from a specific material to statistical representation of a class or sub-class of materials, and
- Uniform data presentation and graphic output to Excel spreadsheets.

LeachXS Lite can be used for data management and analysis for results from leaching tests carried out on a wide range of materials and waste types (e.g., secondary or recycled materials, stabilized waste and construction materials). LeachXS Lite currently is supplied with a database of leaching test results generated under the USEPA program for characterization of coal combustion residues discussed earlier. Leaching test results included in the published reports was for more than 70 coal combustion residue samples but was limited to 13 constituents while the supplied LeachXS Lite database provides further data for approximately 40 constituents in most cases. A separate, compatible database is under development for cementitious materials from the Cementitious Barriers Partnership (CBP) program with support from DOE Office of Environmental Management.⁴

⁴ See www.CementBarriers.org.

SCENARIO MODELING

Data obtained from LEAF testing can be used to develop a chemical speciation fingerprint (CSF) for specific materials that contains the available fractions of constituents for leaching, along with a selection of relevant solid mineral phases and solid solutions, hydrous ferrous oxide (HFO) surfaces for adsorption, clay surface adsorption and organic matter partitioning. The resulting CSF then can be used in conjunction with specific mass transfer models and boundary conditions to evaluate field scenarios. LeachXS, as a result of the CBP and other research programs contains a flexible set of modeling tools for both CSF development and scenario modeling summarized as follows.

Developing a Chemical Speciation Model for Specific Materials

Chemical Speciation and Solubility Indices

Solubility index for possible minerals are evaluated in comparison to liquid-solid partitioning (LSP) data obtained from pH-dependent tests or L/S-dependent test for initial identification of potentially relevant mineral phases based on LSP data.

Laboratory Test Simulations

pH-dependence Test and LSP Simulation

Based on identified mineral phases and other parameters (e.g., availability values, carbon fractionation, etc.), a chemical speciation fingerprint (CSF) is developed to simulate results obtained from pH dependence tests. The parameters of this model can be varied to assess changes to LSP due to L/S (e.g., low L/S ratios found in field conditions), solution chemistry (e.g., high ionic strength solutions in cementitious materials), redox conditions (e.g., oxidation of reductive materials), the amount of iron hydr(oxide) and clay surfaces available for sorption, and the amount/fractionation of particulate organic matter (POM) and DOC.

Monolith Leaching and Mass Transport Rate Test Simulation

The CSF may be combined with diffusion models to determine mass transport parameters (e.g., effective tortuosity values, diffusivity) based on results from mass transport tests. The mass transport model segments a monolithic solid (i.e., a true monolithic form or a compacted granular material compacted to act like a monolith) into a series of layers from the external boundary to the interior core (Fig. 2). Within each layer, the monolith segment is divided into aqueous and solid phases defined by the CSF. Local equilibrium between phases in the segment is calculated at each time step to account for changes in pH and local composition based on dissolved constituent mass transport between the layers by diffusion through the liquid phase. The external surface of the monolith is simulated as being in contact with a well-mixed bath of finite volume which is refreshed at time intervals defined by the leaching test conditions. This laboratory simulation model also can be used to evaluate the impacts to release rates from changes in eluate volume, eluate chemistry (e.g., influx of acid or sulfate attack), and layering of material composition and properties within a monolith (e.g., a carbonated surface layer with an un-carbonated core).

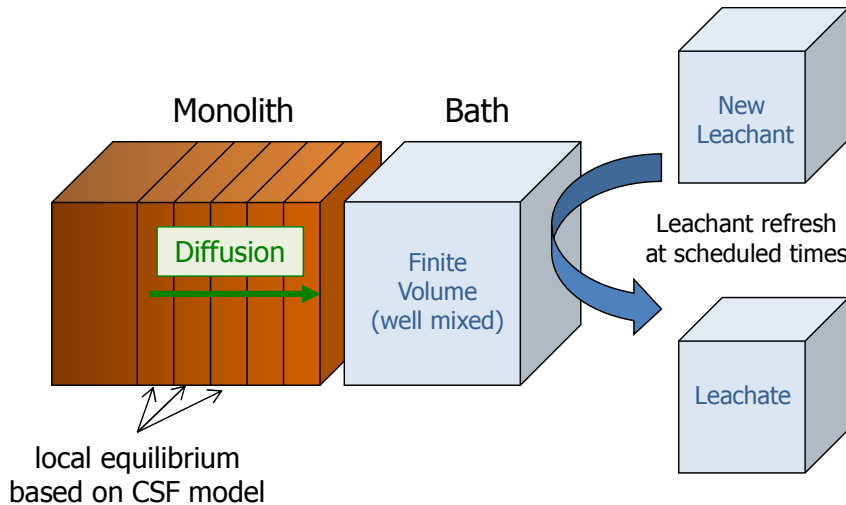


Fig. 2. Mass transport model (laboratory simulation) scenario.

Percolation Test Simulation (mobile-immobile zones)

The CSF and percolation parameters may be used to evaluate the results of percolation column tests using the conceptual model of mobile and immobile zones. The conceptual model (Fig. 3) consists of two zones segmented along the flow path, with one zone containing a mobile fluid phase in local equilibrium with the solid phase and the second zone containing an immobile fluid phase in local equilibrium with the solid phase. Within each column segment, each of the mobile and immobile zones are well mixed (i.e., uniform distribution of constituents within each of the solid phase and liquid phase orthogonal to the flow direction), and the mobile and immobile zones exchange dissolved constituents based a mass transfer coefficient that can be considered an effective diffusion distance. This model can be used for a first-order approximation of the effects of preferential flow in a percolation system, as well as the impacts of changes in redox and influent solution chemistry on the leaching of constituents.

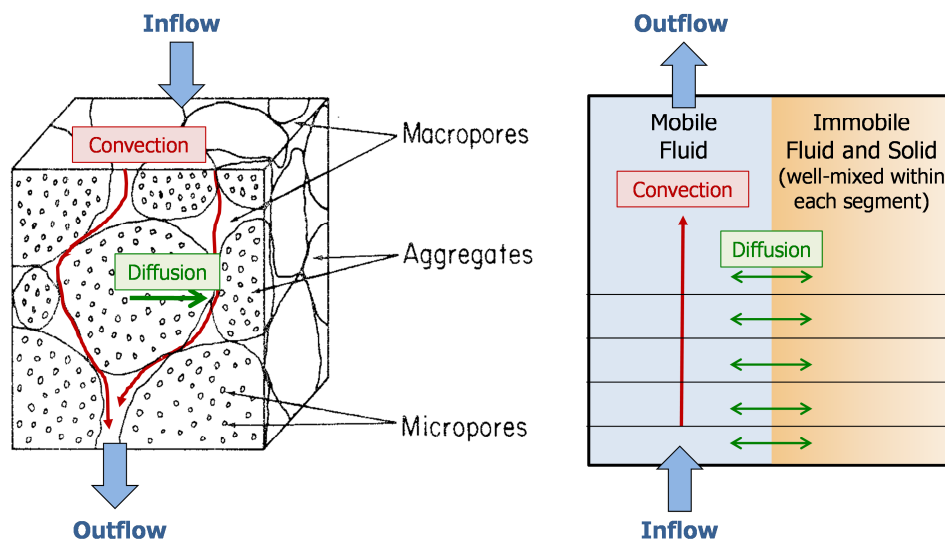


Fig. 3. Conceptual model of percolation with mobile and immobile zones shown for soil aggregates (left; from [20]) and as a 1-dimension approximation in ORCHESTRA (right).

Prediction Scenarios Modeling (Monolithic Diffusion)

Leaching

One dimensional diffusion from a monolith where system size, material layers, time frames, water contact and composition at the boundary can be varied to represent field scenarios. Unsaturated cases also can be simulated but without consideration of gas phase transport and reaction processes (e.g., oxygen or carbon dioxide gas phase transport and reaction).

Leaching with Carbonation and Oxidation

This model is analogous to the Leaching model above but also allows for consideration of gas phase transport and reaction processes to consider impacts of carbonation and oxidation [21].

Sulfate Attack with Leaching

This simulation allows coupling of physical degradation through sulfate attack on cementitious materials with leaching [22,23].

Prediction Scenarios Modeling (Percolation)

Mobile-Immobile Zones Dual Regime Leaching

This simulation is analogous to the percolation column test (mobile-immobile zones) model but with adaptation appropriate for evaluating field scenarios.

Percolation with Radial Diffusion Leaching

The CSF and percolation parameters also may be used for evaluation of field scenarios using the conceptual model of percolation with radial diffusion from porous solid particles. The conceptual model (Fig. 4) consists of two zones segmented along the flow path, with one zone containing a mobile fluid phase in local equilibrium with the solid phase and the second zone containing porous spheres with an immobile fluid phase (in contrast to the previous model using an immobile zone which was well mixed). Mass transport within the spheres occurs by diffusion through the fluid phase with the boundary condition of equal fluid composition at the interface between the sphere surface and the mobile zone, and no diffusion at the center of the spheres [24,25]. Thus, fluid phase constituents can diffuse into and out of the spheres based on concentration gradients, and within the spheres local solid-liquid equilibrium is maintained at each radial layer within the sphere. Within each column segment, each of the phases in the mobile zone is well mixed (i.e., uniform distribution of constituents within each of the solid phase and liquid phase orthogonal to the flow direction) and in local equilibrium between the solid and liquid phases. This approach is more accurately reflective of systems where the diffusion gradients within the immobile zone control release to the mobile zone. Thus, this model is sensitive to overall percolation flow rate and can be used to reflect the impact of fast infiltration that does not reach complete equilibrium between mobile and immobile zones. This model can also be used to evaluate leaching under the effects of preferential flow, cracking in monoliths, varying flow conditions (e.g., intermittent flow, different flow rates), and solution chemistry.

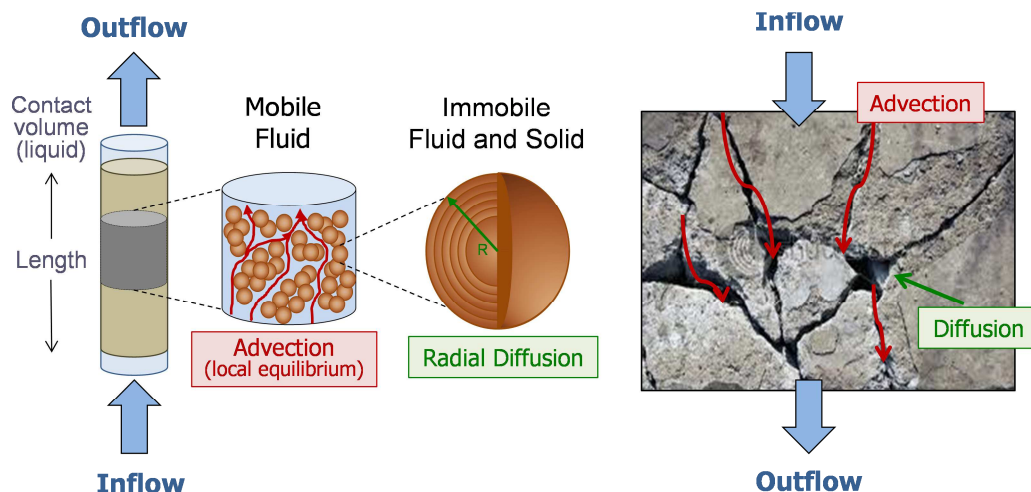


Fig. 4. Conceptual model of percolation with radial diffusion in the immobile zone shown as an up-flow column (left) and as flow through cracks in concrete (right).

CONCLUSIONS

Important components of the current LEAF development effort include: (i) formalization of leaching characterization test methods into standard methods, (ii) interlaboratory, round-robin testing for methods validation, (iii) documentation of the technical basis for test methods and application of specific mass transfer models for field scenarios, (iv) development of data management and evaluation tools, and (v) example application of the LEAF scenario evaluations for a range of applications. Subsequent steps will likely include development of guidance for application to specific types of evaluations and application to a wider range of materials. Continued development and use of LEAF will result in better risk-informed environmental decision making for waste disposal and beneficial use of secondary materials.

REFERENCES

1. Kosson, D. S., H. A. v. d. Sloot, F. Sanchez, and A. C. Garrabrants (2002). An Integrated Framework for Evaluating Leaching in Waste management and Utilization of Secondary Materials. *Environmental Engineering Science*, 19(3): 159-204.
2. Kosson, D.S., Sanchez, F., Kariher, P., Turner, L.H., Delapp, R. and Seignette, P. (2009). Characterization of Coal Combustion Residues from Electric Utilities – Leaching and Characterization Data. USEPA, EPA-600/R-09/151.
3. Thorneloe, S. A.; Kosson, D.S.; Sanchez, F.; Garrabrants, A.C.; Helms, G. (2010) Evaluating the Fate of Metals in Air Pollution Control Residues from Coal-Fired Power Plants. *Environ. Sci. Technol.*, 44 (19): 7351–7356.
4. van der Sloot, H.A., Kosson, D.S., Garrabrants, A.C., Arnold, J. , 2012. The impact of coal combustion fly ash used as a supplemental cementitious material on the leaching of constituents from cements and concretes. EPA/600/R-12/704, October 2012.
5. Kosson, D.S., Garrabrants, A.C., DeLapp, R., and van der Sloot, H.A. (2013). pH-dependent leaching of constituents of potential concern from concrete materials containing coal combustion fly ash. *Chemosphere*, in press.
6. Garrabrants, A.C., Kosson, D.S., DeLapp, R., van der Sloot, H.A. (2013). Effect of coal combustion fly ash use in concrete on the mass transport release of constituents of potential concern. *Chemosphere*, in press.

7. Sundaram, S.K., Chung, C-W, Parker, K.E., Kimura, M.L, Valenta, M.E., Burns, C.A., Pitman, S.G., Um, W., Chun, J., Westsik, J.H. (2011) Secondary Waste Form Development and Optimization-Cast Stone. PNNL-20159 Rev.1.
8. van der Sloot, H.A. , Meeussen, J.C.L. , Kosson, D.S. , Hjelmar, O. (2010). Use of chemical speciation and reactive transport to predict leaching from coal combustion residues in construction applications. *Proc. of the Second International Conference on Sustainable Construction Materials and Technologies*, Ancona, Italy.
9. van der Sloot, H.A., Meeussen, J.C.L., van Zomeren, A., Kosson, D.S. (2006) Developments in the characterisation of waste materials for environmental impact assessment purposes. *J. of Geochemical Exploration*, 88(1-3):72-76.
10. van der Sloot, H. A., van Zomeren, A., Meeussen, J. C. L., Seignette, P.F.A.B., Bleijerveld, R., 2007. Interpretation of test method selection, validation against field data, and predictive modelling for impact evaluation of stabilised waste disposal. *J. of Hazardous Materials*, 141:354-369.
11. Dijkstra, J.J., Meeussen, J.C.L., van der Sloot, H.A., Comans, R.N.J. (2008). A consistent geochemical modelling approach for the leaching and reactive transport of major and trace elements in MSWI bottom ash. *Applied Geochemistry*, 23(6):1544-1562.
12. Sanchez, F., Mattus, C., Morris, M. and Kosson, D.S. (2002). Use of a new leaching test framework for evaluating alternative treatment processes for mercury contaminated soils. *Environmental Engineering Science* 19(4):251-269.
13. Garrabrants, A.C., Sanchez, F., and Kosson, D.S. (2003). Leaching model for a cement mortar exposed to intermittent wetting and drying. *AIChE Journal*, 49(5):1317-1333.
14. van der Sloot, H.A., Dijkstra, J.J., Seignette, P.F.A.B., Hjelmar, O. and Spanka, G. (2009). Evaluation of a horizontal approach to assess the possible release of dangerous substances from construction products in support of requirements from the Construction Products Directive –emphasis on MSWI bottom ash. 3rd BOKU Waste Conference, Vienna, Austria.
15. Garrabrants, A.C., Kosson, D.S., van der Sloot, H.S., Sanchez, F., and Hjelmar, O. (2010). “Background information for the Leaching Environmental Assessment Framework (LEAF) Test Methods,” EPA-600/R-10/170, U.S. Environmental Protection Agency, Air Pollution Prevention and Control Division, December 2010.
16. Garrabrants, A.C., Kosson, D.S., Stefanski, L., DeLapp, R., Seignette, P.F.A.B., van der Sloot, H.A., Kariher, P., Baldwin, M., 2012a. Interlaboratory validation of the leaching environmental assessment framework (LEAF) method 1313 and method 1316. EPA-600/R-12/623, September 2012.
17. Garrabrants, A.C., Kosson, D.S., DeLapp, R., Kariher, P., Seignette, P.F.A.B., van der Sloot, H.A., Stefanski, L., Baldwin, M., 2012b. Interlaboratory validation of the leaching environmental assessment framework (LEAF) method 1314 and method 1315. EPA/600/R-12/624, September 2012.
18. EPA (1992) Guidance for Methods Development and Methods Validation for the RCRA Program. U.S. Environmental Protection Agency, <http://www.epa.gov/osw/hazard/testmethods/pdfs/methdev.pdf>.
19. van der Sloot, H. A., Seignette, P.F.A.B., Meeussen, J.C.L. , Hjelmar, O. and Kosson, D.S. (2008) A database, speciation modelling and decision support tool for soil, sludge, sediments, wastes and construction products: LeachXS™- Orchestra. In: Venice Symposium, Third International Symposium on Energy from Biomass and Waste, Venice, Italy.
20. van Genuchten, M.Th. and Dalton, F.N. (1986) Models for simulating salt movement in aggregated field soils. *Geoderma*. 38:165-183.
21. Brown, K.G., Arnold, J., Sarkar, S., Flach, G., van der Sloot, H.A., Meeussen, J.C.L. and Kosson, D.S. (2013). Modeling carbonation of high-level waste tank integrity and closure. EPJ Web of Conferences, vol. 56, 05003.

22. Sarkar, S., Mahadevan, S., Meeussen, J.C.L., van der Sloot, H., and Kosson, D.S. (2010). Numerical simulation of cementitious materials degradation under external sulfate attack, *Cement and Concrete Composites*, 32(3):241-252.
23. Sarkar, S., Mahadevan, S., Meeussen, J.C.L., van der Sloot, H.A. and Kosson, D.S. (2012). Sensitivity analysis of damage in cement materials under sulfate attack and calcium leaching. *J. of Materials in Civil Engineering*, 24(4):430440.
24. van Beinum, W., Meeussen, J. C. L., Edwards, A. C., and van Riemsdijk, W. H., Transport of ions in physically heterogeneous systems; convection and diffusion in a column filled with alginate gel beads, predicted by a two-region model, *Water Research*, 34(7), 2000, pp. 2043-2050.
25. Sarkar, S., Kosson, D.S., Meeussen, H., van der Sloot, H.A., Brown, K, and Garrabrants, A.C. (2013). A dual regime reactive transport model for simulation of high level waste tank closure scenarios. WM2013 Conference, Feb. 24-28, Phoenix, Az.

DISCLAIMER AND ACKNOWLEDGEMENTS

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This paper was prepared with the financial support by the U. S. Department of Energy, under Cooperative Agreement Number DE-FC01-06EW07053 entitled 'The Consortium for Risk Evaluation with Stakeholder Participation III' awarded to Vanderbilt University. This research was carried out as part of the Cementitious Barriers Partnership supported by U.S. DOE Office of Environmental Management. The opinions, findings, conclusions, or recommendations expressed herein are those of the authors and do not necessarily represent the views of the U.S. Environmental Protection Agency, Department of Energy or Vanderbilt University.