Pacific Northwest NATIONAL LABORATORY

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DVZ-RPT-125

Assessment and Simulation of Particulate Transport for Delivery of Solid Amendments into the Subsurface

FY25 Status Report

September 2025

Katherine Muller
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U.S. DEPARTMENT OF ENERGY

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Summary

For particulate-based amendments to be viable for field-scale remediation at the Hanford Site (e.g., the 200-DV-1 Operable Unit), amendment particles need to be delivered a reasonable distance away from an injection well to provide cost effective in situ treatment. Field-scale particle transport models can estimate spatial deposition of amendment particles in the subsurface, which is critical for developing an overall remediation strategy. However, field-scale particle simulations are currently limited due to insufficient simulation capability and a lack of experimental data to validate and parameterize particle transport models. During this fiscal year, the following progress has been made toward a field-scale particle transport modeling evaluation: (1) in addition to the two particle transport models implemented last FY, four additional particle transport models have been implemented within PFLOTRAN; (2) a Python-based pre-screening tool was finalized, enabling users to quickly estimate the particle radius of influence (ROI) for any given particle-amendment system; and (3) an initial compatibility assessment was completed using both particle transport simulations deployed through the pre-screening tool, in conjunction with general guidelines to (a) identify the most suitable amendment particle sizes for various Hanford sediments and (b) evaluate amendment-delivery fluid compatibility.

The preliminary compatibility assessment revealed that for amendment delivery success to the various Hanford target formations, amendment particle sizes will likely need to be smaller than the amendment sizes tested in the DV-1 treatability study. It is recommended that amendment particles be decreased in size, or alternative smaller size amendments be obtained from the manufacture, prior to any further experimental testing. Also, preliminary testing suggests that xanthan gum may be the most broadly compatible delivery fluid. Planned laboratory experiments will be instrumental in validating and refining the PFLOTRAN particulate transport model formulations, ultimately enabling predictive capabilities to facilitate the design of field-scale amendment delivery systems.

This work consists of acquiring new theoretical or experimental knowledge. The information associated with this report should not be used as design input or operating parameters without additional qualification.

Introduction

Nine technologies were evaluated for their ability to reduce long-term contaminant mobility in the 200-DV-1 Operable Unit (DOE/RL 2019) at the Hanford Site. Five of these technologies are solid-phase amendments, including stannous apatite, two bismuth-based materials, and two iron-based materials. Laboratory results indicate that these particulate amendments hold promise for sequestration of primary and some co-contaminants, but technical challenges remain related to the ability to inject and strategically emplace these materials throughout the subsurface (PNNL-35432 2023).

While these laboratory experiments focused primarily on evaluating the efficacy of treatments, there have been ancillary lessons learned about potential field implementation (e.g., observed plugging and particulate amendment injection distances achieved in 1-D columns). Additional testing, evaluation, and simulations are necessary to fully consider aspects related to amendment delivery and implementation at the field scale. Developing tools and simulation capabilities to evaluate the delivery of solid amendments will enable informed decision-making when assessing the feasibility of particulate amendments and choosing associated parameters for field-scale implementation.

Particle Transport Model Implementation in PFLOTRAN

To advance simulation capabilities, various particle transport models have been integrated within the reactive transport simulator PFLOTRAN.[1] These models include a colloid filtration theory (CFT) model and five additional particle transport models (M1-M5), each with increasing levels of complexity (Table 1). As of fiscal year (FY) 2025, all models (CFT and M1-M5) have been fully implemented in PFLOTRAN. However, to use particle transport models in accurate simulations, they must be validated and parameterized with experimental data.

Table 1. Particle transport models and PFLOTRAN implementation status.

Model	Mechanisms Modeled	Implementation Status
Colloid filtration theory	Deposition	Complete (FY24)
M1	Attachment with blocking (one-site)	Complete (FY24)
M2	Straining with blocking (one-site)	Complete (FY25)
M3	Attachment with blocking and straining (one-site)	Complete (FY25)
M4	Attachment with blocking and straining (two-site)	Complete (FY25)
M5	Attachment with a maximum surface concentration (one-site)	Complete (FY25)

Particle Transport Pre-screening Tool

In parallel to particle transport model development, a pre-screening tool was finalized this FY. This tool provides quick estimations of expected particle injectability, particle deposition rate, and radius of influence for any given particle suspension into a target porous media. This approach enables estimates based solely on simplified laboratory measurements, facilitating the down-selection of viable solid amendments for field-scale application. CFT is frequently used as a simplified model that lumps all particle transport mechanisms into a single first order deposition rate, k_d, to describe overall particle retention. Using CFT, the pre-screening tool calculates the deposition rate from the input parameters and then runs a 1-D reactive transport simulation with PFLOTRAN to estimate the potential particle transport distance.

Using this tool, a series of simulations (n = 1000) were performed across a broad range of input parameters using Latin hypercube random multiparameter sampling. This approach offers insight into

[1] PFLOTRAN: A Massively Parallel Reactive Flow and Transport Model for describing Subsurface Processes (www.pflotran.org)

which factors have the greatest influence on the particle deposition rate. Such insights can aid in identifying key aspects to consider when optimizing amendment suspension formation and system design.

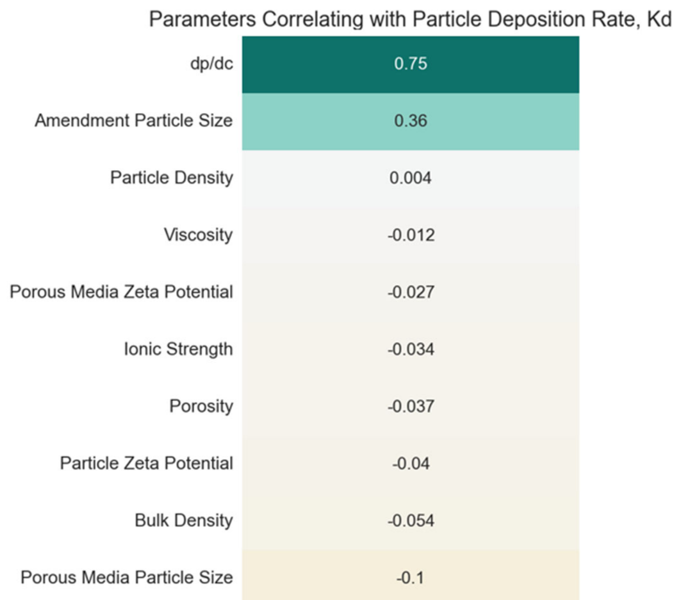

Figure 1. Correlation coefficients between particle deposition rate, k_d, and various amendment suspension properties.

The particle deposition rate is most strongly correlated with the ratio of amendment particle size (amendment particle diameter, d_p) to sediment particle size (collector particle diameter, d_c) (i.e., d_p/d_c) (Figure 1) and amendment particle size. Larger particle deposition rates result in a shorter anticipated transport distance. This finding supports the notion that decreasing amendment particle sizes will likely have the greatest impact on improved delivery outcomes. Additional size recommendations are detailed below. While this preliminary assessment was conducted on a wide range of parameters, system-specific measurements are needed to constrain particle transport predictions for individual amendment-porous media combinations.

Initial Amendment Compatibility Assessment for Successful Delivery

Particle Size Assessment

Particle size analysis was performed on the solid phase DV-1 amendments, as well as relevant Hanford sediments. Amendments include laboratory synthesized Sn(II) apatite (ground and finely ground), laboratory synthesized bismuth oxyhydroxide (BOH), commercially obtained bismuth subnitrate (BSN; Sigma-Aldrich), sulfur modified iron (SMI; SMIwater), and zero valent iron (ZVI; Alfa Aesar). Sediments included a Hanford B Complex sediment (BY), sediment from the Cold Creek Unit – gravel (CCUg), Cold Creek Unit perching zone sand (PZSd), Hanford formation (Hfm) sand collected from Pasco, WA, and two sediments from the Hanford S Complex (S9-batch and S9-column). These were chosen as field-relevant sediments that also cover a range of grain sizes.

Amendment particle size distributions were compared to the Hanford sediments and evaluated using two general guidelines commonly applied in the remediation community to assess the delivery feasibility

(Table 2). First, amendment particle sizes should be less than 0.1 – 0.3 of the sediment pore throat diameter (Bear, 1972). Second, sediment d_{50} should be 1 to 2 orders of magnitude larger than the d_{50} of the amendment. It is important to note that these guidelines are highly simplified, as they only consider particle straining and assume homogenous porous media properties, but do remain a practical tool for quickly estimating suitable amendment particle size ranges for various formations of interest. Table 2 provides a score between 0 and 4 for each sediment-amendment combination, where larger numbers indicate higher likelihood of successful delivery.

Based on this analysis, none of the tested amendments in their current state are suitable for injection into PZSd sediments, and only low suitability was found in BY or CCUg sediments. All amendments, except SMI, would likely be suitable in the Hanford formation. Results indicate that all amendments may benefit from additional processing before injection, such as mechanical grinding to achieve smaller final particle sizes or sieving to obtain only the finer fraction of the amendments. For some amendments, like ZVI, obtaining smaller particle size amendments from manufacturers may also be an option (e.g., nano-ZVI).

Table 2. Applicability of amendment particle sizes for various Hanford sediment formations. Higher scores are better.

Sediment Name	Particle Amendment					
	BSN	BOH	Sn(II)-Apatite (ground)	Sn(II)-Apatite (finely ground)	SMI	ZVI (6-10 µm)
BY	3	2	2	2	0	2
CCUg	2	2	2	2	0	2
PZSd	0	0	0	0	0	0
Hfm	4	4	3	4	0	4

Notes:

(a) Compatibility scores based on points (out of 4). For the pore throat rule, 1 pt if acceptable at the d_{50} value or 2 pts if acceptable using the d_{10} value. For the order of magnitude rule, 2 pts if acceptable using the d_{50} value.

(b) Pore throat rule: $< 0.1 – 0.3\ d_{pore}$ (Bear 1972).

(c) d_{50} comparison: Sediment d_{50} should be 1-2 orders of magnitude larger than the d_{50} of the amendment.

Particle Suspension Stability Analysis

Preliminary batch experiments were conducted to begin evaluating the stability of particulate suspensions across various delivery fluids and concentrations. The results, summarized in Table 3, are colored-coded to facilitate interpretation: green indicates the suspension was stable or kinetically stable, suggesting it is likely injectable, whereas red denotes instability, indicating the suspension would likely be difficult to inject into the subsurface. In some instances, at very high delivery fluid concentration, the suspension formed a gel-like phase, which would likely hinder injectability (shown in blue). While particle size compatibility is critical to consider for delivery as demonstrated above, the ability to create stable amendment suspensions of reasonable amendment concentrations is also paramount. Although, delivery fluids can be tailed to specific amendment-delivery fluid combinations of interest, results in Table 3 suggest xanthan gum as a delivery fluid may be broadly compatible across the particle amendments of interest. Further, these results begin to outline amendment concentrations that likely can be successfully suspended with delivery fluids.

Table 3. Summary of particle suspension stability results.

	Delivery Fluid			
	Xanthan Gum		Guar Gum	
Amendment	800 mg/L	4000 mg/L	800 mg/L	1600 mg/L
Tin-apatite-ground	Stable	Not measured	Unstable	Unstable
BOH	Likely unstable	Not measured	Unstable	Unstable
BSN	Kinetically stable – likely unstable	Not measured	Stable	Stable
SMI	Unstable	Stable-formed gel	Unstable	Not measured
mZVI	Kinetically stable	Stable-formed gel	Unstable	Not measured

Notes:
(a) Data and analysis are currently For Information Only (FIO).
(b) Classification of suspension stability: If A/A_0 at 30 min is > 0.75 = Stable; 0.5 - 0.75 = Kinetically Stable; 0.25 - 0.5 = Likely Unstable; < 0.25 = Unstable.
(c) A is the absorbance measurements and A_0 is the initial absorbance measurement.

Future Activities

The next steps to enable the viable use of particulate-based amendments for field-scale remediation at the Hanford Site are as follows:

- Measure additional relevant parameters of DV-1 amendments, delivery fluids, and porous media properties required for continuation of pre-screening efforts. This effort may include additional preparation of amendments, such as grinding, sieving, or obtaining new materials to decrease particle size.
- After down-selection of amendments based on pre-screening results, conduct multi-scale laboratory injection experiments for model verification and parameterization.
- Model laboratory experiments to parameterize and evaluate the utility of the implemented particle transport models to describe behavior.
- Implement two-phase immiscible flow mode in PFLOTRAN to account for the viscous nature of the delivery fluids.
- Build simplified 3-D model of the Hanford 200 Area to simulate particle injections at the field scale.

Quality Assurance

This work was performed in accordance with the Pacific Northwest National Laboratory Nuclear Quality Assurance Program (NQAP). The NQAP complies with the DOE Order 414.1D, *Quality Assurance*. The NQAP uses NQA-1-2012, *Quality Assurance Requirements for Nuclear Facility Application*, as its consensus standard and NQA-1-2012, Subpart 4.2.1 as the basis for its graded approach to quality. Any data presented in this document is preliminary, FIO, and subject to revision. This work emphasized acquiring new theoretical or experimental knowledge. The information associated with this report should not be used as design input or operating parameters without additional qualification.

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Pacific Northwest
National Laboratory

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