Pacific Northwest National Laboratory

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Cross-Site Transfer System at Hanford: Long-Term Strategy for Waste Acceptance

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under Contract DE-AC06-76RLO 1830

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Executive Summary

This report summarizes the results of a technical panel review of the current methodology for acceptance of the waste for transport through the Hanford Replacement Cross-Site Transfer System (RCSTS). An existing document, *Defining Waste Acceptance Criteria for the Hanford Replacement Cross-Site Transfer System* (Hudson 1996), was reviewed, and this report is issued as a complement to that report rather than a replacement. The methodology proposed in that document was refined based on the recommendations of the panel. The refinements were focused around predicting and preventing the three main modes suspected of plugging the existing CSTS: precipitation, gelation, and particle dropout, or settling. The proposed analysis will require an integration of computer modeling and laboratory experiments to build a defensible case for transportability of a proposed slurry composition for a given tank. This predictive analysis will be validated by recirculating the actual tank waste, in-tank and in-farm, prior to transport. This validation step is considered to be an essential element of the waste acceptance methodology. The panel's recommendation was that the probability of success of waste transfer would be greatly improved by integrating the predictive analysis with real-time control during the operation of the RCSTS.

The long-term objective is to convert the technical contents of this report into a "turn-key" analysis package with clear and simple set of protocols for its use by operating personnel. The short-term objective is to produce a defensible analysis package. To reach our long-term objective we will optimize the methodology with sensitivity studies, validation, prioritization, and experience. The optimization process will most likely entail reducing the complexity of the methodology by eliminating those components of analysis and characterization that do not have a significant impact on the final results.

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1.0 Introduction

A new, replacement cross-site transfer system (RCSTS) has been constructed at the Hanford Site to replace the existing pipelines that hydraulically connect the 200 West and 200 East areas. The objective of this, the W-058 project, is to transfer the waste slurries through the pipeline while preventing any incident that would plug or block this pipeline with solid particulates or solidified waste. Precipitation of solids, which may result in gelation and promote settling of solid particles, has been speculated to be the main cause of plugging in the existing cross-site transfer system pipelines. To better understand these processes and the history of past plugging incidents, a review of some laboratory experiments has been provided.^(a,b,c) In the 1993 report, McKay provides a list of observations on why the existing pipelines at Hanford are plugging, as described by former tank farm operators and managers.

Much is understood about the behavior of the waste slurries from characterization of the species that constitute the waste and rheological measurements of the mixtures. Recent characterization of Tank 241-SY-102 slurries (Onishi et al. 1996a) using transmission electron microscopy (TEM) showed that the major crystalline phases include plate-like γ - Al_2O_3 particles and needle-like $\text{FeO}(\text{OH})$ particles. Other crystalline constituents found were hydroxylapatite, $\text{Ca}_5(\text{PO})_3(\text{OH})$, and gibbsite, $\text{Al}(\text{OH})_3$. The most noteworthy results were associated with the rheology of the mixture. It was found that a large population of amorphous silica and iron hydroxide primary particles exist in the nanometer size range and tend to control the rheology of the mixture. The aggregates formed between the crystalline and amorphous solids yielded a mixture with both rheopectic and thixotropic behavior. Further, the initially shear-thinning (almost a Bingham plastic) mixture became fairly Newtonian after a 1:1 dilution with NaNO_3 solution. However, this result is considered specific to SY-102 waste. Recent ball rheometer results show that dilution of the settled SY-101 sludge with the supernatant liquid from the same tank results in a shear-thinning behavior with a behavior index of approximately 0.5 (Stewart et al. 1996). Thus dilution may not always result in a Newtonian behavior, as also noted in the 1994 review document.^(a)

Another important result relevant to transporting waste slurry across the RCSTS is the solids density. Onishi et al. (1996a) found that the solids in SY-102 waste have densities of approximately 2.8 g/mL. This finding contrasts with the assumption of 1.8 g/mL used in Hudson (1996). Accurate knowledge of this parameter is of paramount importance in determining the velocity at which particles begin to settle. Underestimating this parameter may result in settling and, in turn, pose a potential for pipeline plugging. TEM graphs reveal that the solid particles are agglomerates of a large number of finer, nanometer to micron-sized primary particles, and the formation geometry and size of these agglomerates depend strongly on the

(a) McKay RL. 1993. *TWRS Retrieval Technology Project Slurry Transport - Plugging Investigation*. Pacific Northwest Laboratory, Richland, Washington.

(b) McKay RL, FF Erian, CJ Call, and EA Daymo. 1994. *Slurry Transport of Hanford Tank Wastes: Open Technical Issues and Recommended Actions*. DSTRTP-CY94-012, Pacific Northwest Laboratory, Richland, Washington.

(c) McKay RL, CJ Call, and EA Daymo. 1994. *Methodology for Defining the Appropriate Tank Waste Properties for Transport*. DSTRTP-CY94-031, Pacific Northwest Laboratory, Richland, Washington.

solution chemistry, temperature, and shear field (LaFemina et al. 1995). As a result, characterization density becomes a non-trivial issue and should be treated with care.^(a)

Several studies have been performed that summarize the set of equations most appropriate for calculating critical velocity and pressure drop across RCSTS (Hudson 1996; Onishi and Hudson 1996).^(b, c, d, e) Most of these calculations are based on Newtonian or pseudo-Newtonian formulations. Although the philosophy was consistent for conservatism in calculating critical velocity, it is the view of the current authors that over-conservatism in the calculations would result in decision-making complications at the validation stage (this step will be described in detail later in this report). An accurate prediction is more desirable from the standpoint of understanding the processes and possible lack of consistency with experimental validation. If conservatism is to be built into the critical velocity and pressure drop, it would be to our advantage to do so as a safety factor after the analysis has been completed. In that case, one hopes to have more control on the margin of safety and the associated risk. Further, the degree of confidence in the predictions and the estimated margin of safety improves substantially by performing validation tests.

This report was prepared based on the review of an expert scientific panel, which brought together expertise in five key areas: hydrodynamics, rheology, interfacial science, analytic chemistry, and aqueous phase chemistry (equilibria and kinetics). The function of the panel was to 1) review the proposed methodologies for waste acceptance for RCSTS operation and 2) make recommendations about beneficial improvements to the current methodology. The approach proposed by Hudson (1996), which was found to be the most direct approach for defining the waste acceptance criteria, was the focus of this review.

It is believed that precipitation may lead to an increase in pressure drop, formation of a gel in the pipeline, or enhancement in the settling rate of particles. That is, pipeline blockage may not be a direct consequence of precipitation. From a phenomenological perspective, pressure drop is the result of momentum exchange (loss) from the fluid to the pipe walls through the wall shear stress. An increase in the pressure drop is the result of increases in either the effective fluid viscosity or the wall shear rates. Precipitation during transport is expected to increase the concentration of solids in the slurry, which manifests itself in escalation of the effective viscosity of the slurry (Chang and Powell 1993). On the other hand, formation of high-aspect ratio solid particles, such as highly hydrated sodium phosphate crystals, potentially

-
- (a) As will be discussed further in this report, this quantity cannot be directly characterized. Thus, most of the measurements will lead indirectly to information on agglomerate density.
- (b) Letter, JD Hudson to JE Van Beek, Pacific Northwest National Laboratory, May 10, 1996.
- (c) McKay RL. 1993. *TWRS Retrieval Technology Project Slurry Transport - Plugging Investigation*. Pacific Northwest Laboratory, Richland, Washington.
- (d) McKay RL, FF Erian, CJ Call, and EA Daymo. 1994. *Slurry Transport of Hanford Tank Wastes: Open Technical Issues and Recommended Actions*. DSTRTP-CY94-012, Pacific Northwest Laboratory, Richland, Washington.
- (e) McKay RL, CJ Call, and EA Daymo. 1994. *Methodology for Defining the Appropriate Tank Waste Properties for Transport*. DSTRTP-CY94-031, Pacific Northwest Laboratory, Richland, Washington.

leads to entanglements that, in effect, form clusters. These large clumps or clusters, much like large-scale turbulent structures and eddies, tend to increase the near-wall shear rates by redistributing the velocity profile across the pipe.^(a)

Precipitation will change the density, size, and shape (fractal dimension) of the solid agglomerates. Precipitated solids will nucleate on the surface of the agglomerates within the slurry. If the aggregation during flow is reaction-limited, denser agglomerates may form; if diffusion-limited, agglomerates may be fluffier.^(b) Denser agglomerates would pose enhanced settling potential, while larger and fluffier agglomerates could potentially gel. Gelation is the point at which the material develops a yield strength. Although from a solids transport point of view, a slight yield strength is desirable to prevent settling of solids (Duckworth et al. 1986), simple theoretical arguments as well as measurements show that the effective viscosity of the fluid and the wall shear rate increase simultaneously (Metzner and Reed 1955; Soto and Shah 1976; Park et al. 1989). Settling increases the effective shear rates near the wall by reducing the local flow cross-sectional area. Again, both of these effects tie into the pressure drop via an increase in the overall wall shear stress in a manner similar to that described in the previous paragraph.

Given the above perspective, how do we approach this problem from an engineering problem solving point of view? Can we propose a methodology that is robust enough to cover all the failure modes? This report presents an overall approach that the review panel considers robust: a computational chemistry code that predicts and prevents formation of undesirable solid species for a specific waste composition. This code will also provide quantitative data on the slurry that would otherwise not be easily attainable from characterization. Focused characterization efforts, in addition to providing most of the input parameters to the analysis codes, will also be able to predict conditions under which gelation may occur. These are conditions that will be avoided during transport. The characterization results will then be used in a hydrodynamic analysis code, using well-accepted correlations for defining the waste form to be transferred and the operating conditions within the system specifications. A more detailed picture of the methodology is sketched in Section 2. Section 3 described the approach to the analysis; the parameters needed from characterization are outlined in Section 4. Details of the validation phase are presented in Section 5. The control function is the subject of Section 6, and Section 7 summarizes the strategy. References cited are in Section 8, and the appendix contains a look forward to planned documentation.

(a) Of course, another adverse effect may very well be plugging of narrow orifices such as valve openings when the cluster size reaches that of the pipe diameter.

(b) Rector DR and BC Bunker. 1995. *Effect of Colloidal Aggregation on the Sedimentation and Rheological Properties of Tank Waste*. TWRSP-95-027, Pacific Northwest Laboratory, Richland, Washington.

2.0 Overview of Methodology

The general methodology proposed by Hudson (1996) was considered to be an appropriate framework upon which further refinement can be constructed. The recommended general approach comprises the following stages:

- (a) predictive analysis
- (b) validation using laboratory and field experiments
- (c) control analysis.

The purpose of the predictive analysis is to define the operating criteria for a transfer through RCSTS. It is through performing this stage of analysis that the problem areas to be avoided for waste transportability may be defined, and a set of operating conditions and waste-conditioning criteria may be proposed. The final results will be formalized in a Slurry Transport Waste Acceptance Report (STWAR). STWAR will be considered the guideline for successful operation of the RCSTS for a given waste tank slurry.

Figure 2.1 is the flow diagram of the steps taken before issuing a tank-specific STWAR. This diagram is a modified version of Figure 2.1 in Hudson (1996). The most recent relevant tank characterization data will be gathered. Based on the preliminary constituencies information, phase I analysis will be performed to make some go/no-go decisions on a particular slurry composition to be transported before performing more accurate characterization and phase II analysis for prediction of the appropriate operating conditions.

The equilibria phase chemistry will be predicted using a computational chemistry code such as GMIN. This analysis will determine whether and to what extent some of the known undesirable phases, such as boehmite and apatite, could exist in the slurry during transport. Such prediction, if accurate, would eliminate the need to perform laboratory investigation of precipitation and phase speciation. Further, it is difficult to fully quantify the species present in the system using laboratory measurements. Transmission electron microscopy will yield information on which phases are present, but it is not able to quantify the amounts of these species.

If the proposed slurry to be transferred is not acceptable from a phase chemistry standpoint, a new slurry is identified by adding diluent and testing the final mixture for phase chemistry using the chemical modeling. This process is repeated until a suitable slurry is identified. The resulting slurry is tested for transportability, i.e., the pressure drop and critical velocity should be acceptable. The slurry that meets the transportability tests from both hydrodynamic and phase chemistry viewpoints is submitted. Details of the analysis approach are covered in Section 3.

Additional (limited) tank-specific characterization tests will be performed at this point to meet the retrieval and transport needs. The parameters needed from characterization are spelled out in Section 4. However, the specific characterization needs will be decided for each tank based on what is available from the tank characterization reports. As more experience is gained from the waste cross-site transfer operations, characterization needs are expected to be reduced and streamlined. The results will be issued formally in a Slurry Transport Characterization Report (STCR).

Figure 2.1. Methodology for the Proposed Waste Acceptance Criteria [Slurry Transport Proposal Record (STPR), Slurry Transport Characterization Report (STCR), Slurry Transport Waste Acceptance Report (STWAR)]

A new set of calculations will be performed once better data have been obtained. The final calculations will establish the composition of waste to be transported and the operating conditions for safe transport. This concludes the predictive analysis phase. A report will then be issued proposing the slurry transport conditions to be tested in the in-tank and in-farm recirculation loops prior to transport.

The validation phase is proposed to take place in two forms: 1) laboratory tests that validate the overall methodology using controlled environment and 2) field tests with actual waste to be transported to validate tank-specific waste acceptance criteria. More details on this topic are provided in Section 5.

The structure of the control element is the same as that proposed by Hudson (1996). The control function is responsible for documenting and integrating all prior activities; it uses that information to specify controls that apply to the transfer process and authorizes the transfer. This function also monitors the process for any unusual or unplanned system or slurry behavior and provides procedural and systems support for contingencies. A framework for this function is described in Section 6. The product of the control function is the STWAR, which documents the basis for safe transfer, specifies the applicable controls, and authorizes system use. The appendix provides a preliminary list of the contents of a typical STWAR.

Finally, control analysis will be necessary to ensure that operating parameters are maintained within design specifications. This analysis will use information from the process monitoring function as input and evaluate system performance. If the system is found to be behaving within the expected and acceptable range of performance, no corrective action will be pursued. However, if the process monitoring information point toward a range of operation outside of the acceptable domain, then corrective actions will be recommended. This corrective action may be control of the pump speed or in-line dilution of the slurry. The analysis is described in Section 6.

3.0 Analysis

A transport analysis will determine the suitability of a waste for cross-site transfer through the RCSTS. This analysis uses the available information (for Analysis I) and measured data obtained during characterization (for Analysis II) to evaluate whether the RCSTS is capable of transporting the slurry at a sufficient velocity to ensure that solids will be in suspension during the transfer.

If the waste is determined to be unsuitable for the cross-site transfer, this analysis will then identify an alternative waste option and determine its acceptability. The transport analysis will consist of available data on waste characterization, chemical modeling, physical property determination, and transportability assessment.

As mentioned in Section 2, two groups of analysis are required: 1) aqueous phase chemistry and 2) hydrodynamic transport. The procedures for performing these analyses are discussed in this section, which provides an overview of the types of analyses that will be performed prior to issuing the STWAR. Because this report complements rather than replaces the one previously issued by Hudson (1996), many of the details provided in that report are prerequisite and will not be duplicated in this report.

3.1 Aqueous Phase Chemistry Analysis

Tank wastes may undergo complex chemical reactions (e.g., aqueous reactions and solid precipitation/dissolution) and physical processes/changes (transport, diffusion, changes in densities/viscosities of supernatant, slurry and sludge) during various tank operations (e.g., jet-mixing, adding diluents, waste retrieval, and cross-site transfer). An October 1996 workshop, "Hanford Tank Wastes: Technical and Scientific Issues," organized by PNNL concluded that potential chemical species/reactions and waste rheology are critical to addressing technical issues and in successfully conducting these tank waste operations.

In order to determine how much of the tank waste can be retrieved and thus safely transferred through the RCSTS, we need to know not only the physical characteristics of the wastes (e.g., density, viscosity, solid concentrations), but also the chemical species and their potential chemical reactions (e.g., what Al, Na, P solids and aqueous species exist in a tank and what will happen to them?).

3.1.1 Selection of Chemical Codes

The chemical modeling will determine aqueous, solid, and gaseous chemical species of the waste. The purpose of the chemical modeling is twofold: first, to identify potential troublesome chemical solid formations, which would pose the possibility of blockage of the cross site transfer line; and second, to provide mass fraction of solids and liquid density as a function of temperature and waste composition. These results will then be used for subsequent transport analysis to evaluate the feasibility of the cross-site transfer. This process is also used to identify potential alternative wastes if the original waste is judged unsuitable for cross-site transfer.

The chemical reaction codes should be able to handle characteristics of the Hanford tank wastes, including high ionic strength; high pH; high sodium concentration; large amounts of aluminum, sodium, and iron solids; and high temperature. There are many chemical codes

available; however, most of those (e.g., MINTEQ, WATEQ, and MINEQL) are based on the mass-action approach and are not applicable to these high ionic conditions (Onishi and Hudson 1996). Many chemical models have been previously reviewed by various researchers (Felmy et al. 1983, Jenny et al. 1984, Onishi et al. 1995). As indicated by some of these reports, there are four codes that can predict chemical reactions under high ionic strength conditions like the Hanford tank conditions: EQ3/6, ESP, GMIN (Felmy 1990), and TEMPEST (Onishi et al. 1996b).

The EQ3/6 code is a reaction-path model and has primarily been applied to high-level waste in deep geological repositories. It is widely accepted for geological applications; however, it is a very large computer code that requires large computational resources.

ESP is a user-friendly equilibrium/kinetic chemical code based on the Bromley's model (Bromley 1973). It has a relatively large thermodynamic database. GMIN provided some of database values for ESP. Its database, however, appears to have some thermodynamic inconsistencies, and many of its database values for high ionic strength conditions are not traceable to specific studies/papers.

GMIN, an equilibrium chemical code based on a constrained minimization of Gibbs free energy, uses the Pitzer equations for aqueous phase modeling. It is applicable to high ionic strength conditions. The thermodynamic database of GMIN covers many of the dominating chemical reactions occurring in the Hanford tanks, but it is not very large at this time. Pacific Northwest National Laboratory has been expanding its database to widen its applicability to the tanks. One of the strong points of GMIN is that all thermodynamic databases built into the code are traceable to specific studies and papers, providing scientific defensibility to the model results.

The TEMPEST code (Onishi et al. 1996a), which contains GMIN as one of the submodels, is a time-varying, three-dimensional model that simulates both physical movements (e.g., slurry, solid, and gas movements) and equilibrium/kinetic chemical reactions and the associated changes on some waste properties and rheology.

The GMIN code is a good candidate for the chemical modeling. We also suggest using TEMPEST when kinetics and interactions of chemical reactions and physical mechanisms that occur during cross-site transfers are important factors in transferring the waste in the pipeline.

The GMIN code calculates the chemical composition of systems composed of aqueous phases, pure solid phases, solid-solution phases, adsorbed phases, and gas phases. In the aqueous phase modeling, the excess solution free energy is modeled using the Pitzer equations (Harvie et al. 1987), which are valid to high ionic strengths. The Davies equations (Felmy 1990) can also be used as an option in the GMIN code. In GMIN, the activity coefficients for non-ideal, solid-solution phases are calculated using parameters of a polynomial expansion in mole fraction of the excess free energy of mixing. The free energy of adsorbed phase species is described by the triple-layer, site-binding adsorption model. The mathematical algorithm in GMIN is based on a constrained minimization of the Gibbs free energy (Snoeyink and Jenkins 1980; Harvie et al. 1987). This approach is more numerically stable and reliably converges to a free energy minimum, compared with more common chemical equilibrium codes based on the mass-action approach (Felmy 1990), such as MINTEQ, WATEQ, and MINEQL. Although GMIN does not calculate the liquid density, with some modification it can determine the liquid density theoretically based on calculated aqueous species concentrations.

For the kinetic reactions, the following rate law, R , for chemically reactive solid i is commonly used (Steeffel and Lasaga 1994):

$$R_i = \frac{d[C_{si}]}{dt} \quad (3.1.1)$$

and

$$R_i = \left\{ \text{sign of } \left(\ln \frac{Q_i}{K_i} \right) \right\} \{k_{i1} + k_{i2}(\text{solids surface area})\} \left\{ 1 - \frac{Q_i}{K_i} \right\}^{m_i} \quad (3.1.2)$$

where

$[C_{si}]$	= molality of solid, i
K_i	= equilibrium constant
k_{i1}, k_{i2}'	= kinetic rates of a solid
m_i	= constant
Q_i	= activity product.

The above kinetic expression can be used for both elementary and non-elementary reaction cases. By assuming $m_i = 1$ in the Equation 3.1.2, TEMPEST uses the following kinetic reaction equations:

$$R_i = \frac{d[C_{si}]}{dt} = \{k_{i1} + k_{i2}[C_{si}]\} \left\{ 1 - \frac{Q_i}{K_i} \right\} \quad (3.1.3)$$

where $k_{i2} = b_{si}k_{i2}'$.

The corresponding rate law for associated aqueous species, w_j , then becomes

$$R_{wj} = \frac{d[C_{wj}]}{dt} = a_i \{k_{i1} + k_{i2}[C_{si}]\} \left\{ 1 - \frac{Q_i}{K_i} \right\} \quad (3.1.4)$$

$$R_{wj} = a_i \frac{d[C_{si}]}{dt} \quad (3.1.5)$$

where

a_i	= moles of species in one mole of solid
$[C_{wj}]$	= molality of aqueous species, j

Values of K_i and Q_i are calculated by the equilibrium chemical modeling portion of the modified TEMPEST at every time step when chemical reactions are simulated. Thus these values are changing with time during the simulation period.

3.1.2 Data Requirements for Chemical Modeling

Two types of data are required for the chemical modeling: first, thermodynamic data are required for relevant aqueous, solid, and gaseous chemical reactions. Specifically these data are

- standard chemical potential with temperature

- Pitzer ion-interaction parameters for binary systems
- Pitzer ion-interaction parameters for common ion ternary systems
- Pitzer ion-interaction parameters for neutral species
- parameters (charges of zero and b planes) used for the triple-layer, site-binding adsorption model
- Henry's Law constant.

The second type is model input of chemical and physical data for tank conditions:

- initial chemical conditions of a waste
 - names of potential chemically reactive aqueous, solid, and gaseous species
 - molality and density of solids
 - molality of individual aqueous species or total aqueous species
 - temperature and pressure
- kinetic reaction rates (if kinetic reactions are also modeled)
 - zero-th order kinetic rate, k_{i1}
 - the first-order kinetic rate, k_{i2} .

3.1.3 Chemical Modeling

The chemical modeling will identify and determine amounts of chemical species of aqueous, solid, and gaseous phases under various temperature and waste compositions. It also predicts the total mass of aqueous and solid chemicals. By reflecting various chemical reactions of aqueous and solid phases, the predicted solid mass will then be converted to a solid volume fraction with known densities of solids under a variety of temperatures and waste compositions. The solid volume fraction is a main parameter for the transport analysis to estimate critical velocity and pressure drop for waste pipeline transport. With some modifications, the chemical code GMIN can also predict liquid density thermodynamically for that condition. Other needed work is expanding the thermodynamic database for chemical reactions relevant to Hanford tank conditions, regardless of the chemical code (or codes) to be used. Furthermore, computer codes (e.g., GMIN and TEMPEST) should be tested with known conditions to validate them under Hanford conditions before applying them to RCSTS waste acceptance.

Three levels of chemical modeling will be performed, depending on the availability of the chemical data. In the first level the chemical information of the tank waste consists of only elemental analysis. In this case, chemical modeling will be conducted to predict potential aqueous chemical species that are in equilibrium conditions with assumed solids. This modeling is potentially the largest uncertainty on the model results. In the second level, aqueous chemical analysis is performed, but the only the elemental analysis of the solids is available. In this case, the chemical modeling will be performed by assuming the particular solids are present in the slurry. In the third level, the chemical species of the solids are identified by measurement (say with TEM), and analytical solution results are available. In this case, the chemical modeling is potentially the most accurate. Thus, one of the characterization efforts will be to obtain these chemical data so the chemical modeling will be accurate.

3.2 Hydrodynamic Transport Analysis

Most of the analysis covered in this section is similar to what was covered in Hudson (1996) and Onishi and Hudson (1996). However, three modifications are considered: 1) pressure drop calculation uses more general correlations proposed for non-Newtonian fluids; 2) critical velocity calculations are based on settling of particles in a generalized Newtonian fluid; 3) a more rigorous approach is used for critical velocity calculations.

3.2.1 Mixture Density and Concentration

The current analysis requires that we separate the solid particles into two different groups: 1) those that are part of the carrier fluid (vehicle) or homogeneous fraction, $C_{v, \text{hom}}$, and 2) those large enough to potentially settle during pipeline transport, or the heterogeneous fraction, $C_{v, \text{het}}$. The total solids volume fraction is the sum of (1) and (2). The homogeneous fraction is expected to increase the viscosity and density of the mixture, as will be discussed further in the following sections. For particle settling calculations or for calculation of the critical velocity, the following mixture density will be used:

$$\rho_M = \rho_L \left[1 + C_{v, \text{hom}} \left(\frac{\rho_s}{\rho_L} - 1 \right) \right] \quad (3.2.1)$$

In term of mass fraction of solids, Equation 3.2.1 changes to

$$\rho_M = \frac{\rho_L}{\left[1 - \chi_{s, \text{hom}} \left(1 - \frac{\rho_L}{\rho_s} \right) \right]} \quad (3.2.2)$$

3.2.2 Dependence of Viscosity on Solids Loading

Definition of viscosity in a solid-liquid slurry such as a ceramic suspension is more complex. The viscosity of a mixture is a function of the continuous phase viscosity, electrolyte concentration and ionic charge, the dispersed phase particle size distribution, particle surface properties, number density of solid particles, and the form of particle-particle and fluid-particle interactions.

Literature suggests that the key parameter affecting a suspension viscosity is the concentration of solid particles. This relationship was first modeled by Einstein (1906) who proposed that the relative viscosity of the mixture changes linearly with solids concentration:

$$\mu_r = \frac{\mu_s}{\mu_l} = 1 + 2.5\phi \quad (3.2.3)$$

where ϕ is the solids volume fraction, μ_s is the slurry viscosity, and μ_l is the viscosity of the continuous phase liquid. It has been shown that this relationship works well for a dilute suspension of hard, spherical, and noninteracting particles (Rutgers 1962).

For most slurries of interest, the concentration of solid particles is high enough to promote collision and strong hydrodynamic coupling between the particles. At low to moderate solids loading, the viscosity increase is due to fluid volume exclusion. The presence of the particles that cannot participate in gradual shearing increases the effective shearing rate between the particles, and, as a result, a higher dissipation rate takes place. At high concentrations, solid particles tend to interact at an even closer range, further increasing the shear resistance and energy dissipation.

Several investigators have developed empirical correlations (Shook and Roco 1991) as well as derived models based on theoretical arguments (Frankel and Acrivos 1967; Brady and Bossis 1988; Shapiro and Probstein 1992; Chang and Powell 1993). A commonly used two-parameter empirical model is the Krieger-Dougherty equation (Barnes and Holbrook 1993):

$$\mu_r = (1 - \phi/\phi_m)^{-B\phi_m} \quad (3.2.4)$$

where ϕ_m is the maximum packing fraction and B is the intrinsic viscosity, which is 2.5 for spherical particles. Frankel and Acrivos (1967) derived the functional dependence of viscosity on concentration by modeling the flow between the gaps of neighboring particles and the associated increase in energy dissipation:

$$\mu_r \approx \frac{9}{8} \left\{ \frac{(\phi/\phi_m)^{1/3}}{1 - (\phi/\phi_m)^{1/3}} \right\}, \quad (\phi/\phi_m) \rightarrow 1 \quad (3.2.5)$$

The performance of Equation (3.2.5) was shown to improve at the asymptotic limit of maximum packing of monodisperse solid spheres. Chong et al. (1971), based on extensive investigation of rheological behavior of multimodal suspensions of glass spheres, proposed the following equation:

$$\mu_p = \left[1 + 0.75 \left(\frac{\phi/\phi_m}{1 - \phi/\phi_m} \right) \right]^2 \quad (3.2.6)$$

where again, ϕ_m is the maximum packing fraction, which, besides the physical geometrical arrangement (staggered or aligned lattice structure), depends on the size distribution and modality of the particles. Two parameters that have been cited in the literature are size ratio, λ (the ratio of the largest to smallest particle diameters), and volume fraction of the smaller particles, ξ (Chang and Powell 1993). It has been shown that ϕ_m is approximately 0.74 for a monomodal suspension of spherical particles, although the experimental values are usually below 0.7. As λ or ξ increase, the maximum packing fraction, ϕ_m , increases above the monodisperse maximum packing fraction. Experiments have shown that the relative viscosity at high-volume fractions is directly linked to the maximum packing fraction associated with a particular size ratio, λ , and small sphere volume fraction, ξ (Shapiro and Probstein 1992). Both experimental data and computational results show that the maximum viscosity reduction occurs at $\xi \sim 0.25$ to 0.35 (Shapiro and Probstein 1992, Chang and Powell 1993). Chang and Powell showed that this is the range within which the mean cluster size (or agglomerate size) is the minimum.

Side-by-side comparison of various correlations (Figure 3.1) shows that the correlations provided by Krieger and Dougherty match very well with that of Chong et al. (1971). However, the correlation by Frankel and Acrivos (1967) tend to under-predict the relative viscosity at high concentrations, and over-predict the viscosity at the intermediate concentrations. Thus the method of Chong et al. (Equation 3.2.6) will be adopted for this purpose, which is expected to work well when a broad size distribution exists, such as in the Hanford tank wastes. The parameter ϕ_m , or the maximum packing fraction, is determined for each specific waste from the characterization tests, as will be described in the following section. Equation (3.2.6) is not intended as a stand-alone modeling of the viscosity, and its use is perceived to eliminate the need to perform extensive characterization of the slurry at various concentrations. Instead, the slurry viscosity is measured at a few concentrations, and the data points are fit into the correlation of Equation (3.2.6).

Figure 3.1. Comparison Between the Various Correlations and Experimental Data for SY-102 Sludge (Onishi and Hudson 1996)

3.2.3 Density of Solid Particles

The solids in the waste slurry may range from large particles that settle independently to colloidal particles, which network to form porous aggregates. The colloidal aggregate density used for settling calculations is the composite of both the solid particles and interstitial liquid densities within the aggregate. The range of settling densities is determined by allowing the slurry to settle and the different solid forms to separate. The density of the large particles is obtained by characterizing a sample taken from the bottom of the sediment. A rough estimate of the aggregate density is obtained by measuring the mixture density of the top layer of the settled slurry after removing the drainable liquid.

3.2.4 Critical Velocity

Transporting solids by fluids in pipelines is used to handle raw materials, materials in process, and finished products in various industries, including agriculture, paper, oil, food, chemicals and mining. Common applications include conveyance of coal and ores; disposal of tailings, dredged sediment, and other waste products; solid waste and cement slurries, and pneumatic conveyance of grains (Vanoni 1975). There are four regimes of solid-fluid mixture transport: homogeneous flow, heterogeneous flow, saltation, and stationary bed flow. Because it gives the largest amount of solids transport per unit energy expended, the heterogeneous flow is normally the most economical and the one most often used in the slurry operation.

Because it is so important, extensive research efforts (e.g., Durand 1953; Zandi and Govatos 1967; Shook 1969; Hanks and Sloan 1981; Wani et al. 1982) have been concentrated on this heterogeneous flow regime. There are several review books and papers on the critical velocity above which all solid particles are in suspension and pressure loss (Govier and Aziz 1972; Shook and Roco 1991; Vanoni 1975; Wani 1986). Turian et al. (1987) also provide a rather comprehensive review of the correlations commonly used for estimating the critical velocity, including the well-known correlation by Durand and the modified Durand correlation proposed by Zandi and Govatos (1967). However, the Zandi and Govatos formula was developed for relatively coarse solid particles, and its accuracy is no better than the Durand formula (Vanoni 1975; Wani 1986). Hudson (1996) recognized the limitations of his proposed approach and presents a calculation based on the correlation proposed by Zandi and Govatos.

There are no generally accepted criteria/formulas to describe critical velocity and pressure loss. This is especially true for non-Newtonian flow, since most of the past studies were developed for Newtonian flows having relatively large (upper tens, hundreds, and thousands of microns) solid particles. Since many of Hanford sludges contain large fractions of fine (submicrons to low tens of microns) particles (Onishi et al. 1996a), we recommend, at least initially, using the following three empirical correlation formulas to estimate the critical velocity, above which all solid particles will be in suspension (either as a homogeneous or heterogeneous vertical distribution): Oroskar and Turian (1980) and Wani et al. (1982) for Newtonian flows; and Hanks (1986), based on a Wasp et al. (1963) concept for non-Newtonian flows. The Hanks model, similar to the Wasp's model, requires iteration of computation.

The two Oroskar and Turian (1980) formulas provide generally accurate predictions of critical velocity for narrowly varying particle sizes (Wani 1986). The Wani et al. (1982) equation was developed for slurries with widely varying particle sizes and thus provides reasonably accurate predictions for these conditions. Errors expressed by root-mean-square values from experimentally measured critical velocities for various pipe diameters and particle sizes are 59% for Zandi and Govatos, 52% for Durand, 50% for Wasp et al., 26 and 22% for Oroskar and Turian, and 15 and 22% for correlation models of Wani et al., reported in Wani (1986).

However, the applicabilities of Oroskar and Turian and Wani et al. models to non-Newtonian flow cases are not well established. Introducing the concept of "vehicle," a homogeneous flow with fine particles carrying large particles with a heterogeneous distribution, Wasp (1963) provided a way to handle a mixture of fine and coarse solid particles, in which fine particles are mixed homogeneously and produce a non-Newtonian flow, and coarse particles are mixed heterogeneously in this homogeneous mixture of fine particles and liquid. The Hanks model is a further improvement on the vehicle concept and was applied to wastes from Oak Ridge National Laboratory. We describe these three correlation models below.

3.2.4.1 The Oroskar and Turian Model

Oroskar and Turian (1980) developed two correlation models, one partially based on a theory (Equation 3.2.7) and the other (Equation 3.2.8) purely based on a regression analysis of available data (Shook and Roco 1991; Wani 1986). We recommend Equation 3.2.7.

$$V = [5C_v(1 - C_v)^{2n-1} \left(\frac{D}{d} \right) \left(\frac{D\rho_L \sqrt{gd(s-1)}}{\mu_L} \right)^{\frac{1}{8}} \frac{1}{x}]^{\frac{8}{15}} \sqrt{gd(s-1)} \quad (3.2.7)$$

$$\frac{V}{\sqrt{gd(s-1)}} = 1.85C_v^{0.1536} (1 - C_v)^{0.3564} \left(\frac{D}{d} \right)^{0.378} N_{Re}^{0.09} x^{0.3} \quad (3.2.8)$$

where

- C_v = solid volume fraction
- D = pipe diameter
- d = solid diameter
- g = gravitational acceleration
- n = hindered settling velocity exponential constant as a function of pipeflow Reynold's number, N_{Re}
- N_{Re} = Pipeflow Reynold's number defined and $= DV\rho_L/\mu_L$
- s = density ratio of solid to liquid ($= \rho_s/\rho_L$)
- V = critical velocity, above which all solids are in suspension
- μ_L = dynamic viscosity of liquid
- ρ_L = liquid density
- ρ_s = solid density.

The hindered setting velocity, u_s , is calculated as

$$u_s = u_t(1 - C_v)^n \quad (3.2.9)$$

and the factor, x , is given by

$$x = \frac{2}{\sqrt{\pi}} \left[\frac{2}{\sqrt{\pi}} \gamma \exp\left(-\frac{4\gamma^2}{\pi}\right) + \int_{\gamma}^{\infty} \exp\left(-\frac{4\gamma^2}{\pi}\right) d\gamma \right] \quad (3.2.10)$$

where

- u_t = an unhindered particle setting velocity
- u_s = hindered setting velocity
- γ = ratio of hindered particle settling velocity to the critical velocity (u_s/V).

3.2.4.2 The Wani Model

Wani et al. (1982) developed a series of correlation models. Based on the comparisons of predictions and measured critical velocities, we recommend using the following two formulas depending on the pipeline Reynold's number, N_{Re} :

For the Stokes range ($N_{Re} < 0.1$)

$$\frac{V^2}{gd(s-1)} = 2.3 \times 10^{-4} N_{Re}^{0.27} C_{ov}^{0.973} C^{1.67} C_v^{0.307} \quad (3.2.11)$$

For the intermediate range ($0.1 < N_{Re} < 10$)

$$\frac{V^2}{gd(s-1)} = 7.7 \times 10^{-6} N_{Re}^{0.0014} C_{ov}^{-1.25} C^{4.77} C_v^{0.272} \quad (3.2.12)$$

where

- C = Hazen-Williams pipe roughness factor
- C_{ov} = coefficient of variation = $100/d_w$
- C_v = solid concentration (volume)
- d_w = weighted mean particle diameter.

3.2.4.3 The Wasp Model

The correlations are much more scarce for turbulent flow of a slurry containing a significant amount of colloidal particles. Hanks (1986) provides some of the highlights of the issues that exist in this case. The colloidal suspension in the first place results in a broader size distribution that could potentially span from the primary particles in the nanometer scale to agglomerates in the order of several hundred microns. The difficulty is that the smaller particles contribute to changing the rheology while the larger particles slip. To improve the previous methods, Wasp (1963, 1977) introduced the concept of "vehicle," which is the portion of the slurry that remains homogeneous.

Youngblood et al. (1994) and Hylton et al. (1994) at Oak Ridge National Laboratory (ORNL) performed a series of tests using simulated waste for the ORNL Melton Valley Storage Tanks and Hanford single-shell tanks over a wide range of solids concentration and temperature. Almost all the fluids tested had a reported yield stress. They were unable to find a minimum or critical velocity at which the solids would settle. The calculations based on the procedure developed by Hanks (1986) agreed closely with the experimental data for all the different test conditions and fluid rheology. In one set of tests, where large, 500-micron solid glass beads were used as the heterogeneous solids, they observed slip. The critical velocity in

this case was in the laminar flow regime. Based on these observations of the most relevant set of results, we concluded that a critical velocity for a fluid with yield stress is of no consequence. As a result, as far as the critical velocity calculations is concerned, we will only consider the power law fluids. The Newtonian rheology is one of the special cases of this model.

One form of constitutive equation for a yield power law fluid, known as the Herschel-Bulkley equation, may be written as

$$\tau = \tau_y + K\dot{\gamma}^n \quad (3.2.13)$$

where τ is the shear stress (Pa), τ_y is the yield stress, K is the consistency factor (Pa.sⁿ), $\dot{\gamma}$ is the shear rate (1/s), and n is the behavior index. For flow of a yield power law fluid the most widely accepted Reynold number is defined as

$$Re_p = \frac{\rho_M d^n V^{2-n}}{8^{n-1} K} \quad (3.2.14)$$

where V is the velocity, d is the characteristic dimension, and ρ_M is the mixture density. This definition is different from Hudson's (1996) in two ways: 1) the mixture density is used, and 2) the viscosity includes a factor of 8^{n-1} , which, if ignored, causes the Reynold number to be under-predicted and the drag coefficient over-predicted. On the basis of drag coefficient alone the critical velocity would then be under-predicted. However, other parameters come into play for critical velocity calculations, which will be discussed in the following sections.

Several investigators have reported that the definition of Stoke's drag coefficient for a non-Newtonian fluid would remain the same as that for a Newtonian fluid provided that a new parameter is used instead of a Reynold number (Hanks 1986). For a power law fluid, the definition provided in Equation (3.2.15) is the most appropriate:

$$C_D = \frac{24}{Re_p} \quad (3.2.15)$$

Re_p is the particle Reynold number found by using the settling velocity and diameter of the larger particles. The settling velocity of the particles may be found from the definition of drag coefficient combined with Equations (3.2.8) and (3.2.9):

$$V_s = \left[\frac{g \Delta \rho d^{1+n}}{18 (8^{n-1}) K} \right]^{1/n} \quad (3.2.16)$$

The line of demarcation for the particle size distribution (below which the particles are assumed as being a part of the "vehicle" or remain homogeneous within the carrier fluid mixture and above which the particles are assumed to be settling) is part of the decision that must be

made based on an empirical settling criterion. For example, Youngblood et al. (1994), use the computer program YPLPIPE developed by Hanks (1978) to compute the critical velocity. However, the decision on what portion of the solids contribute to the "vehicle" and what portion is heterogeneous is performed manually by analyzing the particle size distribution (PSD). Their model is based on an 85% criterion, that is, the critical diameter for the calculation is the 85th percentile. Hudson (1996) uses an 80th percentile as the representative particle size for the settling calculations. It appears that the largest size found in the PSD is the most logical particle diameter for these calculations, especially since Equation (3.2.16) reveals that the dependence of settling velocity on particle diameter is as shown below:

$$V_s \propto d^{1+\frac{1}{n}} \quad (3.2.17)$$

Equation 3.2.17 suggests that if the behavior index is 0.5, which is not unreasonable for the tank wastes at Hanford at volume fractions higher than 5%, the settling velocity increases with the cube of the diameter. For Newtonian fluids this relationship is diameter to the second power. Therefore, the larger particles will tend to settle faster than expected. Based on this argument, the criterion for critical velocity calculations will be based on the 90th percentile (Vanoni 1975). The density and viscosity of the carrier mixture would then be, for all practical purposes, what is measured, including most of the solids. Some modification to the viscosity will be incorporated based on Equation (3.2.6). This approach simplifies the characterization process considerably.

There are few actual correlations for calculating deposition critical velocity that have been specifically developed and tested for non-Newtonian fluids. Although Hanks and Sloan (1981) provide a fairly sophisticated methodology for computing the critical velocity for a generalized non-Newtonian slurry, their approach uses many of the same principles on turbulence in a Newtonian fluid. Given that turbulence in non-Newtonian pipe flows is considerably different than in their Newtonian counterparts (Park et al. 1989), such mixing of the Newtonian turbulence with non-Newtonian rheology will not necessarily improve the outcome of the correlations. Therefore, no added benefits are perceived by using their method here.

Hanks (1986) demonstrates that the Durand model, which is very similar to what Hudson (1996) uses, grossly over-predicts the pressure drop and critical velocity in the pipe. He showed that the Wasp model (1977) of vehicle, as discussed earlier, more closely predicts the flow characteristics. Thus we consider the vehicle approach, which is somewhat similar to the notion of hindered settling, to be the best method available. That is essentially why the mixture rheology and density were used for calculating the particle drag coefficient and settling velocity.

3.2.5 Pressure Drop Calculations

To calculate the pressure drop for heterogeneous slurries, that is, for slurries where some or all of the solid particles are treated as a separate phase, as discussed in the previous section. The increase in the head loss due to the presence of solid particles is of interest:

$$i = i_L (1 + \phi C_{v,het}) \quad (3.2.18)$$

In Equation (3.2.18), the parameter f is the dimensionless excess head loss, i_L is the head loss in the vehicle or the homogeneous (particle-free) fluid, and $C_{v,het}$ is the solids volume fraction as defined in Section 3.2.1. In addition to the calculation of the head loss using an appropriate correlation, as will be discussed later, a correlation needs to be used for predicting the dimensionless excess head loss as a function of the characteristics of the heterogeneous phase. Some discussion on this topic was already covered in Section 3.2.3 in relation to the critical velocity calculations. In this section, we concern ourselves only with calculations of the homogeneous phase pressure drop.

Two different friction factors are commonly used for calculating the head loss in a pipe. To avoid confusion, we will provide these two definitions and choose one for the subsequent calculations. Fanning friction factor is related to the wall shear stress in the pipe:

$$f = \frac{\tau_w}{\frac{1}{2} \rho_M V^2} \quad (3.2.19)$$

where τ_w is the wall shear stress and V is the mean velocity in the pipe. Darcy friction factor is linked to the pressure drop across a known length of the pipe:

$$f_D = \frac{\Delta p}{\frac{1}{2} \rho_M V^2} \quad (3.2.20)$$

It is not too difficult to verify that the relation between Darcy and Fanning friction factors is governed by the following equation:

$$f_D = 4f \quad (3.2.21)$$

Among several correlations proposed by various investigators (Shook and Roco 1991), the most general correlation that holds for power law, Bingham or yield power law fluids, is the relationship recommended by Torrance (1963):

$$\frac{1}{\sqrt{f}} = \frac{2.687}{n} - 2.949 + \left(\frac{1.966}{n} \right) \ln \left[(1 - \zeta) \text{Re}_p f^{1-n/2} \right] + \left(\frac{0.682}{n} \right) (5n-8) \quad (3.2.22)$$

where f is defined in Equation (3.2.19), Re_p is defined in Equation (3.2.14), and $\zeta \equiv \tau_y / \tau_w$. Note that $\zeta = 0$ for a power law fluid. Equation (3.2.22) is used for turbulent flow in a smooth pipe and is solved iteratively in conjunction with Equation (3.2.19). A simple root-finding technique such as the Newton-Raphson method can be used for this purpose. Although alternative correlations are proposed for rough pipes, it has been shown that at sufficiently high Reynold numbers ($\text{Re}_p > 3000$) only slight variations in the friction factor have been observed

(Shook and Roco 1991). Further, the change in the friction factor, if any, is usually in the conservative direction. Thus it is sufficient to use Equation (3.2.22) to calculate pressure drop in the pipe.

Other losses exist in the system that result from fittings, bends, and hydraulic components (valves, flow meters, etc.) and will be covered under a "minor losses" correction factor. Although approximate correction factors exist for simple fluids in terms of equivalent length of the pipe, such correction factors are even more approximate for slurries. As a result, the zero-th order approximation will assume that losses through the pipe are dominant and that negligible minor losses exist. A first-order correction may be made based on the in-field validation test results (described in Section 5).

4.0 Characterization

As discussed in Section 2, a set of tank-specific characterization tests will be performed to meet the retrieval and transport needs. The parameters needed from characterization are listed in this section. However, the specific characterization needs will be decided for each tank based on what is available from the tank characterization reports. The results will be issued formally in a slurry transport characterization report (STCR).

4.1 Property Measurement Approach

The goal of the transport analysis is to provide a technical basis for the decision about what are the safe conditions for operation of the RCSTS. Some of the data quality objectives relevant to this assessment are described in Fowler (1995) and Bloom and Nguyen (1995). Because the properties of this slurry can vary with temperature and constituent concentrations, the characterization function should provide information on the quantities and their expected variations. The transport analysis will be performed such that a range of expected operating conditions can be obtained based on the expected variation in the properties. This expected operating range of the system will, in turn, be matched against the system capacity to ensure safe and trouble-free performance. Data will be gathered on physical properties and chemical properties and constituencies.

4.2 Physical Properties

Physical property measurements are required for each of the following: liquid density, mixture density, solids mass fraction, density of centrifuged solids, mass fraction water of the centrifuged solids and a particle size distribution. While there is some redundancy of information in these requirements, they can be used to determine the overall uncertainty associated with the characterization.

4.2.1 Density Measurements

Measurement of the density is somewhat complicated by the fact the solid particles are agglomerates (contain interstitial liquid) and the shape, size, and amount of these solids changes as a function of the amount of precipitation and dissolution, as described in Section 3. The steps used to clarify the density measurements are described in the following subsections.

4.2.1.1 Mixture Density

This mixture contains three different components, drainable liquid, interstitial liquid, and solids. Mixture or bulk density of the proposed slurry will be obtained by a suitable analytical method at the endpoints of the temperature range indicated in the STPR. These data will be used to validate the modeling results. The actual mixture density at the operating conditions is predicted from the models described in Section 3.1.

4.2.1.2 Drainable Liquid Density

Measurement of the drainable liquid density, ρ_L , from the proposed slurry will be obtained by a suitable analytical method. The drainable liquid is obtained after allowing the solid particles to settle. The supernatant liquid will be considered as the drainable liquid. This parameter is commonly available from tank characterization reports (TCR). However, this data are commonly provided at room temperature, and it is believed that the density of a given carrier liquid varies as a function of temperature (see Section 3.1 for explanation). Thus, to the extent possible, filtration and density measurements will be obtained for the endpoints of the temperature range indicated in the STPR. These data would help validate the information obtained from the chemical modeling, as described in Section 3.1.

4.2.1.3 Density of Centrifuged Solids

After removing the drainable liquid from the top of the settled bed, two samples will be obtained from the settled bed—one from the top and another from the bottom. The mixtures of solids and interstitial liquid will then be centrifuged to the highest rpm achievable in the centrifuge. Then the density of the centrifuged solids will be determined by helium pycnometry. This solid density will be assumed to be the density of the solid particles at the maximum packing fraction, ϕ_m , where the interstitial space is filled with liquid at the density of ρ_L . These data can be used to calculate the solids density, ρ_s , used in the analysis.

4.2.1.4 Density and Mass Fraction of Interstitial Liquid

The density of liquid obtained from the centrifuged solids will be determined using the same method employed in Section 4.2.1.3. The mass fraction of this liquid can also be determined and can be used for determining the density of solid particles or agglomerates, as described in Section 3.2.3.

4.2.2 Solids Mass Fraction

The solids mass fraction will be determined by thermal gravimetric analysis of the centrifuge filtered solids, or by a similar method, such that the errors associated with residual liquids are minimized. Measurements will be obtained at the endpoints of the temperature range. These measurements will then be used for validating the analysis of Section 3.1.

4.2.3 Particle Size Distribution

Particle size distribution measurement is required to determine the settling rates and transport conditions. The focus of this measurement is the larger-size components of the slurry. However, it is expected that due to friability, the particle diameter will shift toward a smaller size distribution during cross-site transfer. However, the somewhat conservative approach, without loss of generality, is to assume no degradation of particles occurs during transfer. Any technique capable of detecting particles larger than 1 μm (such as light obscuration or sedimentation-based methods) that provides a size distribution weighted by volume (or mass) would be suitable for this determination. The reported result of this test is a plot of the probability distribution for particle size weighted by volume (or mass).

4.2.4 Rheological Properties

A great deal of emphasis is placed on the rheological characterization because of the impact these variables have on the transport analysis. The two set of quantities required from the rheological characterization are the liquid viscosity and the parameters that characterize the mixture rheology.

4.2.4.1 Liquid Viscosity

Drainable liquids will be characterized for viscosity at the endpoints of the temperature range. This liquid is most likely Newtonian. Therefore, the viscosity at limited range of shear rates will be measured (1 to 100 s⁻¹). The viscometric configuration must be capable of accurately measuring fluid viscosities from approximately 0.0003 Pa-s (0.3 cP) to 0.05 Pa-s (50 cP). Because the transport analysis can be very sensitive to variations in this quantity, errors in this measurement should be no larger than 10% of the reading.

4.2.4.2 Mixture Rheology

Three sets of rheological tests will be performed (see details in Onishi and Hudson 1996): 1) controlled rate experiments, where the shear stress as a function of shear rate is determined (flow curve); 2) time-dependent viscosity experiments, where the changes in viscosity as a function of time for different shear rates are measured (viscosity-time curve); 3) oscillatory experiments, where the amplitude of storage and loss terms of the complex shear modulus as a function of small amplitude oscillation frequency are measured. These tests will cover a range of dilutions with an appropriate diluent, defined according to the waste type and preliminary modeling of Section 3.1.

These experiments are designed to 1) provide the range of viscosity obtained during a steady-state shearing process in the pipeline, 2) provide information suitable to obtain a constitutive relation similar to that of Equation (3.2.7), and 3) provide information on whether the mixture will be rheopectic or thixotropic.

If the mixture is found to be rheopectic to the point that the viscosity is found to continually increase with shearing time (Onishi and Hudson 1996), the proposed slurry will be rejected and declared unacceptable. An alternative slurry will be identified and proposed based on laboratory studies on dilution.

If the solution is thixotropic, that is, its viscosity drops as a function of shearing time, the slurry will be acceptable. The maximum viscosity detected in the measurements will be used for pressure drop measurements. The minimum viscosity, detected during a shearing time equivalent to the expected transient time of the waste in the RCSTS, will be used to calculate the critical velocity.

4.3 Chemical Properties and Constituencies

The analyses of chemical properties largely follow the requirements of the Waste Compatibility Data Quality Objectives (Fowler 1995). The information from these tests, along with the variations in physical properties associated with the specified temperature range, are used to determine when problematic phase changes might occur. As noted in Section 3.1, the

chemical modeling accuracy will increase as the following three chemical analyses are conducted. Many TCRs include at least elemental analysis results, and some include an ion analysis, but they rarely include information on a solid chemical analysis.

4.3.1 Elemental Analysis

An elemental analysis for solids will be performed by inductively couple plasma (ICP) or other suitable method.

4.3.2 Ion Analysis

Ion chromatography (IC) will be used to measure the quantity of ionic constituents in the solid and liquid phases. Analyses will be performed at the endpoints of the temperature range. These results will be used to improve or confirm the chemical modeling results, as discussed in Section 3.1.

4.3.3 Solid Chemical Analysis

Transmission electron microscopy (TEM) will be used to identify major solids in the sludge. The results will be used to improve or confirm chemical modeling results, as discussed in Section 3.1.

4.4 The Slurry Transport Characterization Record (STCR)

The product of the characterization effort is the Slurry Transport Characterization Report (STCR). This report contains all of the characterization information needed for the transport analysis. If certain tests described above were not performed because the information was already available, the report will also include the references to the prior characterization efforts from which the information was taken. The report should provide sufficient detail that the control function will be able to verify that the test results are meaningful and applicable to the transport analysis. Where appropriate, appendixes should be provided that present the raw data (and their uncertainties) from which the final data are derived.

5.0 Validation of Methodology

As mentioned in Section 2, two types of analyses will be required to ensure successful predictive analysis for establishing the operating conditions and for monitoring or control analysis during the field operation. The predictive analysis was described in Section 3. This section describes the procedure that is proposed for validation of the methodology and tank-specific slurry transport waste acceptance criteria.

5.1 Predictive Analysis Validation

The purpose of this validation methodology is to ensure that the calculations performed *a priori* will closely match the actual operating conditions, say within a certain acceptable variance or uncertainty range. For validation of the analysis methodology, three different sets of tests can be performed: 1) sensitivity analyses to determine important parameters and their effects on the waste transferability assessment, 2) laboratory tests using a set of surrogate slurries, and 3) field tests using the actual wastes prior to transport. The following validation/evaluation process describes the actual waste validation methodology.

Two types of sensitivity analyses will be performed. First, sensitivity analyses of the models/formulations described in Section 3 will be performed to determine, in general terms, important processes and parameters on tank waste cross-site transfer before these methodologies are used for RCSTS waste acceptance determination. These results will direct characterization efforts (see Section 3) to address 1) the chemical and physical processes/parameters that will require extra care in their determination, and 2) the potential uncertainty of the waste transferability assessment. The second type of sensitivity analysis will be performed for each waste composition selected for transfer to evaluate the potential error bounds of the assessment, so that this degree of uncertainty will be built into the determination of the RCSTS operating conditions for each specific tank waste.

Since it is very difficult to make surrogate slurries for laboratory testing with all the chemical and physical properties, tailored laboratory tests may be conducted with simulated slurries under conditions known to cause significant changes in chemical and physical property/rheology (e.g., formation of boehmite, high Bi-P mixture, hydrate sodium phosphates at various temperatures). These experiments will be designed to address specific questions and to provide data for testing the viability of the analysis methodologies presented in Section 2. A few very carefully selected scale models with proper physical and chemical properties may be useful for validating the methodologies.

The field tests will use data from three different sources: 1) in-tank recirculation loop, 2) in-farm recirculation loop, and 3) pipe section up to the first booster pump. The pressure drop and flow rate are measured during tests 1 and 2 for various flow rate conditions. The solids volume fraction is inferred from the in-tank mixing and dilution information. The target is to use the in-tank recirculation loop for building the pump performance curves, the in-farm recirculation loop for measuring the pressure drop over a known length of the pipe, and the pipe section up to the first booster pump for verification of the in-farm recirculation loop data and a redundancy check.

The pump performance curves provided by pump manufacturers are commonly produced by using water as a working fluid. Some adjustment to these curves is commonly

recommended for fluid viscosities higher than water. However, the true characteristics of the pump (pressure head, H , versus flow rate, Q , and pump speed, N) may be quite different depending on the rheological properties, size distribution of the solid particles, and the solids loading of the actual slurry used (Shook and Roco 1991). Such slurry-dependent performance curves may be constructed by using the in-tank recirculation loop and are expected to be similar to the curves N_1 , N_2 , and N_3 shown in Figure 5.1. The curve S_1 is the system characteristic curve whose intersection with the pump performance curve represents the expected operating point of the system. For example, if the pump is operated at N_2 , then the operating point would be at OP in Figure 5.1. The angle formed between the pump performance curve and system characteristic curve, α , is commonly used as a measure of stability in system operation. The larger this angle, the more stable the operating conditions are expected to be. Note that as the operating point approaches the critical velocity ($Q < Q_{\min}$), the angle α becomes smaller and may change to negative, at which point the system performance becomes unstable. It is thus clear that optimization of the operating point depends strongly on the pump performance as well as the system characteristic curves making in more critical to perform in-field validation tests.

The shaded area on this figure represents the safe operating envelope for the system. The boundaries of this envelope are constrained by the minimum flow rate (to maintain a velocity above the critical velocity), maximum flow rate allowed (based on pump and pipe erosion, cavitation, and other factors), maximum available pressure head by the pump, and minimum pump speed (based on pump efficiency and performance). The in-field recirculation tests will be used to generate the initial system characteristic curve and determine whether the operating point lies within the desired operating envelope. Some flexibility exists in adjusting the pump speed to maintain the operating point within this envelope.

Figure 5.1. Conceptual Plot of Pump Performance Curves Versus the System Characteristic Curve (OP represents the desirable operating point of the system)

At the operating flow rate, the pressure drop for each condition (in-tank, in-farm, etc.) will be measured. The pressure drop is then plotted versus the equivalent length of the pipe for each given test condition. Some uncertainty in the pressure drop and equivalent length of the pipe will exist, although the latter may be more dominant. A conceptual plot is provided in Figure 5.1 as an example. The subscripts IT, IF, and BP stand for items 1 through 3 described above. The solid line on this figure is the result of the predictive analysis. If the predictive analysis adequately calculates the pressure drops, then the solid line overlaps with the data points, as shown in Figure 5.2. In that case, the expected pressure drop across the entire cross-site transfer system, Δp_{RCSTS} , can be predicted from the analysis model. As long as Δp_{RCSTS} is smaller than the pressure head available from the pump (divided by the specified safety factor), then trouble-free operation is expected.

Another advantage of performing these validation tests is to enable selection of the most suitable model and correlation among those presented in Section 2 (e.g., critical velocity formulas). After performing several of these tests for different waste types, the correlations whose data and analysis results do not match closely will be eliminated.

5.2 Control Analysis

The purpose of this analysis is to provide a real-time monitoring approach of the operation of the transport pipeline from the beginning of the transfer process. It is expected that such analysis will provide the means to prevent the onset of flow blockage in the pipeline. A brief description of the control analysis is provided in this section. However, since this step is

Figure 5.2. Conceptual Plot of Validation Curves for Pressure Drop Versus Equivalent Pipe Length (the solid line represents the analysis results)

under development, more detailed description of the control analysis procedure will be covered in a separate report after the control analysis framework has been fully established and the analysis code has been completed.

This type of analysis can be made rather simple using only the data obtained during operation of the RCSTS. One procedure is as follows: the pressure drop across the length of the RCSTS is equivalent to the gauge pressure at the inlet to the RCSTS, since the outlet side is open to the atmospheric pressure at the receiving tank. This pressure can be monitored in real-time and plotted. As the waste is introduced into the RCSTS (initially filled with water or a diluent), this pressure increases due to an increase in the mixture viscosity and dissipation over that of water (Figure 5.3). It is expected that, for trouble-free operation, this pressure increases linearly as a function of time as long as the flow rate remains constant. Using this approach, one can predict how much pressure drop will take place by the time the entire RCSTS is filled with tank waste. In other words, if the pressure drop increases rapidly, as shown by the dotted lines, the pressure drop would possibly exceed what is available. Such a situation may be avoided by dilution or an alternative solution approach, *a priori*. We recommend laboratory validation of this type of analysis.

Figure 5.3. Conceptual Plot of the Control Analysis Output

6.0 Operational and Procedural Control

This section is very similar to that reported in Hudson (1996). We will provide the same information in this report for emphasis.

6.1 The Slurry Transport Waste Acceptance Report

The Slurry Transport Waste Acceptance Report (STWAR) provides the technical bases for safe transfer using the RCSTS, including all applicable controls. Concurrence from signature authorities for this report constitute signature authority for system use within the documented provisions.

6.2 Control and Documentation

The control function authorizes use of the system; thus it is responsible for reviewing and documenting all previous activities. From the viewpoint of the control function, these activities provide the information necessary to build the technical basis for safe transfer using the RCSTS. The technical basis for the proposed slurry transfer, as well as the bases for safety issues resolution, are described in the Slurry Transport Waste Acceptance Report (STWAR) discussed briefly below.

The control function uses the information from the previous activities to specify all controls that will be applicable for the transfer. The set of controls is documented in the STWAR and is unique to the proposed transfer. Typically, these controls will specify the anticipated waste constituency information that was included in the Slurry Transport Proposal Record (STPR), such as the particular solid or slurry waste, the diluent, the diluent ratio, and the temperature range. Controls related to the transport analysis, such as the minimum transport velocity and maximum allowable pressure drop, are also included here. All other required controls related to on-line or in-line measurements, such as mixture viscosity, pressure loss, and solids mass fraction, will also be included.

6.3 Authority

The control function is also responsible for gaining (and giving) signature authority for use of the RCSTS. This function may also specify controls that differ from those recommended by the transport analysis. Where needed, this can be accomplished by increasing the safety margin to decrease the risk of system loss or safety incident.

6.4 Process Monitoring and Systems Support

Once signature authority is gained, the control function will provide procedural and systems support for the transfer, including monitoring the transfer process, providing procedural and systems support for contingencies such as out-of-specification occurrences, and integrating the activity with retrieval and receiver tank operations.

6.5 Control Analysis

Please see Section 5.2 for a description of the detection methodology and its validation. This type of control analysis may be used in conjunction with an analysis similar to that provided in Section 3 during pipeline operation to determine the most suitable next best set of operational conditions if an unacceptable operating condition is detected.

7.0 Summary

A methodology has been proposed in this document for predicting and validating the safe operating condition of the replacement cross-site transfer system (RCSTS) pipeline at Hanford. The methodology was focused around predicting and preventing the three main suspected modes of plugging of the existing CSTS: precipitation, gelation, and particle dropout, or settling. The proposed analysis will be an integration of computer modeling and laboratory experiments to build a defensible case for transportability of a proposed slurry composition for a given tank. Such predictive analysis will also be validated by recirculating the actual tank waste, in-tank and in-farm, prior to transport. A summary of the steps required for defining the operating criteria for a transfer is provided below. These steps will be transformed into a "turn-key" analysis package with a clear and simple set of protocols for its use by operating personnel. This methodology will follow in the near future.

The steps for predictive analysis are as follow:

- 1) Accumulate data from all existing TCRs on the particular slurry to be transported from a given tank. The information of interest is described in Section 4 (Characterization) under physical and chemical properties, and the data requirements are provided in Section 3.1.2. Some of the desired information may not be found in the TCRs and will be obtained later.
- 2) Use the chemical properties and constituencies data as input into a computational chemistry code such as GMIN (see Section 3.1.3) to predict aqueous and solid species with their respective mass concentrations under various waste transfer conditions (temperature, dilution ratios, etc.).
- 3) If the solids predicted from the previous step are not among the known problematic solids (BiP, aluminates, etc.) perform a hydrodynamic transport analysis in accordance with in Section 3.2. (Some of the steps are provided below.)
 - 3-1) Convert species mass concentrations to volume fractions and determine density based on characterization data.
 - 3-2) From physical properties data of Section 4, calculate the mixture density (Equation 3.2.2).
 - 3-3) Using Equation (3.2.6) and the data from TCRs, formulate the dependence of viscosity on solids concentration (using maximum packing fraction). If sufficient data are not available, use a zero-th-order approximation of viscosity. Correct as more data become available.
 - 3-4) Determine the density of solid particles (or agglomerates) using the TCR data. Details of this type of calculations are provided in Onishi et al. (1996a). If sufficient data are not available, use an approximate value based on the solids found in TCRs.
 - 3-5) Use the Oroskar and Turian model to calculate the critical velocity, as depicted by Equations (3.2.7) through (3.2.10). Also calculate the critical velocity using the Wani model (Equations 3.2.11 and 3.2.12) and the Wasp model (Equations 3.2.13 through 3.2.16).

- 3-6) Compare the results and determine the average and standard deviation of the critical velocity.
- 3-7) Use Equation (3.2.22) to calculate the friction factor and (3.2.18) to determine the pressure drop in the pipe for a range of velocities higher than the critical velocity.
- 3-8) Based on the data obtained from the two previous steps, 3-6 and 3-7, determine whether the transfer conditions are acceptable.
- 4) If transfer conditions are acceptable for the proposed slurry, evaluate the tank characterization data to see if they include all the necessary data on the proposed slurry. If such data are available, then we are ready for the validation step; otherwise, further characterization is required on the proposed slurry to obtain some limited information on the physical and chemical properties. Once these data are available, repeat steps 2 and 3 to improve the calculation results. Go to step 5 when done with these calculations.
- 5) Perform validation experiments as described in Section 5.1.

8.0 References

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Appendix

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Slurry Transport Waste Acceptance Report (STWAR)

Appendix

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Slurry Transport Waste Acceptance Report (STWAR)

Slurry Transport Waste Acceptance Report for Tank 241-XX-XXX

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