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Simulant Development for Hanford Double-Shell Tank Mixing and Waste Feed Delivery Testing

PA Gauglitz
DN Tran
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September 2012



Pacific Northwest
NATIONAL LABORATORY

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Pacific Northwest National Laboratory
Richland, Washington 99352

Executive Summary

The U.S. Department of Energy Office of River Protection manages the River Protection Project, which has the mission to retrieve and treat the Hanford tank waste for disposal and close the tank farms. Washington River Protection Solutions, LLC, is responsible for a primary objective of this mission: to retrieve and transfer tank waste to the Hanford Waste Treatment and Immobilization Plant (WTP). A mixing and sampling program with four separate demonstrations is currently being conducted to support this objective and also to support activities in a plan for addressing safety concerns identified by the Defense Nuclear Facilities Safety Board related to the ability of the WTP to mix, sample, and transfer fast-settling particles.

Previous studies have documented the objectives, criteria, and selection of nonradioactive simulants for these four demonstrations. The identified simulants include Newtonian suspending liquids with densities and viscosities that span the range expected in waste feed tanks. The identified simulants also include non-Newtonian slurries with Bingham yield stress values that span a range that is expected to bound the Bingham yield stresses of waste in the feed delivery tanks. The previous studies identified candidate materials for the Newtonian and non-Newtonian suspending fluids, but did not provide specific recipes for obtaining the target properties, and information was not available to evaluate the compatibility of the fluids and particles or the potential for salt precipitation at lower temperatures.

The purpose of this study is to prepare small batches of simulants in advance of the demonstrations to determine specific simulant recipes, to evaluate the compatibility of the liquids and particles, and to determine whether the simulants are stable for the potential range of test temperatures. The objective of the testing, which is focused primarily on the Newtonian and non-Newtonian fluids, is to determine the composition of simulant materials that gives the desired density and viscosity or rheological parameters.

Recipes for five Newtonian liquids were developed to match low and high targets for density and viscosity and to match a typical density and typical viscosity target. The recipes were developed using aqueous solutions of sodium thiosulfate or sodium thiosulfate and glycerol to match the density and viscosity targets for four of the five targets. Sodium thiosulfate was the preferred salt because it is nonhazardous and inexpensive. An aqueous solution of sodium bromide, which gives lower viscosities in concentrated solutions, was selected as a preferred material for a high-density/low-viscosity target. The effect of temperature on viscosity was determined for all the solutions; the solutions including glycerol are the most temperature sensitive. All of these solutions were stable (no salt precipitation after about a day) down to 10°C. There was only one liquid/particle compatibility issue observed during the testing, and this was when a specific gibbsite material was added to a solution of glycerol in water (low-density/high-viscosity target). For this mixture, slurries always stayed cloudy during settling and would form settled layers that were difficult to resuspend. This recipe was reformulated by adding 0.1 wt% sodium thiosulfate, which altered the particle behavior, and the settling and resuspension results were much improved.

For the non-Newtonian slurries, simulant recipes were developed using slurries of kaolin clay in water or kaolin clay in sodium thiosulfate solutions. For the kaolin in sodium thiosulfate solutions, the proportions of both the sodium thiosulfate and kaolin were adjusted to obtain slurries with Bingham yield stresses of 1, 3 and 10 Pa having a constant density (matching the high-density Newtonian target). For the kaolin-in-water slurries, the density was not adjusted but was comparable to the low-density

Newtonian target. The effect of temperature on the Bingham yield stress and consistency were determined and slurries were stable (no salt precipitation) down to 10°C.

Acknowledgments

The authors would like to acknowledge K. Pat Lee and Mike Thien of Washington River Protection Solutions for their technical involvement in defining simulant targets and evaluating candidate recipes, and also thank Matthew Ahrendt for his support in providing samples of the simulant materials from the batches obtained for the demonstration tests. The authors also would like to thank Beric Wells for his technical peer review and Maura Zimmerschied for her technical editing.

Acronyms and Abbreviations

DI	deionized
EPK	Edgar plastic kaolin, from Edgar Minerals division of The Feldspar Corporation
g	gram
in	inch
KA	KitchenAid®
m	meter
MB	Magic Bullet®
min	minute
mL	milliliter
Pa	Pascal
PSD	particle size distribution
QA	quality assurance
s	second
SS	stainless steel
VSR	variable speed re-cycling
WTP	Hanford Waste Treatment and Immobilization Plant

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1.0 Introduction

The U.S. Department of Energy Office of River Protection manages the River Protection Project, which has the mission to retrieve and treat the Hanford tank waste for disposal and close the tank farms (Certa et al. 2011). Washington River Protection Solutions, LLC is responsible for a primary objective of this mission: to retrieve and transfer tank waste to the Hanford Waste Treatment and Immobilization Plant. Washington River Protection Solutions is currently conducting a mixing and sampling program to support this objective (Thien and Sexton 2012). The mixing and sampling program is also being conducted to support the activities identified in the implementation plan for addressing comments from the Defense Nuclear Facilities Safety Board on waste mixing (DOE 2011). This program includes four experimental demonstrations that use nonradioactive simulants: a small-scale mixing demonstration, a remote sampler demonstration, scouting studies at the Savannah River National Laboratory, and full-scale transfer pump testing (Thien and Sexton 2012).

Lee et al. (2012) documents the objectives, criteria, and selection of simulants for use in these demonstrations. Further definition of the simulant targets and materials for specific testing is given in individual test plans (Lee 2012a, 2012b, 2012c). The identified simulants include Newtonian suspending liquids with densities and viscosities that span the range expected in waste feed tanks based on an evaluation of actual waste data and planned blending in the feed delivery tanks. A variety of particles with different size distributions and densities were specified for addition to the Newtonian liquids to create appropriate slurries for testing. The identified simulants also include non-Newtonian slurries with Bingham yield stress values that span a range that is expected to bound the Bingham yield stresses of waste in the feed delivery tanks. Specific large and dense particles were again specified for addition to the non-Newtonian materials to create slurries for testing. Lee et al. (2012) identified candidate materials for the Newtonian and non-Newtonian suspending fluids and the added particles, but did not provide specific recipes for obtaining the target properties for the Newtonian and non-Newtonian suspending fluids. In addition, existing information was not available to evaluate the compatibility of all the fluids and particles or the potential for salt precipitation at lower test temperatures for the candidate materials.

The purpose of this study is to prepare small trial batches of simulants in advance of the demonstration tests to determine specific simulant recipes, to evaluate the compatibility of the liquids and particles, and to determine whether the simulants are stable for the potential range of test temperatures. The objective of the testing, which is focused primarily on the Newtonian and non-Newtonian suspending fluids, is to determine the composition of selected simulant materials that give the desired density and viscosity or rheological parameters. Results will also be shown for the evaluation of the compatibility of the fluids and particles and the effect of temperature on the simulants.

In the following subsections we discuss the quality assurance (QA) level for these scoping tests and the simulant recipes. Section 2 discusses the target values for the bulk density and viscosity or Bingham properties for the simulants. Section 3 describes the experimental method and materials used in developing the simulant recipes and characterizing the simulants. Section 4 summarizes the final recipes and results of testing, and Section 5 provides an overall summary.

1.1 Quality Assurance

In conducting this work, Pacific Northwest National Laboratory (PNNL) followed the basic requirements that are implemented for all work conducted in accordance with the U.S. Department of Energy Prime Contract DE-AC05-76RL01830 at PNNL. The demonstrations described in Section 1.0 are conducted using a higher level of QA requirements and the testing conducted in this study does not meet this higher QA level. Accordingly, the intent of this simulant development effort is to provide recipes as guidance for the demonstration tests; the demonstrations will need to mix and characterize simulant batches according to the appropriate QA procedures for those demonstrations.

2.0 Simulant Targets

Target values of density and viscosity for Newtonian liquids and Bingham parameters for non-Newtonian fluids are given in Lee et al. (2012). Following simulant selection guidelines for tank waste (ASTM 2010), the target values were determined from an evaluation of actual waste data and planned waste blending in the feed delivery tanks for the parameters that are important for the planned testing. Table 2.1 shows the Newtonian-liquid targets and also describes the priority property for the simulant to match. For the typical-density/typical-viscosity target, the selection in the table is identified in Lee (2012b) and represents a multicomponent caustic salt solution used in previous mixing studies (Adamson et al. 2010) that is representative of Hanford supernatant liquid. For the two low-viscosity targets and the typical-density/typical-viscosity target, the priority is to match the target density because the fluid density is likely to have a more significant role in jet mixing than the viscosity (Wells et al. 2012). For the low-density/high-viscosity target, while the fluid density is certainly important, the purpose of this simulant is to explore the impact of elevated viscosity. For this simulant, the priority is to match the target viscosity and be reasonably close to the low-density target.

Aqueous solutions of salts and glycerol were specified in Lee et al. (2012) as candidates for producing Newtonian liquids with the target densities and viscosities. In general, glycerol was selected for increasing the viscosity (it also changes the density) and salts were selected for adjusting the density (salts also affect the viscosity). A variety of salts are candidates, but for the planned demonstration tests a preference was given to salts solutions that are nonhazardous and readily available at a reasonable cost. Sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3$) and sodium bromide (NaBr) were identified as specific candidates in Lee et al. (2012). Because of the lower purchase and disposal costs, sodium thiosulfate was the preferred salt and was used for all the recipes with one exception. For the high-density/low-viscosity target, a sodium thiosulfate solution at the target density would give a viscosity that is significantly higher than the target of 1 mPa·s (CRC 2011). A sodium bromide solution for the same target density gives much lower viscosity and was selected for the high-density/low-viscosity simulant.

Table 2.2 gives the target densities and Bingham yield stresses for the non-Newtonian slurries. The overall range for the Bingham yield stress is described in Lee et al. (2012) and the specific 3 Pa target is given by Lee (2012a). Lee et al. (2012) do not give specific values for the Bingham consistency, but note that the consistency should be appropriate for the yield stress based on the actual waste data presented in their Appendix B. For the majority of the data in Appendix B of Lee et al. (2012), the Bingham consistency is between 1 and 10 mPa·s. The high-density target for the non-Newtonian slurries was selected by Lee (2012a) to match the high-density Newtonian fluid. The low-density target was selected to be comparable to the low-density Newtonian liquid as comparable as can be achieved with kaolin-in-water slurries (Lee 2012a).

Lee et al. (2012) and Lee (2012a) specified slurries of kaolin clay in water and kaolin clay in sodium thiosulfate solutions as candidates for giving the target densities and Bingham parameters. In general, kaolin was selected for increasing the Bingham yield stress (kaolin also affects the density) and sodium thiosulfate was selected for adjusting the density (it also changes how the kaolin increases the yield stress). Some limited testing was done with kaolin in sodium bromide solutions, but the lower purchase and disposal cost of sodium thiosulfate made it the preferred salt for these recipes.

Table 2.1. Targets for Newtonian-Liquid Simulants

Simulant (Density/Viscosity)	Targets from Tank Waste Data ^(a)		Comments
	Density (g/mL)	Viscosity (mPa·s)	
Low/Low ^(a)	1.1	1	Simulant priority is to match target density
Low/High ^(a)	1.1	8	Simulant priority is to match target viscosity
High/Low ^(a)	1.37	1	Simulant priority is to match target density
High/High ^(a)	1.37	15	Simulant target is to match both density and viscosity
Typical/Typical ^(b)	1.29	2.6	Simulant priority is to match target density

(a) Targets as defined in Lee et al. (2012) except the Typical/Typical simulant

(b) Typical/Typical target is to match multicomponent caustic salt solution used in previous mixing studies (Adamson et al. 2010) that is representative of Hanford supernatant liquid

Table 2.2. Bingham Yield Stress and Density Targets for Non-Newtonian Suspending Fluid

Simulant (Bingham Yield Stress)	Targets from Tank Waste Data	
	Density (g/mL)	Bingham Yield Stress (Pa)
Low ^(a)	1.37 ^(b)	1
Middle ^(a)	1.37 ^(b)	3
High ^(a)	1.37 ^(b)	10
Low ^(a)	1.1 ^(c)	1
Middle ^(a)	1.1 ^(c)	3
High ^(a)	1.1 ^(c)	10

(a) overall range described in Lee et al. (2012) and specific 3 Pa target given by Lee (2012a)

(b) density target selected to match the high-density Newtonian fluid (Lee 2012a)

(c) density target is to be comparable to low-density Newtonian liquid (as comparable as can be achieved with kaolin-in-water slurries) (Lee 2012a)

3.0 Experimental Method and Materials

Trial batches of simulants were prepared to develop simulant recipes to match the target properties for specific Newtonian and non-Newtonian simulants given in Section 2. The primary properties measured were the density and viscosity or Bingham parameters. The materials for these tests were obtained as subsamples from large batches that will be used in the planned demonstration tests. In addition to measuring the primary parameters, tests were conducted to evaluate the compatibility of the suspending fluids with the planned base and spike simulant particles. For the non-Newtonian simulants, one suite used the addition of salt to kaolin slurries to achieve a high-density target while a second suite used kaolin in water. For these tests, the presence of the salt significantly increased the Bingham parameters in addition to increasing the density. For these kaolin slurries, the particle size distribution (PSD) of the kaolin was determined to evaluate whether the presence of salt changed the kaolin particle size in a way that might make it difficult to compare the behavior of the kaolin slurries with and without salt in the demonstration tests. Testing results are summarized in Section 4.

3.1 Simulant Materials and Preparation

Various solutions and slurries were prepared to evaluate rheological properties and to observe process characteristics in an attempt to determine potential large-scale slurry preparation, mixing, and/or suspension problems. Additional particles were also added as needed for compatibility studies. The preparation was different for the Newtonian and non-Newtonian fluids and the specifics are discussed below.

3.1.1 Newtonian Liquids

The chemicals used for mixing the Newtonian liquids are given in Table 3.1. See Appendix A for specific details about the materials used for evaluation. Most of the solutions were hand mixed (shaken) in both nominal 250 g and nominal 500 g batches using 250 mL and 500 mL clear acrylic bottles, respectively. In all cases, when a solution contained glycerol, an attempt was made to dissolve all chemical additives in the water component of the solution prior to the addition of the glycerol. During early scoping tests, warm tap water was run over mixing containers to help attain 100% dissolution of sodium thiosulfate pentahydrate. Note that in a few cases, chemical additives that did not go into solution in the low-weight-percent water fraction went into solution without additional heat when sufficient glycerol was added.

Both sodium thiosulfate pentahydrate and anhydrous sodium thiosulfate salts were evaluated. Endothermic and exothermic reactions were experienced respectively, though not to the same degree. The endothermic reaction of the hydrated sodium thiosulfate was much more discernable than the exothermic reaction of the anhydrous form. It is reasonable to believe that the relatively easy dissolution of the solutions containing anhydrous sodium thiosulfate may have been enhanced by the exothermic dissolution of this salt.

Table 3.1. Materials Used in Newtonian Liquid Simulants

Material	Supplier	Chain of Custody Identification	Sample Identification
Pasco City Water	N/A	ES-RSD-051 ES-SSMD-062	RSD-272 SSMD-792, -793, -794, and -795
Glycerol	Silver Fern	ES-SSMD-062	SSMD-791
Sodium Thiosulfate (anhydrous)	Brainerd Chemical Company	ES-SSMD-062	SSMD-790
Sodium Bromide	Albemarle Corporation	ES-SSMD-063 ES-RSD-051	SSMD-799 RSD-273

For the high-viscosity/high-density Newtonian-liquid target given in Table 2.1, an aqueous single-component solution of sodium thiosulfate, sodium bromide, or glycerol will not match both the density and viscosity targets (CRC 2011). To match both of these targets, the approach will be to add both glycerol and sodium thiosulfate to water and then adjust their proportions to match both the density and viscosity targets. Information is not available in the literature to estimate viscosity as a function of composition; viscosity results for various mixtures will be discussed in Section 4. A simple model for density of glycerol and sodium thiosulfate mixtures can be estimated by assuming an ideal mixture of pure glycerol with a salt solution whose composition is given by the water and salt content of the mixture. The bulk density of the combination of glycerol and sodium thiosulfate solution is given by the following:

$$\rho_{g-s} = \frac{1}{(1-x_g)/\rho_{ss} + x_g/\rho_g} \quad (3.1)$$

where

ρ_{g-s} = the density of the glycerol/ sodium thiosulfate solution

x_g = the mass fraction of glycerol in the bulk mixture

ρ_g = the density of pure glycerol

ρ_{ss} = the density of the sodium thiosulfate solution whose composition is given by the water and salt content of the mixture

The density for the solution of water and sodium thiosulfate can be determined from literature data (CRC 2011), and the following linear equation fits the data for sodium thiosulfate mass fractions between 0.2 and 0.4:

$$\rho_{ss} = 0.9638 + 1.044x_{\text{salt in ss}} \quad (3.2)$$

where $x_{\text{salt in ss}}$ is the mass fraction of sodium thiosulfate in the salt solution whose composition is given by the water and salt content of the mixture. The target density for the high-viscosity/high-density simulant is 1.37 g/mL and Equations 3.1 and 3.2 can be used to calculate the concentrations of glycerol and sodium thiosulfate that will give this density. The measured density of various mixtures used for viscosity testing will be compared to the model estimates in Section 4.

3.1.2 Non-Newtonian Fluids

Testing was conducted with simulants that are slurries of Pasco City water and Edgar plastic kaolin (EPK, Feldspar Corporation) with sodium thiosulfate or sodium bromide. Table 3.2 shows the materials used in this study and also refers to chain of custody documentation containing additional product information, which is located in Appendix A.

Most of the non-Newtonian slurries were prepared using a 300 watt *UltraPower* KitchenAid® mixer in nominally 500 g batches that were transferred primarily to 500 mL clear acrylic bottles after mixing. A standard approach for mixing the slurries was developed during testing. Typically, dry components were added to the mixing bowl followed by fluid components. The mixer was then turned on to a mixing speed set point of 2 (maximum speed set point is 10) for 5 minutes. The mixing set point was increased to 4 and mixing continued for an additional 15 minutes. The mixer arrangement utilized a stainless steel mixing bowl and a compatible ceramic coated paddle.

Prior to measuring rheological properties, the slurry was presheared. The preeminent method of preshearing involved returning the slurry to the KitchenAid mixer for an additional 5 minutes at a mixing speed set point of 4.

As can be expected, the intensity of the mixing and the subsequent preshearing has an impact on rheology. An alternate, a more intense, method of preshearing involving a 250 watt, model MB1001C, Magic Bullet® blender was also evaluated prior to analysis. The previously mixed slurry was placed in the Magic Bullet and processed for 1 minute at high speed (the Magic Bullet has only one speed, which is higher than speeds of the KitchenAid mixer). It should be noted that other mixing techniques were used during testing and that all mixing and preshearing steps are identified for each specific experiment. In test and/or sample nomenclature, “KA” refers to use of the KitchenAid mixer and “MB” refers to the use of the Magic Bullet. “KA/MB” indicates initial mixing with the KitchenAid and preshearing with the Magic Bullet.

Table 3.2. Materials used in Non-Newtonian Liquid Simulants

Material	Supplier	Chain of Custody Identification	Sample Identification
Pasco City Water	N/A	ES-RSD-051 ES-SSMD-062	RSD-272 SSMD-792, -793, -794, and -795
EPK Kaolin	Feldspar Corporation ZEMEX Industrial Minerals, Inc.	ES-SSMD-063	SSMD-796
Sodium Thiosulfate (anhydrous)	Brainerd Chemical Company	ES-SSMD-062	SSMD-790
Sodium Bromide	Albemarle Corp.	ES-SSMD-063 ES-RSD-051	SSMD-799 RSD-273

For the high-density non-Newtonian fluid target given in Table 2.2, mixtures of only kaolin in water will not match the target density. To match the density and Bingham yield stress targets, the approach will be to add both kaolin and sodium thiosulfate to water and then adjust their proportions to match both targets. Information is not available in the literature to estimate Bingham yield as a function of both salt

and kaolin concentration; rheology results for various mixtures will be discussed in Section 4. The density of a composite slurry of kaolin and sodium thiosulfate can be estimated by assuming an ideal mixture of the kaolin particles with a salt solution whose composition is given by the water and salt content of the mixture. The bulk density of the slurry is given by the following (Gauglitz et al. 2010):

$$\rho_{k-s} = \frac{1}{(1 - x_k) / \rho_{ss} + x_k / \rho_k} \quad (3.3)$$

where

- ρ_{k-s} = density of the kaolin-sodium thiosulfate slurry
- X_k = the mass fraction of kaolin in the bulk mixture
- ρ_k = kaolin particle density (2.65 g/mL, Gauglitz et al. 2010)
- ρ_{ss} = the density of the sodium thiosulfate solution whose composition is given by the water and salt content of the composite slurry

The density, ρ_{ss} , for an aqueous solution of sodium thiosulfate can be determined from literature data (CRC 2011). Equation 3.2 can be used to estimate the density of an aqueous solution of sodium thiosulfate that has a mass fraction, $x_{\text{salt in ss}}$, in the range of 0.2 and 0.4. The target density for the high-density non-Newtonian slurries is 1.37 g/mL and Equations 3.3 and 3.2 can be used to calculate the concentrations of kaolin and sodium thiosulfate that will give this density. For the non-Newtonian simulants, the measured densities for the specific simulant recipes are given Section 4.

3.1.3 Base and Spike Simulant Particles

Table 3.3 and Table 3.4 show the base and spike particles used in this study to evaluate the compatibility of the particles in the Newtonian and non-Newtonian fluids. The base particles are the components given in Lee et al. (2012) for the low-base simulant and the high-base simulant. The spike particles in Table 3.4 are a single size from each of materials planned for use as spike particles in the demonstration testing (Lee 2012a). The compatibility experiments included observing the settling behavior of the base and spike particles. Table 3.5 lists the particle densities and estimates of the median particle diameter, d_{50} .

Table 3.3. Materials Used as Base Simulant Particles in Newtonian Liquids

Material	Supplier	Chain of Custody Identification	Sample Identification
Gibbsite 3431 (large)	Huber	ES-SSMD-067	SSMD-833
Gibbsite (small)	Nalbatec	ES-SSMD-064	SSMD-801
Sand (small, Sil-Co-Sil 250)	US Silica	ES-SSMD-063	SSMD-797
Sand (Large, NJ6)	US Silica	ES-SSMD-064	SSMD-800
Zirconium Oxide	Washington Mills Electro Corp	ES-SSMD-063	SSMD-798
Stainless Steel Powder	Pellets, LLC	ES-SSMD-064	SSMD-802

Table 3.4. Materials Used as Spike Particles in Newtonian and Non-Newtonian Fluids

Material	Manufacture	Chain of Custody Identification	Sample Identification
Soda Lime Glass Beads	Walter Stern, Inc.	ES-SSMD-064	SSMD-803
Stainless Steel (1/16")	Pellets LLC	ES-SSMD-064	SSMD-804
Tungsten Carbide Balls (1/16")	Tungsten Heavy Powder, Inc.	ES-SSMD-068	SSMD-834

Table 3.5. Approximate Particle Properties

Particle	d ₅₀	Density (g/mL)	Reference
Gibbsite (small) APYRAL® 40CD	1.3 µm	2.42	Lee et al. (2012)
Gibbsite 3431 (large)	10 µm	2.42	Lee et al. (2012)
Sand (small, Sil-Co-Sil 250)	40 µm	2.65	d ₅₀ estimated from vendor data Density from vendor data
Sand (Large, NJ6)	520 µm	2.65	d ₅₀ estimated from vendor data Density for typical sand
Zirconium Oxide	6 µm	5.7	Lee et al. (2012)
Stainless Steel Powder	112 µm	8	Lee et al. (2012)
EPK Kaolin	5 µm	2.65	d ₅₀ from Figure C.2 Density - Gauglitz et al. (2010)
Soda Lime Glass Beads	2 mm	2.49	Vendor Data
Stainless Steel (1/16")	1/16 in.	8	d ₅₀ from vendor data Density from Lee et al. (2012)
Tungsten Carbide Balls (1/16")	1/16 in.	14	d ₅₀ from vendor data Density from Lee et al. (2012)

3.2 Viscosity and Rheology

Viscosity and rheological measurements were performed using a Haake RS600 rheometer operated with RheoWin software (Thermo Electron Corporation). The RS600 rheometer was equipped with a low-inertia torque motor and coaxial cylinder measurement geometry. The drive shaft of the motor was centered by an air bearing, which provides virtually frictionless transmission of the applied torque to the sample. Viscosity and rheological analyses were conducted at various temperatures ranging from 15 to 30°C, which was the estimated potential range of test temperatures. Each rheogram (flow curve) was obtained by shearing the sample at a controlled rate increasing from 0 (zero) to 1000 s⁻¹ for 5 minutes, holding constant at 1000 s⁻¹ for 1 minute, following by shearing at a controlled rate decreasing from 1000 to 0 s⁻¹ (zero) for 5 minutes. Prior to measuring a flow curve, each sample was gently shaken by hand and sheared at a constant rate of 250 s⁻¹ for 3 minutes. The purpose of pre-measurement mixing was to make sure that the material being analyzed was homogenized and representative of the sample.

Typically, for Newtonian simulants, one rheogram (flow curve) was obtained from one sample aliquot. For non-Newtonian simulants, two or three rheograms were obtained from one sample aliquot. A flow curve represents shear stress as a function of shear rate. For Newtonian simulants, the Newtonian Model, shown in Equation 3.4, was used to fit the data. For non-Newtonian simulants, the Bingham Plastic Model, shown in Equation 3.5, was used for data quantification (Mewis and Wagner 2012). Unless specified otherwise, all of the Newtonian simulant data was fitted to the entire range of testing shear rates of 0 to 1000 s⁻¹. For the non-Newtonian simulants, the selected shear rate range for data fitting was 250 to 700 s⁻¹, unless specified otherwise. For non-Newtonian simulants, the Bingham yield stress and Bingham consistency values from the down-ramp of the second flow curve are reported and discussed in this document.

$$\tau = \eta * \gamma \quad (3.4)$$

where τ = shear stress in Pa
 γ = shear rate in s⁻¹
 η = viscosity in Pa·s

$$\tau = \tau_0 + \eta_p * \gamma \quad (3.5)$$

where τ = shear stress in Pa
 τ_0 = critical shear stress or Bingham yield stress in Pa
 γ = shear rate in s⁻¹
 η_p = Bingham consistency in Pa·s

3.3 Bulk Density

Bulk density of the selected simulants was measured using certified glass pycnometers (Wilma LabGlass) and a calibrated balance. A pycnometer is a volumetric flask with a known volume that is specifically designed for density measurements. Prior to density measurements, a performance check of the balance was performed using a 10 g and/or a 50 g certified check weight (manufactured by Rice Lake and calibrated and certified by Quality Control Services, Inc.). The balance performance-check result was recorded in a density measurement bench sheet. After the balance performance check, the tare weight of the pycnometer to be used was obtained and recorded in the density measurement bench sheet. The pycnometer was then filled with the simulant fluid to be measured. The gross weight of the pycnometer containing the simulant fluid was obtained and recorded in the density measurement bench sheet. The net weight of the simulant fluid was calculated by subtracting the pycnometer tare weight from the gross weight of the pycnometer containing the simulant. The bulk density of the simulant fluid was calculated using Equation 3.6. Unless specified otherwise, all density measurements were carried out at room temperature. Room temperature associated with each density measurement was also measured using a calibrated thermocouple and thermocouple readout and recorded in the density measurement bench sheet.

$$\rho = \frac{M}{V} \quad (3.6)$$

where ρ = bulk density in g/mL
 M = net weight of the simulant fluid in g
 V = volume of the simulant fluid in mL

3.4 Compatibility of Suspending Fluids and Particles

Two types of qualitative tests were conducted to evaluate the compatibility of the base and spike particles with the Newtonian and non-Newtonian suspending fluids. The first evaluation was to determine the settling behavior of the particles in the suspending fluids. The second evaluation was to determine the intensity of mixing required to re-suspend layers of particles that had settled overnight, or for seven days. For the first evaluation, approximately 200 mL of each simulant mixture of suspending fluid and base and spike particles, at concentrations that are described below in Table 3.6 through Table 3.9, were added to a 250 mL graduated cylinder. The cylinder was then closed and shaken and the settling rate of the simulant particles was observed over a period of about 8 hours. The settling measurements were conducted for four consecutive days for each sample to determine whether there were changes in settling behavior over this duration. For the second evaluation, each graduated cylinder was allowed to continue settling undisturbed overnight after the last settling measurement was recorded each day. The intensity of shaking needed to resuspend the settled layer was determined for each graduated cylinder on the following day prior to continuing settling measurements. For the resuspension testing, in addition to the daily re-suspension measurement from each graduated cylinder that was also used for settling evaluation, one additional graduated cylinder of each simulant mixture was prepared and allowed to settle undisturbed for seven days prior to performing the same resuspension evaluation. The intensity of shaking was controlled by progressing through the following sequential steps until the settled layer and large spike particles were observed to move.

- Level 0 Swirl in a circular motion - heavy stainless steel (SS) beads are not stuck to the bottom (applicable to bottle 7 day test only).
- Level 1 Rock vertical to horizontal gently at least 20 times - heavy SS beads are not stuck to the bottom.
- Level 2 Shake gently side to side ~2 shakes per second, at least 20 times - heavy SS beads are not stuck to the bottom.
- Level 3 Shake vigorously side to side ~4 shakes per second, at least 20 times - heavy SS beads are not stuck to the bottom.

Table 3.6 shows the composition of the Newtonian liquids used for the compatibility evaluation. The compositions of these solutions are the same as the final recipes discussed in Section 4. Each of these liquids was combined with the solid particles for the low-base simulant and high-base simulant given by Lee (2012a). Lee (2012a) also specifies the total quantity of base and spike particles, and the compatibility tests used 15 wt% solids loading to match the high-solids loading tests. Table 3.7 provides the compositions of slurries of the base and spike particles in Newtonian suspending fluids that were used in compatibility tests. For simplification, the first row of the table lists the short name for each simulant mixture that will be used when discussing the testing results in Section 4.

Table 3.6. Compositions of Newtonian Liquids used for Evaluating Particle and Liquid Compatibility

Component	Newtonian Simulant					
	Low Density Low Viscosity	Low Density High Viscosity	Low Density High Viscosity (w/Na ₂ S ₂ O ₃)	High Density Low Viscosity	High Density High Viscosity	Typical Density Typical Viscosity
Sodium Thiosulfate Anhydrous	12.0%	-	0.1%	-	33.4%	31.5%
Glycerol	-	53.0%	53.0%	-	19.5%	-
Sodium Bromide	-	-	-	37.0%	-	-
Pasco City Water	88.0%	47.0%	46.9%	63.0%	47.1%	68.5%

Table 3.8 shows the compositions of the non-Newtonian slurries used for the compatibility evaluation. The compositions of these slurries are the same as the final recipes discussed in Section 4 for the simulants with a target Bingham yield stress of 3 Pa. Each of these slurries was combined with quantities of spike particles, as described in Lee (2012a), to have the same quantity of spike particles as the Newtonian liquids with the same density. Table 3.9 provides the compositions of slurries and spike particles for these tests, including one test that had a higher quantity of spike particles to make observation easier.

Table 3.7. Compositions of Slurries of Newtonian Liquids and Base and Spike Particles for Compatibility Testing

	Short Names for Simulant and Base Particle Mixtures ^(a)									Typ/Typ/Lo	Typ/Typ/Hi
	Lo/Lo/Lo	Lo/Lo/Hi	Lo/Hi/Lo	Lo/Hi/Hi	Hi/Lo/Lo	Hi/Lo/Hi	Hi/Hi/Lo	Hi/Hi/Hi	Typ/Typ/Lo		
Newtonian Liquid	Lo Density Lo Visc.	Lo Density Lo Visc.	Lo Density Hi Visc.	Lo Density Hi Visc.	Hi Density Lo Visc.	Hi Density Lo Visc.	Hi Density Hi Visc.	Hi Density Hi Visc.	Hi Density Hi Visc.	Typ Dens. Typ Visc.	Typ Dens. Typ Visc.
	both with and without 0.1 wt% Na ₂ S ₂ O ₃										
Newtonian Liquid ^(b)	85%	85%	85%	85%	85%	85%	85%	85%	85%	85%	85%
Small Gibbsite	14.25%	-	14.25%	-	14.25%	-	14.25%	-	14.25%	-	-
Large Gibbsite	-	0.43%	-	0.43%	-	0.43%	-	0.43%	-	-	0.43%
Small Sand	-	4.99%	-	4.99%	-	4.99%	-	4.99%	-	-	4.99%
Large Sand	-	2.99%	-	2.99%	-	2.99%	-	2.99%	-	-	2.99%
Zirconium Oxide	-	1.14%	-	1.14%	-	1.14%	-	1.14%	-	-	1.14%
SS Powder	-	4.70%	-	4.70%	-	4.70%	-	4.70%	-	-	4.70%
Soda Lime Glass	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%
SS Beads	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%
Tungsten Carbide Beads	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%

(a) Each short name specifies the density and viscosity of the Newtonian liquid, and type of base particle, respectively (Lo = Low, Hi = High, Typ = Typical)

(b) Each test mixture contained 85 wt% Newtonian liquid and 15 wt% solids loading to match the high-solids loading tests specified in Lee (2012a).

Table 3.8. Compositions of Non-Newtonian Slurries used for Evaluating Particle and Liquid Compatibility

Non-Newtonian Simulant		
Target Density	1.37 g/mL	1.1 g/mL
Target Yield Stress	3 Pa	3 Pa
Component		
Sodium Thiosulfate Anhydrous	24.9%	-
Kaolin	14.5%	22.5%
Pasco City Water	60.6%	77.5%

Table 3.9. Compositions of Non-Newtonian Slurries and Spike Particles for Compatibility Testing

Non-Newtonian Simulant			
Target Density	1.37 g/mL	1.37 g/mL	1.1 g/mL
Target Yield Stress	3 Pa	3 Pa	3 Pa
		(100x each spike)	
Non-Newtonian Simulant	99.04%	51.7%	99.04%
Soda Lime Glass (2 mm)	0.32%	16.1%	0.32%
Stainless Steel (1/16")	0.32%	16.1%	0.32%
Tungsten Carbide Spheres (1/16")	0.32%	16.1%	0.32%

3.5 Temperature Stability

The effect of temperature on the final recipes of Newtonian and non-Newtonian simulants recommended for demonstration tests was observed down to 10°C to determine the stability of the selected simulants at the potential lowest test temperature. Temperature stability studies were carried out in a temperature-controlled water bath. The simulant fluids were placed in the water bath at 15°C. The

simulant slurries were visually checked for precipitation after 20 to 24 hours. After visual check for precipitation, the water bath temperature set point was lowered to 10°C. Visual inspection was performed again after 20 to 24 hours to check for precipitation.

3.6 Clay Particle Size Distribution

PSD measurements were made using a Microtrac S3000 Particle Size Analyzer that has a full size range of 0.02 to 2000 µm (Microtrac, Inc.). The Microtrac S3000 Particle Size Analyzer uses laser diffraction technology. Prior to performing analyses on slurry samples, a number of measurements were made at different instrument settings to determine the appropriate settings. Measurements were then made on three selected slurries using what were considered the best settings. As is typical for PSD measurements, the results are affected by the choice of instrument settings. The instrument analysis program was set up to analyze the particle size using volume distribution with standard progression. The analysis was performed between the size range of 0.021 and 1408 microns with a run time of 30 seconds and an average of 3 runs used to determine each analysis result.

Three kaolin clay slurries were prepared for PSD analysis as shown in Table 3.10. Samples 071012WCB01 and 071012WCB02 were stirred in a KitchenAid mixer at mixing speed set point of 2 for 5 minutes, followed by an additional 15-minute mixing at speed set point of 4. Sample 071012WCB03 was hand mixed by placing the kaolin and water in a poly bottle and shaking for about 2 minutes. All three samples were split equally into two containers for each sample. Preshearing was performed on Samples 071012WCB01 and 071012WCB02. One split sample was presheared using a Magic Bullet for 1 minute and the other split was presheared with the KitchenAid mixer at mixing speed set point of 4.

Table 3.10. Kaolin Slurries for PSD Analysis

Slurry ID	Kaolin (wt%)	Na ₂ S ₂ O ₃ (wt%)	Pasco City Water (wt%)	DI Water (wt%)
071012WCB01	14.5	24.9	60.6	-
071012WCB02	22.5	-	77.5	-
071012WCB03	22.5	-	-	77.5

Ten samples were analyzed. The samples were analyzed as provided and after sonication with an ultrasonic horn as indicated in Table 3.11. For those subsamples treated with the ultrasonic horn, an aliquot was prepared and then “sonicated” intermittently for a nominal 20 seconds with a MICROGON Ultrasonic Cell Disrupter (PNNL-assigned property # R104106). The set point for the ultrasonic horn was 12, resulting in 10 to 12 watts (root mean square) directed into the slurry of ~0.2 g of sample material in ~10 mL of deionized (DI) water, Pasco City water or 29 wt% sodium thiosulfate diluent contained in an ~25 mL poly beaker. The diluent used was dependent upon the corresponding analytical carrier solution. Sample material was transferred from the poly beaker to the analyzer using a transfer pipette. The transfer pipette was also used to keep the sample material suspended in the poly beaker between analyses. The variable speed re-cycling (VSR) pump integral to the analyzer was set at 45.

Table 3.11. Preshearing and Sonication for Kaolin Slurry Samples

Sample ID	Slurry ID	MB Pre-Shear	KA Pre-Shear	Sonicated	Analytical Carrier Solution
KA/MB Salts/Sonicated	071012WCB01	x		x	29 wt% Na ₂ S ₂ O ₃ ^(a)
KA/MB Salts	071012WCB01	x			29 wt% Na ₂ S ₂ O ₃ ^(a)
KA/KA Salts/Sonicated	071012WCB01		x	x	29 wt% Na ₂ S ₂ O ₃ ^(a)
KA/KA Salts	071012WCB01		x		29 wt% Na ₂ S ₂ O ₃ ^(a)
KA/MB Kaolin/Sonicated	071012WCB02	x		x	Pasco City Water
KA/MB Kaolin	071012WCB02	x			Pasco City Water
KA/KA Kaolin/Sonicated	071012WCB02		x	x	Pasco City Water
KA/KA Kaolin	071012WCB02		x		Pasco City Water
Hand Kaolin/Sonicated	071012WCB03			x	DI Water
Hand Kaolin	071012WCB03				DI Water

(a) 29 wt% is the concentration of sodium thiosulfate in water within the slurry

Multiple runs were performed on each sample to evaluate potential particle breakdown or agglomeration during the analysis. In addition, the VSR pump speed set point was varied down to 30 and up to 60 in an effort to determine whether heavy particles were settling in the VSR pump. It was determined that there was minimal particle breakdown over time and the VSR pump speed had negligible effect on the PSD; these runs were not reported. Table 3.12 gives parameters used in the sample analysis setup.

Table 3.12. Microtrac Instrument Parameters for PSD Measurements of Kaolin in Water and in a Sodium Thiosulfate Solution

Setup Name: Kaolin	Setup Name: Kaolin (Na ₂ S ₂ O ₃)
Setzero time: 30 seconds	Setzero time: 30 seconds
Run time: 30 seconds	Run time: 30 seconds
Run number: 3	Run number: 3
Particle: Kaolin	Particle: Kaolin
Particle transparency: Trans	Particle transparency: Trans
Particle refractive index: 1.57 ^(a)	Particle refractive index: 1.57 ^(a)
Particle shape: irregular	Particle shape: irregular
Fluid: water	Fluid: 29.1 wt% Na ₂ S ₂ O ₃ in water
Refractive index: 1.33 ^(b)	Refractive index: 1.40 ^(b)
Progression: geometric root 8	Progression: geometric root 8

(a) Malvern (2007)
(b) CRC (2011)

4.0 Results and Final Recipes

Experiments were conducted to determine the compositions of aqueous solutions and slurries that have densities and viscosities or rheological parameters that match the simulant targets given in Section 2. For the Newtonian liquids, the mixtures were primarily single components in water and the desired targets were readily obtained by adjusting the concentrations of the single components. For these mixtures, the priority for the simulant was matching either the fluid density or viscosity target. For the parameter that was not specifically matched, testing was needed to confirm that the value was comparable to simulant target. One of the Newtonian simulants involved adding two components to water, and here both the density and viscosity targets could be matched. For this mixture, additional measurements and analyses were needed to determine how to vary the concentration of the two added components to match both properties. For the non-Newtonian simulants, the high-density target involved adding both kaolin clay and a salt to water to match both the density and Bingham yield stress targets.

The primary temperature for developing simulant recipes was 20°C, which was selected as a representative value for the planned demonstration tests. The actual test temperature is expected to vary, so the viscosity or Bingham parameters were also measured at 15° and 25°C. For the high-viscosity/high-density simulant that uses glycerol, the viscosity was also measured at 30°C because glycerol solutions are known to be somewhat temperature sensitive (CRC 2011). Because many of the simulants are concentrated salt solutions, there is a potential that the salt may precipitate at lower temperatures. Accordingly, the stability of the solutions was determined at 10°C, which was selected as a temperature that should be lower than that of the demonstration tests. After the recipes were developed to match the target properties, tests were conducted to evaluate the interaction of base and spike simulant particles with the suspending Newtonian and non-Newtonian fluids. In one case, a combination of a specific type of base particle and Newtonian liquid formed overly strong settled layers, and the recipe for this Newtonian simulant was modified by adding a small concentration of sodium thiosulfate.

Table 4.1 gives a summary of the final recipes for the Newtonian simulants, the specific simulant targets, and the measured properties for these recipes at 20°C. The table also shows the priorities for matching the specific density and/or viscosity targets. All of these recipes were stable down to 10°C as discussed below. Table 4.2 gives the final recipes for the non-Newtonian simulants, the specific simulant targets, and the measured properties at 20°C. These slurries were also stable down to 10°C as discussed below. Specific tests were not conducted to evaluate long-term degradation of these simulants, but there was no indication of degradation in test samples over a period of a couple weeks. In the subsections below, detailed testing results for trial batches of these simulants are given. For the non-Newtonian simulants, the full rheograms are given in Appendix B.

Table 4.1. Newtonian Liquid Simulants

Simulant (density/viscosity)	Targets from Tank Waste Data ^(a)		Simulant Properties (20°C) ^(c)		Simulant Recipes
	Density (g/mL)	Viscosity (mPa·s)	Density (g/mL)	Viscosity (mPa·s)	
Low/Low ^(a)	1.1	1	1.098	1.62	12 wt% Na ₂ S ₂ O ₃ 88 wt% water Simulant priority is to match target density
Low/High ^(a)	1.1	8	1.135	7.96	53 wt% glycerol 0.1 wt% Na ₂ S ₂ O ₃ 46.9 wt% water Simulant priority is to match target viscosity
High/Low ^(a)	1.37	1	1.370	2.00	37 wt% NaBr 63 wt% water Simulant priority is to match target density
High/High ^(a)	1.37	15	1.368	14.6	19.5 wt% glycerol 47.1 wt% water 33.4 wt% Na ₂ S ₂ O ₃ Simulant target is both matched density and viscosity
Typical/Typical ^(b)	1.29	2.6	1.284	3.60	31.5 wt% Na ₂ S ₂ O ₃ in water Simulant priority is to match target density

(a) Targets as defined in Lee et al. (2012) except Typical/Typical simulant

(b) Target is to match multi-component caustic salt solution used in previous mixing studies (Adamson et al. 2010) that is representative of Hanford supernatant liquid

(c) Viscosity was measured at 20°C; the density is expected to be insensitive to temperature and was measured at ambient temperature that varied between 17° and 21°C

Table 4.2. Non-Newtonian Slurry Simulants

Simulant (Bingham Yield Stress)	Targets from Tank Waste Data		Simulant Properties (20°C) ^(d)			Simulant Recipes
	Density (g/mL)	Bingham Yield Stress (Pa)	Density (g/mL)	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)	
Low	1.37 ^(b)	1 ^(a)	1.36	1.1	7.3	9.5 wt% Kaolin 29.6 wt% Na ₂ S ₂ O ₃
Middle	1.37 ^(b)	3 ^(a)	1.36	3.7	8.1	14.5 wt% Kaolin 24.9 wt% Na ₂ S ₂ O ₃
High	1.37 ^(b)	10 ^(a)	1.34	11	10	20.0 wt% Kaolin 19.9 wt% Na ₂ S ₂ O ₃
Low	1.1 ^(c)	1 ^(a)	1.13	1.2	3.4	20 wt% Kaolin
Middle	1.1 ^(c)	3 ^(a)	1.15	2.6	4.0	22.5 wt% Kaolin
High	1.1 ^(c)	10 ^(a)	1.19	11	6.0	26.5 wt% Kaolin

(a) Lee et al. (2012) define range as up to 10 Pa; target values of 1, 3, and 10 Pa are selected to span this range and to support planned testing (Lee 2012a)

(b) Density target selected to match High/High Newtonian liquid as part of planned testing (Lee 2012a)

(c) Density target is to be comparable to low-density Newtonian liquid; as comparable as can be achieved with kaolin-in-water slurries (Lee 2012a)

(d) Bingham parameters measured at 20°C; the density is expected to be insensitive to temperature and was measured at ambient temperature that varied between 17° and 21°C

4.1 Newtonian Simulant Density and Viscosity

Simulant recipes to meet targets for the Newtonian liquids were obtained with single-component aqueous solutions for all but the high-density/high-viscosity target. The specific recipes for these four simulants at 20°C are given in Table 4.1. For the single-component solutions, the simulant target was achieved by adjusting the concentration of each single component. Figure 4.1 shows the effect of temperature on viscosity for these four of the simulant recipes. The most temperature-sensitive recipe is the low-density/high-viscosity simulant that is a mixture of glycerol in water. The final recipe for this simulant has a small amount of sodium thiosulfate added to improve the behavior of the small gibbsite base particle in this liquid (see Section 4.2). The temperature dependence of viscosity was measured for the initial recipe for this simulant without the sodium thiosulfate but this temperature dependence should be essentially the same for the final recipe.

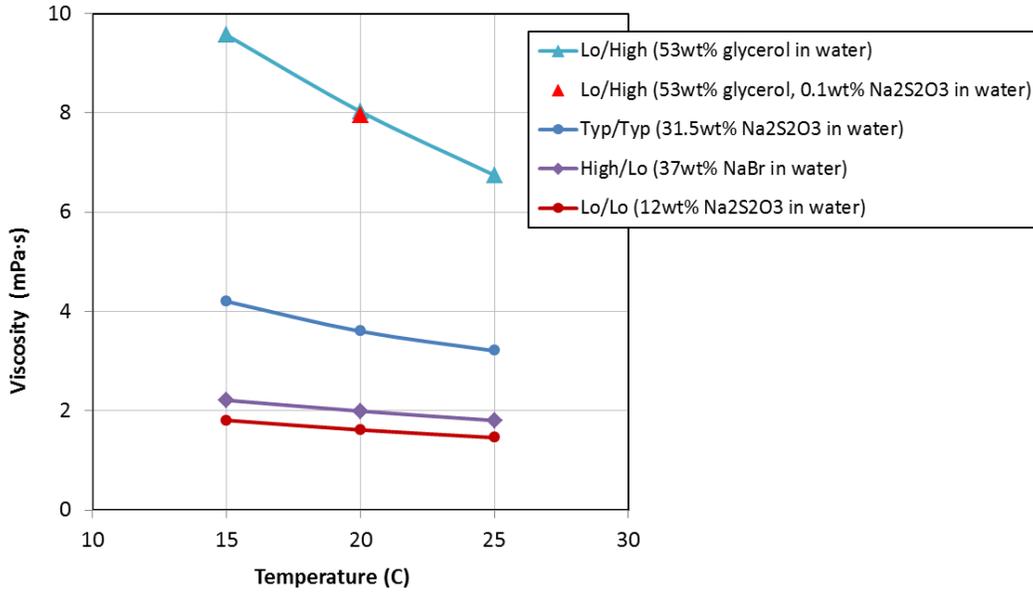


Figure 4.1. Effect of Temperature on Viscosity for Newtonian Simulants

For the high-density/high-viscosity target, glycerol and sodium thiosulfate were added to water to adjust both the viscosity and density. Four solutions were prepared that were selected to have progressively higher viscosities and also, by using Equations 3.1 and 3.2 to select compositions, have a density of 1.37 g/mL. Two additional solutions were prepared that have slightly higher or lower densities than the 1.37 g/mL target value to get a better set of data for evaluating the density predictions and to support development of the viscosity model discussed below. Table 4.3 gives the measured densities of the solutions and shows a comparison to the calculated densities.

Table 4.3. Densities of Aqueous Solutions of Glycerol, Sodium Thiosulfate and Water and Comparison to Calculated Density

Glycerol (wt%)	Na ₂ S ₂ O ₃ (wt%)	Calculated Density (g/mL)	Measured Density ^(a) (g/mL)	Difference Measured to Calculated (%)
17	34.1	1.37	1.365	- 0.4
19.5	33.4	1.37	1.368	- 0.1
22	32.7	1.37	1.367	- 0.2
25.5	31.8	1.37	1.367	- 0.2
17	32.7	1.35	1.351	+ 0.1
22	34.1	1.38	1.381	+ 0.1

(a) The density is expected to be insensitive to temperature, provided there is no salt precipitation, and was measured at ambient temperature that varied between 17 and 21°C

Table 4.4 gives the viscosities of the glycerol and sodium thiosulfate solutions as a function of temperature. To allow viscosities to be estimated for intermediate temperatures and concentrations, a

simple model was used to correlate the viscosity data. Base on literature data for similar solutions, exponential dependencies were used for the glycerol and sodium thiosulfate concentrations and for the effect of temperature. This model was fit to the data in Table 4.4 using a least-squares method and the resulting correlation is given below.

$$\mu_{gs} = 0.2904(e^{7.838 x_g})(e^{9.571 x_s})(e^{-0.0398 T}) \quad (4.1)$$

where μ_{gs} = viscosity of aqueous glycerol and sodium thiosulfate solutions
 x_g = mass fraction glycerol in bulk mixture
 x_s = mass fraction of sodium thiosulfate in bulk mixture
T = Temperature (°C)

Table 4.4. Effect of Temperature on the Viscosity of Aqueous Solutions of Glycerol and Water

Glycerol (wt%)	Na ₂ S ₂ O ₃ (wt%)	Temp (°C)	Measured Viscosity (mPa·s)
17	34.1	15	15.88
		20	12.80
		25	10.61
		30	8.98
17	32.7	15	13.70
		20	11.22
		25	9.38
		30	7.98
22	34.1	20	19.39
		25	15.46
		30	12.95
22	32.7	15	20.75
		20	16.61
		25	13.61
		30	11.59
19.5	33.4	15	17.79
		20	14.63
		25	12.20
25.5	31.8	30	10.04
		15	25.19
		20	19.90
		25	16.31
		30	13.58

Figure 4.2 shows the viscosity data and correlation results for the four solutions with a density of 1.37 g/mL. The final recipe for the high-viscosity/high-density target is a solution of 19.5 wt% glycerol and 33.4 wt% sodium thiosulfate, and this solution gives a viscosity that matches the target of 15 mPa·s at 20°C. For higher or lower test temperatures, different recipes can be selected to achieve the 15 mPa·s target.

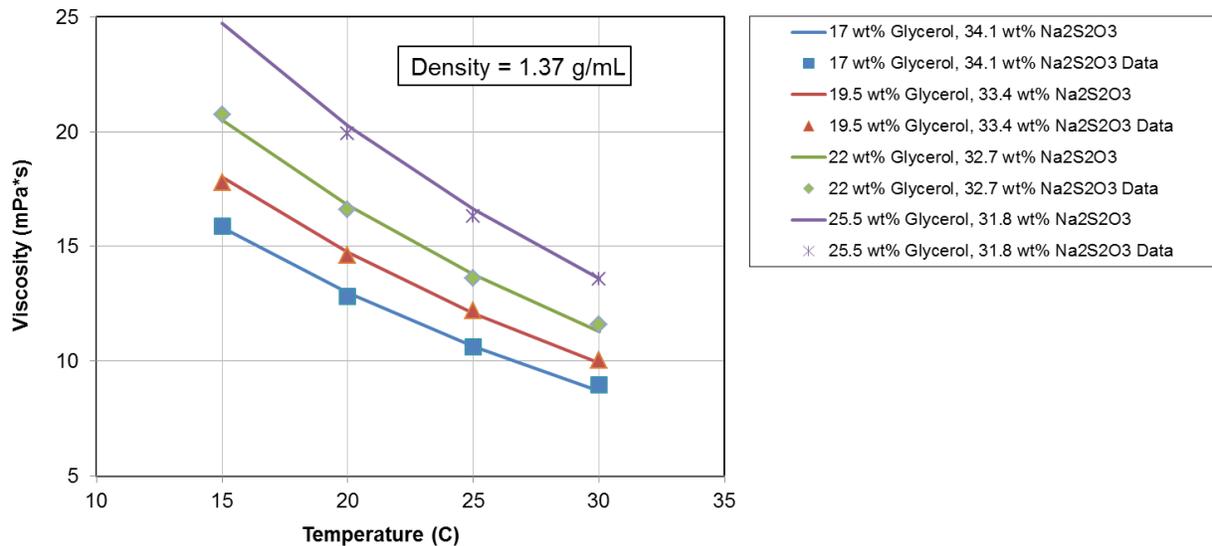


Figure 4.2. Experimental Results and Model Fit for the Effect of Temperature on Viscosity with the Solution Composition Adjusted to give a Density of 1.37 g/mL

4.2 Compatibility of Base and Spike Particles in Newtonian Simulants

Two qualitative evaluations were conducted to evaluate whether the base and spike particles are compatible with the Newtonian fluids. For the first evaluation, the settling behavior of the particles was measured in each Newtonian simulant. These settling tests were repeated for four successive days to detect any significant changes in settling behavior over time. Ideally, the base and spike particles will settle based on the size and density of the particles and the viscosity and density of the suspending fluid. For the second evaluation, the intensity of mixing needed to resuspend layers that had settled overnight, or for seven days, was determined. The resuspension tests give an indication of unexpectedly strong settled layers that might form in test tanks, piping, and pumps if the slurry is not continuously mixed.

Figure 4.3 shows an example of the settling behavior for the low base simulants for the first day of settling experiment (see Table 3.3 and Table 3.5 for small gibbsite). The low-base simulant particles all settle, though at different rates, in each of the Newtonian simulants with the exception of the low-density/high-viscosity liquid (curve labeled as lo_hi_lo), which did not show an observable settling layer. The addition of 0.1 wt% sodium thiosulfate to this glycerol-water solution changed the particle behavior and a settling layer below clear liquid was observed. The settling behavior of the final low-density/high-viscosity recipe (curve labeled as lo_hi_lo+0.1wt% Na₂S₂O₃) is also shown in Figure 4.3. For the low-base simulant and the five final Newtonian recipes (see Table 4.1), the settling behavior shows reasonable results for the low-base particle in the different viscosity and density liquids. The settling behavior was measured on four consecutive days and the settling results were essentially the same on each day. Equivalent settling tests were conducted with the high-base simulant in the five Newtonian liquids. For the high-base simulant, it was difficult to observe a settling upper layer though the rapidly settling

particles were easily seen collecting at the bottom of the graduated cylinders. In these tests, the low-density/high-viscosity simulant again seemed unusual and always remained cloudy during the day-long settling tests. The revised recipe with 0.1 wt% sodium thiosulfate gave improved and more consistent behavior.

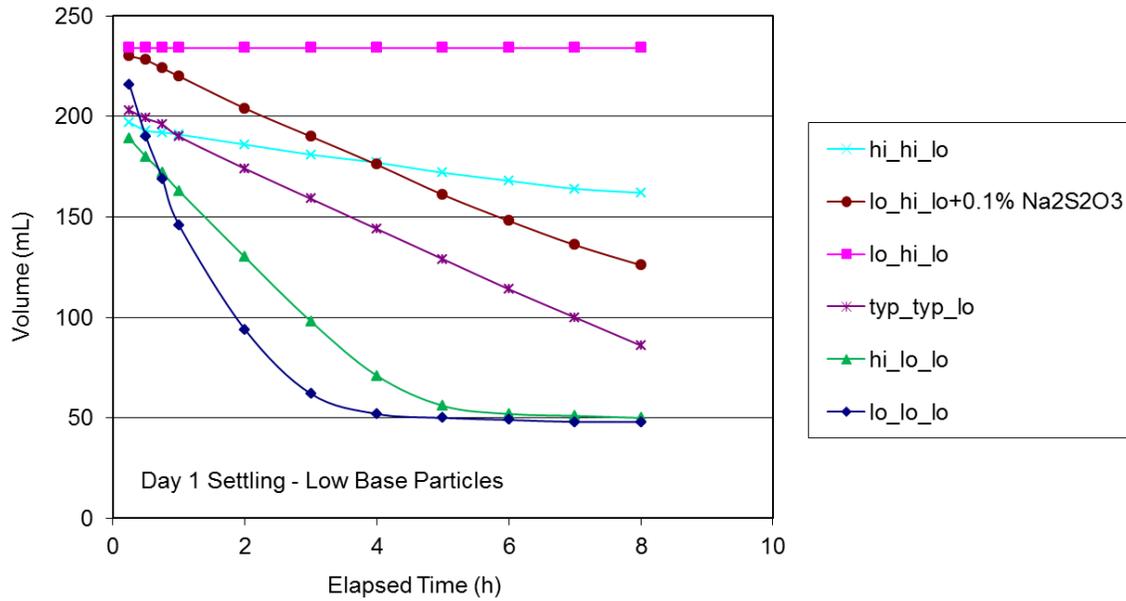


Figure 4.3. Settling Behavior of Low-Base Simulant Particles in the Newtonian Liquids

Table 4.5 shows the results of the resuspension tests for the base and spike particles in the Newtonian liquids. The results for the low-density/high-viscosity liquid without 0.1 wt% sodium thiosulfate show the difficulty in resuspending settled layers of both low-base and high-base particles in this liquid. The final recipe for the low-density/high-viscosity target with the addition of 0.1 wt% sodium thiosulfate has improved resuspending behavior, especially for the low-base particles. For the low-density/low-viscosity and low-density/high-viscosity fluids, the results show that it is more difficult to resuspend settled layers of the low-base particles than settled layers of the high-base particles. In contrast, the results for the typical-density/typical-viscosity liquid show that it is more difficult to resuspend a settled layer of the high-base particles. Overall, these resuspension testing results give a qualitative indication of the potential for strong settled layers that might form in tanks, piping, and pumps if the slurry is not continuously mixed.

Table 4.5. Resuspension Results for Newtonian Liquids with Base and Spike Particles

	Low Density Low Viscosity	Low Density High Viscosity	Low Density High Viscosity (0.1 wt% Na ₂ S ₂ O ₃)	High Density Low Viscosity	High Density High Viscosity	Typical Density Typical Viscosity
	High Base	High Base	High Base	High Base	High Base	High Base
Day 1	1+	2+	2	1+	-	2
Day 2	1	2	2	2	2	2
Day 3	1	2	2	1	2	1+
Day 4	1	2	2	1+	2	2
Day 7	1	1 (20)	2	1	1 (16)	1

	Low Density Low Viscosity	Low Density High Viscosity	Low Density High Viscosity (0.1 wt% Na ₂ S ₂ O ₃)	High Density Low Viscosity	High Density High Viscosity	Typical Density Typical Viscosity
	Low Base	Low Base	Low Base	Low Base	Low Base	Low Base
Day 1	1+	2	2	1	2	1
Day 2	2	3+	2	1	2	1
Day 3	2	3+	2	1	2	1
Day 4	3	3++	2	1	1+	1
Day 7	2+	3	2	1+	1+	2

1 - rock vertical to horizontal gently at least 20 times - heavy SS beads are not stuck to the bottom

2 - shake gently side to side ~2 shakes per second at least 20 times - heavy SS beads are not stuck to the bottom

3 - shake vigorously side to side ~4 shakes per second at least 20 times - heavy SS beads are not stuck to the bottom

"+" - did not suspend at specific level but suspended 1 to 2 iterations into the next level

(#) – number in parentheses indicates the number of rotations or shakes when motion occurred for specific tests that were very close to the next-higher level of intensity

4.3 Non-Newtonian Simulant Density and Rheology

Figure 4.4 and Figure 4.5 compare the Bingham yield stress and Bingham consistency, respectively, as a function of weight percent of kaolin for various aqueous slurries of kaolin and sodium thiosulfate with a controlled slurry density of 1.37 g/mL that were prepared by different mixing methods that varied the shear rate and mixing duration. The KitchenAid mixing represents a low-shear mixing method while the Magic Bullet mixing represents a high-shear mixing method. The recommended recipes of non-Newtonian kaolin-sodium thiosulfate simulants are represented by the green filled-triangles. The results in Figure 4.4 and Figure 4.5 show that increasing the mixing shear rate would noticeably increase

the Bingham yield stress but would only slightly increase the Bingham consistency of these selected non-Newtonian kaolin-thiosulfate simulants. This implies that variations in measured Bingham yield stress of the demonstration testing samples using the recommended recipes may be expected due to different mixing shear rates. The intensity of shearing in the demonstration testing is not known, so changes to the slurry concentrations may be needed to achieve the desired Bingham yield stress targets.

Figure 4.6 and Figure 4.7 compare the Bingham yield stress and Bingham consistency, respectively, as a function of weight percent of kaolin, for various aqueous slurries of kaolin with a slurry density ranging from 1.13 to 1.19 g/mL that were prepared by different mixing methods that varied the mixing shear rate. Unlike the case of the aqueous kaolin-thiosulfate simulants, increasing mixing shear rate would noticeably increase both the Bingham yield stress and the Bingham consistency of these selected non-Newtonian kaolin simulants. This implies that variations in measured Bingham yield stress and Bingham consistency of the demonstration testing samples using the recommended recipes may be expected due to different mixing shear rates. The intensity of shearing in the demonstration testing is not known, so changes to the slurry concentrations may be needed to achieve the desired Bingham yield stress targets.

Figure 4.8 and Figure 4.9 compare the Bingham yield stress and Bingham consistency, respectively, as a function of weight percent of kaolin for various slurries of kaolin in water, and with sodium thiosulfate and sodium bromide. For the kaolin slurries with salt, the bulk slurry density is near the high-density target of 1.37 g/mL. The results in Figure 4.8 and Figure 4.9 show that for an equal weight percent of kaolin in aqueous kaolin slurry, addition of sodium bromide or sodium thiosulfate salt increases the Bingham yield stress and Bingham consistency of the slurry. The sodium thiosulfate addition produces a greater increase in the Bingham properties of the kaolin slurry than the addition of sodium bromide.

Figure 4.10 shows the Bingham consistency as a function of Bingham yield stress for various slurries of kaolin in water, and with sodium thiosulfate and sodium bromide. This data shows that at the same Bingham yield stress, sodium thiosulfate addition gives a greater increase in the Bingham consistency than the addition of sodium bromide. Accordingly, additions of different salts would allow parametric testing to isolate the role of the Bingham consistency of aqueous kaolin clay slurries.

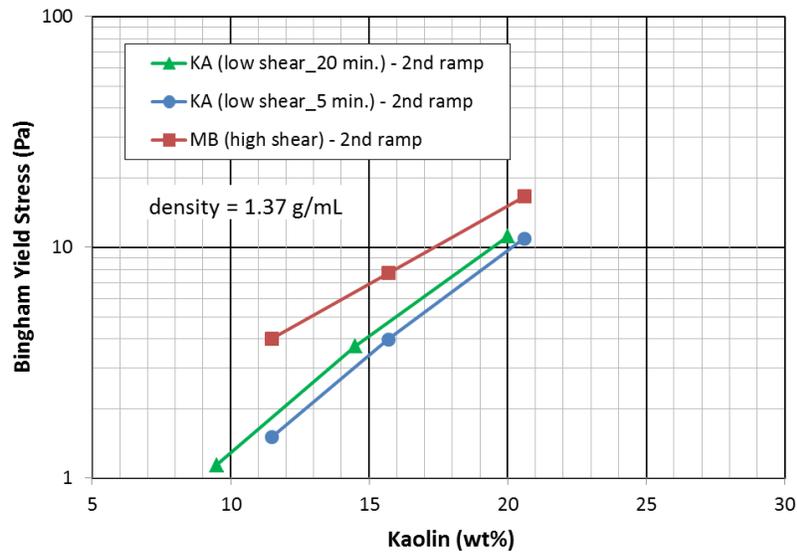


Figure 4.4. Bingham Yield Stress for Aqueous Slurries of Kaolin and Sodium Thiosulfate with a Slurry Density of 1.37 g/mL

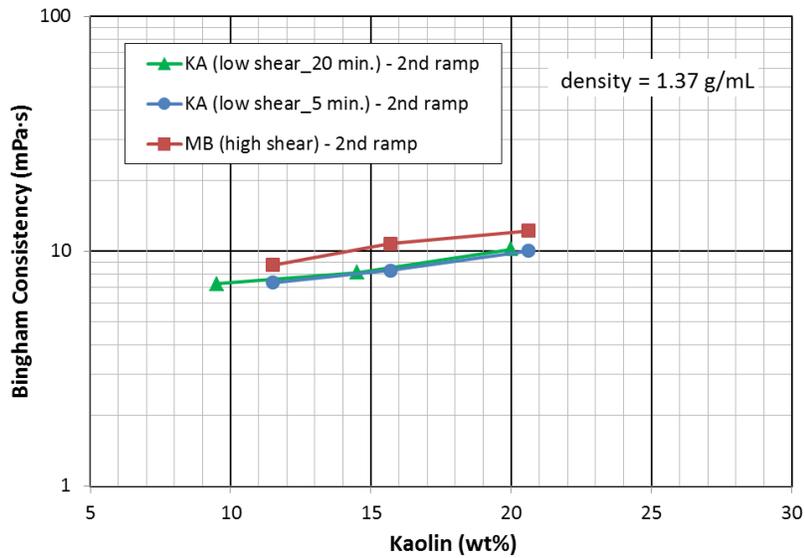


Figure 4.5. Bingham Consistency for Aqueous Slurries of Kaolin and Sodium Thiosulfate with a Slurry Density of 1.37 g/mL

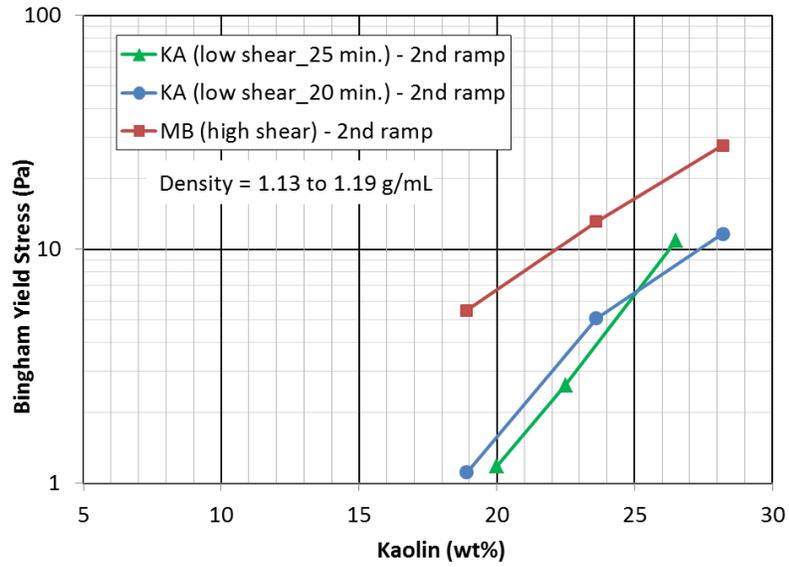


Figure 4.6. Bingham Yield Stress for Aqueous Slurries of Kaolin with Slurry Densities that Vary from 1.13 to 1.19 g/mL

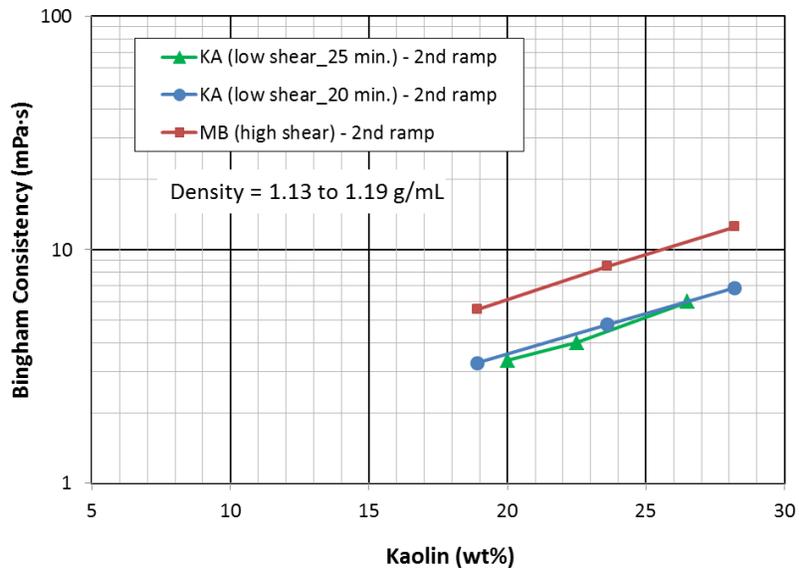


Figure 4.7. Bingham Consistency for Aqueous Slurries of Kaolin with Slurry Densities that Vary from 1.13 to 1.19 g/mL

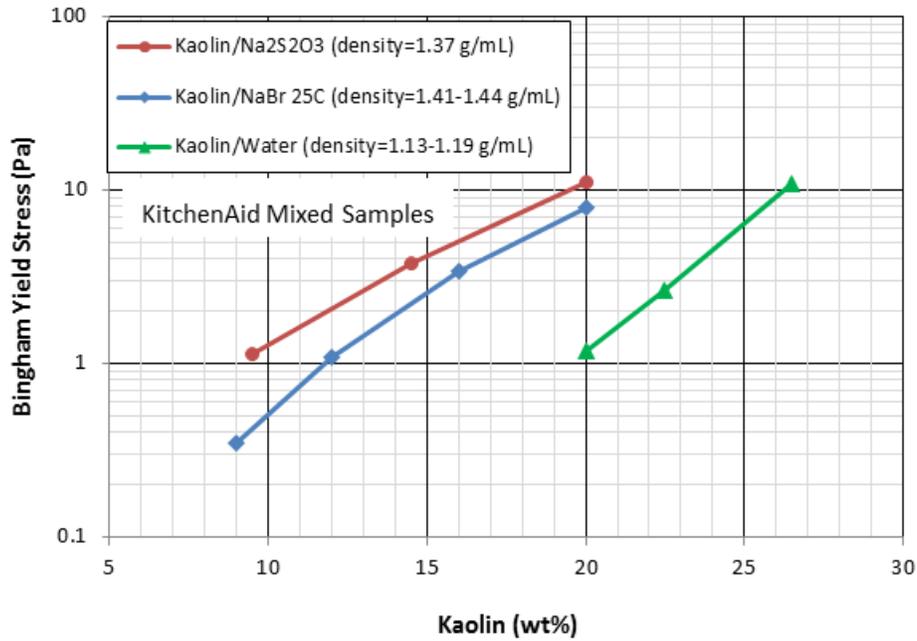


Figure 4.8. Comparison of Bingham Yield Stresses for Kaolin Slurries in Water and with Sodium Thiosulfate and Sodium Bromide

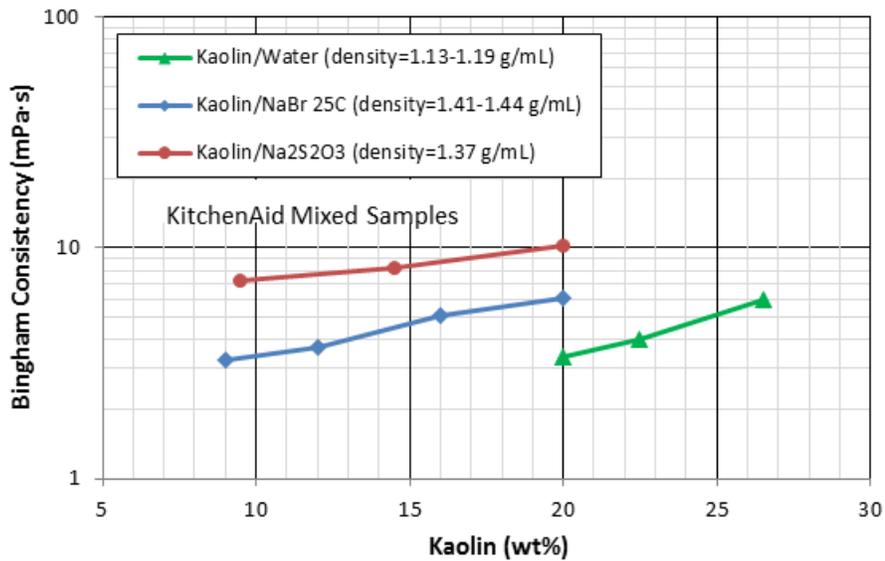


Figure 4.9. Comparison of Bingham Consistencies for Kaolin Slurries in Water and with Sodium Thiosulfate and Sodium Bromide

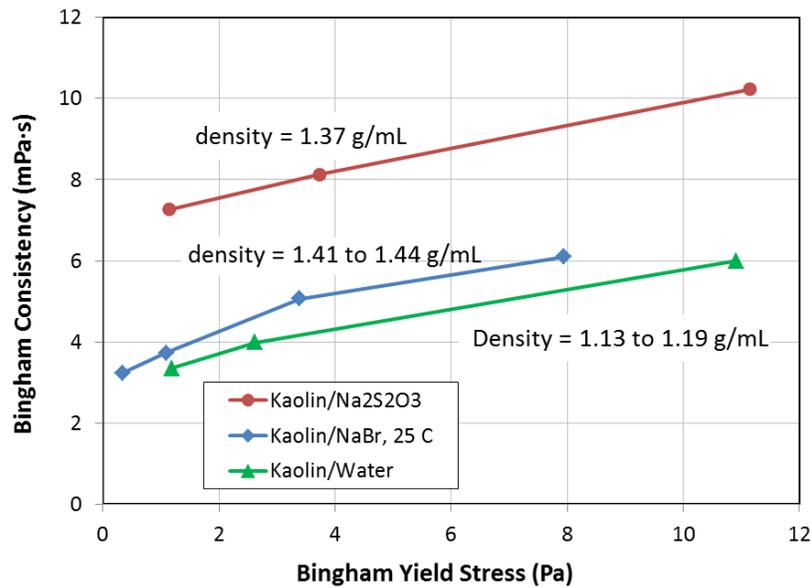


Figure 4.10. Bingham Consistency versus Yield Stress for Kaolin Slurries in Water and with Sodium Thiosulfate or Sodium Bromide

Table 4.6 shows the effect of temperature on Bingham yield stress and Bingham consistency for the recommended recipes of non-Newtonian kaolin-sodium thiosulfate simulants. The result shows essentially no effect of temperature on the Bingham yield stress and minimal temperature effect on Bingham consistency for these simulants. However, the small temperature effect on the Bingham consistency is insignificant because the variations are within an acceptable range of experimental error. In addition, there was no salt precipitation observed in the temperature stability studies described in Section 3.6. This result indicates the stability of the recommended non-Newtonian kaolin-sodium thiosulfate simulants for the potential range of test temperatures.

Table 4.6. Effect of Temperature on Bingham Yield Stress and Consistency for Slurries of Kaolin and Sodium Thiosulfate in Water

Temp (°C)	Simulant Composition: Kaolin/Na ₂ S ₂ O ₃ in Pasco City Water					
	9.5 wt% kaolin 29.6 wt% Na ₂ S ₂ O ₃		14.5 wt% kaolin 24.9 wt% Na ₂ S ₂ O ₃		20 wt% kaolin 19.9 wt% Na ₂ S ₂ O ₃	
	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)
20	1.1	7.3	3.7	8.1	11	10.2
25	1.1	6.2	3.4	7.3	10	10.0
30	1.1	5.6	3.5	6.9	11	8.5

Table 4.7 presents the effect of temperature on Bingham yield stress and Bingham consistency for the recommended recipes of non-Newtonian aqueous kaolin simulants. The experiment was done in duplicate to verify the results and the testing protocol. Both data sets show essentially no effect of temperature on the Bingham yield stress and Bingham consistency for these non-Newtonian aqueous

kaolin simulants. The small variations in the Bingham yield stress and Bingham consistency measurements are within an acceptable range of experimental error. This result indicates the stability of the recommended non-Newtonian aqueous kaolin simulants for the potential range of test temperatures.

Table 4.7. Effect of Temperature on Bingham Yield Stress and Consistency for Slurries of Kaolin in Water

Simulant Composition: Kaolin in Pasco City Water						
Temp (°C)	20.0 wt% kaolin		22.5 wt% kaolin		26.5 wt% kaolin	
	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)	Bingham Yield Stress (Pa)	Bingham Consistency (mPa·s)
20	1.6	3.5	3.4	4.3	11	6.5
	1.3	3.3	3.3	4.0	12	6.1
25	2.2	3.6	5.0	4.4	11	5.8
	1.3	3.1	3.9	3.7	12	6.0
30	1.7	3.1	4.8	3.9	11	5.6
	1.6	3.3	5.2	4.1	14	5.8

4.4 Compatibility of Spike Simulant Particles in Non-Newtonian Slurries

Table 4.8 shows the resuspension test results for spike particles in one slurry of kaolin and sodium thiosulfate and one slurry of kaolin in water. Because there was a very small quantity of the spike particles, an additional test was done with 100-fold higher quantity of the spike particles in the kaolin-sodium thiosulfate simulant to have a better opportunity to evaluate the compatibility of the spike particles and the slurry. The results in Table 4.8 show that all the slurries were easily resuspended and there is no indication of unexpected behavior such as unexpectedly strong settled layers.

Table 4.8. Resuspension Results for Spike Particles in Non-Newtonian Simulant

	14.5% Kaolin 24.9% Na ₂ S ₂ O ₃	14.5% Kaolin 24.9% Na ₂ S ₂ O ₃ (100x Spike)	22.5% Kaolin
Day 1	1	1	1
Day 2	0+	0+	0+
Day 3	0+	1	1
Day 4	0+	0+	1
Day 7	0+	1	0+

0 - swirl in a circular motion - heavy SS beads are not stuck to the bottom (applicable to bottle 7-day test only)

1 - rock vertical to horizontal gently at least 20 times - heavy SS beads are not stuck to the bottom

"+" - did not suspend at specific level but suspended 1 to 2 iterations into the next level

4.5 Non-Newtonian Kaolin Slurry Particle Size Distributions

The addition of sodium thiosulfate or sodium bromide significantly increased the Bingham yield stress of the kaolin slurries, as shown in Figure 4.8, in addition to increasing the slurry density. An overall objective of the planned demonstration tests is to compare testing results using different non-Newtonian and Newtonian fluids (Lee 2012a, 2012b, 2012c). While many previous studies have used kaolin slurries, and also bentonite and kaolin/bentonite mixtures, for waste simulants (Lee et al. 2012), there are no studies that have evaluated the suitability of kaolin slurries with high salt concentrations for use as waste simulants. For the planned demonstration tests, it is important that the addition of sodium thiosulfate does not change the kaolin slurry in an unexpected manner that makes the recipes with kaolin and salt a poor selection for a non-Newtonian waste simulant. An additional slurry characterization method that can be used to evaluate the role of salt on the slurry is to measure the PSD of the kaolin particles with and without sodium thiosulfate; a summary of these measurements is given below. The rheology results in Section 4.3 also showed a significant effect of preshearing on the rheology of the slurries. PSD measurements comparing different intensities of preshearing can be helpful in understanding how shearing in the pumps and flow system for the demonstration tests might change the rheology of the kaolin slurries. PSD results for different slurry preshearing are also shown below. More detailed PSD data are provided in Appendix C for the results shown in this section.

Figure 4.11 shows a comparison of PSDs for all the kaolin slurries where the samples were sonicated prior to the PSD measurement to disperse aggregates. Sonicated samples generally give results showing the PSD of the primary particles. Figure 4.11 shows similar PSDs regardless of whether there was no preshearing (Hand Kaolin/Sonicated) or preshearing was done with only the KitchenAid mixer (KA/KA Kaolin/Sonicated) or with the Magic Bullet (KA/MB Kaolin/Sonicated). Additionally, the hand-mixed kaolin slurry without preshearing shows essentially identical PSD to that of the KitchenAid-mixed followed by KitchenAid-presheared slurry. The results also show a more narrow distribution for the slurries with sodium thiosulfate but there was no shift in the PSD to lower or higher particle sizes with the added salt. Figure 4.12 shows a similar comparison for unsonicated slurries; the addition of sodium thiosulfate has only a minor effect on the kaolin PSD. Typically, a reduction in the PSD corresponds to an increase in the Bingham yield stress for a slurry. Accordingly, because the sodium thiosulfate did not shift the PSD of the kaolin, the addition of the salt increases the rheology of the kaolin slurries by some other mechanism such as particle attractions, or equivalently making the kaolin particles more cohesive.

Figure 4.13 shows the effect of preshearing intensity on the PSD of unsonicated slurries of kaolin in water and kaolin with sodium thiosulfate addition. These results show that preshearing the kaolin slurry with the Magic Bullet, which provides higher shearing intensity than that provided by the KitchenAid mixer, shifts the PSD to smaller sizes. A reduction in PSD is consistent with the observation of increased Bingham yield stress with the Magic Bullet preshearing. Moreover, the KitchenAid preshearing shows essentially no effect on the PSD of the kaolin slurry as compared to that of the hand-mixed slurry without preshearing. Figure 4.14 shows that sonication also shifts the PSD to smaller sizes and this shift is similar to the reduction caused by the Magic Bullet preshearing. Finally, Figure 4.15 shows that sonication does not further reduce the PSD of samples presheared with the Magic Bullet, so the Magic Bullet preshearing probably gives the smallest PSD and highest Bingham yield stress for strong preshearing.

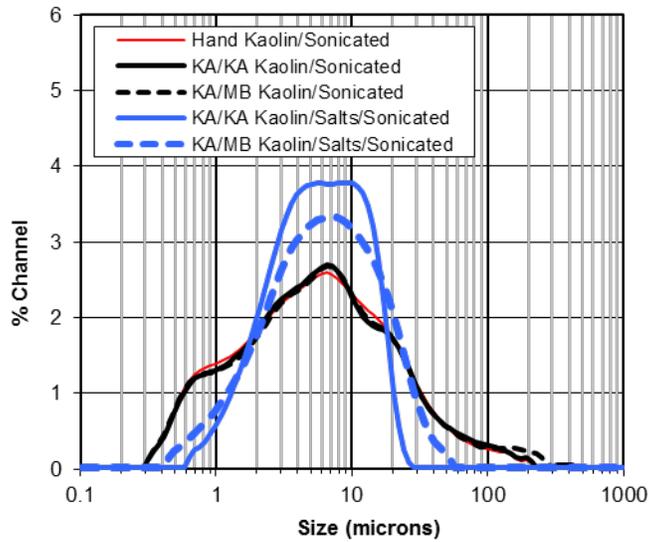


Figure 4.11. Kaolin PSDs for all Sonicated Slurries having Different Preshearing for Slurries of Kaolin in Water and Kaolin in Sodium Thiosulfate Solutions

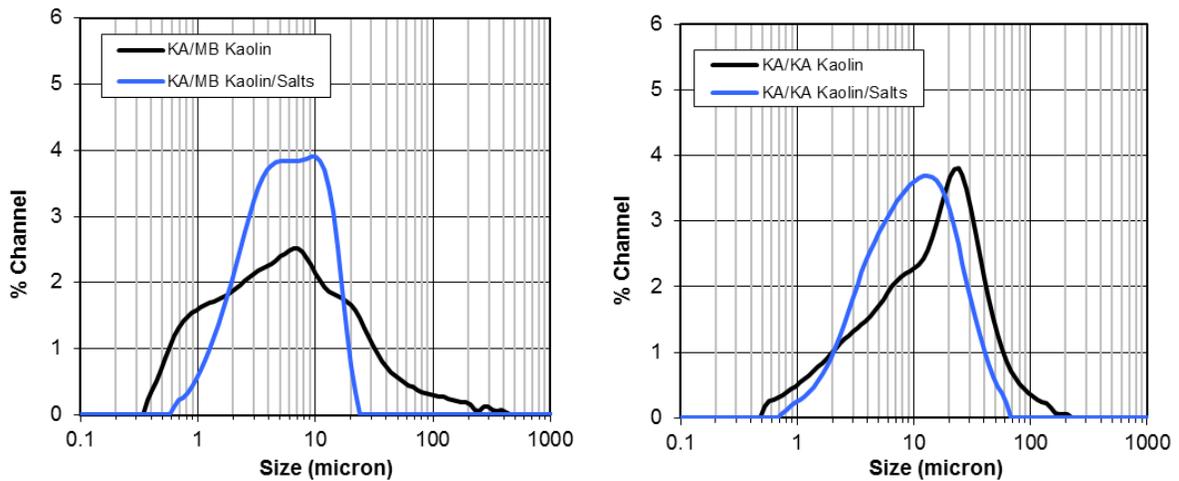


Figure 4.12. Effect of Sodium Thiosulfate on Kaolin PSDs for Unsonicated Kaolin Slurries with Different Preshearing Left: KitchenAid-Mixed/Magic Bullet-Presheared; Right: KitchenAid-Mixed/KitchenAid-Presheared

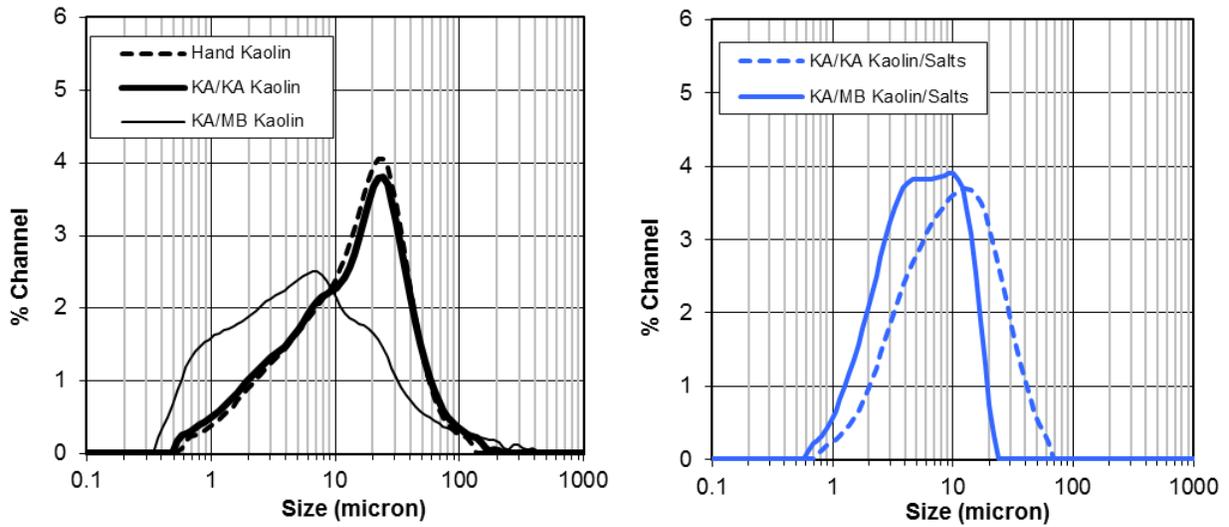


Figure 4.13. Effect of Preshearing on Kaolin PSDs for Unsonicated Slurries of Kaolin in Water (left) and Kaolin in Sodium Thiosulfate Salt Solution (right)

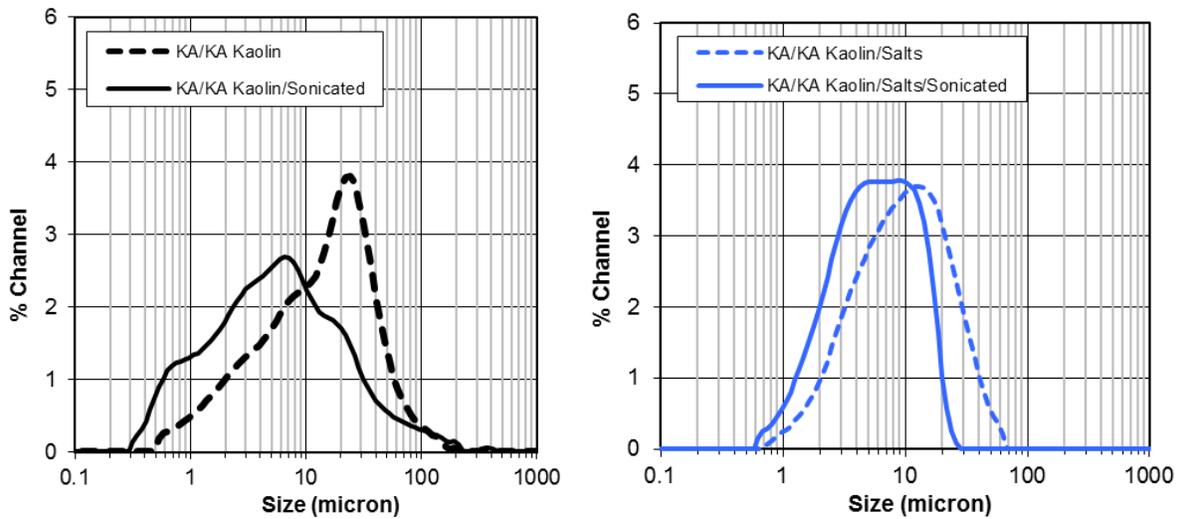


Figure 4.14. Effect of Sonication on Kaolin PSDs for Slurries of Kaolin in Water (left) and Kaolin in Sodium Thiosulfate Salt Solution (right)

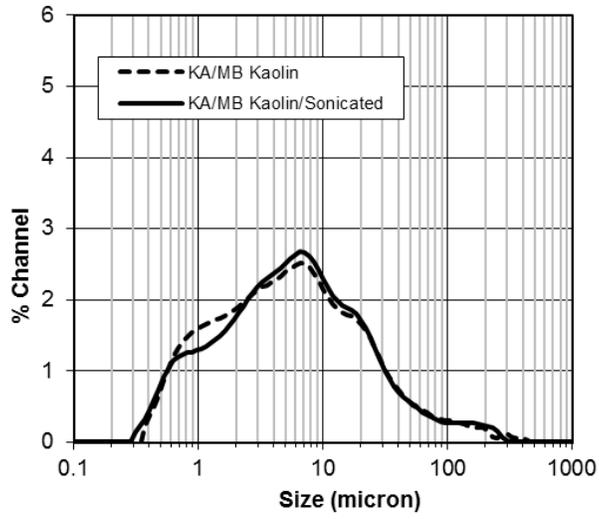


Figure 4.15. Effect of Sonication on Kaolin PSDs for Slurries of Kaolin in Water Presheared with the Magic Bullet Mixer

5.0 Summary and Conclusions

This report summarizes the results of laboratory studies on small trial simulant mixtures to determine specific simulant recipes, to evaluate the compatibility of the liquids and particles, and to determine whether the simulants are stable for the potential range of test temperatures. The objective of the testing, which is focused primarily on the Newtonian and non-Newtonian suspending fluids, is to determine the composition of selected simulant materials that give the desired density and viscosity or rheological parameters.

For the Newtonian liquids, simulant recipes were developed using aqueous solutions of sodium thiosulfate or sodium thiosulfate and glycerol to match the density and viscosity targets for four of the five targets. Sodium thiosulfate was the preferred salt because it is nonhazardous and inexpensive. An aqueous solution of sodium bromide, which gives uniquely low viscosities in concentrated solutions in comparison to other salt solutions, was selected as a preferred material for a high-density/low-viscosity target. The effect of temperature on viscosity was evaluated because the glycerol/thiosulfate solutions are somewhat temperature sensitive. All of these solutions were stable (no salt precipitation after about a day) down to 10°C.

There was only one liquid/particle compatibility issue observed during the testing, and this was when one type of the gibbsite material was added to a solution of glycerol in water (low-density/high-viscosity target). For this mixture, slurries always stayed cloudy during a day of settling, and clear liquid above a settling layer, which is the expected behavior, was not observed. This mixture would still give settled layers, and these layers were quite difficult to resuspend. This recipe was reformulated using 0.1 wt% sodium thiosulfate, which altered the particle behavior, and the settling and resuspension results were much improved.

For the non-Newtonian slurries, simulant recipes were developed using slurries of kaolin clay in water or kaolin clay in sodium thiosulfate solutions. For the kaolin in sodium thiosulfate solutions, the proportions of both the sodium thiosulfate and kaolin were adjusted to obtain slurries at a constant density (matching the high-density Newtonian target) for Bingham yield stresses between 1 and 10 Pa. For the kaolin-in-water slurries, the density was not adjusted but was comparable to the low-density Newtonian target as desired. The effect of temperature on the Bingham yield stress and consistency were determined and slurries were stable (no salt precipitation after about a day) down to 10°C. Compatibility tests with spike particles did not identify any concerns for slurries of either kaolin clay in water or kaolin clay in sodium thiosulfate solutions. PSD measurements of the kaolin slurries showed that both intense preshearing and sonication reduce the PSD of the kaolin particles and the addition of sodium thiosulfate does not shift the PSD of the kaolin.

6.0 References

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Appendix A

Simulant Materials Provided to Pacific Northwest National Laboratory

Appendix A

Simulant Materials Provided to Pacific Northwest National Laboratory

Pacific Northwest National Laboratory received batches of dry simulant particles, dry salts, glycerol, and Pasco city water from Energy Solutions. The figures below show the Chain of Custody forms for these materials and give a brief description of each material.

C H A I N O F C U S T O D Y					
Chain of Custody No.: ES-SSMD-064	Chain of Custody Date: 2012-05-17	Chain of Custody No. issued by (Name): Gregg Dillingham	Ship to: Phil Gauglitz APEL 350 Hills Street, Richland, WA	Phone: 509.372.5146	
Simulant(s): the samples listed below contain these simulant constituents				Purchase Order #: N/A	
<ol style="list-style-type: none"> 1. Sand (Large, NJ6), US Silica Company, Lot #BKBG 1168958-1 2. Gibbsite (Small), Nabaltec, Lot #0230102/1.1 3. Stainless Steel Powder, Pellets LLC, Lot #7/8 4. Soda Lime Glass Beads, Walter Stern Inc, No Lot # 5. 1/16" Stainless Steel, Pellets LLC, Lot #L632-4-29 					
Sample Date	Sample Identification	Sample Description	Quadrant	Analyze Sample for:	
2012-05-17	SSMD-800	1.0 kg Large Sand	N/A	N/A	
2012-05-17	SSMD-801	2.0 kg Small Gibbsite	N/A	N/A	
2012-05-17	SSMD-802	1.0 kg Stainless Steel Powder	N/A	N/A	
2012-05-17	SSMD-803	0.5 kg 2mm Soda Lime Glass Beads	N/A	N/A	
2012-05-17	SSMD-804	0.5 kg 1/16" Stainless Steel	N/A	N/A	
Sent By (Signature) / Organization: Energy Solutions		Date: 5-18-12	Time: 1035	Received By (Signature) / Organization: - PNNL	
				Date: 5/18/12 10:40AM	Time: 10:40AM

A.2

Figure A.1. Chain of Custody Form for Sand, Gibbsite, Stainless Steel Powder and Pellets, and Soda Lime Beads

C H A I N O F C U S T O D Y

Chain of Custody No.: ES-SSMD-062	Chain of Custody Date: 2012-05-07	Chain of Custody No. issued by (Name): Lester Jollie	Ship to: Phil Gauglitz APEL 350 Hills Street, Richland, WA	Phone: 509.372.5146
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Simulant(s): the samples listed below contain these simulant constituents 1. Glycerin USP, Silver Fern, Lot # 12-2147 2. Sodium Thiosulfate, Brainerd Chemical Company, Lot # 120101 3. Pasco City Water, no Lot	Purchase Order #: N/A
--	-------------------------------------

Sample Date	Sample Identification	Sample Description		Analyze Sample for:
			Quadrant	
2012-05-07	SSMD-790	Sodium Thiosulfate (33.016 lb)	N/A	N/A
2012-05-07	SSMD-791	Glycerin-USP (22.035 lb)	N/A	N/A
2012-05-07	SSMD-792	Pasco City Water (~5 gallons)	N/A	N/A
2012-05-07	SSMD-793	Pasco City Water (~5 gallons)	N/A	N/A
2012-05-07	SSMD-794	Pasco City Water (~5 gallons)	N/A	N/A
2012-05-07	SSMD-795	Pasco City Water (~5 gallons)	N/A	N/A

Sent By (Signature) / Organization: Energy Solutions <i>L Jollie</i>	Date: 2012-05-07	Time: 0945	Received By (Signature) / Organization: <i>W Buehler</i>	Date: 5/7/11	Time: 1011
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A.3

Figure A.2. Chain of Custody Form for Glycerin, Sodium Thiosulfate, and Pasco City Water

C H A I N O F C U S T O D Y

Chain of Custody No.: ES-SSMD-063	Chain of Custody Date: 2012-05-10	Chain of Custody No. issued by (Name): Rodney Jochen	Ship to: Phil Gauglitz APEL 350 Hills Street, Richland, WA	Phone: 509.372.5146
---	---	--	--	---------------------

Simulant(s): <u>the samples listed below contain these simulant constituents</u>	Purchase Order #:
<ol style="list-style-type: none"> EPK Kaolin, The Fledspar Corp., Lot K125116 Sand (small, Sil-Co-Sil 250), U.S. Silica Company, Lot # 50042012 Zirconium Oxide, Washington Mills Electro Minerals Corp, Lot# L-309077 Sodium Bromide, Albemarle Corp. Lot# SINOWT01090407 	N/A

Sample Date	Sample Identification	Sample Description		Analyze Sample for:
			Quadrant	
2012-05-10	SSMD-796	EPK Kaolin (22.0 lbs)	N/A	N/A
2012-05-10	SSMD-797	Sand (small, Sil-Co-Sil 250) (2.2 lbs.)	N/A	N/A
2012-05-10	SSMD-798	Zirconium Oxide (4.4 lbs.)	N/A	N/A
2012-05-10	SSMD-799	Sodium Bromide (6.6 lbs.)	N/A	N/A

Sent By (Signature) / Organization:  Energy Solutions G. Dillingham	Date: 5-10-12	Time: 1602	Received By (Signature) / Organization: 	Date: 5/10/12	Time: 1630
--	-------------------------	----------------------	--	-------------------------	----------------------

A.4

Figure A.3. Chain of Custody Form for EPK Kaolin, Sand, Zirconium Oxide, and Sodium Bromide

C H A I N O F C U S T O D Y

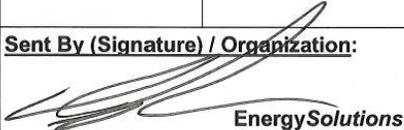
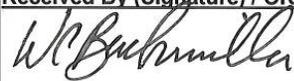
Chain of Custody No.: ES-RSD-051		Chain of Custody Date: 2012-04-09		Chain of Custody issued by (Name): Brian Brown		Ship to: APEL Richland Wa		Phone:	
Simulant(s): the samples listed below contain these simulant constituents 1. Sodium bromide Albemarle corp 2. Pasco city Water								Purchase Order #: N/A	
Sample Date	Sample Identification	Sample Description			Analyze Sample for:				
		Raw Simulant	Lot #	Quadrant					
2012-04-09	RSD-273	Sodium Bromide	SINOWT01090407	N/A	For analytical requirements contact Matt Vanatta 509-375-9520, mwvanatta@energysolutions.com.				
2012-04-09	RSD-272	Pasco City Water (no lot #)							
Sent By (Signature) / Organization:  Energy Solutions		Date: 4-9-12	Time: 1130	Received By (Signature) / Organization: 		Date: 4/9/12	Time: 1255		

Figure A.4. Chain of Custody Form for Sodium Bromide and Pasco City Water

A.5

C H A I N O F C U S T O D Y

Chain of Custody No.: ES-SSMD-067	Chain of Custody Date: 2012-06-11	Chain of Custody No. issued by (Name): Kristin Van Andel	Ship to: Phil Gauglitz APEL 350 Hills Street, Richland, WA	Phone: 509.372.5146
---	---	--	--	----------------------------

Simulant(s): the sample listed below contains this simulant constituent 1. Gibbsite 3431 (large), manufacturer: Huber, Lot No. 0230102/1.1, Noah Catalog No. R6011	Purchase Order #: N/A
--	-------------------------------------

Sample Date	Sample Identification	Sample Description	Quadrant	Analyze Sample for:
2012-06-11	SSMD-833	Gibbsite 3431 (large) (516grams)	N/A	N/A

Sent By (Signature) / Organization: <i>ke</i> Energy Solutions	Date: 6/11/12	Time: 13:15	Received By (Signature) / Organization: <i>Swanhaul</i> (PNNL)	Date: 6/11/12	Time: 1:50 pm
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A.6

Figure A.5. Chain of Custody Form for Gibbsite

C H A I N O F C U S T O D Y

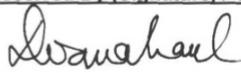
Chain of Custody No.: ES-SSMD-068		Chain of Custody Date: 2012-06-13		Chain of Custody No. issued by (Name): Kristin Van Andel		Ship to: Phil Gauglitz APEL 350 Hills Street, Richland, WA		Phone: 509.372.5146	
Simulant(s): the sample listed below contains this simulant constituent							Purchase Order #:		
1. Tungsten Carbide Balls (1/16"), Tungsten Heavy Powder, Inc., Lot No. RM120421-1							N/A		
Sample Date	Sample Identification	Sample Description		Analyze Sample for:					
			Quadrant						
2012-06-13	SSMD-834	Tungsten Carbide Balls (1/16") (500 grams)	N/A	N/A					
Sent By (Signature) / Organization:  Energy Solutions		Date: 6/13/12	Time: 14:00	Received By (Signature) / Organization:  PNNL		Date: 6/13/12	Time: 2:25 pm		

Figure A.6. Chain of Custody Form for Tungsten Carbide Balls

A.7

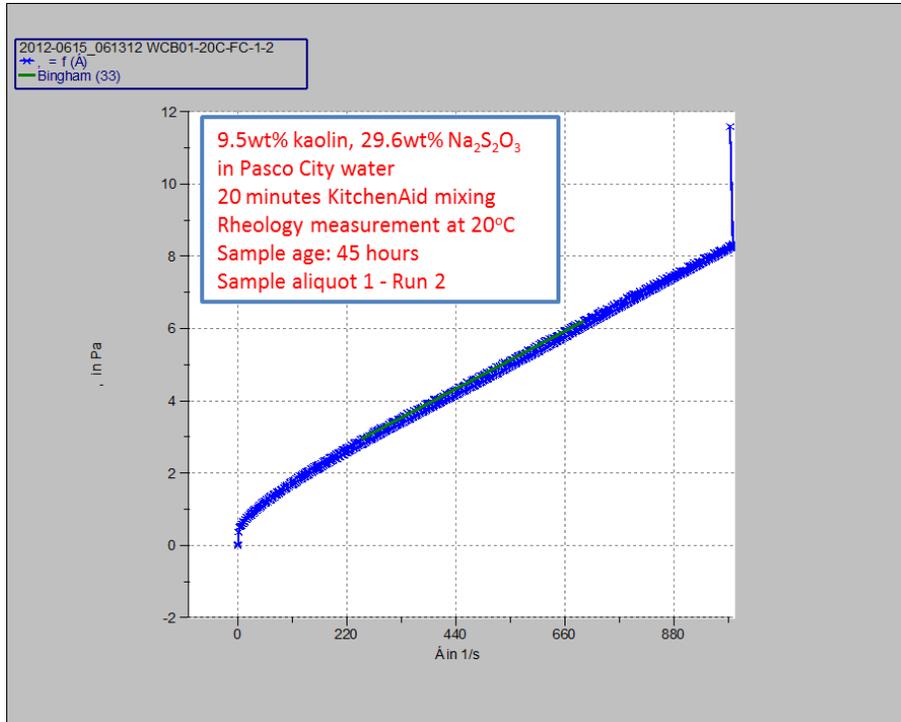
Appendix B

Rheograms for Kaolin Clay Slurries

Appendix B

Rheograms for Kaolin Clay Slurries

The measured rheograms for the final non-Newtonian kaolin clay simulant recipes are shown below. These plots are for the second down-ramp, and this data is used to determine the Bingham yield stress and consistency provided in Section 4.



FigureB.1. Rheogram (Second Down-Ramp) for 9.5 wt% Kaolin and 29.6 wt% $\text{Na}_2\text{S}_2\text{O}_3$ in Pasco City Water (Simulant Target Properties of 1 Pa, 1.37 g/mL)

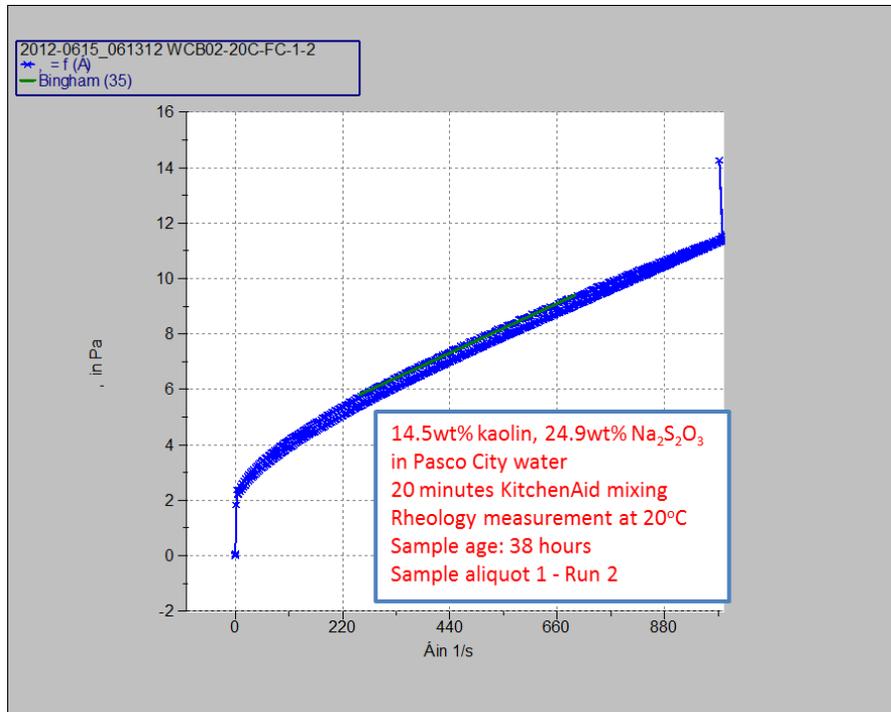


Figure B.2. Rheogram (Second Down-Ramp) for 14.5 wt% Kaolin and 24.9 wt% Na₂S₂O₃ in Pasco City Water (Simulant Target Properties of 3 Pa, 1.37 g/mL)

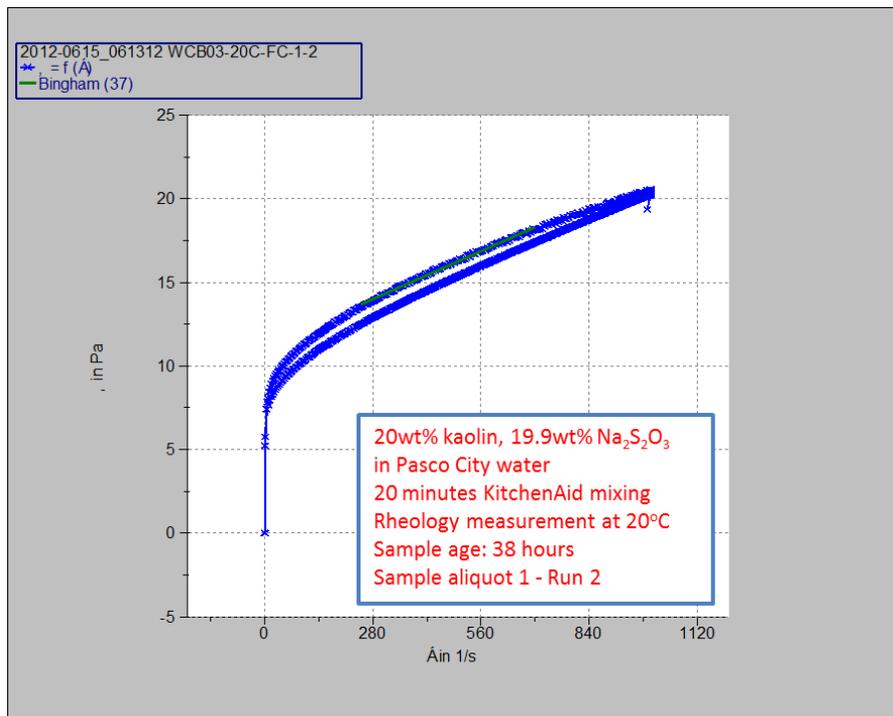


Figure B.3. Rheogram (Second Down-Ramp) for 20.0 wt% Kaolin and 19.9 wt% Na₂S₂O₃ in Pasco City Water (Simulant Target Properties of 10 Pa, 1.37 g/mL)

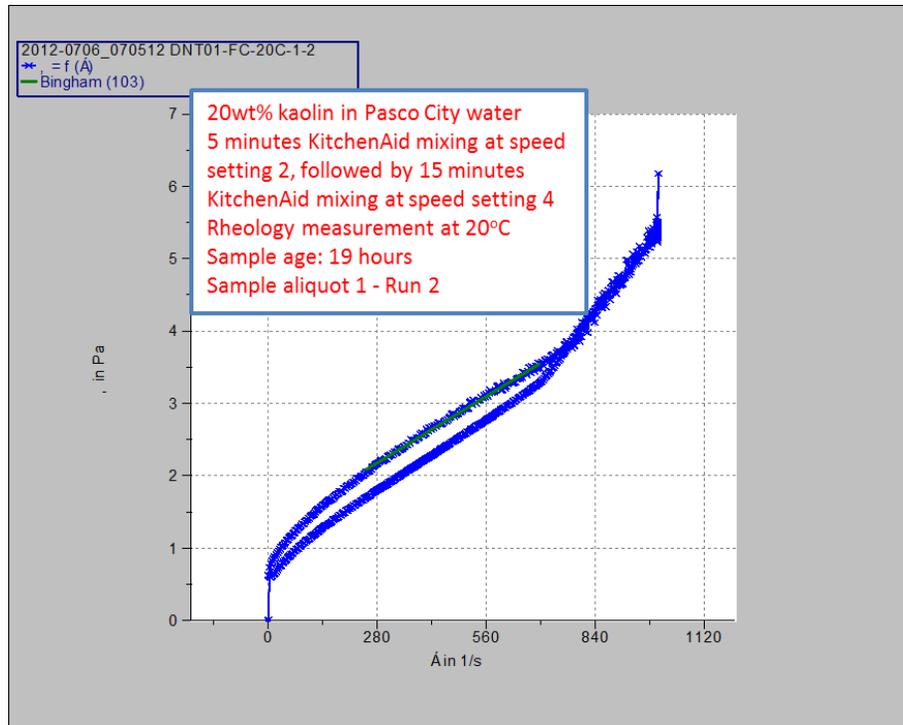


Figure B.4. Rheogram (Second Down-Ramp) for 20.0 wt% Kaolin in Pasco City Water (Simulant Target Properties of 1 Pa, 1.1 g/mL)

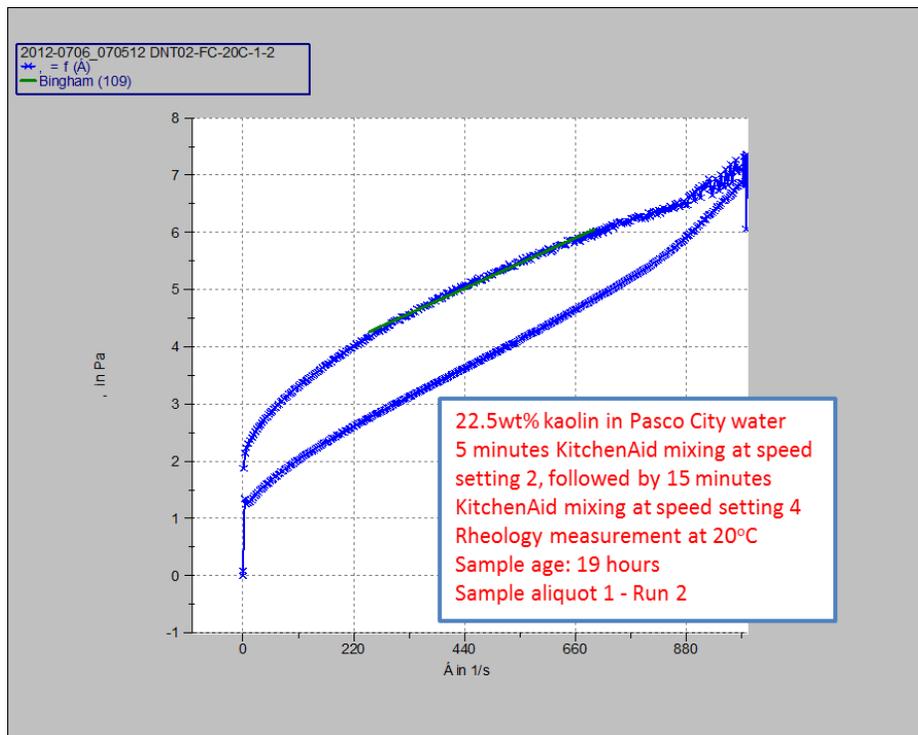


Figure B.5. Rheogram (Second Down-Ramp) for 22.5 wt% Kaolin in Pasco City Water (Simulant Target Properties of 3 Pa, 1.1 g/mL)

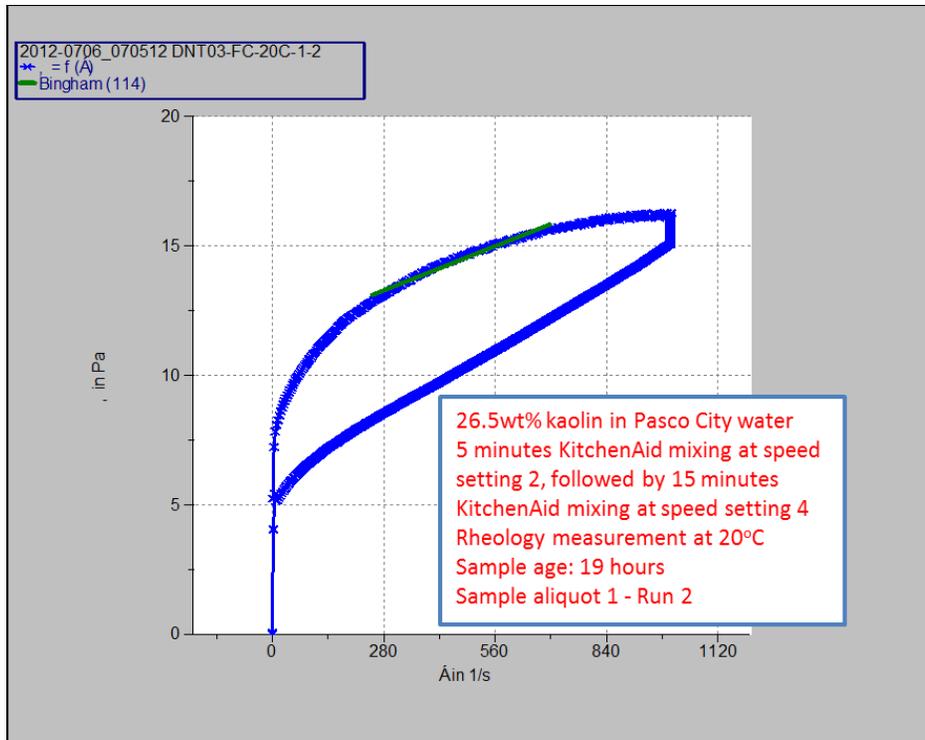


Figure B.6. Rheogram (Second Down-Ramp) for 26.5 wt% Kaolin in Pasco City Water (Simulant Target Properties of 10 Pa, 1.1 g/mL)

Appendix C

Particle Size Distributions for Kaolin Clay in Simulant Recipes

Appendix C

Particle Size Distributions for Kaolin Clay in Simulant Recipes

The figures below show the Microtrac instrument output for each of the kaolin slurry samples reported in Section 4.

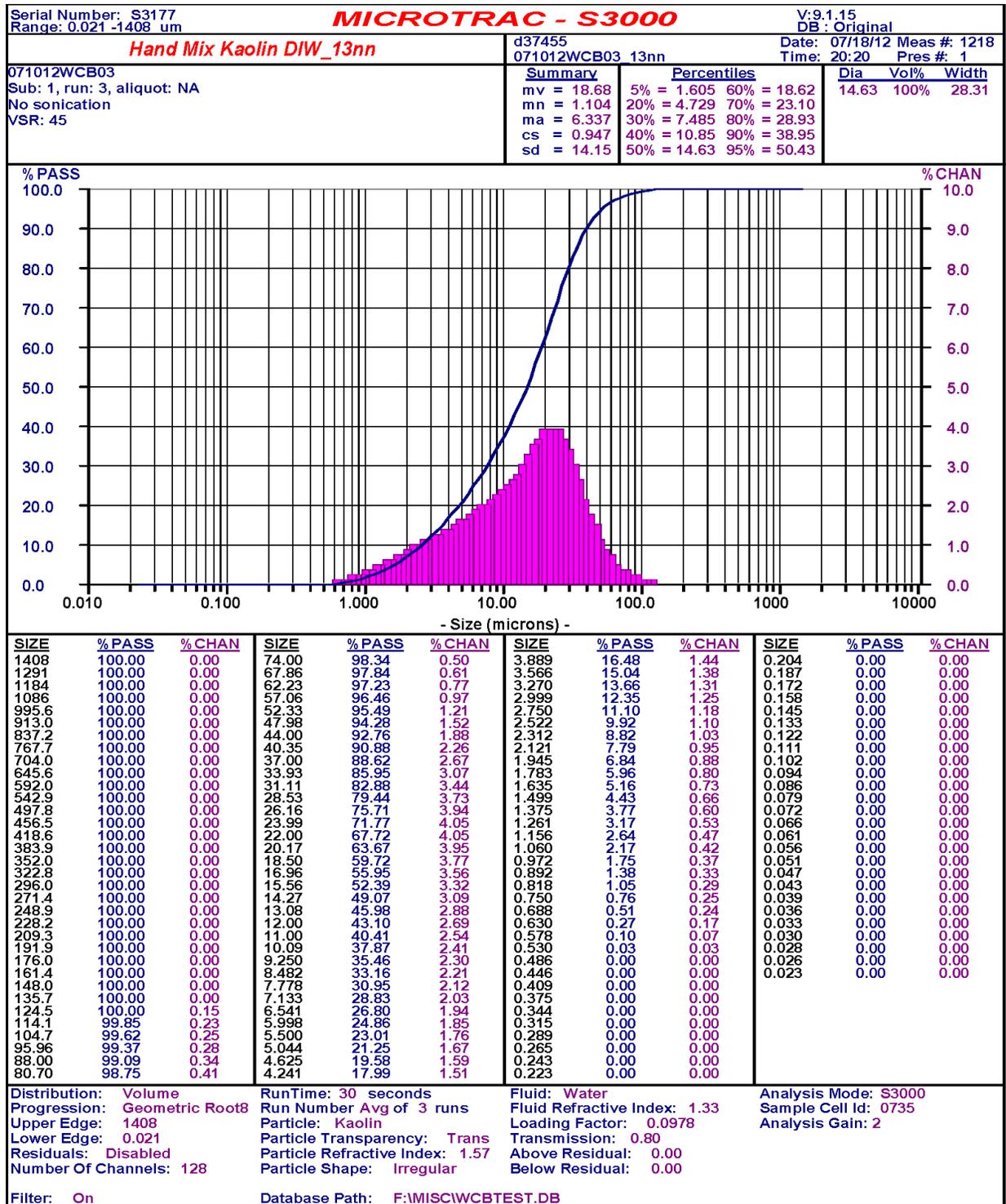


Figure C.1. Particle Size Distribution for the Hand-mixed Slurry of Kaolin Clay in DI Water without Sonication (see Hand Kaolin in Table 3.9, Section 3.6)

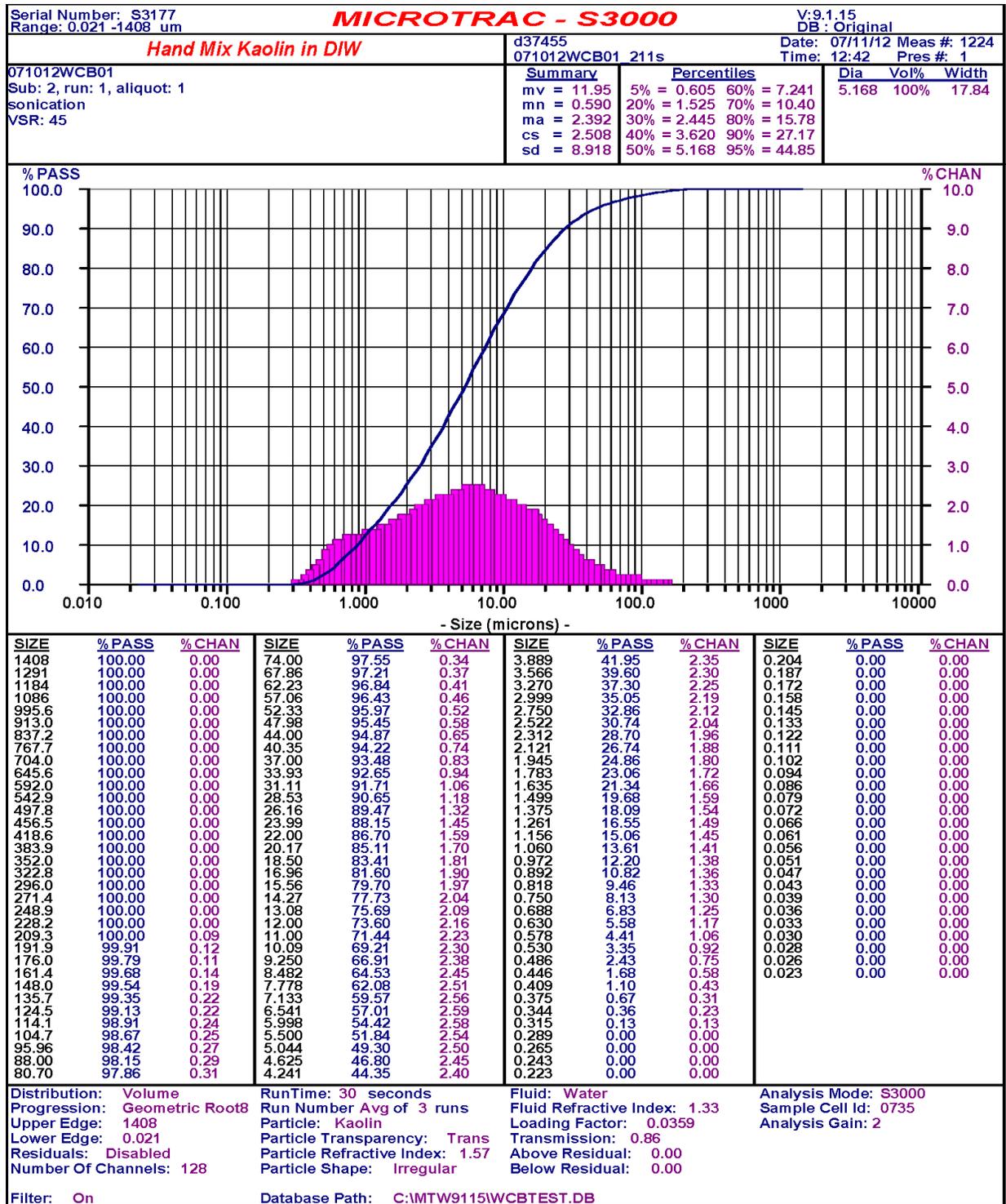


Figure C.2. Particle Size Distribution for the Hand-mixed Slurry of Kaolin in DI Water with Sonication (see Hand Kaolin/Sonicated in Table 3.9, Section 3.6)

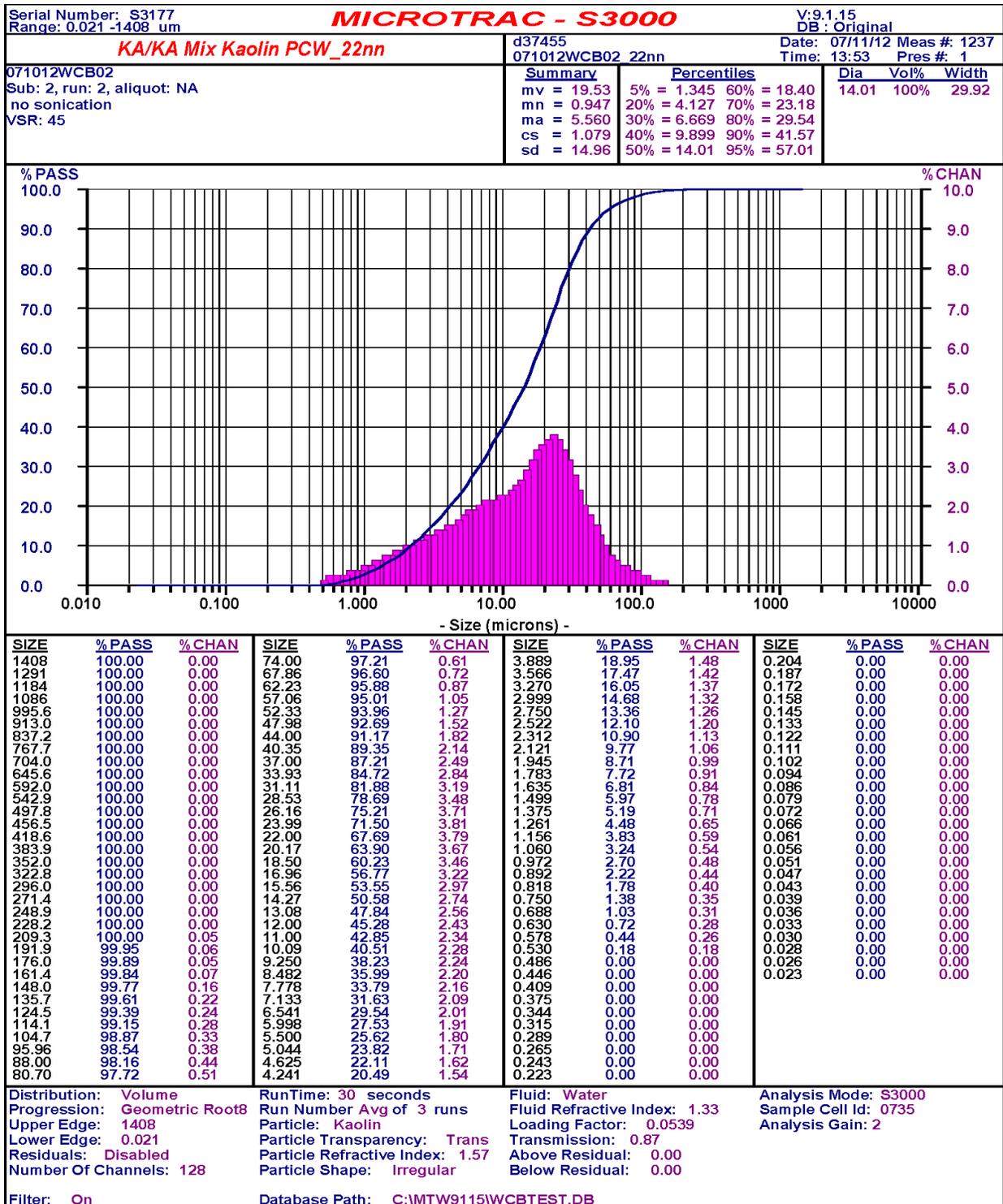


Figure C.3. Particle Size Distribution for KitchenAid-Mixed/KitchenAid-Presheared Slurry of Kaolin Clay in Pasco City Water without Sonication (see KA/KA Kaolin in Table 3.9, Section 3.6)

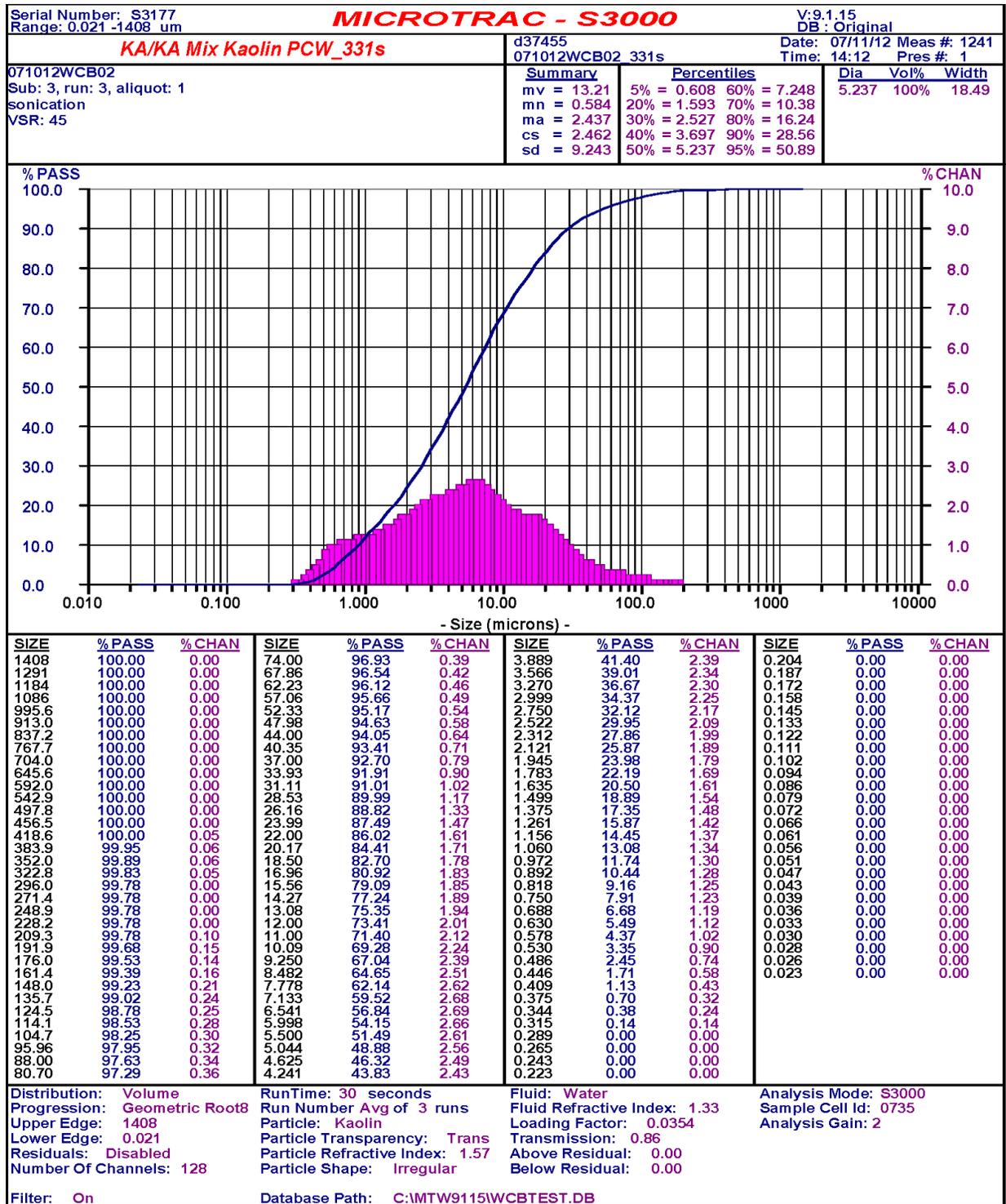


Figure C.4. Particle Size Distribution for KitchenAid-Mixed/KitchenAid-Presheared Slurry of Kaolin Clay in Pasco City Water with Sonication (see KA/KA Kaolin/Sonicated in Table 3.9, Section 3.6)

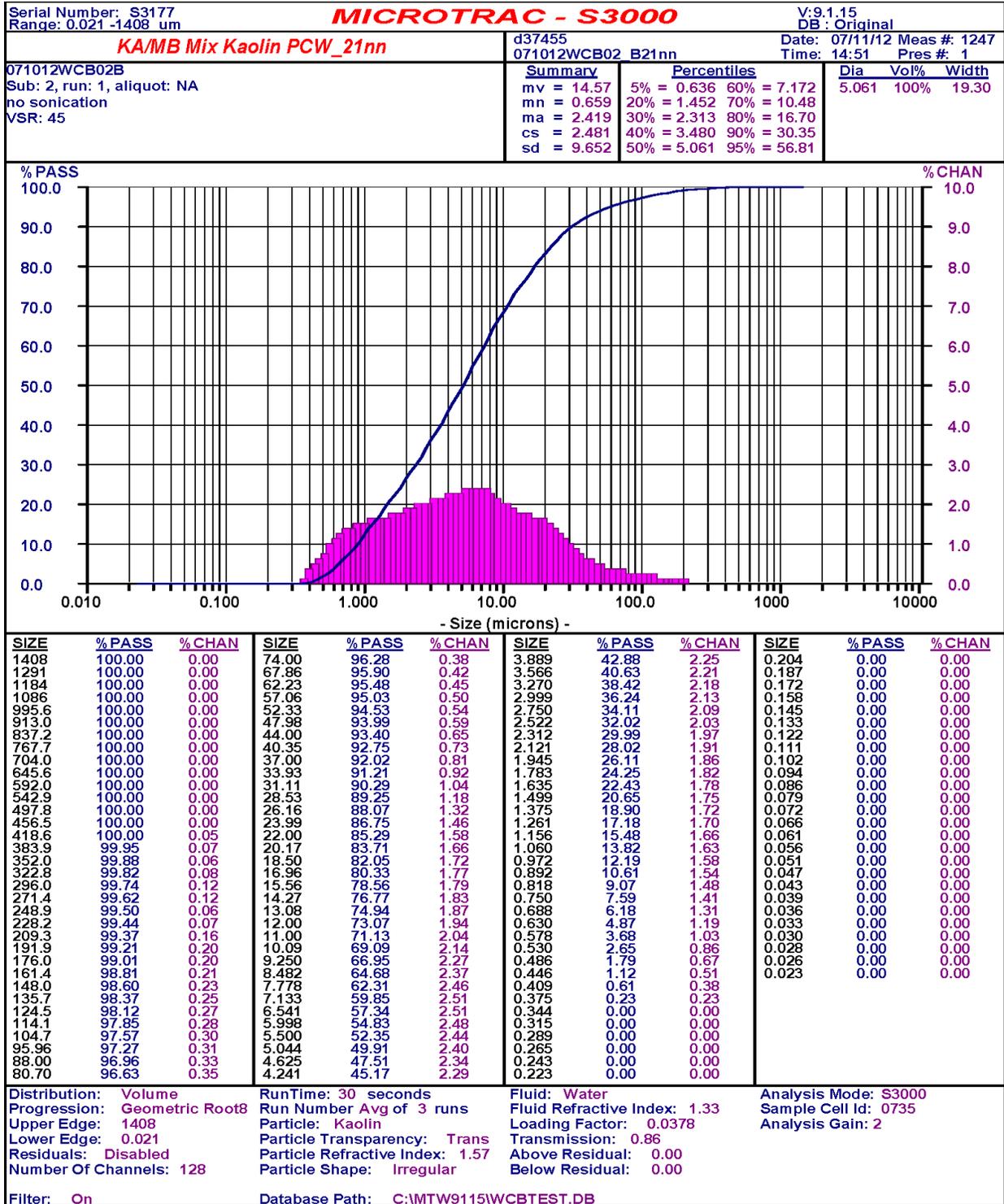


Figure C.5. Particle Size Distribution for KitchenAid-Mixed/Magic Bullet-Presheared Slurry of Kaolin Clay in Pasco City Water without Sonication (see KA/MB Kaolin in Table 3.9, Section 3.6)

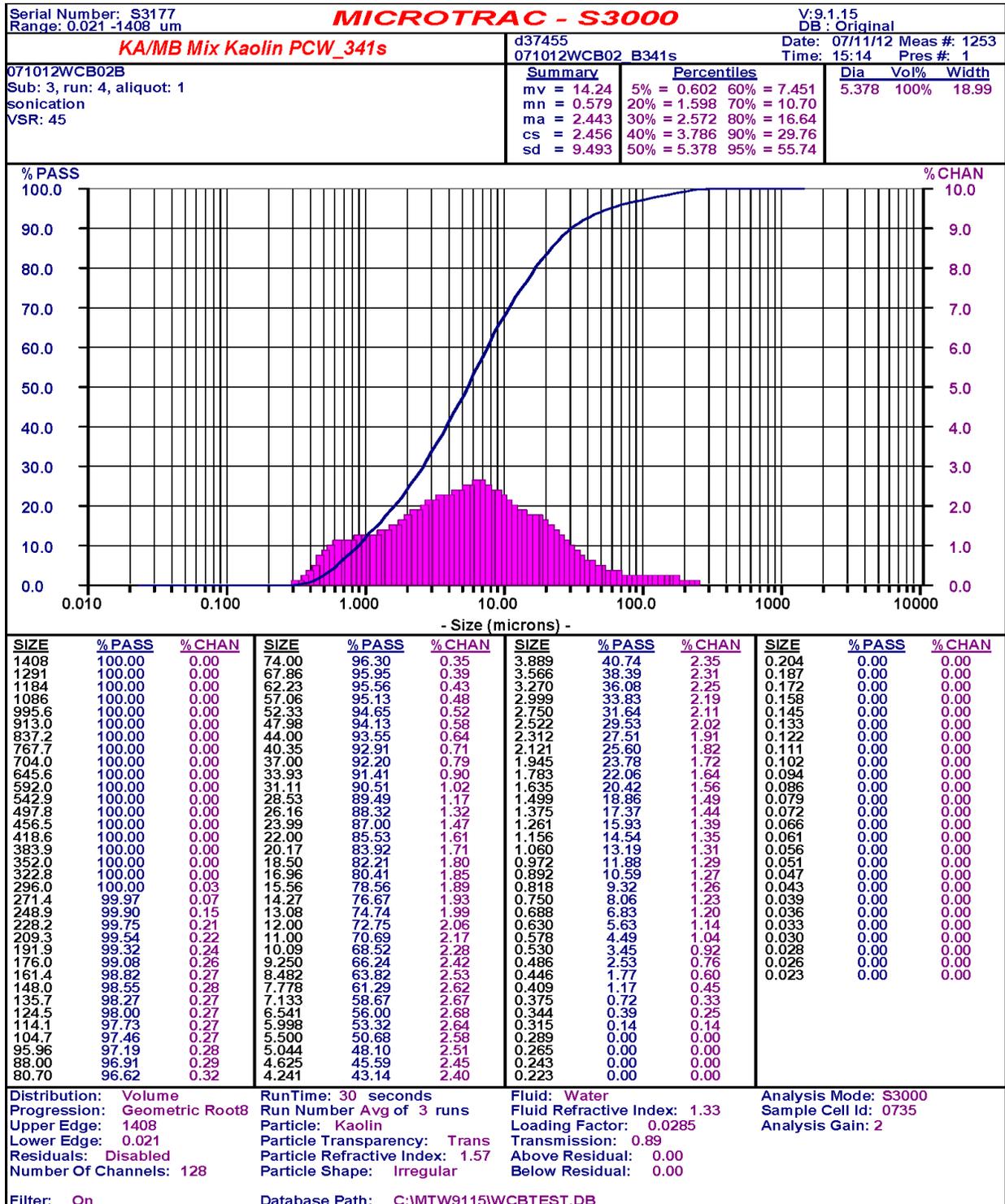


Figure C.6. Particle Size Distribution for KitchenAid-Mixed/Magic Bullet-Presheared Slurry of Kaolin Clay in Pasco City Water with Sonication (see KA/MB Kaolin/Sonicated in Table 3.9, Section 3.6)

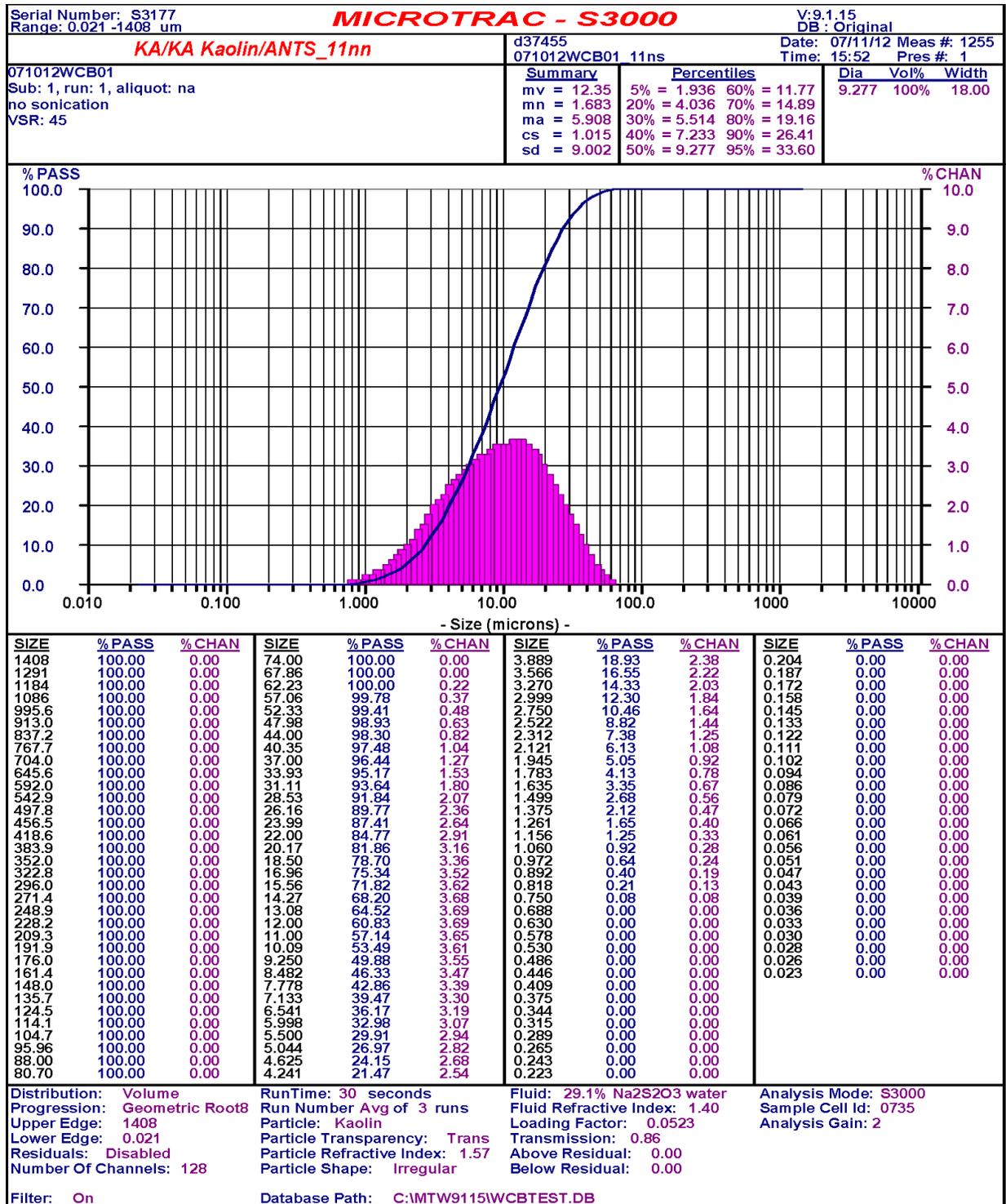


Figure C.7. Particle Size Distribution for KitchenAid-Mixed/KitchenAid-Presheared Slurry of Kaolin Clay in Pasco City Water with 24.9 wt% Anhydrous Sodium Thiosulfate; without Sonication (see KA/KA Kaolin/Salts in Table 3.9, Section 3.6)

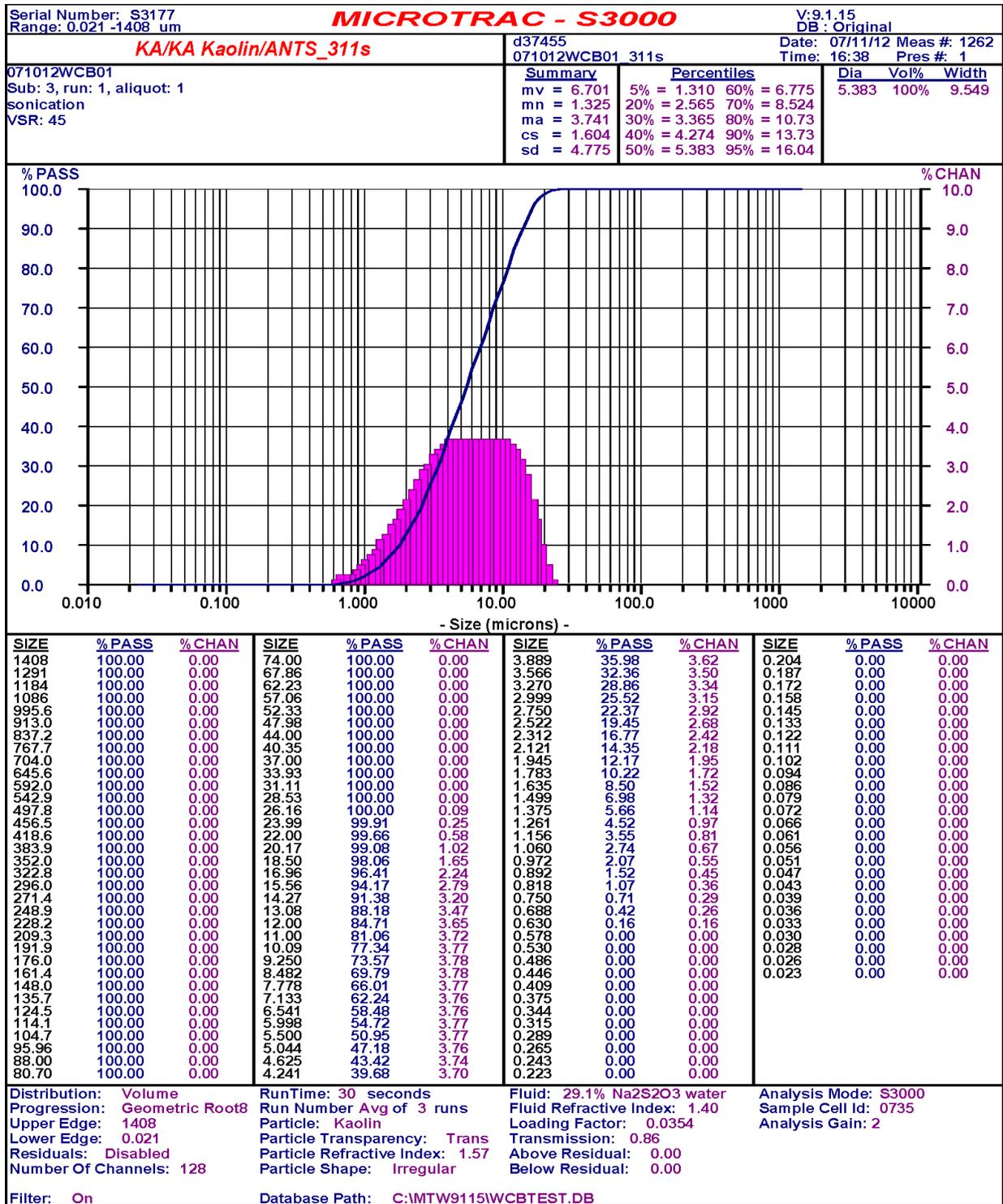


Figure C.8. Particle Size Distribution for KitchenAid-Mixed/KitchenAid-Presheared Slurry of Kaolin Clay in Pasco City Water with 24.9 wt% Anhydrous Sodium Thiosulfate; with Sonication (see KA/KA Kaolin/Salts/Sonicated in Table 3.9, Section 3.6)

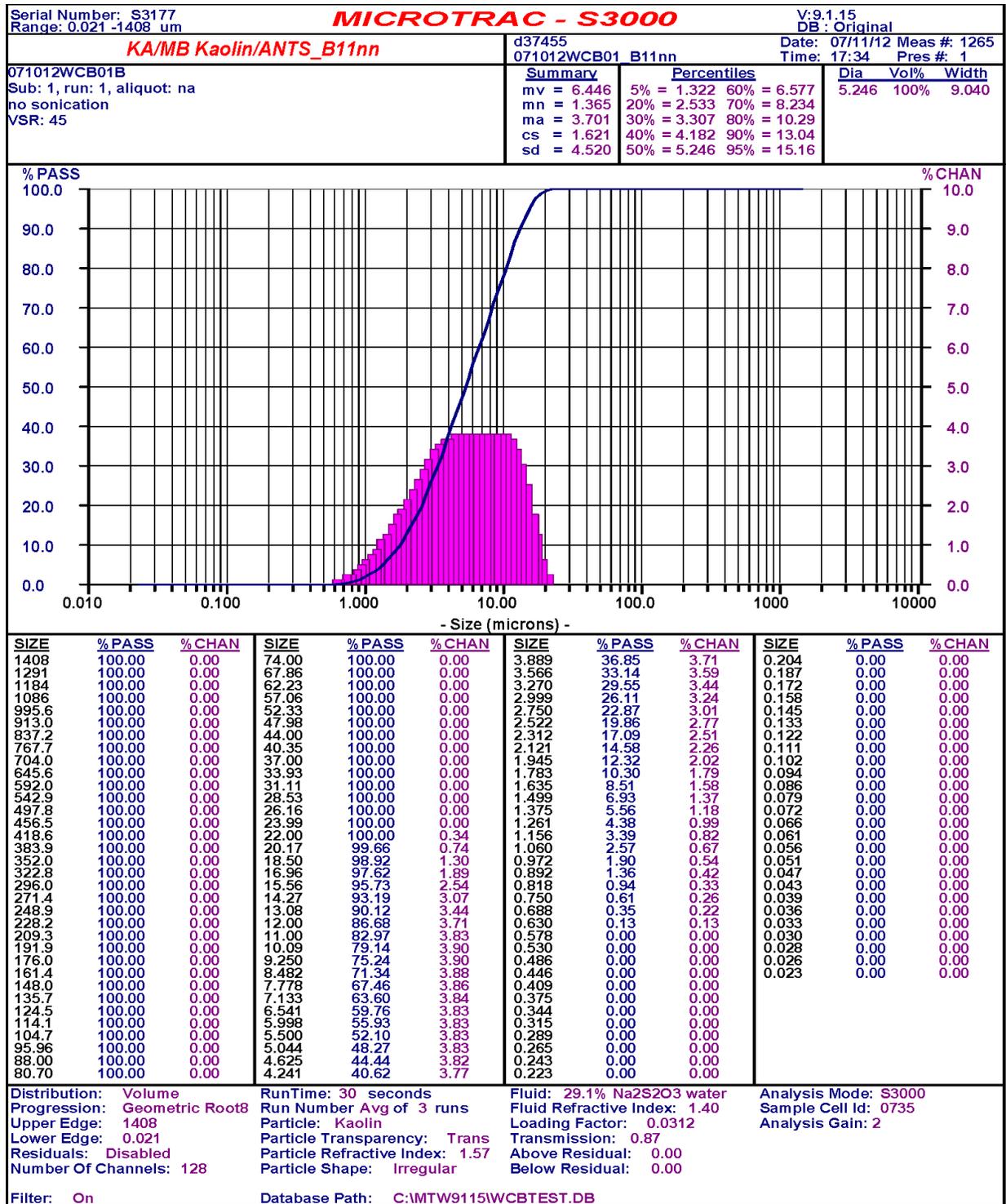


Figure C.9. Particle Size Distribution for KitchenAid-Mixed/Magic Bullet-Presheared Slurry of Kaolin Clay in Pasco City Water with 24.9 wt% Anhydrous Sodium Thiosulfate; without Sonication (see KA/MB Kaolin/Salts in Table 3.9, Section 3.6)

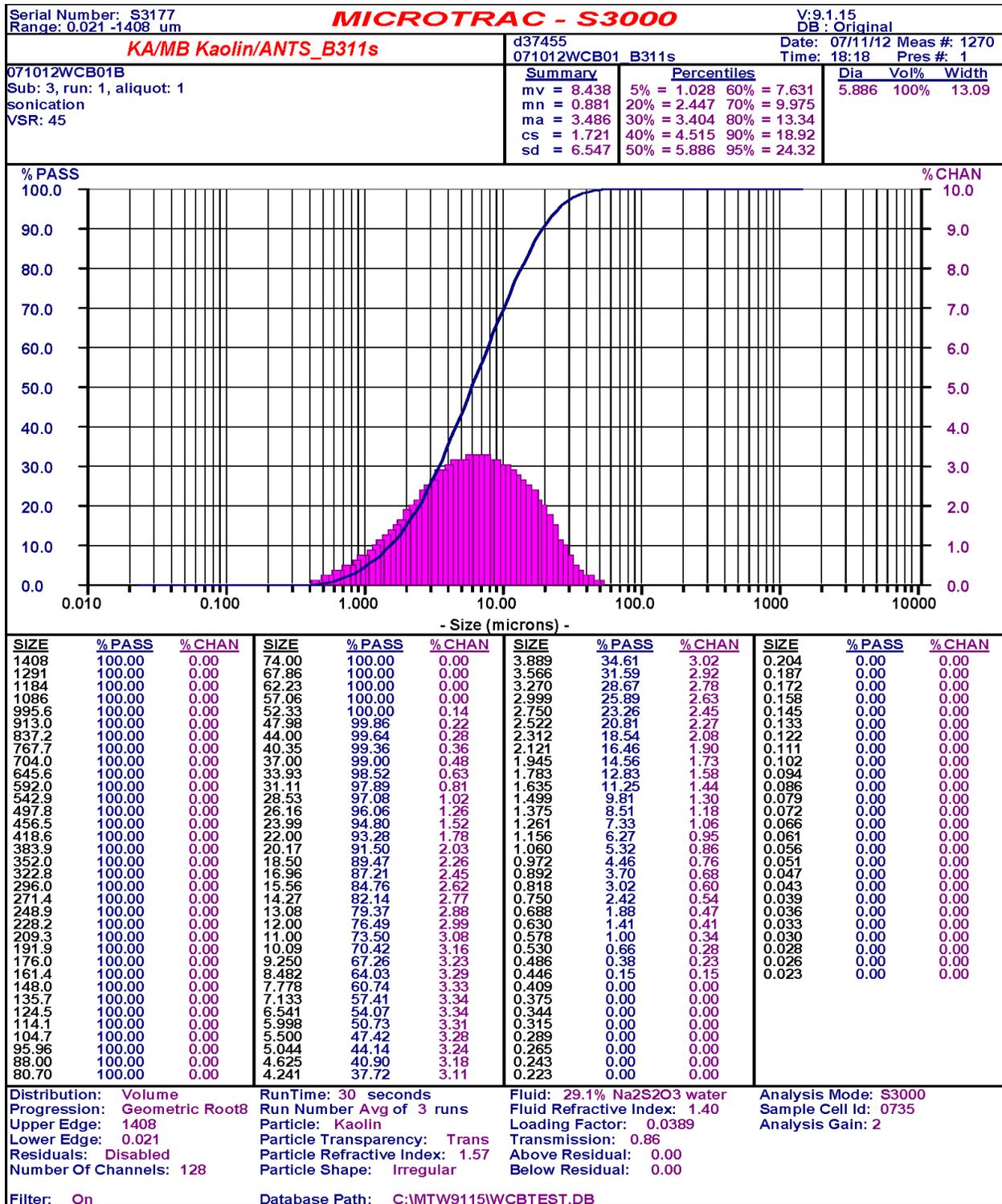


Figure C.10. Particle Size Distribution for KitchenAid-Mixed/Magic Bullet-Presheared Slurry of Kaolin Clay in Pasco City Water with 24.9 wt% Anhydrous Sodium Thiosulfate; with Sonication (KA/MB Kaolin/Salts in Table 3.9, Section 3.6)

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