This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.

DISCLAIMER

WSRC-TR-2005-00495
Revision 0

PARTICLE SIZE ANALYSIS OF SIMULANT SLUDGE SLURRIES AND TANK 40 RADIOACTIVE SLUDGE SLURRY

D. R. Click
S. McWhorter
C. J. Bannochie

October 16, 2005

Unclassified
Does not contain
Unclassified Controlled
Nuclear Information

ADC &
Reviewing Official: L M Chandler
Name and Title mgr Analytical Development
Date: 12-22-05

Analytical Development
Savannah River National Laboratory
Aiken, SC 29808

Prepared for the U.S. Department of Energy Under Contract Number DEAC09-96SR18500

SRNL
SAVANNAH RIVER NATIONAL LABORATORY

This page intentionally left blank

DISCLAIMER

Printed in the United States of America

Prepared For
U.S. Department of Energy

This page intentionally left blank

Key Words: *method developent, sieving, particle size*

Retention: Permanent

PARTICLE SIZE OF SIMULANT SLUDGE SLURRIES AND TANK 40 RADIOACTIVE SLUDGE SLURRY

D. R. Click
S. McWhorter
C. J. Bannochie

October 16, 2005

Analytical Development
Savannah River National Laboratory
Aiken, SC 29808

Prepared for the U.S. Department of Energy Under Contract Number DEAC09-96SR18500

SRNL
SAVANNAH RIVER NATIONAL LABORATORY

This page intentionally left blank

REVIEWS AND APPROVALS

AUTHOR(S):

Daniel R Click — 12/16/05
D. R. Click, Analytical Development — Date

A. McWhorter — 12/20/05
S. McWhorter, Analytical Development — Date

C J Bannochie — 16 Dec 05
C. J. Bannochie, Process Engineering Technology — Date

TECHNICAL REVIEWERS:

Arthur Jurgensen — 12/19/05
A. R. Jurgensen, Analytical Development — Date

APPROVERS

L M Chandler — 12-22-05
L. M. Chandler, Manager, Analytical Development — Date

Connie C. Herman — 12/19/05
C. C. Herman, Manager, Process Engineering Technology — Date

Frank Pennebaker — 12/16/05
F. M. Pennebaker, Manager, Spectroscopic Research and Instrumental Analysis — Date

This page intentionally left blank

EXECUTIVE SUMMARY

As part of the PET (Process Engineering Technology) simulant development program, an investigation was initiated to determine the best method for particle size analysis of both simulant and radioactive sludge slurries using a physical separation method (i.e. sieving) or a laser light diffraction method such as Microtrac or Lasentec. The experiments performed in this investigation were completed with Sludge Batch 3 simulant and radioactive sludge slurries. The long-term goals of the investigation were: (1) to build a database of the size of particles in actual radioactive waste tank sludge and determine the impact of particle size on rheology, and (2) to aid in the development of simulant slurries that closely match the particle size and rheological behavior of radioactive sludge.

The goals of this document are to:

1. Present and summarize results of the three previously mentioned analysis methods and recommend the best method for PET and the DWPF (Defense Waste Processing Facility) to measure particle size in sludge slurries;

2. Recommend further laboratory work and suggest experimental protocol for testing of simulant and radioactive sludge slurries and give references to how particle size determination is done at other DOE sites.

Several factors have been shown to influence the particle size of simulant and radioactive sludge slurries such as chemical composition, temperature, ionic strength, mechanical mixing or passage through pumps, and sonication.[1,2] This work focuses on determining the best method for particle size analysis of simulant sludge slurries and radioactive sludge slurries received at SRNL but does not address the following important issues:

1. Uncertainty of the agglomeration and de-agglomeration phenomena during transport and settling of actual radioactive waste;

2. Densities of the individual or agglomerated particles measured during particle size analysis.

For the sieving method, the investigation included design changes and material changes (i.e., substituting Teflon for polycarbonate) of the sieving setup which had been previously tested in the laboratory[3] and preliminary testing of the method and equipment in a remote environment (the SRNL shielded cells mock-up area) using different simulant slurries. The final phase of the sieving method involved analysis of actual radioactive waste from a Sludge Batch 3 Tank 40 sample in the SRNL shielded cells.

Investigation of the Microtrac method included analysis of both non-radioactive and radioactive sludge slurries and developing suspending mediums in which to analyze these slurries.

The Lasentec method trial was conducted using ground glass frit and ground glass frit added to non-radioactive Sludge Batch 4 SRAT product.

This page intentionally left blank

TABLE OF CONTENTS

This page intentionally left blank

LIST OF FIGURES

LIST OF TABLES

This page intentionally left blank

LIST OF ACRONYMS

AD	Analytical Development
DI	De-Ionized
DOE	Department of Energy
DWPF	Defense Waste Processing Facility
CETL	Clemson Environmental Technologies Laboratory
FBRM	Focused Beam Reflectance Measurement
IC	Ion Chromatography
ICP-ES	Inductively Coupled Plasma Emission Spectroscopy
HDPE	High Density Polyethylene
NIST	National Institute of Standards and Technology
PET	Process Engineering Technology
PSD	Particle Size Distribution
SB	Sludge Batch
SEM	Scanning Electron Microscopy
SRM	Standard Reference Material
SRNL	Savannah River National Laboratory
USC	University of South Carolina

This page intentionally left blank

1.0 INTRODUCTION AND BACKGROUND

For each sludge batch that is processed in DWPF (Defense Waste Processing Facility), SRNL (Savannah River National Laboratory) performs a characterization of the sludge slurry. The sludge slurry is analyzed for chemical composition, radionuclide composition and physical characteristics (i.e., weight percent solids and rheology). The information obtained from this characterization is used in radioactive demonstrations of the DWPF process (subsequent to characterization of non-radioactive sludge simulants) to evaluate potential processing issues and identify DWPF processing strategies.

There are several characteristics that can influence the processing of a sludge batch, and one of these is the rheology of the sludge slurry. Rheology measurements are performed to ensure that the slurry can be transferred and mixed properly. The flow curves (i.e., shear rate vs. shear stress) generated from the rheology measurements are modeled to obtain the yield stresses and consistencies of these samples. The yield stresses and the consistencies are then compared to the design bases for the DWPF to determine if the feed may pose potential processing problems (e.g., pumping/ transfer problems).

To facilitate understanding of the rheology (i.e., yield stress and consistency of a sludge slurry sample) of a given sludge batch the parameters that influence the rheology must be measured on both non-radioactive and radioactive sludge slurry samples (e.g., pH, temperature, chemical composition, particle size, etc.). Particle size has been shown to affect rheological behavior in studies using non-radioactive sludge slurries and radioactive sludges.[4,5]

Sieving is a widely used and inexpensive technique for measuring particle size. The relative simplicity of the technique, low investment, good reliability, and broad size range make it attractive. To date, there has been one sieving experiment performed on DWPF radioactive sludge slurry to determine the particle size at SRNL.[6] The experiment was somewhat successful, but the majority of particles (~60%) were smaller than 20 microns. This document will describe the final stages of scoping studies performed to develop a sieving method for determining particle size and distribution in non-radioactive sludge simulant and the implementation of the method into the SRNL shielded cells for determination of the particle size in the Sludge Batch 3 Tank 40 sample.

Currently, there is an abundance of Microtrac particle size data for the non-radioactive sludge slurries although the experimental protocol was limited in scope. Conversely, minimal particle size information is available for radioactive sludge slurries and that information has only been obtained using a Microtrac instrument. However, no systematic study has been done to address the factors that affect the particle size measurements such as pH of the suspending medium, extent of dissolution upon dilution of a sample, sonication, sample flow rate in the sample chamber of the instrument and the variations encountered among different instruments at SRNL (see references 5 and 7 for some studies at other DOE sites). This report attempts to address some of these issues with Microtrac analysis.

Lasentec, another laser diffraction technique, has found many applications for particle size measurements including on-line monitoring of many kinds of industrial processes.[8,9] To date, Lasentec has not been used to measure particle size of radioactive sludge slurries.

This page intentionally left blank.

2.0 EXPERIMENTAL

Material

The simulant sludge slurries given to AD for sieving were labeled USC (University of South Carolina) Tank 40 Drum 3 and CETL (Clemson Environmental Technologies Laboratory) Sludge Batch 2. Elemental composition is given elsewhere.[3] The weight percent insoluble solids content of the simulant slurries used for sieving were measured and found to be 14.3% and 18.1% on a dry basis, respectively. AD was also supplied with a sub-sample of radioactive Tank 40 sludge slurry, which is Sludge Batch 3. The insoluble solids content of the slurry sub-sample was measured and found to be 18.1%. Elemental composition is given elsewhere.[10]

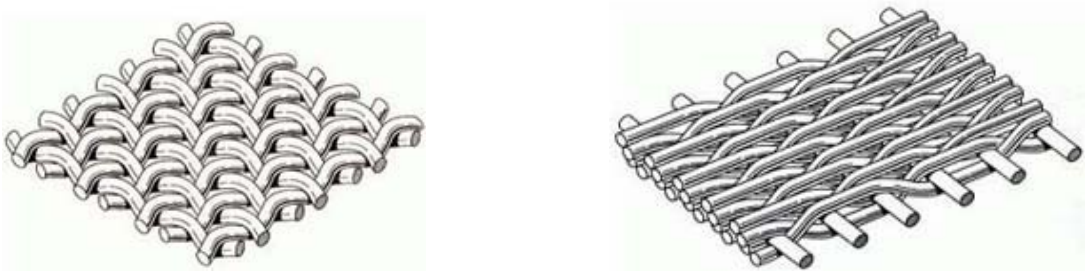
Microtrac testing was completed with Batch 3 Tank 40 Test 3C sludge and Batch 3 Tank 40 Test 2. The elemental composition of the simulant slurries are given elsewhere.[4, 11] Microtrac testing was also done with a sub-sample of the same radioactive Tank 40 sludge slurry mentioned above.

Lasentec evaluation was completed using Frit 418 and simulated DWPF SRAT Product SB4-8. There is no particular significance to this SRAT product versus any other SRAT product that could have been used in these experiments.

The Microtrac NIST 1984 standard reference material (SRM) was provided by David Missimer (AD) and is a mixture of tungsten carbide and cobalt, ranging in particle size from 9.5 to 26.5 μm by Microtrac analysis.

Sieving Apparatus

Analytical Development of SRNL developed a wet sieving system for determining particle size of simulant slurry and radioactive sludge slurries. The system works by re-circulating slurry supernate or water, which has passed through the sieves and collected in the glass bottom which is suspended by a high density polyethylene (HDPE) sieve holder, back up to the top sieve by the use of a pump while the sieves are being shaken by a sieve shaker. The HDPE sieve holder has a 150 mL reservoir and a hole for the use of vacuum. The vacuum hole is protected by a funnel inside the sieve holder so that the sieved liquid drains into the reservoir. There is also a small hole in the side near the bottom of the sieve holder which serves as a drain so that supernate/water can be pumped out and re-circulated. The vibratory sieve shaker is an Analysette 3 model manufactured by Fritsch. The shaker can be operated in two modes; normal and micro mode. The micro mode is used for sieves 20 μm and smaller. The sieves used during these experiments were made by Newark Wire Cloth Company and conform to ASTM specification E-11. The plain weave mesh of the sieves size 20 μm and greater result in square shape openings and below 20 μm the twill Dutch weave of the mesh results in openings which are probably more wedged or elliptical in shape. The wedged shape openings makes it more difficult for particles to pass through. It is for this reason that the sieve holder was developed with vacuum capability. Pictures in Figure 2.1 demonstrate the differences in the weave of the wires of the sieves.



A B

Figure 2.1 (A) Representative of a plain weave mesh used with sieves 20 μm and larger and results in square openings in the mesh. (B) Representative of a twill Dutch weave used with sieves with aperture openings samller than 20 μm which results in wedged shaped or elliptical type openings in the mesh.

Sieving Procedures

NIST 1984 Standard

Triplicate analysis of the NIST 1984 standard was performed on the bench-top in a laboratory to test the re-designed sieving system and in the SRNL mock-up area. The NIST 1984 standard was mixed by rotating and turning the bottle end-over-end twenty times and an ~1 g portion was weighed into a beaker. The contents of the beaker were transferred into a 25 μm sieve which was the top sieve of a stack consisting of a pre-weighed 25 μm sieve, a 20 μm sieve, and a 16-18 (165x1400 mesh) μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min. At the completion of the cycle the sieves were rinsed and set in the oven to dry overnight. Three additional sieves were assembled and then an ~1 g portion of the NIST standard was weighed into a beaker. The contents of the beaker were transferred into a 12-14 (200x1400 mesh) μm sieve which was the top sieve of a stack consisting of a pre-weighed 12-14 μm sieve, an 11-12 (250x1400 mesh) μm sieve and an 8-9 (325x2300 mesh) μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min. At the completion of the cycle the sieves were rinsed and set in the oven to dry overnight.

CETL Sludge Batch 2 Simulant Slurry

Duplicate sieving experiments were completed with CETL Sludge Batch 2 simulant slurry in the following manner. Approximately 5 g of simulant slurry was added to a weighing cup, diluted in 50 mL of water and poured into a pre-weighed, pre-wetted 150 μm sieve which was the top sieve of a stack consisting of the pre-weighed and pre-wetted (with DI water) 150 μm sieve, a 75 μm sieve and a 45 μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min while water was re-circulating. The top sieve was then rinsed and removed. The wet sieving system was re-assembled and the sieves where shaken for an additional 5 mins. This pattern was repeated down to the last sieve in the three sieve stack. Each sieve was dried overnight at 50 ºC and re-weighed. An additional aliquot of sample of simulant slurry was then weighed (~1 g) and added to a weighing cup, diluted in 50 mL of water and poured into a pre-weighed pre-wetted 32 μm sieve which was the top sieve of a stack consisting of the pre-weighed and pre-wetted (with DI water) 32 μm sieve, and a 25 μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min while water was re-circulating. The top sieve was then rinsed and removed. The wet sieving system was re-assembled and the sieves where shaken for an additional 5 mins. This

pattern was repeated for each sieve in the stack. Each sieve was dried overnight at 50 ºC and re-weighed. An additional aliquot of simulant slurry was then weighed (~0.5 g) and added to a weighing cup, diluted in 50 mL of water and poured into a pre-weighed, pre-wetted 16-18 μm sieve which was the top sieve of a stack consisting of the pre-weighed and pre-wetted (with DI water) 16-18 μm sieve, a 12-14 μm sieve, and an 11-12 μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min while water was re-circulating. The top sieve was then rinsed and removed. The wet sieving system was re-assembled and the sieves where shaken for an additional 5 mins. This pattern was repeated for each sieve in the stack. Each sieve was dried overnight at 50 ºC and re-weighed. An additional aliquot of simulant slurry was then weighed (~0.5 g) and added to a weighing cup, diluted in 50 mL of water and poured into a pre-weighed, pre-wetted 8-9 μm sieve which was the top sieve of a stack consisting of the pre-weighed and pre-wetted (with DI water) 8-9 μm sieve, a 6-7 (450x2750 mesh) μm sieve, and an 5-6 (510x3600 mesh) μm sieve. The vibratory sieve shaker was put in micro mode, set at an interval time of 10 s and a total sieve time of 5 min while water was re-circulating. The top sieve was then rinsed and removed. The wet sieving system was re-assembled and the sieves where shaken for an additional 5 mins. This pattern was repeated for each sieve in the stack. Each sieve was dried overnight at 50 ºC and re-weighed.

Pre-weighed sieves of the same size as those used in experiments above were stacked and then the corresponding supernate from CETL Sludge batch 2 simulant was poured through the sieves and allowed to drain out. The sieves were shaken off by hand then dabbed with a Kimwipe™ before being dried overnight at 50 ºC and re-weighed. This progression was repeated twice. The average weight of dissolved solids that collected on each size sieve was then subtracted from the difference of weight obtained before and after sieving the simulant slurry with supernate.

Particle size analysis was conducted on two additional sludge slurries, one non-radiaoactive and one radioactive in the manner above. The size of sieves used for each of the experiments and the approximate amount of sludge slurry used is given in Table 2.1. More specific experimental details involving each type of sludge slurry are given in the paragraphs after Table 2.1.

Table 2.1 Sludge slurries sieved, sieves used and mass of slurry sieved.

Slurry	Sieves Used (μm)	Mass of Slurry Sieved (g)
UCS Tank 40 Simulant	32, 25, 20	~1
	16-18, 12-14, 11-12	~0.5
	8-9, 6-7, 5-6	~0.5
Sludge Batch 3 Tank 40 Radioactive Sludge	150, 75, 45	~5
	32, 25, 20,	~1
	16-18, 12-14, 11-12	~0.5
	8-9, 6-7, 5-6	~0.5

USC Tank 40 Simulant Slurry

Duplicate experiments using USC Tank 40 simulant were conducted in both the SRNL shielded cells mock-up area and the SRNL high-level shielded cells in the following manner.

Pre-weighed sieves of the same size as those used in experiments above were stacked and then corresponding supernate from USC Tank 40 simulant was poured through the sieves and allowed to drain

out. The sieves were shaken off by hand then dabbed with a Kimwipe™ before being dried overnight at 50ºC and then re-weighed. This progression was repeated twice. The average weight of dissolved solids that collected on each size sieve was then subtracted from the difference of weight obtained before and after sieving the simulant slurry with supernate.

Sludge Batch 3 Tank 40 Radioactive Sludge

Multiple sieving experiments were completed on Sludge Batch 3 Tank 40 Radioactive Sludge simulant slurry in the following manner. The experimental protocol above was not suitable for analysis of the radioactive sludge primarily because too many sieves were being used and the coefficient of variation was too large since different sets of sieves were being used interchangeably. A quick histogram graph of the percent relative standard deviation of the weight of cumulative mass below a certain sieve *versus* the sieve size in microns revealed large percent RSDs for many of the sieves and these were removed from the sieve stack for the future experiments. In addition, several sieves were taken from the stack based upon the first five experiments and the lack of large particle present in the sludge slurry. Two additional experiments were conducted in the manner above with the 25-, 20-, 8-9- and 5-6 μm sieves, and the same sieves were used for each experiment. In addition, four experiments were done using only the smallest size sieve. A ~0.5 g sample or ~1 g sample was sieved through the smallest sieve available which is the 5-6 μm sieve. The results from these experiments are in the results section.

Sieve Cleaning

The sieves were cleaned by removing as much dried slurry as possible using a water rinse. Then, the sieves were submerged in a 2% nitric acid solution for a period of five minutes, rinsed with DI water and transferred to a 1 M oxalic acid solution for five minutes. The sieves were then rinsed with DI water and placed sieve foil side up in a sonicator bath for no longer than three minutes (damage may result to the sieves if left in sonicator longer than three minutes). The sieves were again rinsed with DI water and if there was any resulting residue on the sieves, the process was repeated. The repeatability of the experiments suggests the sieve cleaning protocol is sufficient.

Microtrac

Non-radioactive samples were measured on a Microtrac S3000 particle size analyzer on the laboratory bench-top. The Microtrac measures particle diameter by measuring the scattered light from a laser beam projected through a stream of the fluid carrying the diluted sample particles. The amount and direction of the light scattered by the particles is measured by an optical detector array and then analyzed to determine the size distribution of the particles. The data is stored into volume bins, where a bin is defined by an upper and lower diameter and data is then normalized to 100% after the measurement is complete. The range of the non-radioactive instrument is 0.026 to 1408 μm. Radioactive samples were measured on a Microtrac X-100 particle size analyzer in a radioactive hood. The size range of the radioactive instrument is 0.688 to 704 μm.[12] The Microtrac software is able to calculate the mean number, mean surface area, mean volume, number distribution and volume distribution data.

The experimental protocol for Microtrac analysis of non-radioactive samples (Batch 3 Tank 40 Test 2 and Batch 3 Tank 40 Test 3C Simulant Sludge) involved sub-sampling of the sample to be analyzed by stirring with an impeller and sampling at approximately the same depth each time. This was followed by sonication of half of the samples, and then dilution of the non-radioactive sample in the suspending medium (water or simulated supernate) in the sample chamber of the Microtrac analyzer. Samples of the Sludge Batch 3 Tank 40 radioactive sludge were diluted in a suspending medium, and in some cases

sonicated, prior to analysis and left to sit overnight before decanting and weighing out the resulting wet solids. The solids were then diluted in the sample chamber of the Microtrac analyzer in a suspending medium and analyzed.

Lasentec

The Lasentec is an in-line analyzer for measuring chord-length distribution of suspended solid particles. The chord length and particle size are not equivalent terms but there is a direct correlation between the two. The instrument uses a technique known as Focused Beam Reflectance Measurement (FBRM) to provide continuous in-process and real-time measurement of the rate and degree of change of the particle dimension and particle count. The focal point of the laser beam, which is just outside the probe window, is rotated around the window at a linear velocity of 2 m/s. When the focal point intersects the edge of a particle, the particle begins to backscatter light and this continues until the focused beam has reached the edge of the particle. The backscatter is collected by the FBRM optics and converted into an electronic signal. A unique discrimination circuit is then used to isolate the time period of backscatter from one edge of an individual particle to its opposite edge. This time period (t) is multiplied by the scan speed (v), to yield a distance of chord length $\boldsymbol{c}$ according to the following equation:

$$\boldsymbol{c} = \nu \times t$$

The chord length c in Equation 3.1 is the straight-line distance between any two edges of a particle and is a function of the particle shape. Typically, thousands of chord length are measured per second and counted by the FBRM electronics. The resulting chord length by number distribution is a robust thumbprint of the particle size distribution in the slurry. Any change in the size distribution will have a corresponding change in the chord-length distribution.

The electronics associated with the Lasentec monitor "sort" the measured chord lengths into 30 "bins". The "bins" are on a log scale from 0.8 μm to 1.9 μm and an extended top "bin" for counts greater than 1000 μm. At the end of the user-defined measurement duration (between 2 s and 5 min), the Lasentec software constructs a histogram of the measured chord lengths from the number of particles classified in each "bin".

This page intentionally left blank

3.0 RESULTS

Sieving Experiments

Sieving was chosen as a possible method to determine particle size distribution in sludge slurries. Below are the some of the strengths and limitations associated with sieving.[13]

- Strengths
  - low capital investment
  - minimal sample preparation
  - ease of operation – does not require highly skilled operators
  - broad size range can be analyzed

- Limitations
  - requires long analysis times and times get longer as sieves with finer aperture openings are used
  - automation is limited
  - particles with high aspect ratios (e.g., needle shaped particles) give rise to large uncertainties
  - mechanical motion during sieving can affect repeatability and reproducibility
  - the data obtained is mass fraction coarser than or finer than a particular size, which can be converted to a volume basis
  - large sample size needed compared to laser diffraction techniques

The following results highlight equally well some of the strengths and limitations of sieving. Microtrac data is given for comparison. Since sieving data is based on mass, and Microtrac analysis is based on volume, where Microtrac data is graphed along with sieving data, an important assumption was made. The density of the particles was assumed to be uniform throughout the PSD and therefore, the volume percent of particles in a given range equaled the mass percent in that range.[14]

NIST 1984 Standard

In a previous report, the particle size retention of each of the sieves used was determined by wet sieving the NIST 1984 SRM, drying the solids that remained on each sieve, and analyzing the solids by scanning electron microscopy (SEM). The NIST 1984 SRM, which is used for calibration of the Microtrac instrument, was chosen because of its size distribution, irregularly shaped particle morphology, resistance to fracture on handling and low level of aggregation. It was found that 90% of the particles on each sieve were larger than the largest size particle the sieve was rated for. In other words, 90% of the particles on a 5-6 micron sieve were larger than 6 microns and the results have been calculated using the upper size limit for each sieve. A SEM picture is shown in Figure 3.1 of the NIST 1984 SRM standard in which the different morphology of the particles can clearly be seen.

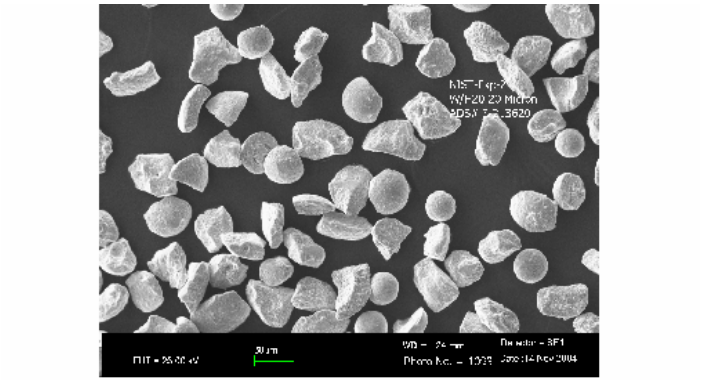


Figure 3.1 SEM of particles collected on 20 μm sieve screen.

The NIST 1984 standard was sieved using different sets of sieves on the bench-top (Figure 3.2), in the SRNL shielded cells mock-up area (Figure 3.3) and in the SRNL shielded cells. Histogram plots of % insoluble solids on sieve *versus* aperture size generated from the sieving data of the NIST 1984 SRM has only one mode of particle distribution as does number data from Microtrac analysis.

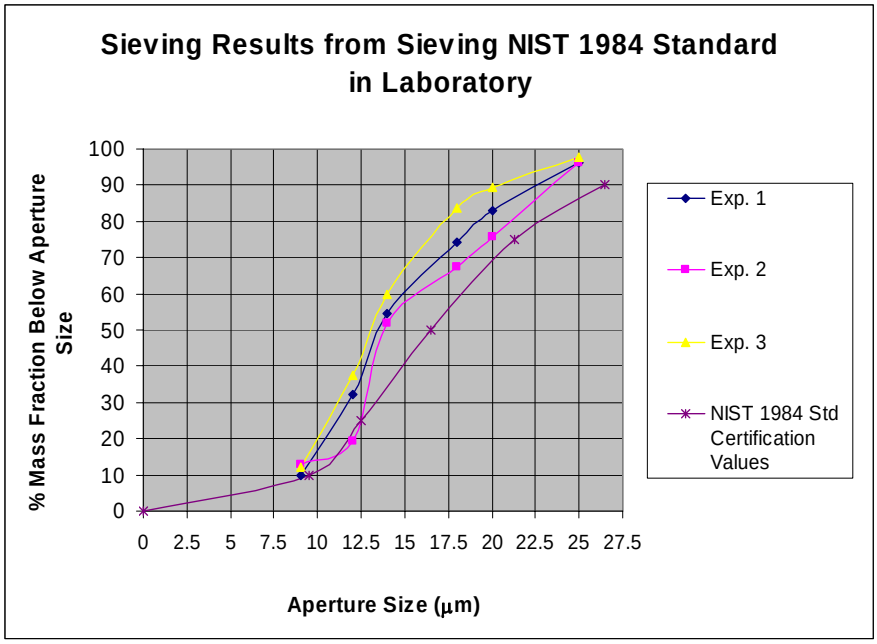


Figure 3.2 Experimental results obtained from sieving NIST1984 standard (% Mass Below Sieve Size *vs* Aperture Size (μm)).

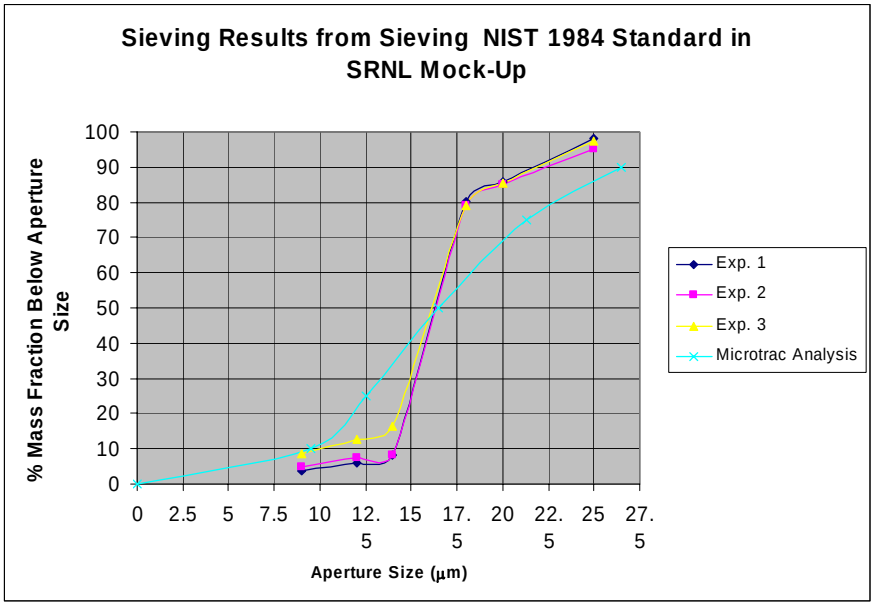


Figure 3.3 Experimental results obtained from sieving NIST1984 standard (% Mass Below Aperture Size *vs* Aperture Size (µm)).

The results obtained upon sieving the NIST 1984 SRM agree with literature data that the coefficient of variance (precision) may be 5-10% when different sets of sieves with large aperture widths are used and greater than 10% when different sieves with small aperture widths are used.[9] Precision was excellent considering the particle morphology. For most of the sieves tested, except the 12-14 µm sieve, the accuracy (as compared to Microtrac volume data) was approximately ±30% of the accepted value. However, for the 12-14 µm sieve the values obtained were as much as ±80% of the accepted value. Larger differences were expected because of the twill dutch weave of the wire of the sieves having aperture openings less than 20 µm. The certification values of the NIST 1984 SRM are given in Figures 3.2 and 3.3. for comparison. The certified values were determined by Microtrac and Scanning Electron Microscopy (SEM) analysis.

USC Tank 40 Drum 3 Simulant Slurry

Experiments 1 and 2 were performed in the SRNL shielded cells mock-up using USC Tank 40 Drum 3 simulant. The experimental results are graphed in Figure 3.4 along with the Phase 1 average result.[3]

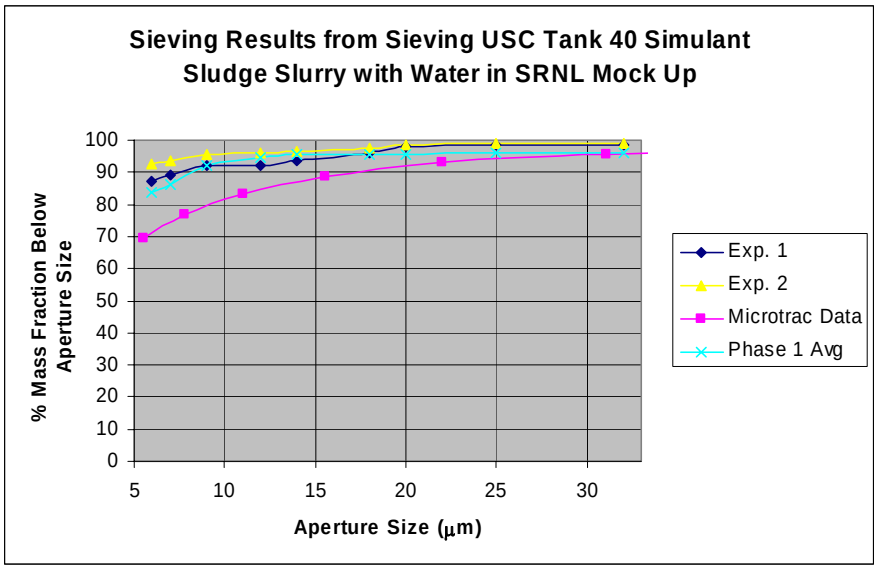


Figure 3.4 USC Tank 40 Drum 3 simulant sieving results using water as re-circulating fluid (% Mass Below Aperture Size *vs* Aperture Size (μm)).

Good repeatability was obtained upon duplicate analysis of USC Tank 40 Drum 3 and the data trend in the same direction as the Microtrac data although this need not be the case because the conditions of analysis are very different. This simulant slurry has a bimodal distribution of particles (based on Microtrac channel data) but this was difficult to discern from the sieving data as a much smaller particle size range was analyzed and the second mode of the bimodal distribution was under 5 μm.

The second set of duplicate experiments was done in an analogous manner to those above except filtered supernate was used as the re-circulating fluid and a dissolved solids correction factor was applied to the data. To obtain the dissolved solids correction factor, supernate was re-circulated through the sieves and then they were dried overnight. The average mass of dissolved solids that were found to be retained by the sieves from two experiments was subtracted from the weight of the dried sieves after sieving the sludge slurry. The data shown in Figure 3.5 is corrected for the dissolved solids in the supernate. When the correction factor was applied to data, if it resulted in negative masses the correction factor was set to 0. The average of the duplicate experiments is similar to the average obtained during Phase 1.

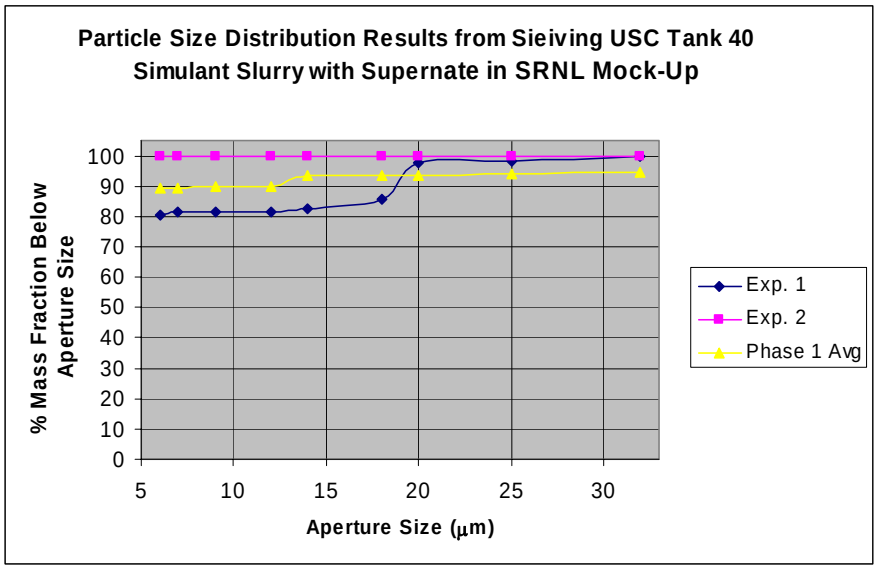


Figure 3.5 USC Tank 40 Test 3 simulant sieving results using supernate as re-circulating fluid (% Mass Below Aperture Size *vs* Aperture Size (μm)).

CETL Sludge Batch 2 Simulant Sludge Slurry

CETL Sludge Batch 2 simulant slurry has large particles in it (based on Microtrac data) and so additional sieves with large aperture openings were implemented and used. The mass of the sludge slurry used for the first three sieves was also increased from ~1 g to ~5 g of simulant slurry. In total, ~7 g of sludge slurry was needed to complete a full experiment. The experimental results are graphed in Figure 3.6.

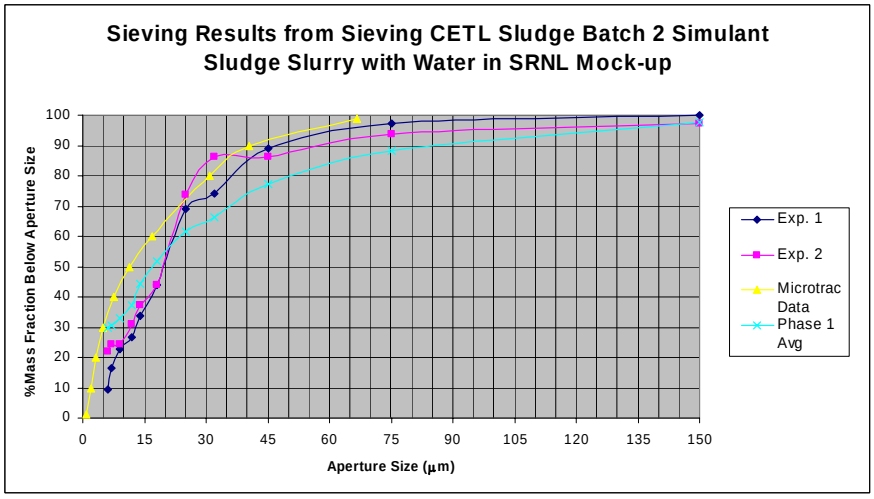


Figure 3.6 CETL Sludge Batch 2 simulant sieving results using water as re-circulating fluid (% Mass Below Aperture Size *vs* Aperture Size (μm)).

Good repeatability was also observed with this simulant slurry between duplicate experiments. However, the bimodal distribution of particles in the histogram plot of number data from Microtrac analysis is not evident in the histogram plot of % insoluble solids on sieve *versus* aperture size of sieve from the data of

the sieving experiment and a majority of the particles in the second mode of the bimodal distribution are smaller than the smallest size sieve used.

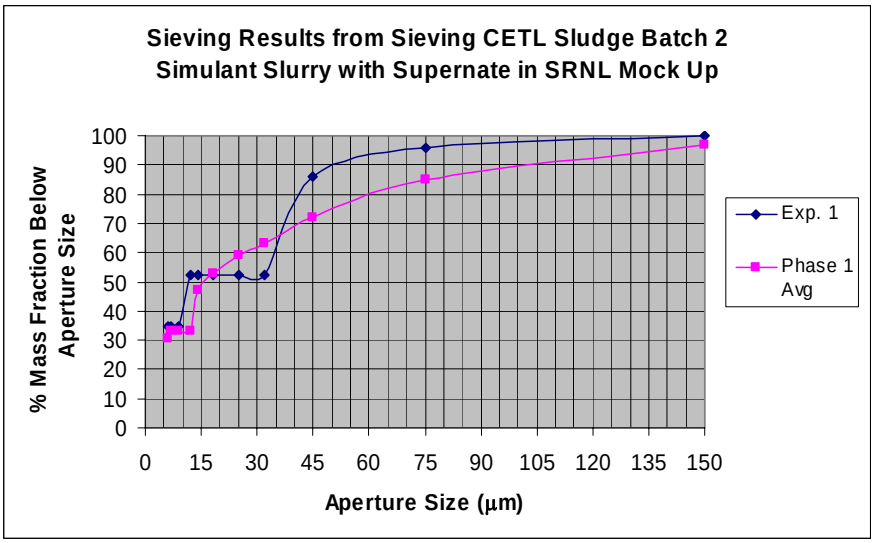


Figure 3.7 CETL Sludge Batch 2 simulant sieving results using supernate as re-circulating fluid (% Mass Below Aperture Size *vs* Aperture Size (μm)).

The results of sieving CETL Sludge Batch 2 slurry with supernate are shown in Figure 3.7 and agree well with the average obtained from Phase 1. Two experiments were attempted but only one was successful due to a faulty sieve and lack of additional material.

It should be noted that it is possible to see multi-modal particle size distribution from sieving experiments but they do not often match the modes from Microtrac analysis because the modes from sieving are based upon volume whereas the modes reflected in the histogram plot of Microtrac data is based upon the number of particles in a specific size channel or bin. Furthermore, the size range analyzed during sieving is usually much smaller which results in lack of detailed analysis of particles outside of the sieve size range.

Sludge Batch 3 Tank 40 Radioactive Sludge

The first set of experiments with Tank 40 radioactive slurry was performed with many sieves (11) because no prior knowledge of the size of particles in the sludge was available. However, the results were not acceptable and there was greater than 50% difference in the results. The large differences observed can be attributed to the use of too many sieves and using different sets of sieves. The coefficient of variance (precision) can be 10% or larger for sieves with small aperture widths which has been documented.[9] The goal here was to obtain ≤10% difference for sieves larger than 20 microns and ≤20% difference for sieves under 20 μm. The difference being the weave of the wire mesh for sieves having aperture widths less than 20 μm (see Figure 2.1). The results of the first five experiments are shown in Table 3.1.

Table 3.1 Experimental results from sieving Sludge Batch 3 Tank 40 radioactive sludge slurry.

Experiment Number	Sieves Used (μm)	% Mass Fraction of Particles Under 6 μm
1	150, 75, 45, 32, 25, 20, 16-18, 12-14, 11-12, 8-9, 6-7, 5-6	67.8
2	150, 75, 45, 32, 25, 20, 16-18, 12-14, 11-12, 8-9, 6-7, 5-6	42.6
3	150, 75, 45, 32, 25, 20, 16-18, 12-14, 11-12, 8-9, 6-7, 5-6	95.1
4	150, 75, 45, 32, 25, 20, 16-18, 12-14, 11-12, 8-9, 6-7, 5-6	85.8
5	150, 75, 45, 32, 25, 20, 16-18, 12-14, 11-12, 8-9, 6-7, 5-6	49.2

A histogram plot of the % RSD of mass of solids colleted on sieves *versus* sieve size in μm revealed large percent RSDs for many of the sieves. The sieves with the greatest variation were removed from the sieve stack and not used in future experiments. SEM experiments were done to determine if the particles in the sludge were needles and if this was contributing to the difficulty encountered during the analysis. The experiment was done using ~0.5 g of sludge which was diluted in 10 ml of water and then the resulting diluted solution was poured onto a 5-6 μm sieve screen. The solids were dried briefly and then a SEM stub was pressed against the screen to lift the particles. The morphology of the particles is varied and agglomerates can be seen in Figures 3.8-3.10 but particles with a high aspect ratio were not seen. Two additional experiments were then conducted using ~0.5 g or ~1 g of slurry and the 25, 20, 8-9 and 5-6 μm sieves. In this case, the same sieves were used in duplicate experiments. The results are shown in Figure 3.11 along with the Microtrac data analysis in water.

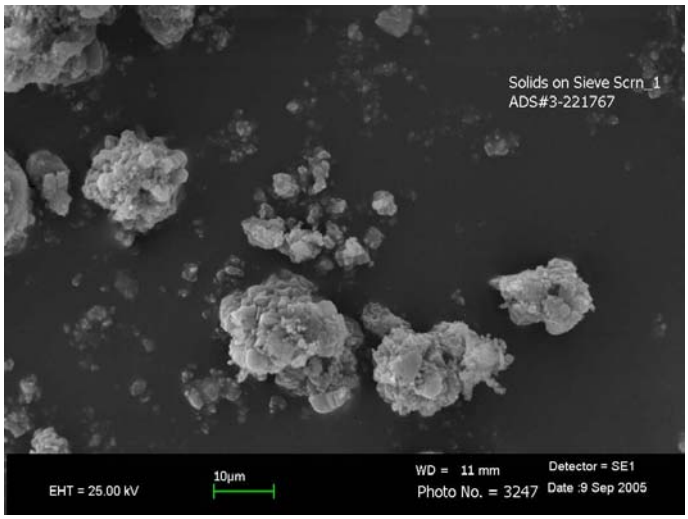


Figure 3.8 SEM of Sludge Batch 3 Tank 40 sludge particles.

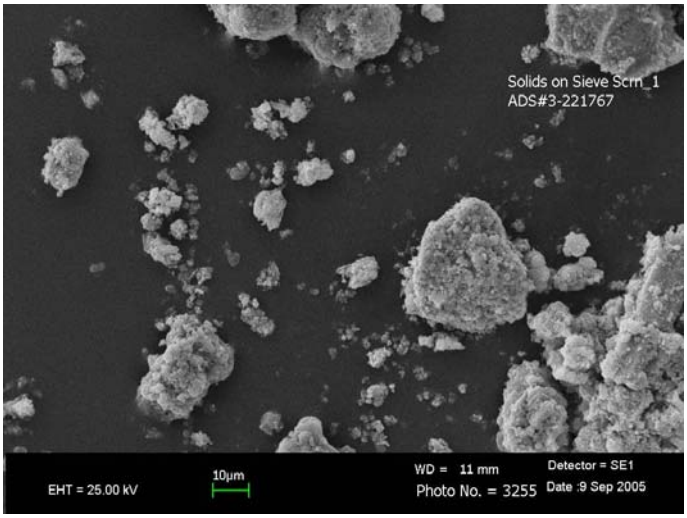


Figure 3.9 SEM of Sludge Batch 3 Tank 40 sludge particles.

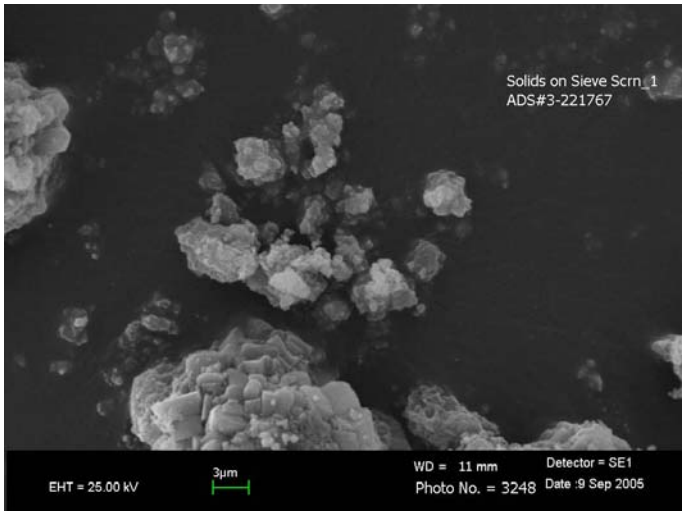


Figure 3.10 SEM of Sludge Batch 3 Tank 40 sludge particles

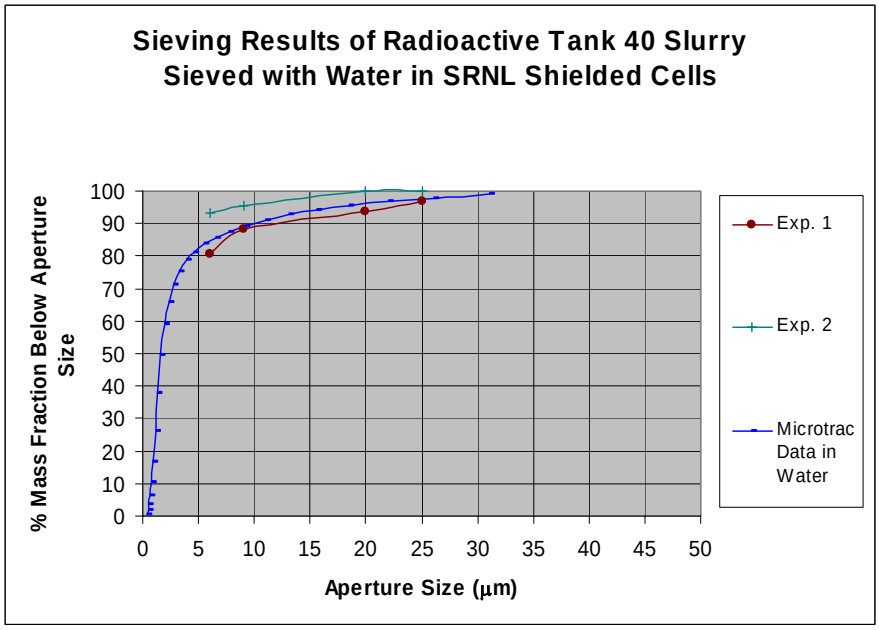


Figure 3.11 Radioactive Tank 40 slurry sieving results.

The data from the second round of experiments is similar to the Microtrac data in the size range analyzed. Furthermore, using less sieves and the same set of sieves resulted in better repeatability. However, the sieve system was originally designed to be used with multiple sets of sieves to cut down on analysis time and using only one set of sieves more than doubled the analysis time. In addition to the second round of experiments above, four experiments were completed using only the smallest size sieve, which is the 5-6 µm sieve and a ~0.5 g sample or ~1 g sample was sieved and the solids were dried. The average results from these experiments suggest that ~85 % of the insoluble particles in the sludge are less than 6 µm, which agrees well with the results shown in Figure 3.11. These experiments were done as a final check of the results graphed in Figure 3.11.

Recommendation of Use

It is recommended that sieving be used as a back-up method to determine particle size in sludge slurries and be used as the primary method when separation and identification of specific particles (>6 µm) needs to be accomplished (e.g. coal particles in the waste).

Microtrac and Lasentec Results

Microtrac and Lasentec are laser diffraction techniques for analyzing particle size and more specific information has been given about these methods in the experimental section. Some general strengths and limitations of laser diffraction methods are given below.[13]

- Strengths of laser diffraction instruments
  - rapid analysis
  - relatively simple sample preparation
  - some instruments can be used for both dry powders and powders in suspension
  - some instruments can be used on-line and off-line
  - relatively inexpensive
  - does not require highly skilled operators

- Limitations of laser diffraction instruments
  - cannot distinguish between dispersed primary particles and agglomerates
  - during Microtrac analysis error may be introduced in the particle size distributions if powder particles deviate from spherical configuration
  - Microtrac instrument design precludes analysis of concentrated suspensions
  - may require knowledge of optical properties of specimen
  - instrument performance and operation is highly dependent on instrument design (e.g., laser sources of different wavelengths, differing number and positions of detectors, sample flow rates)
  - particle size determination limited by wavelength of light
  - requires knowledge of the chemical processes happening upon dilution and changing pH of the sample

Lasentec Results

There were two main objectives for using the Lasentec instrument: (1) obtain particle size on sludge slurries and (2) be able to follow the changes in the particle size of sludge slurry *in situ* throughout the course of a Slurry Receipt Adjustment Tank (SRAT) cycle or alternatively, before and after a SRAT cycle without dilution.

Frit 418 was ground in a planetary mill and the resulting solids submitted for Microtrac particle size distribution measurements. Additionally, two samples of simulated DWPF SRAT Product SB4-8, one at full concentration and one at 25 wt. %, were submitted for Microtrac analysis. SB4-8 is a SRAT product produced by the SB4 development activities at ACTL. There is no particular significance to this SRAT product versus any other SRAT product that could have been used in these experiments.

During the initial experiment, the Lasentec signal for deionized water (DI H_2O) was monitored, as well as the resulting signal for successive additions of ground glass frit; the first at 2 wt. % and the second at 4 wt. %. The purpose of this experiment was to establish if the Lasentec instrument could see changes in the insoluble solids concentration in material being used to investigate the behavior of SRAT product.

The effect of adding ground glass frit to the SRAT product SB4-8 was then determined. The chord length signal was collected for full strength SRAT product and also for a diluted, 25 wt% SRAT product. Additions of ground glass frit were introduced into the diluted SRAT slurry to achieve 2, 4, 8, 10, 12, 14, and 16 wt. % frit. The spectrum following each addition was collected.

The tests performed with 0.0 – 16 wt. % ground glass frit additions to SRAT product SB4-8 diluted to 25 wt. % show that the particle chord lengths and distributions do not change significantly with the addition of further insoluble solids. Apparently, the number of fines, particles at and below 10 μm in size, in SRAT product, and presumably SRAT feed, present a system that is sufficiently challenging to the Lasentec instrument to blind it to further changes in the particle distribution. The instrument does not appear to be useful for *in situ* measurements of particle chord length distributions, nor does it appear useful for measurements of SRAT materials even when diluted to one quarter the original concentration. The actual degree of dilution necessary to restore the system's ability to distinguish changes was not explored.[15]

Recommendation of Use

It is recommended that the Lasentec system not be considered for the purpose of analyzing DWPF feeds and/or SRAT products at this time or for the determination of PSD in simulant and radioactive sludge slurries.

Microtrac Results

The goal of testing Microtrac was to determine if it was the best method to use for particle size analysis and, if so, what protocol was necessary. In order to accomplish this goal, recognition and testing of the factors that influence particle size in sludge slurries before and during analysis had to be completed. No systematic study has been done at SRNL to address factors that affect the particle size measurements such as pH of the suspending medium, extent of dissolution upon dilution of a sample, sonication, sample flow rate in the sample chamber of the instrument, differences encountered when using two different instruments and surface chemistry characteristics of the sludge particles and temperature. This report does not address all of these factors but gives results for the differences in particle size resulting from analysis in water *versus* filtered supernate or simulant supernate, sonication *versus* no sonication and 'hot' instrument (used for radioactive samples) *versus* the 'cold' instrument (used for non-radioactive samples). The sample flow rate of the instrument and the surface chemistry characteristics of the sludge particles were not investigated. All experiments were carried out at ambient temperature. Simulant supernate was prepared to closely match the composition of the actual supernate (based upon IC and ICP-ES analysis) for the two non-radioactive sludge slurries and for the Tank 40 radioactive sludge. The suspending medium was prepared to match the supernate of each of the sludge slurries to keep the pH and dissolved solids close to the measured values of the actual supernate to prevent dissolution upon dilution of the sample or changes resulting from a different pH value. No experiments were done to evaluate the change in particle size as a function of time.

The double axis bar charts and tables are used to summarize data in this section. Double axis bar charts plot % pass and % channel data against size. The blue line in the figures represents the % pass data which is defined as the volume percentage of particles under the size listed on the x axis. The maroon bars represent the % channel data which is defined as the volume percentage of particles in a specific size range. The D_{10}, D_{50} and D_{90} values listed in the tables represent the particle diameter where 10%, 50% or 90% of the particles are smaller than the μm size indicated. The mean diameter (mv) , in μm, of the volume distribution represents the center of gravity of the distribution. It is weighted toward larger particles. The mean diameter (mn), in μm, of the number distribution is calculated using volume distribution data and is weighted toward smaller particles. All experiments were run with the NIST 1984 SRM as a calibration check before the samples were analyzed.

Batch 3 Tank 40 Test 2 Sheared Simulant Slurry

Analysis of Batch 3 Tank 40 Test 2 sheared slurry was accomplished using the non-radioactive Microtrac S3000 instrument and by the following protocol. Thirty sub-samples (~2 g each) were pulled from a 2 L container where an impeller was used to stir the slurry and each sample was taken at approximately the same depth using a modified slurry pipette. Five samples were analyzed in water, five were sonicated in a bath and then analyzed in water, five were analyzed in filtered supernate from Batch 3 Tank 40 Test 2 slurry, five were sonicated in an ultrasonic bath and then analyzed in filtered supernate from Batch 3 Tank 40 Test 2 slurry, five were analyzed in Batch 3 Tank 40 Test 2 simulant supernate and five were sonicated in an ultrasonic bath and then analyzed in simulant supernate. The simulant supernate composition was based upon the analysis of actual filtered supernate from Batch 3 Tank 40 Test 2 filtered supernate and confirmed by re-analysis of the simulant supernate. Figures 3.12-3.17 contain the average particle size results from five measurement experiments, except the experiment done in simulant supernate without

sonication (Figure 3.17), which is an average of three experiments. This sludge slurry was also re-analyzed on the radioactive Microtrac X-100 for comparison. This set of experiments was designed to specifically monitor changes in the data when the suspending medium was changed or the sample was sonicated and the actual PSD obtained by either instrument was not that important. Table 3.1contains a summary of the Microtrac S3000 results.

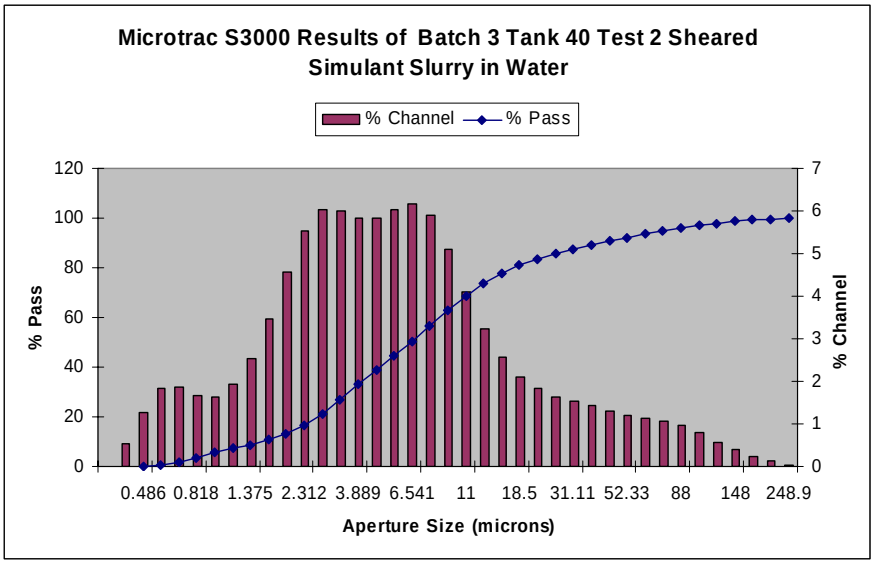


Figure 3.12 Microtrac results of Batch 3 Tank 40 Test 2 sheared sludge slurry analyzed in water.

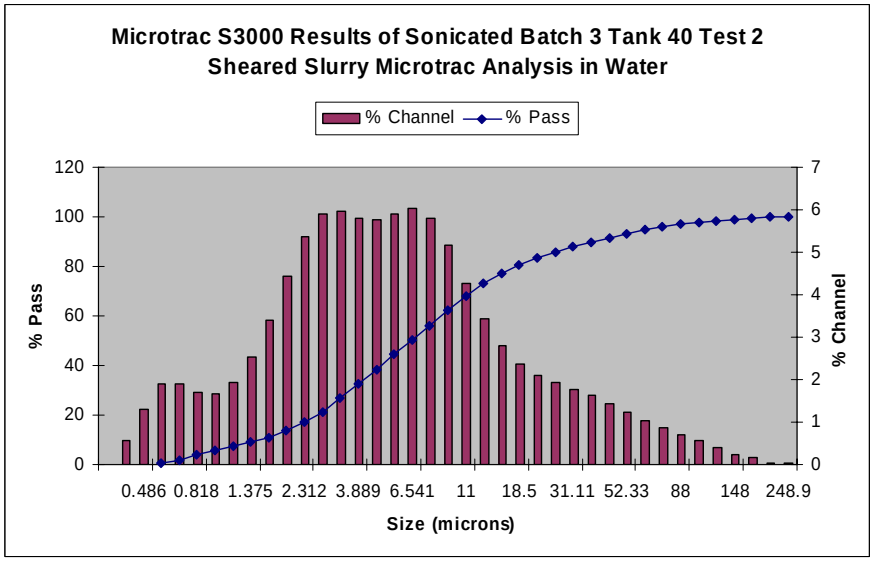


Figure 3.13 Microtrac results of sonicated Batch 3 Tank 40 Test 2 sheared sludge slurry analyzed in water.

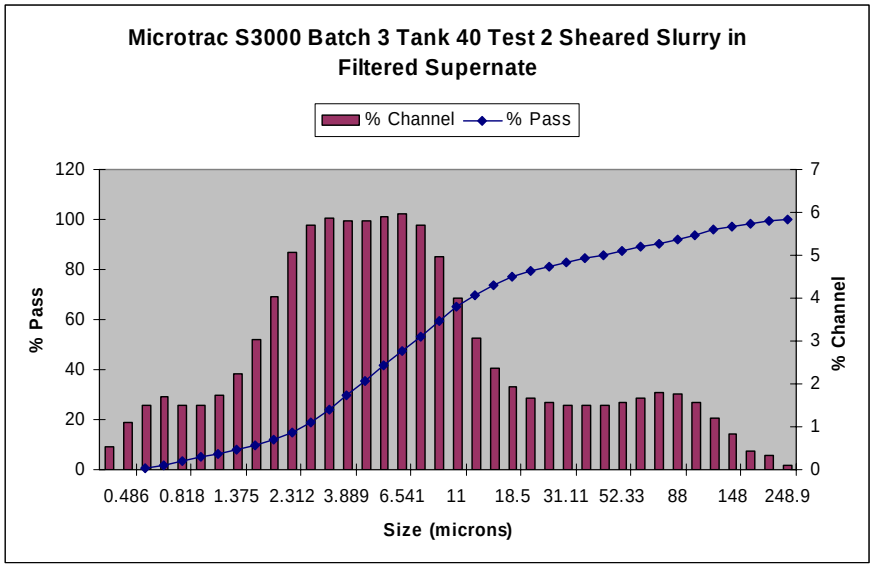


Figure 3.14 Microtrac results of Batch 3 Tank 40 Test 2 sheared slurry analyzed in filtered supernate.

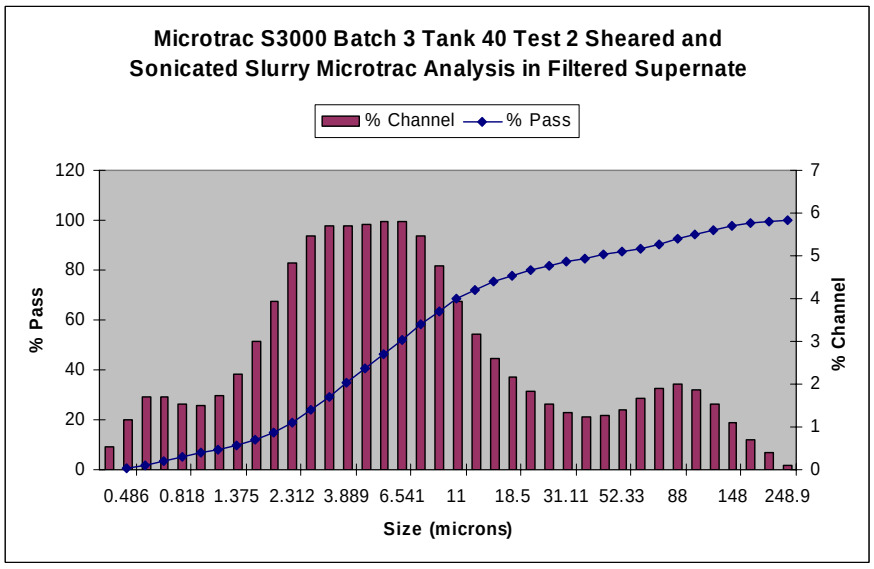


Figure 3.15 Microtrac results of sonicated Batch 3 Tank 40 Test 2 sheared slurry in filtered supernate.

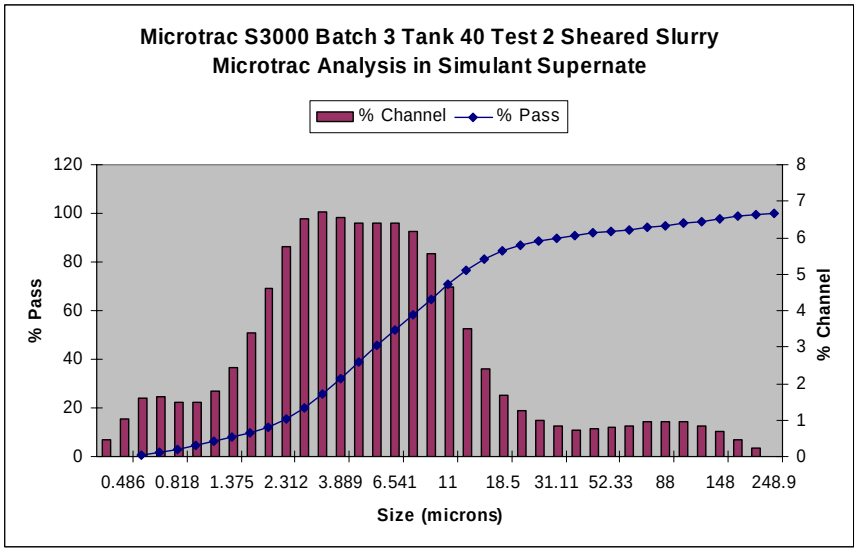


Figure 3.16 Microtrac results of Batch 3 Tank 40 Test 2 sheared slurry in simulant supernate.

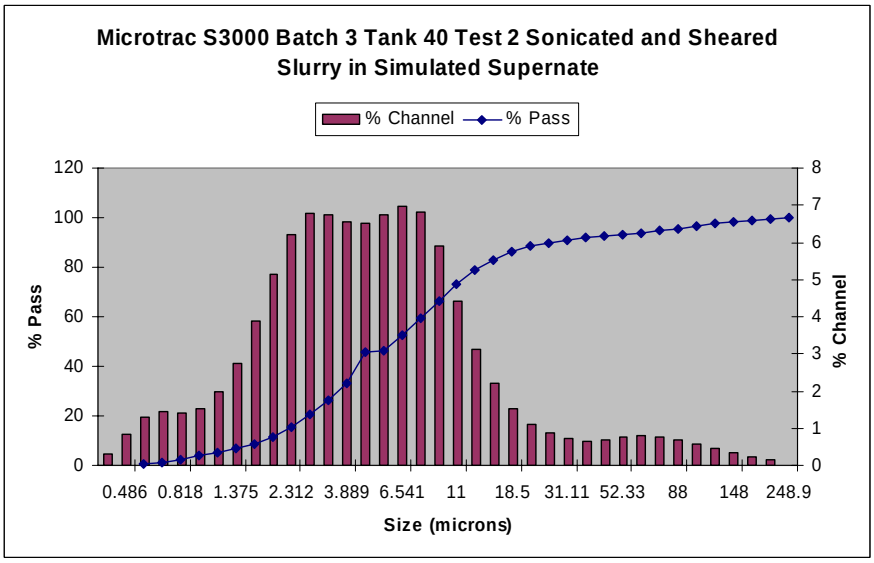


Figure 3.17 Microtrac results of sonicated Batch 3 Tank 40 Test 2 sheared slurry in simulant supernate.

Table 3.2 Microtrac results of Batch 3 Tank 40 Test 2 sheared slurry in different suspending media.

Suspending Medium	Water		Filtered Supernate		Simulant Supernate	
	Not Sonicated	Sonicated	Not Sonicated	Sonicated	Not Sonicated	Sonicated
mv (mean volume in μm)	11.81	10.88	15.78	17.74	11.92	9.59
mn (mean number in μm)	0.65	0.65	0.65	0.65	0.74	0.65
D_{90} (percent less than μm size indicated)	33.87	29.54	49.67	56.43	35.45	26.83
D_{50} (percent less than μm size indicated)	4.64	4.64	5.01	5.17	4.61	4.36
D_{10} (percent less than μm size indicated)	1.09	1.06	1.18	1.17	1.10	1.22

The Microtrac results indicate that shearing has had a large impact on the particle size of this sludge simulant. The suspending medium (filtered supernate *vs* water) has a large effect on the largest size particles as reflected by the D_{90} values listed for each set of experiments but a smaller effect on the smallest particles. Also, the number data indicate that the most numerous particles are those nearest the average particle size and there is not a shift toward a large number of smaller particles with change in suspending medium. There is little difference between the PSD determined in simulant supernate and water. It was expected that the simulant supernate PSD would be more similar to that of the filtered supernate values. It appears that in water and simulant supernate some of the large particles are dissolving. However, the elemental composition and the percent of dissolved solids in the simulant supernate closely matched the filtered supernate composition. The D_{90} values are decreased when the sample is sonicated, albeit a small amount, and water or simulant supernate is used as the suspending medium. The particles were sonicated for one minute to reduce agglomerations of particles into primary particles. Sonication has been shown to be most effective at reducing agglomerations during the first minute of use but if the sample is sonicated more than three minutes it can start to dissolve due to thermal heating caused by the sonication.[9] No additives were added to any of the experiments to prevent re-agglomeration of the particles after sonication. The PSD came out to be very different when this simulant slurry was analyzed on the Microtrac X-100 'hot' instrument. However, the same trends were observed for different suspending media. The largest particles were observed in simulant supernate (Tank 40 RAD Simulant Supernate was used) and the smallest in water.

Analysis of Batch 3 Tank 40 Test 3C slurry was accomplished using the Microtrac S3000 non-radioactive instrument and by the following protocol. Twelve sub-samples (~2 g each) were pulled from a 60 mL container, after shaking the sample bottle vigorously for approximately one minute, using a slurry pipette that had been cut off. Three samples were analyzed with water, three were sonicated in a bath and then analyzed in water, three were analyzed in simulant supernate and three were sonicated in a bath and then analyzed in simulant supernate. The simulant supernate composition was based upon the analysis of actual filtered supernate from Batch 3 Tank 40 Test 3C filtered supernate and confirmed by re-analysis of the simulant supernate. Again, this set of experiments was designed to specifically monitor changes in the data when the suspending medium was changed or the sample was sonicated. The actual PSD obtained by either instrument was not that important, although in theory similar PSDs should be obtained.

Figures 3.18-3.21 contain the average results from five experiments except the experiment done in simulant supernate without sonication, which is an average of three experiments. This sludge slurry was also re-analyzed on the 'hot' Microtrac X-100 instrument for comparison. Table 3.3 contains a summary of the Microtrac results.

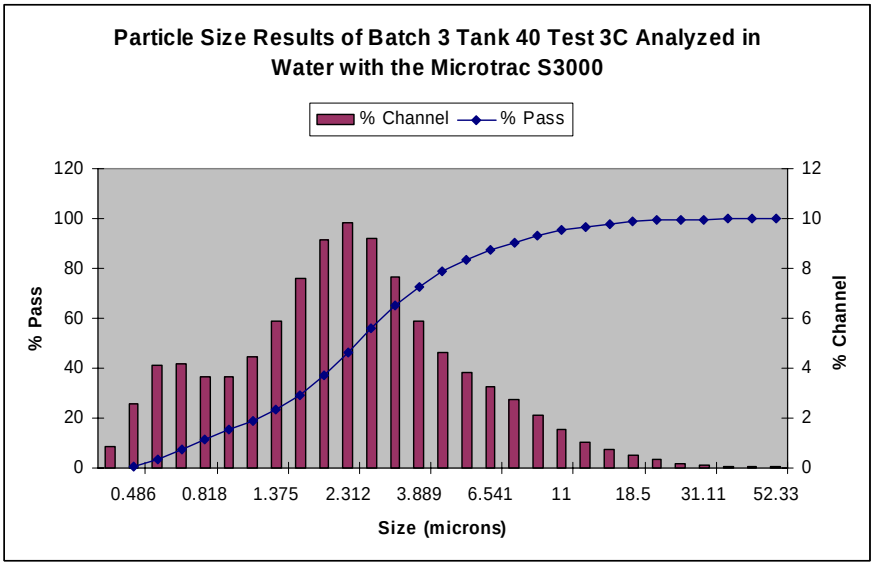


Figure 3.18 Results of Batch 3 Tank 40 Test 3C Analyzed in Water with the Microtrac S3000.

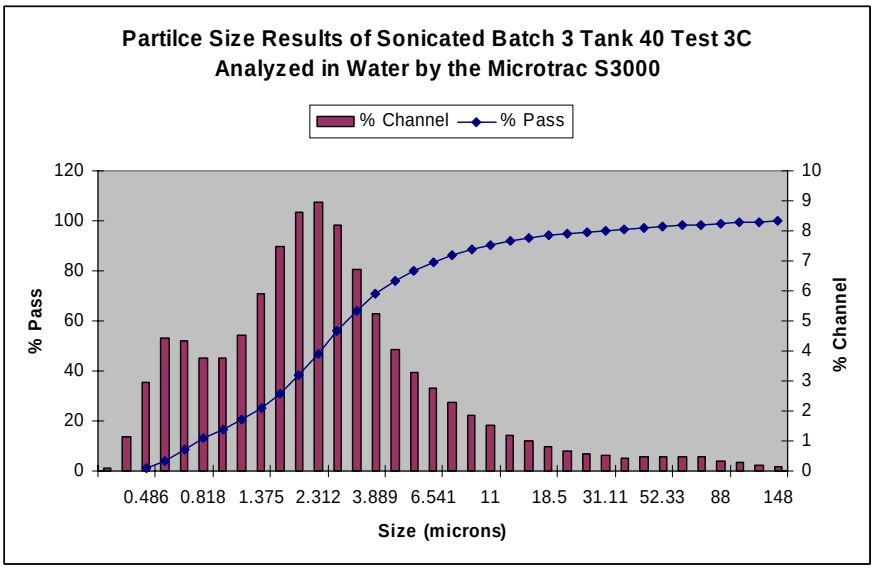


Figure 3.19 Results of Sonicated Batch 3 Tank 40 Test 3C Analyzed in Water with the Microtrac S3000.

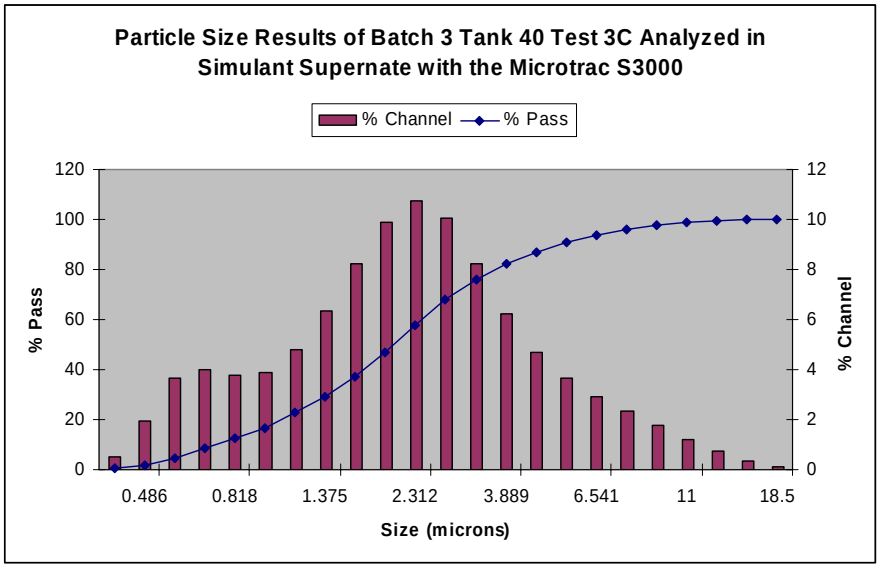


Figure 3.20 Batch 3 Tank 40 Test 3C Analyzed in Simulant Supernate with the Microtrac S3000.

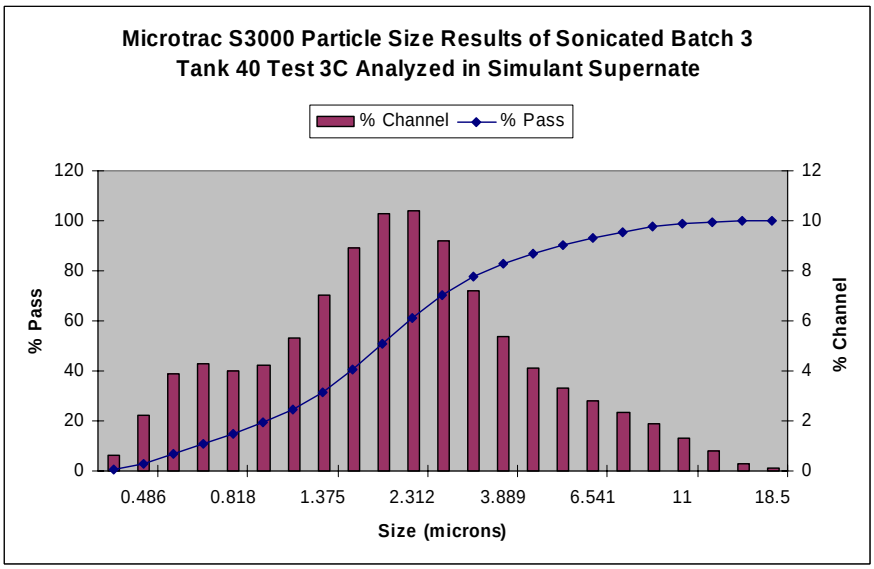


Figure 3.21 Sonicated Batch 3 Tank 40 Test 3C Analyzed in Simulant Supernate with the Microtrac S3000.

Table 3.3 Microtrac S3000 and X-100 results of Batch 3 Tank 40 Test 3C slurry in different suspending media.

Instrument	Microtrac S3000				Microtrac X-100		
Suspending Medium	Water		Simulant Supernate		Water	Simulant Supernate	
	Not Sonicated	Sonicated	Not Sonicated	Sonicated	Not Sonicated	Not Sonicated	Sonicated
mv (mean volume in μm)	3.07	5.24	2.57	2.64	3.81	23.09	14.24
mn (mean number in μm)	0.65	0.62	0.68	0.67	1.85	1.86	2.00
D_{90} (percent less than μm size indicated)	6.34	9.58	5.38	5.35	5.21	86.29	47.92
D_{50} (percent less than μm size indicated)	2.09	2.06	1.92	2.04	2.41	2.622	2.355
D_{10} (percent less than μm size indicated)	0.64	0.61	0.66	0.68	1.65	1.675	1.772

The Microtrac results of the Batch 3 Tank 40 Test 3 simulant sludge slurry are less consistent than results obtained on the sheared simulant slurry. Only a small amount of Batch 3 Tank 40 Test 3C simulant slurry was available and therefore Microtrac analysis was done using only water and simulant supernate not filtered supernate. PSDs change with different suspending medium upon analysis with the Microtrac X-100 but do not significantly with the Microtrac S3000 except for the analysis done in water with sonicated simulant. The data from the Microtrac X-100 indicate the largest particles were affected by a change in the suspending medium. The Microtrac X-100 data suggest sonication changes the PSD. The D_{90}, D_{50} and D_{10} simulant supernate values are smaller for the Microtrac S3000 instrument compared to the Microtrac X-100. The D_{90}, D_{50} and D_{10} values are more similar for the two instruments when water is the suspending medium. Again, in the histogram plot, the most numerous particles are centered near the average particle size and there is not a shift toward a large number of small particles. One thing noted during the radioactive and non-radioactive instrument cross experiments was that the sample flow rates are set differently. The radioactive instrument does not have an adjustable flow rate but the non-radioactive instrument does. The flow rate could have an impact on the particle size of the sludge if, during analysis, the force from the flow is able to break up the particles. In one experiment, the flow rate on the non-radioactive instrument was set to 60% and no difference in the particle size of slurry resulted. The flow rate of each instrument is presumably set at a rate high enough to mobilize the sample but low enough to minimize bubble entrainment. Bubble entrainment becomes more and more of a problem at higher pH values and greater $[OH^-]$. It is recommended that more studies concerning the instrument flow rates be performed.

Sludge Batch 3 Tank 40 Radioactive Sludge Slurry

Sub-samples of the Sludge Batch 3 Tank 40 radioactive sludge (~1 g each) were diluted in a suspending medium (water or simulated supernate) left to sit overnight, then sonicated if necessary prior to decanting

the suspending medium and weighing out the resulting wet solids. Approximately 0.2 g of the concentrated sludge solids were weighed out into a green shielded bottle and removed from the SRNL shielded cells for analysis. The solids were then diluted in the sample chamber of the Microtrac analyzer in a suspending medium and analyzed. In each case, care was given to sub-sampling from a larger vessel. The results are shown in Figure 3.22-Figure 3.24 and summarized in Table 3.4.

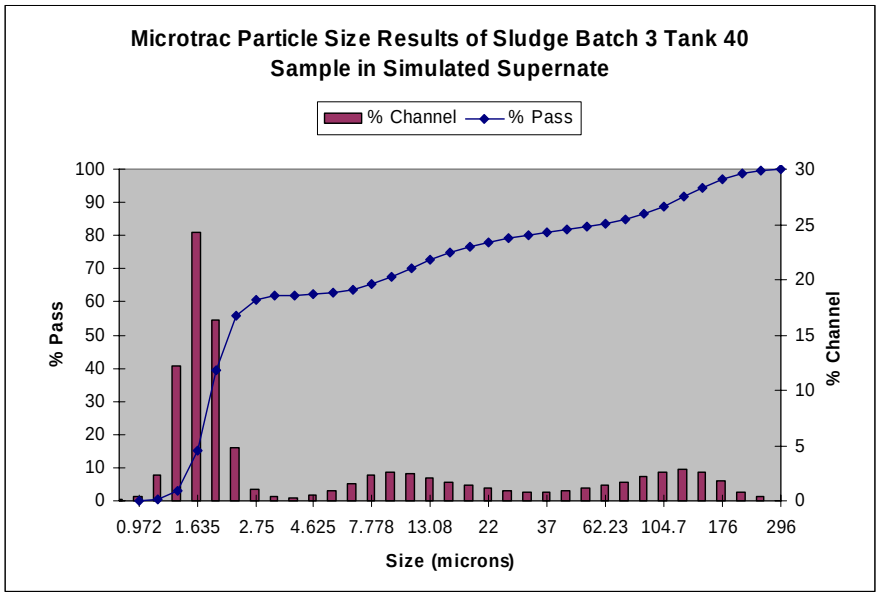


Figure 3.22 Microtrac Results of Batch 3 Tank 40 Radioactive Sludge Analyzed in Simulant Supernate with the Microtrac X-100.

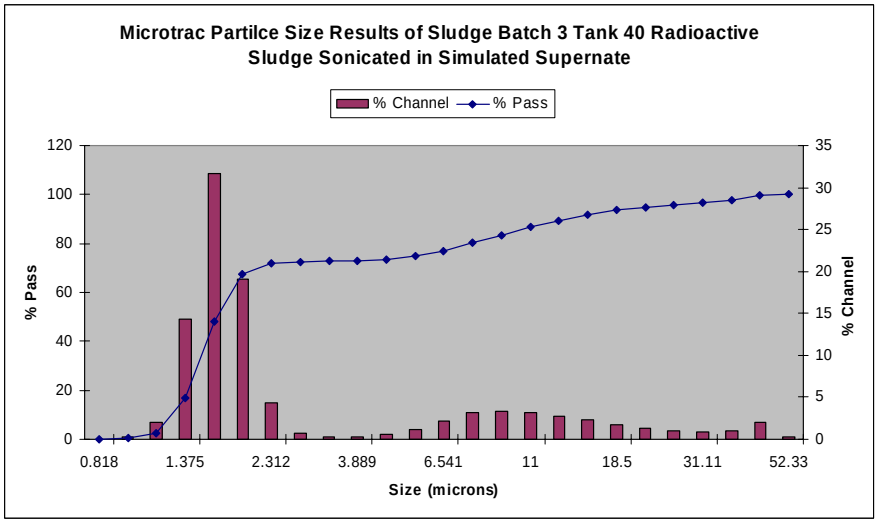


Figure 3.23 Microtrac Results of Sonicated Batch 3 Tank 40 Radioactive Sludge Analyzed in Simulant Supernate with the Microtrac X-100.

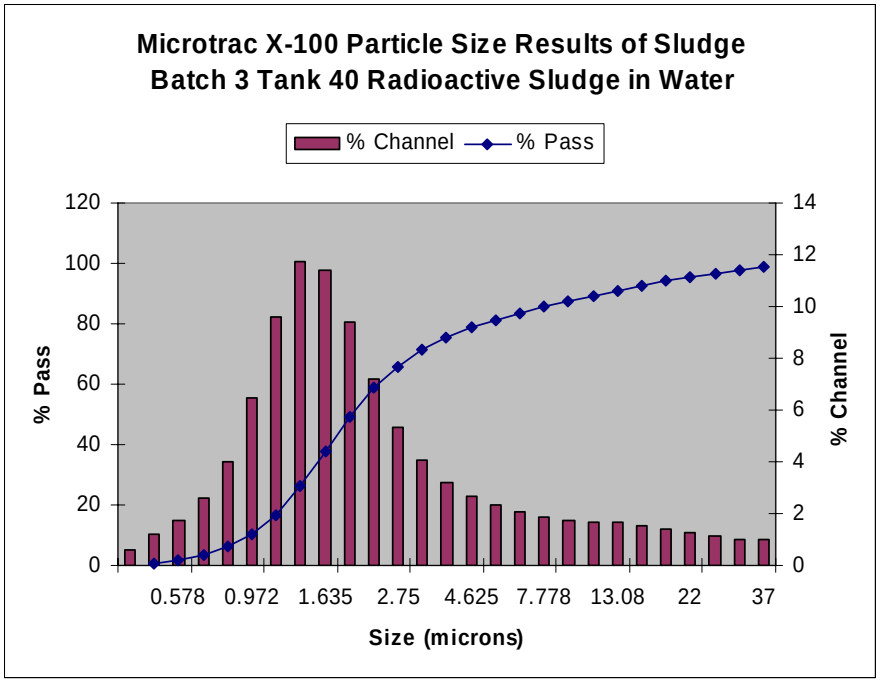


Figure 3.24 Microtrac Results of Batch 3 Tank 40 Radioactive Sludge Analyzed in Water with the Microtrac X-100.

Table 3.4 Summary of Microtrac X-100 Particle Size Results of Tank 40 Radioactive Sludge

Instrument	Microtrac X-100		
Suspending Medium	Water	Simulant Supernate	Simulant Supernate
	Not Sonicated	Sonicated	Not Sonicated
mv (mean volume)	3.854	5.21	22.68
mn (mean number)	0.762	1.45	1.438
D_{90} (percent less than µm size indicated)	9.342	13.71	94.25
D_{50} (percent less than µm size indicated)	1.721	1.66	1.801
D_{10} (percent less than µm size indicated)	0.839	1.30	1.305

The largest PSD was observed in the simulant supernate, as expected, and the smallest particles were observed when water was used as the suspending medium. Sonication can be used to break up the particle agglomerations into primary particles and leave the primary particles intact. Data from the analysis of the sludge slurry in water indicate that agglomerations are broken up and that some dissolution upon dilution may be taking place because the particle size is smaller in every category compared to the sonicated simulant supernate runs. No experiments were performed to monitor changes in particle size as a function of time in water or simulant supernate.

It is worth noting that the NIST 1984 SRM was analyzed for each instrument (Microtrac S3000 and Microtrac X-100) in the suspending medium to be used prior to the analysis of each batch of samples in that suspending medium. In each case, the PSD of the standard was within the range specified. The SRM being used is a robust material and appears to not be affected by changes in the suspending medium. This also indicates that each of the instruments can handle a solution with a high pH (>12 for the filtered supernate and simulant supernate) and accurate results are still obtained.

Recommendation of Use

Microtrac analysis should be used as the primary method for determining particle size of simulant and radioactive sludge slurries. The analysis should be tailored to the information desired and analysis should be done using the radioactive Microtrac X-100 instrument for consistency until differences between the two instruments can be resolved.

4.0 CONCLUSIONS

Sieving, Lasentec and Microtrac have been used to determine particle size in non-radioactive and radioactive sludge slurries. Sieving experiments on non-radioactive and radioactive sludge slurries were performed using water and supernate as the re-circulating medium and the results were similar to one another. It is recommended that sieving be used as a back-up method to determine particle size in sludge slurries and be used as the primary method when separation and identification of specific particles (>6 μm) needs to be accomplished (e.g. coal particles in the waste).

Lasentec experiments were performed on SRAT material where an increased amount of ground glass frit was added to test the ability of the Lasentec instrument to distinguish PSD as the insoluble solids concentration increased. However, particles at and below 10 μm in size in SRAT product and presumably SRAT feed, present a system that is sufficiently challenging to the Lasentec instrument and blind it to further changes in the particle distribution. It is recommended that the Lasentec system not be considered for the purpose of analyzing DWPF feeds and/or SRAT products at this time or for the determination of PSD in simulant and radioactive sludge slurries.

Microtrac analysis of non-radioactive and radioactive sludge slurries were performed using different suspending mediums (water, filtered supernate and/or simulant supernate), and in some cases the samples were subjected to ultra-sonication. Different PSDs of non-radioactive and radioactive sludge slurries were observed on the radioactive Microtrac X-100 instrument depending upon the suspending medium used and if the sample was subjected to ultra-sonication. Smaller PSDs and smaller changes in the PSD were noted when the same simulant sludge slurries were analyzed on the non-radioactive Microtrac S3000 instrument.

Microtrac is the recommended method at the current time for determining particle size of non-radioactive and radioactive sludge slurries. Small sample sizes are needed and both volume and number data can be obtained. However, the suspending medium has an affect on the PSDs and careful consideration should be given to experimental protocol and chemical reactions occurring when using different mediums.

This page intentionally left blank

5.0 RECOMMENDATIONS/PATH FORWARD

Based upon the results obtained to date, the following is recommended.

- Microtrac analysis should be used as the primary method for determining particle size of simulant and radioactive sludge slurries. The analysis should be tailored to the information desired and analysis should be done using the radioactive Microtrac instrument that AD has until differences between the radioactive and non-radioactive instruments can be resolved.

- It is recommended that the Lasentec system not be considered for the purpose of analyzing DWPF feeds and/or SRAT products at this time.

- It is recommended that sieving be used as a back-up method to determine particle size in sludge slurries and be used as the primary method when separation and identification of specific particles needs to be accomplished (e.g. coal particles in the waste) if the particles are above 6 microns.

- It is also noted that clear communication between the customer and operator about the sampling methodology must occur to ensure quality results.

More experimental work is needed to refine particle size analysis on the Microtrac instrument. The following experiments are recommended

- Design experiments to interrogate the agglomeration and de-agglomeration phenomenon suspected to happen when sludge is mixed or transferred between waste tanks.

- Evaluate the use of a global diluent *versus* a tailored simulant supernate or diluent.

- Evaluate changes of the surface of the sludge particles and the resulting chemistry in different suspending media.

- Evaluate the affect that specific ions and anions have on the PSD (i.e., [OH^-], [Na+]).

- Investigate instrument flow rates.

- Evaluate particle size as a function of time in suspending media.

This page intentionally left blank

6.0 REFERENCES

[1]Schramm, G. *A Practical Approach to Rheology and Rheometry*, Gebrueder HAAKE GmBH, D-76227 Karlsruhe, Dieselstrasse 4, Federal Republic of Germany.

[2]Eibling, R.E. **Impact of Simulant Production Methods on the Physical Properties of DWPF Sludge Batch 3 Simulant** WSRC–TR-2004-00578.

[3]Click, D.R. **Scoping Studies to Develop a Method to Determine Particle Size in Simulant Sludge Slurries by Sieving** WSRC-TR-2005-00077.

[4]Eibling, R.E. **Impact of Simulant Production Methods on the Physical Properties of DWPF Sludge Batch 3 Simulant** WSRC-TR-2004-00578**.**

[5]Jewtt, J.R.; Jensen, L. **Assessment of Available Particle Size Data to Support Analysis of the Waste Feed Delivery System Transfer System** RPP 6427 Rev 0.

[6]Fellinger, T.L. **Results of the Shielded Cells Sludge Batch 2 Phase II Testing** SRT-GDP-2003-00078.

[7]Poloski, A.P.; Bredt, P.R.; Swoboda, R.G. **Rheological and Physcial Properties of AP-104 LAW Pretreated Waste and Melter Feed** PNWD-3465 or WTP-RPT-107 Rev 0.

[8]Bontha, J.R.; Colton, N.G.; Daymo, E.A.; Hylton, T.D.; Bayne, C.K.; May, T,H. **Qualification of the Lasentec M600P Particle Size Analyzer and the Red Valve Model 1151 Pressure Sensor** PNNL 13064.

[9]Sistare, F; St. Pierre Berry, L.; Mojica, C.A. **Process Analytical Technology: An Investment in Process Knowledge** *Organic Process Research and Development*, **2005**, 9, 332-336.

[10]E-mail communication from David Koopman, Process Engineering Technology.

[11]Bannochie, C.J., Fellinger, T.L., Pareizs, J.M. **Tank 40 Final SB3 Chemical Characterization Results** WSRC-TR-2005-00049.

[12]Hansen, E; Crawford, C. **Hanford HLW AY102/C106 Pretreated Sludge Physical and Chemical Properties Prior to Melter Feed Processing** WSRC-TR-2004-00394 REVA.

[13]Jillavenkatesa, A.; Dapkunas, S.J.; Lum, L-S. H. **NIST Recommended Practice Guide to Particle Size Characterization** Special Publication 960-1.

[14]Bredt, P.R.; Tingey, J.M.; Schmidt, A.J. **Comiplation and Integration of K Basin Sludge Partcile Size Analysis Data** PNNL-13376

[15]Bannochie, C.J., Daniel, G. **Lasentec Measurements of DWPF Sludge/SRAT Surrogate** SRNL-ITS-2005-00188.

This page intentionally left blank

7.0 ACKNOWLEDGEMENTS

The authors would like to acknowledge Art Jurgensen for the technical review, Terri Fellinger, David Missimer (for providing Microtrac analysis and discussions), Don Blankenship for discussion and Rita Sullivan.