Grain and Floc Size Distribution Analysis of Fine Coal Refuse Slurry

Analyse de la Composition Granulométrique de Grain et Flocons de Fin Déchets de Charbon en Suspension

Cyrus Jedari, Angelica M. Palomino
Department of Civil and Environmental Engineering, University of Tennessee, USA, apalomin@utk.edu

Howard Cyr
Department of Anthropology, University of Tennessee, USA

Eric C. Drumm
Department of Biosystems Engineering and Soil Science, University of Tennessee, USA

Daniel Boles
Geotechnical Engineering Services, S&ME Inc., USA

ABSTRACT: Fine coal refuse (FCR) is a by-product of the coal preparation/crushing process. FCR is typically hydraulically placed in on-site impoundments, and the predominant silt-sized material usually contains significant amounts of coal which yields a low specific gravity. A flocculant is added to the slurry in order to accelerate settlement and subsequent consolidation. The added flocculent complicates characterization of FCR by altering the effective grain size distribution, combining smaller particles into larger flocs. The purpose of this study is to investigate the influence of an anionic polyacrylamide flocculant on the measured grain size distribution of FCR slurry samples using two different analysis methods: (1) traditional hydrometer and sieve analysis and (2) laser diffraction particle size analysis (PSA), with and without flocculant. Results from both the hydrometer and PSA methods indicate the floc stability in the FCR slurry is susceptible to the surrounding chemical environment. The advantages of the PSA method are the method has a greater capability of identifying the presence of flocs and can be applied to FCR slurries in their as-placed condition without any pretreatment.

RÉSUMÉ: Les fin déchets de charbon (FDC) sont produits pendant le processus de préparation/broyage du charbon brut. Ces déchets sont typiquement placés hydrauliquement dans des bassins de retenue sur site et contiennent habituellement des quantités significatives de charbon de faible densité spécifique à cause de leur taille limoneuse. Un floculant est ajouté à la suspension de fin particules de charbon afin d'accélérer leur dépôt et consolidation ultérieure dans le bassin. L'addition du floculant complique la caractérisation du FDC en modifiant la distribution de la taille des grains et en combinant des particules plus petites en flocons de plus grande taille. Le but de cette étude est d'étudier l'influence d'un floculant anionique à base de polyacrylamide sur la distribution granulométrique mesurée des échantillons de FDC en suspension, avec et sans floculant, en utilisant deux méthodes d'analyse différentes: (1) l'analyse traditionnelle du tamis et de l'hydromètre et (2) l'analyse granulométrique laser. Les résultats des deux méthodes indiquent que la stabilité du floc dans la suspension de FDC est sensible à l'environnement chimique environnant. Les avantages de l'analyse granulométrique laser sont que cette technique est susceptible d'identification de la présence de flocs et peut être appliqué à des suspensions de FDC dans leur état initial de déposition sans aucun prétraitement.

KEYWORDS: Particle size analysis, Floc size analysis, laser diffraction, Fine coal refuse

1 INTRODUCTION

According to the U.S. Energy Information Administration Statistics (2016), approximately 900 million tons of coal was produced in the U.S. during 2015, 90 percent of which was used in coal fired power plants to generate 33 percent (~ 1352 million MWh) of the total U.S. power production. Coal is processed to remove some impurities before transporting to the power plants, leaving behind waste materials usually described as coarse or fine refuse. The fine coal refuse (FCR) is typically stored behind on-site impoundments constructed primarily with the coarse refuse. This FCR is typically hydraulically placed in on-site impoundments by pumping in slurry form through a series of pipelines.

Flocculants are added to the fine coal refuse (FCR) slurry in order to aggregate fine particles and accelerate settlement and subsequent consolidation after placement. As a result, the effective particle size distribution of the FCR is altered, potentially impacting the properties of the placed material such as consolidation time, void ratio, and rheology. For example, particle size distribution is one of the most important factors controlling viscosity of coal-water suspensions (Fidleris and Whitmore, 1961; Farris Ferini et al., 1984). However, the ability to accurately measure particle size distribution of FCR, which consist of aggregated flocs, is not straight forward. In this study, the results of traditional hydrometer sedimentation tests for grain size analysis of fine coal refuse were compared with those from a laser diffraction particle size analyzer (PSA).

2 TEST METHODS AND MATERIALS

Traditional methods of grain size analysis are based on the physical separation of coarse grains (e.g. wet or dry sieve technique) and changes in the specific gravity of a soil suspension through time (e.g. hydrometer technique) (Day 1965). Although established geotechnical quantitative techniques exist (ASTM D 422-63), these methods have several disadvantages such as the length of time needed to complete the analyses, the dependency of results on laboratory techniques, and operator error (Syvitski et al., 1991). These

methods also require relatively large sample sizes (tens to hundreds of grams). Moreover, the hydrometer method relies on Stokes' Law to quantify grain size distribution, which introduces assumptions rarely present within natural settings such that all particles are solid spheres and each have the same unit weight (Murthy 2002). These issues make traditional methods less advantageous for rapid and accurate analysis of a large number of particulate samples, especially when dealing with aggregate materials such as FCR, which are artificially produced and contain substantial quantities of carbon.

The coal refuse samples used in this study were obtained as slurry from a coal mine in the Commonwealth of Kentucky (US).

2.1 Hydrometer analysis

As shown in Figure 1, the chemistry of the suspending fluid has a significant influence on the sedimentation behavior of the FCR. Traditional hydrometer tests were performed on the FCR according to ASTM D422 with some modifications to the suspending fluid. FCR samples were analyzed with and without sodium hexametaphosphate (dispersant), as well as with the supernatant fluid collected with the FCR slurry samples to represent the field conditions.

Three separate hydrometer tests were conducted each using a different dispersant fluid. The first hydrometer test was performed using a 40 g/l sodium hexametaphosphate – distilled water solution. The second test was conducted using only distilled water. The third test was conducted using the as-collected supernatant fluid from the FCR slurry samples.

For the first and second tests, 50 grams of oven dried FCR were soaked in the sodium hexametaphosphate solution for 16 hours. The third test used 50 grams of FCR in slurry form. The moisture content of the as-collected FCR slurry was determined previously. Thus, a given volume of FCR slurry had a known mass of solids. The material was then placed in the settling tube and brought to volume (i.e. 1000mL) using the appropriate solution. Hydrometer readings were recorded at time intervals of t = 2, 5, 8, 15, 30, 60, 120, 250, 1,440 minute. Following the 1,440 minute reading, additional hydrometer measurements were taken until the effective diameter (D_{10}) was identified.

Following the hydrometer analysis, each suspension was passed through a series of nested sieves to define the size distribution of particles larger than the sieve #200 (i.e. greater-than-75 μm). The fluid and particles passing the sieve #200 was collected in a plastic bucket. The material retained in each sieve was dried in the oven and weighed.

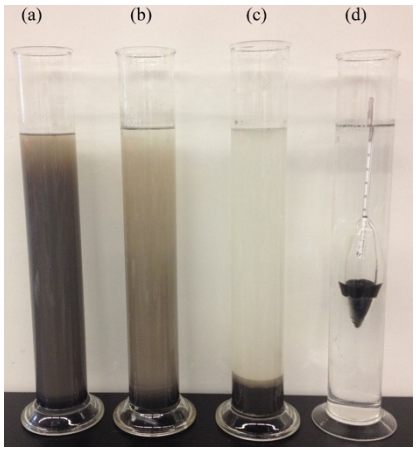

Figure 1. Hydrometer tests with different background solutions (a) FCR with sodium hexametaphosphate (ASTM D422), after 3 weeks of sedimentation, (b) FCR in distilled water after 3 weeks, (c) FCR in the sample supernatant fluid after 24 hours, and (d) tap water.

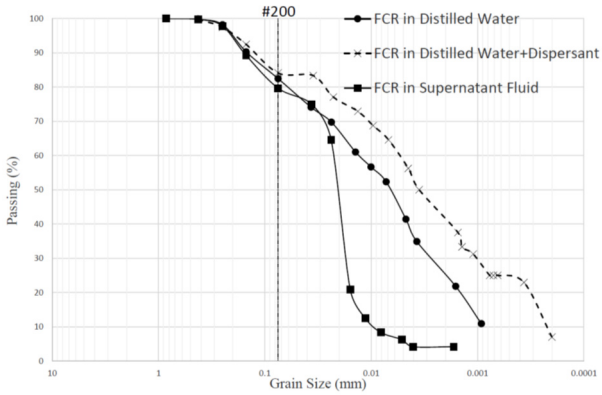

Figure 2. Grain size distribution curves for FCR in different background solutions.

The FCR grain size distribution curves based on hydrometer and sieve analyses are presented in Figure 2. For all cases, approximately 80 % of fine coal refuse is finer than 75 μm (#200), and the D_{50} values for FCR in the solution with sodium hexametaphosphate, distilled water and supernatant fluid are 6.3 μm, 3.5 μm and 20 μm, respectively.

Based on the grain size distribution curves, the majority of the FCR material falls within the silt-size range. These results are in accordance with the findings of Hegazi et al (2004) who reported FCR material to be a non-plastic silt (ML). Note that for particles/flocs greater than 75 μm (retained on sieve #200), the background solution does not have a significant effect but does have a significant influence on particles/flocs passing the sieve #200.

2.2 Laser diffraction analysis

In recent decades, new methods for particle size analysis have been developed. One such method is laser diffraction, which measures particle size distributions by analyzing the angular variation in the intensity of light scattered as it passes through a dispersed particulate sample (Sorti and Balsamo, 2010). For this study, grain size distribution was measured using a Mastersizer 3000 laser diffraction particle size analyzer (Malvern Instruments Ltd). A major advantage of Malvern's laser diffraction technique is the Mastersizer 3000 provides indirect size measurement of a wide range of particles (0.01 to 3500 μm) in a single sample during the same analysis (e.g. Beuselinck et al. 1998). The method also requires very little sample material (between 0.2 and 2.0 g) and short measurement times (approximately 10 to 20 seconds per analysis). Important to this study, when using the wet dispersion cell, repeated measurements can be run on the same sample. This not only provides statistical reproducibility but also allows one to accurately measure real-time changes in the grain size distribution through time (i.e. if the sample aggregates or deflocculates during the course of the analysis).

The Mastersizer 3000 employs a blue and red light dual-wavelength single-lens detection system to measure the degree to which light is diffracted by particles distributed within a medium. As shown in Figure 3, the Mastersizer 3000 is comprised of the dispersion unit, the sample measurement cell, two light sources, and a series of back scatter detectors, focal plane detectors, and side scatter detectors. Material is added to the dispersion unit using either the wet dispersion or dry dispersion modes. The suspended material is then circulated across the measurement cell and illuminated by a 10mW 470 nm blue LED and a 4 mW He-Ne 633 nm red laser. The blue

LED has a greater sensitivity to wide angle diffraction and allows for an accurate measurement of sub-100 nm particles.

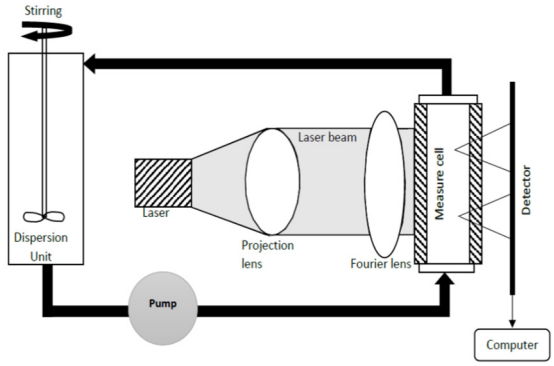

Figure 3. Schematic of laser diffraction method (after Storti and Balsamo 2010).

The red laser provides small angle resolution appropriate for particle sizes greater-than-100 nm. The angular variation in the intensity of scattered light is measured across each detector as energy impulses.

Using Mie theory of light scattering, the Mastersizer 3000 software calculates the particle distribution responsible for creating the measured scatter pattern. Mie theory provides a detailed mathematical description of the correlation between the particle size distribution of a sample and the intensity of the scattered light produced (Wriedt 2012). To accurately calculate the grain size distribution, the Mastersizer 3000 software takes into account the optical properties of both the sample material and the dispersing medium in the Mie algorithm. The source of the optical properties used in this study are discussed below. These properties include the refractive and absorption indices of the sample material and the refractive index of the dispersing medium and are entered into the Malvern software operating procedure prior to analysis.

This study employed the Hydro LV wet dispersion unit, which uses a 600 ml tank equipped with a centrifugal pump system to pass the suspended material through the measurement cell. Three different background solutions including distilled water, the supernatant fluid of the FCR slurry, and a 40 g/L sodium hexametaphosphate solution were used to investigate the influence of the background solution on the particle size distribution and floc size distribution of the FCR. For each test, approximately 600 ml of the specified background solution was added to the dispersion tank and allowed to circulate. The pump speed was then reduced to 0 rpm and between 1 and 2 g of the slurry matrix was added to the dispersion tank. The pump speed was then increased to 2,200 rpm to circulate the suspended material through the measurement cell.

Prior to the analyses, material properties specific to bituminous coal were added to the Malvern software measurement settings following Goodarzi and Murchison (1971). These included a refractive index of 1.8, absorption index of 0.1, and density of 2.21 g/cm^3. A total of six measurements were taken for each background solution to investigate the influence of time and pump speed on floc size formation/breakage. Each measurement was conducted with both the blue LED and the red laser for a duration of 10 seconds for each light. Values of D_{10}, D_{50} and D_{90} were obtained for each of six samples.

Figure 4 shows D_{90} values of FCR in each background solution. D_{90} values are representative of formed floc sizes during the sedimentation process for these cases. As shown in Figure 4, the flocculent/supernatant water suspension has higher D_{90} values in comparison with FCR in distilled water and the dispersant solution. The figure suggests the flocculent in the supernatant water causes an increase in floc size, and hence the D_{90} value, over time. For the FCR samples in distilled water and the dispersant solution, the value of D_{90} decreases with time.

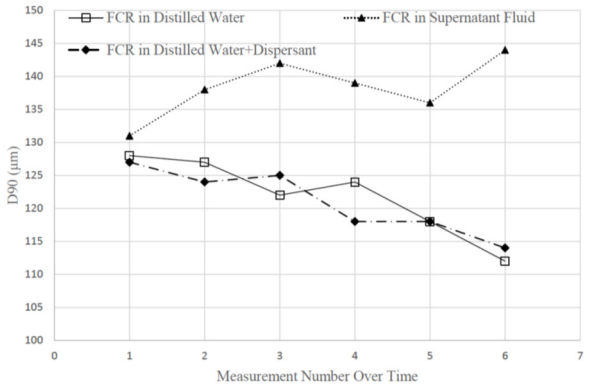

Figure 4. Values of D_{90} in different background solutions.

This trend is consistent with deflocculation, likely due to the decrease in flocculant concentration in the distilled water case and the presence of the deflocculating agent in the dispersant solution case.

3 DISCUSSION

The distribution curves obtained using hydrometer analyses suggest the particle size distribution changes over the duration of each test (Figure 2). The curve for FCR in the sodium hexametaphosphate solution shows two measurement plateaus, one at ~82 % passing and one at ~25 % passing. These plateaus are indicative of an increase in the percentage of finer particles over time (i.e. deflocculation). For the FCR in the background solution of supernatant fluid, a significant drop in percent passing from ~65 % to ~21 % may be the result of a significant increase in the percentage of larger particle sizes, which indicates flocculation is likely occurring between these measurement times. Thus, the effective size distribution of the FCR material does not remain constant through the duration of the test, a break of a fundamental assumption for hydrometer analysis (i.e. that the grain sizes remain constant throughout the test). Therefore, hydrometer analysis is not applicable for FCR suspensions if floc stability is in question.

A summary of the D_{90}, D_{50}, and D_{10} obtained using the PSA method and traditional hydrometer analysis is given Table 1.

Table 1. Values of D_{10}, D_{50} and D_{90} for PSA and hydrometer analysis in different background solutions.

Background Solution	Measurement Method	D_{90}	D_{50}	D_{10}
Dispersant Solution	PSA*	121	3.83	2.02
	Hydrometer	130	3.5	0.23
Distilled Water	PSA*	122	11.2	2.31
	Hydrometer	150	6.3	0.9
Supernatant Fluid	PSA*	138	10.7	2.17
	Hydrometer	151	20	10

*Values obtained using PSA are the average of 6 measurements of the same suspension

Note that the D_{90} values obtained from the PSA method are

smaller than values of D_{90} obtained from the hydrometer method. This difference is likely due to the transformation of the suspended material during the course of the hydrometer analysis as well as some influence from sampling bias (difference between the larger sample size for hydrometer and smaller sample size for PSA).

Based on measurements made using the PSA method, the D_{90} for each suspension either increased or decreased over the six consecutive measurements of the same sample. Yet, the values of D_{10} and D_{50} for the same suspensions did not show any significant change (measurements not shown here). Figure 5 shows the frequency of volume density with particle size for FCR in different background solutions for measurement #1 and measurement #6 for each case (left Y-axis). The change in volume density from measurement #1 to measurement #6 for each case is also shown (right Y-axis). A positive change in volume density from measurement #1 to #6 denotes an increase in the number of particles within a given size range (flocculation), while a negative change denotes a decrease of particles within a given size range (deflocculation). For the FCR in the supernatant fluid, the change in volume density for particle sizes 120 μm to 500 μm indicates flocculation has occurred from measurements #1 to #6, while deflocculation is evident in the size range 3 μm to 120 μm. This shift in grain/floc size distribution occurs only for particle sizes greater than the D_{50} values, which is consistent with the absence of variation in D_{10} and D_{50} values from measurement #1 to #6.

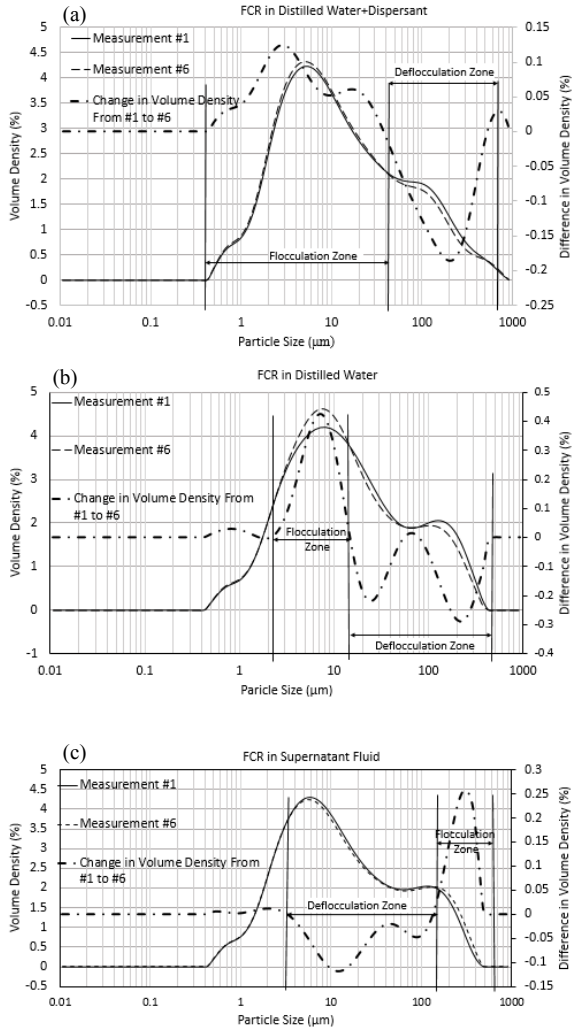

Figure 5. Particle size volume density (right axis) and difference in volume density (left axis) for PSA measurements #1 and #6 for (a) distilled water + dispersant, (b) distilled water, and (c) supernatant fluid.

4 CONCLUSIONS

The grain size distribution curves for three background solutions of fine coal refuse were obtained using traditional hydrometer analysis and laser diffraction with a Particle Size Analyzer. The results show that (1) the background solution chemistry influences the FCR floc stability and alters the grain size distribution over time and (2) the D_{90}, D_{50}, and D_{10} values obtained from the two methods are not in agreement, with larger D_{90} values reported from hydrometer analysis. The differences between the values obtained highlight one important limitaion of hydrometer analysis, which is the assumption the grain size distribution remains constant with time. If grain size changes during the hydrometer analysis, the initial readings represent a different sample than the final readings and may not be comparable. In other words, the hydrometer test may not accurately measure grain size distribution over the measurement period.

Laser diffraction offers several advantages including a short measurement period, small sample size requirement, and the ability to measure a wide range of particle sizes in the same analysis. This study highlights another advantage of the PSA method, the ability to accurately measure changes in particle size over time within the same sample. The ability to capture real-time flocculation or deflocculation means this technique can be applied to grain size distribution studies of FCR slurries in their as-placed condition without any pretreatment.

5 ACKNOWLEDGEMENTS

This research was part of a project funded by Office of Surface Mining Reclamation and Enforcement (OSMRE) of the U.S. Department of the Interior, whose support is gratefully acknowledged. We also thank the Geoarchaeological and Palaeoenvironmental Services Center at the University of Tennessee, Knoxville for its technical support and assistance with the Mastersizer 3000 analyses.

6 REFERENCES

ASTM. 2007. Standard test methods for Particle-Size Analysis of Soils, ASTM D422-63.

Beuselinck, L., Govers, G., Poesen, J., Degraer, G., and Froyen, L. 1998. Grain-size analysis by laser diffractometry: comparison with the sieve-pipette method. Catena, 32, 193–208,

Day, P. R. 1965. Particle fractionation and particle-size analysis. In Methods of Soil Analysis, edited by C.A. Black, pp. 545-567. American Society of Agronomy, Madison.

Ferrrini, F., Battara, V., Donati, E. Piccinini, C. 1984. Optimization of particle grading for high concentration coal slurry, Proc. 9th Int. Conference Hydraulic Transport of Solids in Pipes, Rome, Paper B2.

Fidleris, V., Whitmore, R.L. 1961. The physical interaction of spherical particles in suspensions, Rheol. Acta 1, 573

Goodarzi, F. and Murchison, D. G. 1971. Optical properties of carbonized vitrinites. Fuel, Vol 51, p 322-328

Hegazi, Y., Cushing, A., & Lewis, C. (2004). “Physical, Mechani cal and Hydraulic Properties of Coal Refuse for Slurry Impou ndment Design”, D’Appolonia Engineering, Monroeville, PA

Murthy, V.N.S. 2002. Geotechnical Engineering: Principles and Practices of Soil Mechanics and Foundation Engineering. Marcle Dekker, Inc., New York.

Storti, F. and Balsao, F. 2010. Particle size distributions by laser diffraction: sensitivity of granular matter strength to analytical operating procedures. Solid Earth, 1, 25–48

U.S. Energy Information Administration Statistics (2016), https://www.eia.gov/tools/faqs/faq.cfm?id=427&t=3 accessed January 3, 2016

Wriedt, Thomas 2012. Chapter 2. Mie Theory: A Review. In The Mie Theory: Basics and Applications, Wolfram Hergert and Thomas Wriedt, eds. Springer, New York.