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ANALYSIS OF TECHNIQUES FOR MEASUREMENT OF THE SIZE DISTRIBUTION OF SOLID PARTICLES

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Abstract - Determination of the size distribution of solid particles is fundamental for analysis of the performance several pieces of equipment used for solid-fluid separation. The main objective of this work is to compare the results obtained with two traditional methods for determination of the size grade distribution of powdery solids: the gamma-ray attenuation technique (GRAT) and the LADEQ test tube technique. The effect of draining the suspension in the two techniques used was also analyzed. The GRAT can supply the particle size distribution of solids through the monitoring of solid concentration in experiments on batch settling of diluted suspensions. The results show that use of the peristaltic pump in the GRAT and the LADEQ methods produced a significant difference between the values obtained for the parameters of the particle size model.
Keywords: Gamma-ray attenuation technique; Size distribution of diameters.

INTRODUCTION

The size distribution of particles characterized by small dimensions is usually given in terms of the Stokes diameter (d_{st}), i.e., the diameter of the sphere that falls with the same terminal velocity as the particle in the same fluid and at the same temperature. The distribution of Stokes diameters of solids is obtained in experiments on sedimentation of solids in diluted suspensions (about 0.75% of volume of solids) in which the concentration of solids is monitored in a given fixed position, through sampling and measurement with gravimetric techniques. Experiments with an Andreasen pipette (Allen, 1981) are usually utilized for this purpose.

The Andreasen pipette method has a major drawback: the long duration of the experiments. An alternative technique uses a test tube known as LADEQ (Silva and Medronho, 1986). The LADEQ test tube reduces the time of each experiment considerably. The artifice of producing the continual suction of the suspension through a tube located at the base of the test tube is used. However, even in experiments with the LADEQ test tube, the use of gravimetric techniques for measurement of concentrations results in a big loss of time between the collection of samples and their interpretation. Besides, the LADEQ test tube method is a destructive measurement technique.

In the Andreasen pipette method as well as in the LADEQ test tube method, the Stokes diameter (d_{st}) is calculated with the following equation:

$$d_{st} = \sqrt{\frac{18\mu h}{(\rho_s - \rho_l) g t}} \qquad (1)$$

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where μ is the viscosity of the liquid, ρ_s and ρ_l are respectively the densities of the solids and the liquid, *g* is the acceleration of gravity, *h* is the difference between the heights of the level of the suspension and of the point of collection of the samples and *t* is the time.

The fraction of particles with diameters smaler than the Stokes diameter (Y) is given by the equation

$$Y = \frac{\varepsilon_s}{\varepsilon_{so}} \tag{2}$$

where ε_s and ε_{so} are the volumetric concentrations of solids at any instant t and at t=0, respectively.

An alternative to the gravimetric technique is the use of the gamma-ray attenuation technique (GRAT). The main advantage of the use of this technique is that it is not destructive and allows an immediate analysis of results. Vaz et al. (1996) demonstrated the efficiency of GRAT in determinig the particle size of solids. However, there is a lack of information on the conditions under which the experiments were carried out and, mainly, on the approaches for determining the gamma-ray source to be used in the experiments, taking into account the composition of the solid analyzed.

In this paper we have tried to specify, through theoretical arguments, the best operational conditions so that the GRAT can be used with better results in experiments whose objective is to determine the particle size of solids.

The main objective of this work is to compare the results obtained with two traditional methodologies, GRAT and the LADEQ test tube technique, for determination of the particle-size distribution of powdery solids. The effect of draining the suspension in the two techniques used will also be analyzed.

THEORETICAL FUNDAMENTALS

Gamma-Ray Attenuation

The number of pulses produced by absorption of gamma rays in a scintillation detector is given by Lambert's equation (Gardner and Ely Jr., 1967):

$$R = R_0 \exp(-\sigma X) \tag{3}$$

where R_0 and R are respectively the relative number of monochromatic gamma-ray pulses before and after crossing a physical medium with a surface density X and a mass attenuation coefficient σ. The surface density of the medium is defined as the product of the specific mass (ρ) and the thickness (x):

$$X = \rho x \tag{4}$$

The deviation in the measurements of $X(\delta_x)$ and $R(\delta_R)$ is given with the following equation:

$$\delta_x = \left| \left(\frac{\partial X}{\partial R} \right)_\sigma \right| \delta_R \tag{5}$$

Using Lambert's equation, it is easy to show that

$$\left(\frac{\partial X}{\partial R} \right)_\sigma = -\frac{1}{\sigma R} \tag{6}$$

and

$$\delta_X = \frac{\delta_R}{\sigma R} \tag{7}$$

According to Gardner and Ely Jr. (1967), the error associated with the countdown due to its statistical nature is given by

$$\delta_R = \sqrt{\frac{R}{t_c}} \tag{8}$$

where t_c is the countdown time. Thus, the errors associated with measurement of surface density are given by the equation

$$\delta_x = \frac{1}{\sigma} \sqrt{\frac{\exp(\sigma X)}{R_0 t_c}} \tag{9}$$

The error δ_x will be at a minimum when

$$(\sigma X) = 2 \tag{10}$$

The Separation Process Research Nucleus (SPRN) at the Federal University of Uberlândia has an Americium source (Am^{241}). Its main characteristics are

- Activity, 100 mCi;
- Half-life, 423.6 years;
- Mean energy of the emission, 60 keV.

To study the possibility of using this source in the particle-size analysis of powders, the procedure proposed by Storm and Israel (1970) proved to be useful. These researchers say, "for a substance composed of n chemical elements, the mass attenuation coefficient can be given by the equation"

$$\sigma = \sum_{i=1}^{n} w_i f_i \sigma_{oi} \qquad (11)$$

where w_i is the mass fraction of the element *i* in the molecule of the substance and f_i and σ_i are respectively a characteristic factor and the mass attenuation coefficient of each element *i*.

In Table 1 the theoretical values for the mass attenuation coefficients for niobium oxide, Nb_2O_5; barium sulfate, $BaSO_4$; and water are presented. The f_i values and σ_{oi} were obtained from the article of Storm and Israel (1970).

According to Allen (1981), the initial volumetric concentration of solids in the tested suspension should be less than 1% so that the effects of concentration are not seen when determining of the distribution of Stokes diameters of a powdery material. In Table 2 the values of $(\sigma X)_{s\text{-}susp}$ are presented for solids in aqueous suspensions with 1% volumetric concentrations of barium sulfate and niobium oxide (with 40% impurities that have low absorptions) interacting with monochromatic radiation of 60 keV. These values were calculated from the following equation:

$$(\sigma X)_{s-susp} = 0.01 x \rho_s \sigma_s \qquad (12)$$

Table 1: Mass attenuation coefficients for gamma rays with energy of 60 keV for barium sulfate, niobium oxide and water.

Substance [mol] (g/gmol)	i	Atomic Mass (g/gmol)	w_i	f_i (cm^2/b/g)	s_{oi} (b)	$w_i f_i s_{oi}$ (cm^2/g)	s (cm^2/g)
$BaSO_4$ [233.3]	Ba	137.3	0.5883	0.004385	1940	5.0055	5.1131
	S	32.1	0.1375	0.018780	21.5	0.0555	
	O	16.0	0.2742	0.037640	5.05	0.0521	
Nb_2O_5 [265.8]	Nb	92.9	0.6990	0.064820	619	28.0464	28.1036
	O	16.0	0.3010	0.037640	5.05	0.0572	
H_2O [18.016]	H	1.008	0.1119	0.597500	0.545	0.0364	0.2052
	O	16.0	0.8881	0.037640	5.05	0.1688	

Table 2: Theoretical values of sX for monochromatic gamma rays (60 keV) interacting with 1% volume aqueous suspension of barium sulfate and niobium oxide with x=6.2 cm.

Aqueous Suspension	r_s (g/cm^3)	$(sX)_{s\text{-}susp}$
$BaSO_4$	4.21±0.10	1.381
Nb_2O_5 (60%)	4.03±0.09	4.213

MATERIALS AND METHODS

With the objective of verifying experimentally the theoretical behavior described in the previous section, settling tests were conducted with aqueous suspensions of barium sulfate (ρ_s = 4.21±0.10 g/cm^3) and niobium oxide (ρ_s =4.03±0.09 g/cm^3) at 1% volumetric concentrations. Four types of experiments were carried out:

- Batch settling with GRAT tests without suspension suction or batch settling tests without flow;
- Batch settling with GRAT tests with continuous suction of suspension or batch settling tests with flow using a peristaltic pump.
- Batch settling with the LADEQ test tube with continuous gravitational draining of suspension.

- Batch settling with the LADEQ test tube with continuous suction of the suspension using a peristaltic pump.

Gamma-Rays Attenuation Technique (GRAT)

The system of detection of radiation pulses that is presented in Figure 1 has the following components:

- A photomultiplier valve, which transforms luminous pulses into electric pulses;
- A high-tension source that supplies stabilized tension to the photomultiplier valve;
- A preamplifier, which adjusts the height of pulses originating at the photomultiplier valve so that they can be transmitted to the amplifier.
- An amplifier, which increases the height and the width of the pulses to about 1 μs and 10V, respectively;
- A voltage discriminator that allows establishment of the tension interval in order to be sent to the pulse counter;
- A pulse counter that quantifies the pulses released by the voltage discriminator. The pulse counter module is provided with a timer, which defines the countdown time.

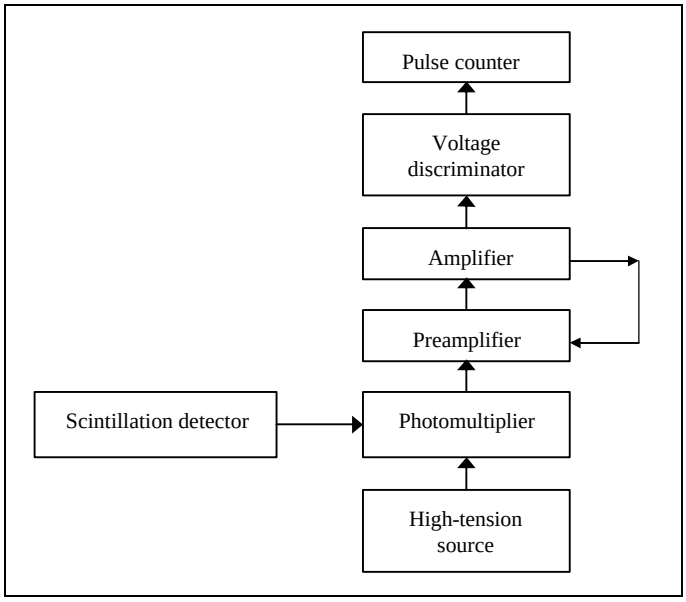

Figure 1: Schematic view of gamma-ray detection system.

The suspension to be analyzed was prepared in a 1-L volumetric balloon, in which the amount of solids was added to produce a 1% volumetric solid concentration in water. To minimize the effects of gathering particles for niobium oxide, one gram of sodium hexametaphosphate was added to each liter of suspension and then the balloon was placed in an ultrasonic bath for one minute. After use of the ultrasonic bath, the content of the volumetric balloon was transferred to a 1-L test tube (diameter of 6.2 cm), under vigorous shaking with a manual axial shaker.

The source-detecting system was positioned directly opposite to the test tube at a height of about 21 cm below the level of the suspension. Measurements were made and a countdown time of 10 s was adopted.

In the settling experiments monitored by GRAT with continuous suction of the suspension, a peristaltic pump was used. Grade fraction values smaller than the Stokes diameter were calculated with the following equation:

$$Y = \frac{\varepsilon_S}{\varepsilon_{So}} = \frac{\ln\left(\frac{R_o}{R}\right)}{\ln\left(\frac{R_o}{R_c}\right)} \tag{13}$$

where R_c is the countdown rate obtained for the initial concentration of the test and R_o, in this case, is the countdown rate obtained for the test tube containing pure water.

LADEQ Test Tube

The standard experiments with the LADEQ test tube were performed with suspensions of barium

sulfate and niobium oxide at 1% volumetric concentrations. The tests were in accordance with the methodology proposed by Silva and Medronho (1986).

Model for Particle Size Distribution

Aiming at adjusting the experimental data, the model of Rosin-Rammler-Bennet (RRB), given by the following equation was used:

$$Y = 1 - \exp\left[-\left(\frac{d}{d'}\right)^{n}\right] \qquad (14)$$

where d is the diameter of the particle and d' and n are the parameters of the model.

RESULTS

The parameters of Equation (14) were found from the data obtained in experiments related to the four types of tests conducted (conventional GRAT, GRAT with flow, conventional LADEQ and LADEQ with flow using a peristaltic pump). For each method three replicates were carried out in order to conduct a statistical analysis by means of hypothesis tests. Data from the particle-size analysis was ajusted with the Rosin-Rammler-Bennet model.

The results obtained in the particle-size analysis of the barium sulfate monitored by the four types of experiments, conventional GRAT, GRAT with flow, conventional LADEQ and LADEQ with flow using a peristaltic pump, are presented in Figure 2.

By observing Figure 2, it can be verified that the experiments monitored by GRAT showed results similar to those obtained with the standard experiments in the LADEQ test tube. In the experiment with suspension flow, the results were slightly affected by problems due to the constant shaking of the test tube, which probably produced small turbulences. Even so, the experiments monitored by GRAT with flow were acceptable as they allowed the size distribution of solids to be obtained in a few minutes, unlike what was obtained with GRAT in the case without flow and in the LADEQ test tube, where the results were only available in several hours or days, respectively.

The results obtained in the particle-size analysis of the niobium oxide monitored by the four types of experiments, conventional GRAT, GRAT with flow, conventional LADEQ and LADEQ with flow using a peristaltic pump, are presented in Figure 3. By observing this figure, it can be seen that, as expected, the experiment monitored by GRAT also had the same behavior as that obtained with the standard experiments in the LADEQ test tube.

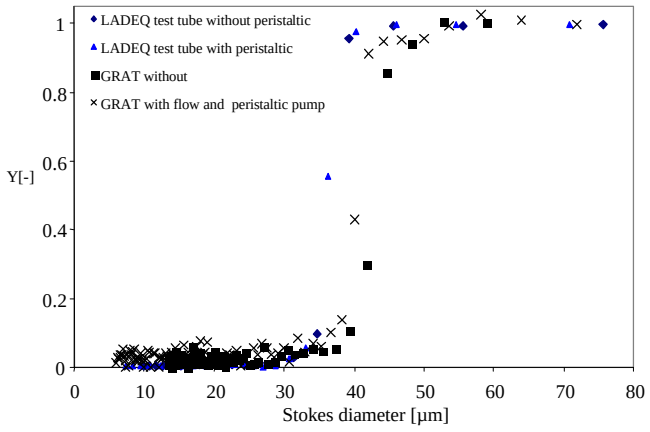

Figure 2: Results obtained with particle-size analysis of the barium sulfate.

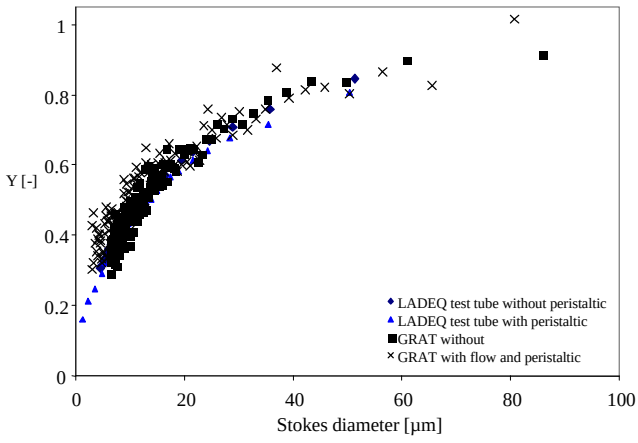

Figure 3: Results obtained with particle-size analysis of the niobium oxide.

The results for the d' and n parameters of the RRB model in the four types of experiments were analyzed to verify that there were significant differences between them. The results of the statistical comparison between more than two treatments for parameters d' and n, respectively are presented in Tables 3 and 4. In these tables it can be seen that the significance level (p level) obtained in the statistical test was less than 5% in all cases (p level<0.05). From these results it follows that there are significant differences between the mediums of the different methods applied.

The results for the hypothesis test applied in comparing the methods of particle size analysis, two

by two, are presented in Table 5. In this statistical test, if the significance level is less than 5% (p level<0.05), then there is a significant difference between the parameters obtained by the two techniques. If it is greater than 5% (p level>0.05), then the parameters are equal.

For the barium sulfate tests, the use of a dispersing agent and an ultrasonic bath during the tests could have broken the particles, thereby modifying the original particle size. So, for this material, we did not use the dispersing agent and ultrasonic bath. The possible agglomeration of particles therefore may have influenced the results presented in Table 5 for the barium sulfate. For the niobium oxide results, important conclusions can be drawn. The peristaltic pump can be influencing the results of GRAT since it is a highly sensitive method. For the conventional GRAT and LADEQ technique, where a peristaltic pump was not used for suction of the suspension, parameter d' of the RRB model was the same.

Use of the peristaltic pump in GRAT and the LADEQ technique resulted in a significant difference between the values obtained for parameter d' for niobium oxide. The use of the peristaltic pump could be effecting GRAT since it is a highly sensitivity technique.

In the suction of the suspension there was no difference between the shapes of the particle size distribution curves for the LADEQ test tube with or without peristaltic pump, where there were no significant differences between the parameter. However, it could again be seen for GRAT that the peristaltic pump modified the shape of the curve, possibly because of the sensitivity of the method to disturbances in the suspension flow.

Table 3: Comparison between more than two treatments – Hypotheses test for d' parameter.

Material	Type of experiments	Parameter d'	Avarage	Deviation	p level
Barium sulfate	LADEQ without peristaltic pump	37.77 37.54 37.35	37.56	0.21	2.29E-09
	LADEQ with peristaltic pump	36.57 36.29 35.96	36.28	0.30	
	GRAT without flow	43.49 43.41 43.74	43.55	0.17	
	GRAT with peristaltic pump	40.87 40.55 40.25	40.56	0.31	
Nobium oxide	LADEQ without peristaltic pump	27.70 27.79 26.71	27.40	0.60	4.26E-07
	LADEQ with peristaltic pump	28.55 28.05 26.79	27.80	0.91	
	GRAT without flow	38.49 37.24 39.65	38.46	1.21	
	GRAT with peristaltic pump	27.06 26.20 27.17	26.81	0.53	

Table 4: Comparison between more than two treatments –Hypotheses test for n parameter.

Material	Type of experiments	Parameter n	Avarage	Deviation	p level
Barium sulfate	LADEQ without peristaltic pump	21.11 21.01 20.88	21.00	0.12	1.00E-06
	LADEQ with peristaltic pump	23.55 22.77 23.32	23.22	0.40	
	GRAT without flow	19.60 20.31 20.92	20.28	0.66	
	GRAT with peristaltic pump	16.81 17.10 16.36	16.76	0.37	
Nobium oxide	LADEQ without peristaltic pump	0.67 0.66 0.66	0.66	0.00	1.00E-07
	LADEQ with peristaltic pump	0.64 0.66 0.66	0.65	0.01	
	GRAT without flow	0.73 0.70 0.70	0.71	0.01	
	GRAT with peristaltic pump	0.50 0.49 0.48	0.49	0.01	

Table 5: Comparison of treatments, two by two - Hypotheses test.

Comparison	Barium sulfate		Niobium oxide	
	p level			
	d'	n	d'	n
LADEQ without and LADEQ with peristaltic pump	0.0038	0.5596	0.0008	0.1725
LADEQ without peristaltic pump and GRAT without flow	0.0000	0.0001	0.1366	0.0012
LADEQ without peristaltic pump and GRAT with peristaltic pump	0.0002	0.2709	0.0000	0.0000
LADEQ with peristaltic pump and GRAT without flow	0.0000	0.0003	0.0028	0.0018
LADEQ with peristaltic pump and GRAT with peristaltic pump	0.0001	0.1788	0.0000	0.0000
GRAT without flow and GRAT with peristaltic pump	0.0001	0.0001	0.0013	0.0000

CONCLUSIONS

From the work presented in this paper, we came to the following conclusions.

The requirement of using low concentrations of solids in the experiments for measurement of Stokes diameters, regardless of the source used (Am^{241}) to present low intensity and emission energy, limits their use in the case of substances having low mass attenuation coefficients. Therefore in order to carry out these experiments accurately, it is essential that the molecule of the solid material has at least one element with a reasonably high mass attenuation coefficient.

The use of GRAT for determination of suspension concentration should be preceded by a careful theoretical analysis of the compatibility of the radioactive source with the experimental system to be used. This type of analysis is fundamental to guaranteeing the precision of the experiments.

In the cases in which the use of GRAT, for a specific radioactive source accurate is not possible, the use of other sources with lower energy is advisable, whereas the mass attenuation coefficient usually increases with the reduction in electromagnetic radiation energy. For this reason, in several cases, a source of LASER, instead of X and gamma rays is used.

For a given gamma-ray source to be used, depending on the experimental system under study, we can adjust the thickness of the physical medium or the initial suspension concentration in order to decrease the margin of experimental error related to the detection system.

An ultrasonic bath or sodium hexametaphosphate cannot be used to separate barium sulfate for separation particles. The agglomeration of particles might have been one of the factors responsible for displacement of the curve for particle size distribution.

According to the hypotheses test, it could be observed that for niobium oxide both the LADEQ and GRAT, without a peristaltic pump, provided identical results for the parameter d' of the RRB model.

GRAT proved to be efficient mainly in terms of the experiments. However GRAT is much more sensitive to disturbances in the suction of suspension at the base and when the peristaltic pump is used.

NOMENCLATURE

f	characteristic factor of each element	(-)
g	acceleration of gravity	($m\ s^{-2}$)
h	difference between the heights of the level of the suspension and of the point of sample collection	(m)
R	countdown rate	(s^{-1})
t	time	(s)
t_c	countdown time	(s)
w	mass fraction	(-)
x	thickness of the physical medium	(m)
X	superficial density of the physical medium	($kg\ m^{-2}$)
Y	smaller fraction than d_{St}	(-)
σ	mass attenuation coefficient	($m^2\ kg^{-1}$)
μ	viscosity of the fluid	($kg\ m^{-1}s^{-1}$)
ρ	density	($kg\ m^{-3}$)
ε	volume concentration	(-)

Subscripts

i	element	(-)
o	initial or pure	(-)
s	Solid	(-)
s-susp	solids in suspension	(-)

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