The PERSEVERANCE of the SEDIGRAPH METHOD of PARTICLE SIZING

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The SediGraph method for particle sizing has been used in laboratories world wide since 1967. Although more 'modern' alternatives have been developed for determining particle size, the SediGraph method continues to satisfy best the needs of certain applications. The reasons for the continued viability of the SediGraph method is the subject of this article.

THE SEDIGRAPH METHOD

The SediGraph method for determining the mass distribution of particles as a function of size (particle size distribution) has been employed in a wide variety of applications since its embodiment into a commercial instrument in 1967. The instrument has undergone improvements in speed, sample handling capability, and data reduction and presentation features since its introduction. The fundamental analytical method, however, has remained the same: particle sizing by the measurement of settling velocity and mass fraction determination by relative absorption of low-energy X-ray.

Although constantly challenged by 'new' technology based on optical and ultrasonic techniques paired with complex data reduction algorithms, the SediGraph method remains the accepted method in many applications. It has been objectively compared to various other particle sizing techniques (1 – 18) and found to be a reliable and repeatable method producing results comparing favorably with 'reference' techniques. The National Institute of Standards and Technology (NIST) in a guide to particle size characterization (19), describes the SediGraph as being based on a robust technique, providing rapid analyses, being well suited for industrial environments, relatively inexpensive, not requiring highly skilled operators, and having the capability to be used over a broad size range with minimal changes.

Particle scientists and technologists currently entering the workplace have an ever-increasing selection of sizing methods from which to choose. The choice must be made with a thorough understanding of the model upon which the measurement technique is based. Since size is seldom measured directly, one needs to understand what characteristics of the particle the instrument is measuring and how this measurement relates to size (20). This article answers those fundamental questions for the SediGraph method. In doing so, it should help the reader understand why the technique is described as 'robust' and why the SediGraph method has persevered over the years.

THEORY OF OPERATION: SIZE DETERMINATION

The SediGraph method is based on two well-established and well-understood physical phenomena—gravitational sedimentation and low energy X-ray absorption.

Stokes' law describes the gravitational sedimentation of a particle as a function of particle diameter. Stokes' law states simply that the terminal settling velocity of a spherical particle in a fluid medium is a function of the diameter of the particle. That is,

$$v = (D/K)^2 \qquad (1)$$

The constant of proportionality, K, accounts for the density of the particle and physical properties of the liquid medium. Expanded, the variables composing K and their relation are shown below.

$$K = [(18\eta)/(\rho - \rho_o)g]^{1/2} \qquad (2)$$

where,

ρ = particle density,
ρ_o = liquid density,

η = liquid viscosity, and
g = gravitational acceleration.

Stokes' law applies rigorously provided that, as the particle settles and displaces the liquid, laminar flow around the particle is maintained. This condition is satisfied as long as

$$(Dv\rho_0/\eta) < 0.3. \qquad (3)$$

The left side of the above expression is known as the Reynolds number. Fluid flowing around the particle maintains a laminar flow pattern as long as the Reynolds number is less than 0.3. The flow pattern becomes turbulent if the relationship between the parameters results in the Reynolds number exceeding 0.3. The energy required to produce turbulence reduces the settling velocity of the particle and the relation between v and D is no longer exactly as described by Equation 1. From the analyst's point of view, the Reynolds number is easily maintained within the proper range simply by modifying the viscosity or density of the dispersion liquid. The SediGraph provides in the software a means of helping the user select the appropriate suspension liquid for the sample being analyzed.

If the velocity (v) of fall is known, then the distance traveled (s) at this velocity is readily calculated from the well-known '*distance equals rate times time*' relationship, or

$$s = vt. \qquad (4)$$

Likewise, if we measure the time required for a particle to fall (settle) a known distance, the velocity is easily determined from Equation 4. With v and K in Equation 2 being known, Stokes law can be used to determine particle size D_t as a function of the time required to settle a known distance *s*.

A simple thought experiment to illustrate this is to consider the elementary case of a spherical particle of known density, but of unknown size being released at the surface of a liquid of known viscosity and having a density less than that of the particle. It accurately can be predicted when the particle will attain a certain level (depth) in the liquid. Likewise, if many particles of various sizes are introduced simultaneously to the surface of a liquid (as illustrated in Figure 1 for a two-particle system), it can be predicted accurately when all particles larger than any given size have fallen below a particular depth.

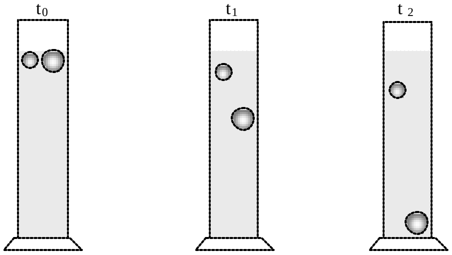

Figure 1. Solid particles of density ρ placed in a fluid medium of density less than ρ will settle with a velocity proportional to their diameter.

Imagine, now, that a homogeneous suspension of particles is allowed to settle as illustrated in Figure 2). In this situation, also, it accurately can be determined the size range of particles that have settled below a specific depth in the container after an elapsed time. It is by this method that the SediGraph determines size information. By recording the elapsed time since the sedimentation began

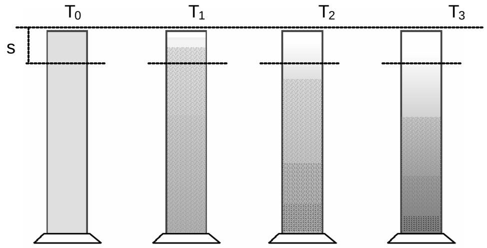

Figure 2. At t_0, the cylinder contains a homogeneous suspension of liquid and particles. At t_1, all particles of size d_1 and larger have settled below level *s* as calculated by Equation 4. Sedimentation continues until t_n at which time all particles of sizes larger than d_n have settled below level *s*. If an X-ray beam is projected through the cell at level *s*, the transmitted energy will increase proportional to the decrease in particle mass in the beam path.

and the depth of the measurement zone within the sample chamber, the size range of particles above and below any vertical position in the sample cell can be determined. This is discussed in more detail below.

THEORY OF OPERATION: MASS FRACTION MEASUREMENT

The SediGraph uses a beam of X-ray collimated into a thin horizontal band to measure directly particle mass concentration in the liquid medium. This is done by first measuring the intensity, I_{max}, of a baseline or reference transmitted X-ray beam that has been projected through the liquid medium prior to the introduction of the sample (see Figure 3). As liquid circulation continues, solid sample is added to the liquid reservoir and mixed until a homogeneously dispersed suspension of solid sample and dispersion liquid is being pumped through the cell.

More X-ray is absorbed by the solid than the liquid, therefore the transmitted X-ray beam is attenuated. Since the mixture of the flowing suspension is homogeneous, the intensity assumes a constant value, I_{min}, for X-ray transmission at full scale attenuation.

Flow of the mixture is stopped and the homogeneous dispersion begins to settle as transmitted X-ray intensity is monitored at a depth of s in Figure 3. During the sedimentation process, the largest particles are first to settle below the measuring zone and, finally, all particles settle below this level leaving only clear liquid. As more and more of the larger particles settle below the measurement zone and are not replaced by the same size particles settling from above, attenuation of the X-ray beam diminishes.

Therefore, during the sedimentation process, the transmitted X-ray intensity increases from I_{min} to I_{max}. Transmitted intensities between the two extremes will be symbolized by I_t and can be expressed simply as

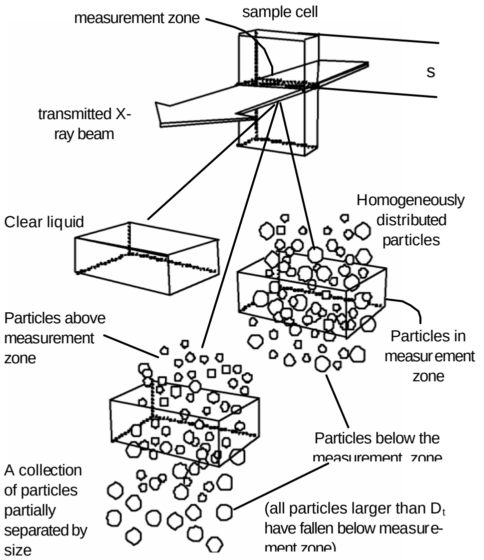

Figure 3. Upper: Diagram of SediGraph sample cell penetrated by a horizontally collimated X-ray beam. The volume of the sample cell through which the X-ray beam passes defines the measurement zone. **Lower:** Measurement zone shown at three stages of an analysis. First, only clear suspension liquid circulates through cell. Next, sufficient sample is introduced to produce a suitable attenuation of X-ray. Finally, circulation is stopped and the suspension is allowed to settle, thus separating the particles by size. Sedimentation continues until all particles have settled below the measurement zone.

$$I_t = I_{max} \times 10^{-kM_t} \qquad (5)$$

where M_t is the mass fraction of particles in the measuring zone at time t, and k incorporates all other variables that affect X-ray absorption. Note in Equation 5, when the mass fraction of particles in the measuring zone equals zero, the exponent equals zero. The 0^{th} power of any number is 1, so, when there are no particles in the measuring zone, I equals I_{max}, as would be expected. At the other extreme (when a homogeneous distribution of particles is in the measurement zone), sample concentration is increased until the attenuation of the transmitted X-ray (I_{min}/I_{max}) provides sufficient span to achieve good measurement resolution over the zero to one-hundred mass percent range.

The numerical value of k in Equation 5 can be determined as follows after measurements of I_{min} and I_{max}.

$$I_{min} = I_{max} \times 10^{-k}$$

$$I_{min}/I_{max} = 10^{-k}$$

$$k = -\log(I_{min}/I_{max}). \quad (6)$$

Of course, these measurement processes are automated in the SediGraph, I_{max} being measured while clear liquid is circulating through the sample cell, and I_{min} after the sample has been introduced and a homogeneous dispersion is being circulated. All other values of I are measured during the sedimentation process after flow has ceased.

APPLICATIONS IN TECHNOLOGY

There is an optimum particle size, or at least a smallest and largest acceptable size, for most things involving particles. Thus, industrial production and processing operations usually require knowledge of the particle size of component materials used in product manufacturing, or of the finished product itself in cases where the finished product is in particulate form. This knowledge is necessary because the particle size of powdered materials and the distribution of sizes can have a profound effect on the strength, density, porosity, and thermal properties of finished goods, and affects the flowability, dissolution rate, optical properties, and interstitial void volume of powdered materials.

An example in particle technology is controlling the surface-to-mass ratio by manipulating particle size. This, in turn, allows control over characteristics of the finished goods, such as the saturation and brilliance of paints, both a function of particle size. Likewise, the setting time of materials such as concrete and dental fillings proceeds in accordance with particle size.

Powders used in manufacturing processes often are mixed into a liquid to form a suspension or slurry as an intermediate process or as the final product. Particle size is an important parameter in predicting the behavior of such a mixture. Since sizing particles by the SediGraph technique also involves dispersing powders in a liquid, the environment in which the particle is analyzed is closely related to that of the application.

APPLICATIONS IN THE EARTH SCIENCES

The benefit of the SediGraph compared to other sizing methods is exemplified in the study of marine silts and sediments. Since the transport of solid components and the subsequent deposition are dependent upon sedimentation rate in water, the SediGraph technique is an ideal method since it, too, is based on sedimentation rate. Any application in which particle size is determined in order to understand settling mechanisms, the SediGraph is the natural choice as the analytical tool.

By techniques such as light scattering, electrozone sensing, and sieving, settling velocities are calculated from particle size and Stokes' law. The SediGraph directly measures settling velocity and from this, calculates particle size. To support applications in which settling velocity is the sought-after value and to eliminate the need to back-calculate settling velocity, the newest model SediGraph reports the distribution of mass by settling velocity.

Investigations of other natural processes benefit from knowledge of particle size distribution, and may be unrelated to settling velocity. As examples, the size characteristics of geologic materials can provide clues to its origin and its weathering. The size of airborne particles affects weather, thus is of interest to climatologists. Historical climatologists study particle depositions in ice cores as evidence of weather patterns over thousands of years in the past.

Agricultural scientists study the grain size of near-surface soils because of its relation to qualities associated with agricultural production, while civil engineers study the grain size of subsurface materials to assess their load-bearing capabilities. Environmentalists use soil and subsoil particle size data to assess the percolation rate, diffusion, and retention characteristics of these granular material beds in order to better manage hazardous substance spills.

SUMMARY

Particle size is studied in many applications of science and technology. It is easy to understand why particle sizing equipment is available in such a variety of configurations since particle sizes can range from nanometers to millimeters and involve essentially any material, liquid or solid, suspended in a liquid or a gas. The SediGraph is only one of many choices (20) and, because of its time-proven capabilities, continues to hold its place as the most relied-upon method of particle sizing in certain applications.

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