PARTICLE SIZE ADJUSTMENT TO INCREASE HOMOGENEITY OF A CHEMICAL GRADE SILICON METAL PREPARED AS A CANDIDATE OF A REFERENCE MATERIAL

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Abstract. *The width of the particle size distribution of particulate materials represents an important factor for the control of the variability of the results obtained in chemical analyses. This work aims at studying the elutriation technique as an adjustment method of the particle size of the material prepared as a candidate of a matrix reference material. The efficiency of the particle size adjustment method was undertaken by means of laser diffraction technique. Using the elutriation process, it could be noticed that the particles were classified in a more efficient way than the traditional dry sieving procedure.*
Keywords: *particle size adjustment; elutriation; powder silicon metal, matrix reference material*

1. INTRODUCTION

Homogeneity is a fundamental characteristics of reference materials once each sample of the material should be representative of the lot prepared for its intended purposes.

From the studies of Ingamells[(1)], it is known that the homogeneity of a particulate material is a function of the particle size of the samples used in the analysis. It is also known that the smaller the number of particles present in the material samples, the smaller will be its homogeneity, due to the probabilistic characteristic of the sampling of solids[(2)]. On the other hand, if extremely small particles are present in the samples, the homogenization processes become more complex and difficult to be implemented due to several factors, such as the formation of agglomerates and effects of adherence of small particles on larger particles, among others. It is also a known fact that, in many cases, the elements to be analyzed are distributed at different proportions, according to the different particle size ranges[(3)]. This is an additional reason why the reference materials producers always look to identify the proper particle size ranges for their products, eliminating the largest and the smallest particles, according to the specific needs. As a general rule reducing the particle size range implies in an improvement of the particulate material homogeneity.

The objective of this work is to study the elutriation technique to reduce the width of the particle size distribution of the powdered silicon metal material, in order to improve the quality of the lot prepared as a candidate of a matrix reference material.

1.1 Elutriation Technique

The experimental elutriation technique is a particle separation system that allows controlling precisely the velocity of the ascending fluid, so that performing the separations of fractions with quite close particle sizes.

Consider a solid particle allowed settling freely in an incompressible fluid. Be **E** the buoyant force that the fluid exerts on the particle, **P** the weight of the sphere and, **A** the drag force that is being formed as occurs the movement of the sphere. The equation of the particle movement is:

$$P - E - A = m_P \cdot \frac{dv}{dt} \qquad (1)$$

The weight of the particle and the buoyant force on the particle are given by:

$$P = m_P \cdot g = r_P \cdot V_P \cdot g = r_P \cdot \frac{p \cdot f_P^3}{6} \cdot g \qquad (2)$$

$$E = r_L \cdot V_P \cdot g = r_L \cdot \frac{p \cdot f_P^3}{6} \cdot g \qquad (3)$$

According to Streeter[(4)] and Bennett[(5)], the expression for the drag force on a spherical particle, in free settling under the condition of a laminar flow against an incompressible fluid, is represented by Equation (4), where R_p = radius of the spherical particle, m_L= liquid viscosity, n = particle velocity

$$A = 6p \cdot R_P \cdot m_L \cdot v = 3p \cdot f_P \cdot m_L \cdot v \qquad (4)$$

Substituting equations (2), (3) and (4) in (1), and performing some algebraic operations, we obtain the expression (5) known as Stokes Law, that supplies a mathematical model for the sedimentation of spherical particles at the laminar flow conditions (r_L= liquid density; r_P = particle density; f_P = diameter of the spherical particle)

$$v = \frac{f_P^2 \cdot g \cdot (r_P - r_L)}{18 \cdot m_L} \qquad (5)$$

Most authors[(5,6)] limit the use of this equation for Re_P <1, but some[(7)] advise its use for Re_P <0.3 and some even for Re_P <0.1 (Reynolds number of the particle $Re_p = f_p \,.\, r_L . v \;/\; m_L$).

When the particles are not spherical, it is necessary to use correction factors related with the sphericity of the particles[(8-11)], in order to adapt the movement law. According to Streeter[(4)], the drag force can be represented in a generalized way by equation (6) where A_{rp} = particle area of the particle projected in the plane normal to the flow.

$$A = \frac{C \cdot A_{TP} \cdot r_L \cdot v^2}{2} \qquad (6)$$

The drag coefficient C is a function of the particle shape and of the Reynolds number of the sedimentation process. We can use a shape factor for the particle called sphericity (ψ). The sphericity is defined as the relationship between the surface area of the sphere with the same volume of the particle and the particle surface area.

$$y = \frac{A_{ee}}{A_P}, \quad para \quad V_{ee} = V_P \qquad (7)$$

In order to obtain the drag coefficients, we can use empiric equations[(9-11)], or even experimentally obtained graphs, as the one developed by Comings[(11)]. From the knowledge of the geometric shape of the particles, it is possible to determine its sphericity and obtain the drag coefficient more appropriate. The non spherical particle velocity is given by equation (8):

$$v = \sqrt{\frac{p}{3} \cdot \frac{g \cdot f_P^3}{C \cdot A_{TP}} \cdot \left(\frac{r_P - r_L}{r_L} \right)} \tag{8}$$

2. METHODOLOGY

Approximately 60 kg of bulk silicon metal, containing 98.5% Si, were supplied by LIASA – Ligas de Alumínio S.A., located at the Industrial District of Pirapora in the Brazilian State of Minas Gerais. The material was ground and dry sieved with sieving openings of 63 and 75 μm. From such operation approximately 32 kg of material were obtained, which was homogenized by the method of manual riffling using a plastic film.

The elutriation technique was studied using the particle size separation unit constructed for this purpose (Figure 1).

Figure 1. Aspect of the particle size separation unit

The system was designed to perform the particle size separation of several particle types and it is quite flexible regarding the possibilities of adjusting the operating flow rate, types of drag fluids and other operational characteristics. The particle size adjustment efficiency is made by comparing the results obtained by dry sieving. By means of the Laser Diffraction method using an equipment from Malvern Instruments, the results of the size distribution of the particles were compared. For the sample dispersion a mixture of water and detergent was used.

3. RESULTS AND DISCUSSION

A sample of silicon metal was taken from the lot in study by sieving between 63 and 75 μm sieves and it was analyzed to know its particle size (Figure 2). The density of the sieved material, $\rho = 2.34\ g/cm^3$, was obtained by the method of helium picnometry. The value obtained was identical to the values found in Perry's Chemical Engineer's Handbook[(7)], for pure crystals of metallic silicon.

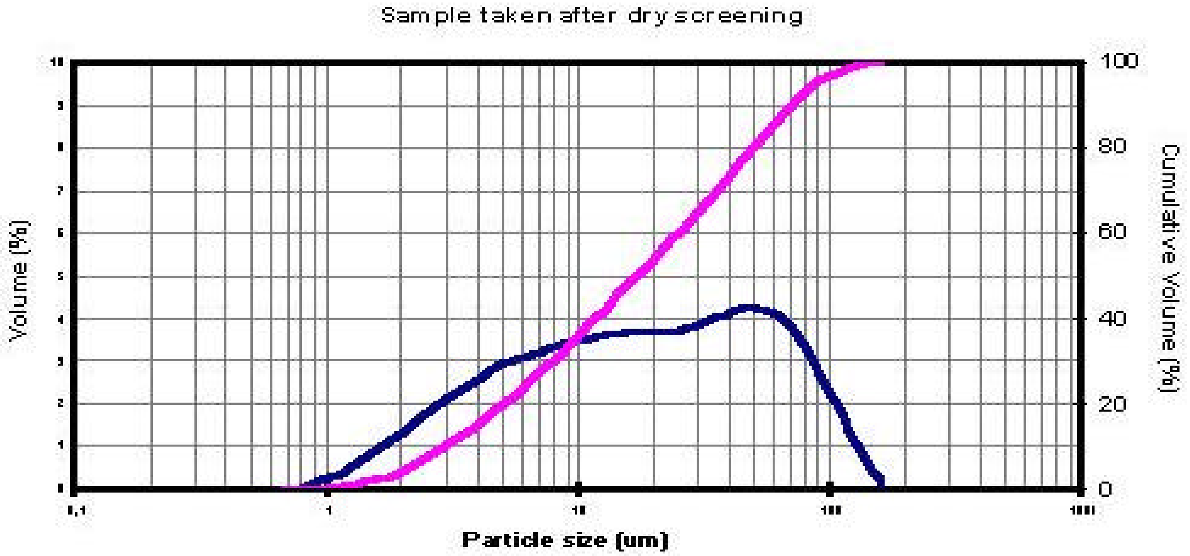

Figure 2– Size distribution of the metallic Si particles in the sample obtained by dry sieving

It is possible to observe that the material does not present particles that characterize a monomodal distribution. The fraction obtained by dry sieving contains a significant amount of particles below 10 µm, a size that seems to be the lower limit of a particle distribution with the maximum point placed at about of 50 µm. This is explained by the tendency of the smaller particles to be agglomerated around others of larger diameter. The small agglomerated particles are blocked by the sieve with smaller opening remaining in the oversize material. Observing with the optical microscope, with magnifications of 100x, the particles of silicon metal presented a shape predominantly symmetrical in the three dimensions, of voluminous character and they did not present a plate-like or needle-like characteristics, not being considerably different from spheres.

To test the elutriation system, two samples of approximately 1000 g, separated by dry sieving in the particle size range from 63 to 75 µm, were taken from the studied lot of the silicon material. Upon the performance of a preliminar test done in the elutriator, it was decided to use the sphericity value equal to 1,0. Consulting the graph[11] relating the drag coefficient and the sphericity, it was possible to obtain parameters to determine the expression that relates the Reynolds number with the drag coefficient and to apply these parameters in the equation (8) of the terminal velocity of the particle. The drag coefficient for the sphericity equal to 1,0 can be represented by equation (9):

$$C = 34{,}892.\ [(\varnothing_P.\ \psi_L.v)/\mu_L\ ]^{-0{,}777} \qquad (9)$$

Using equations (8) and (9), one can calculate the terminal velocity of the particles by means of equation (10).

$$v_t = \{0{,}038213.g.\varnothing_P.[(\rho_P - \rho_L)/\rho_L].[(\varnothing_P.\ \psi_L)/\mu_L\ ]^{0{,}777}\}^{0{,}81766} \qquad (10)$$

With the equation (10) it is possible to determine the terminal velocity of the particles of the indicated interval. With those calculated values and taking into account the settling time of the particles, the graph of Figure 3 was obtained. It allows determining the volumetric flow rates (12.5 L/min and 17.0 L/min) to be used to obtain the desired particle diameter range. The application times were defined as 20 minutes, to guarantee the separation of the borderline particles.

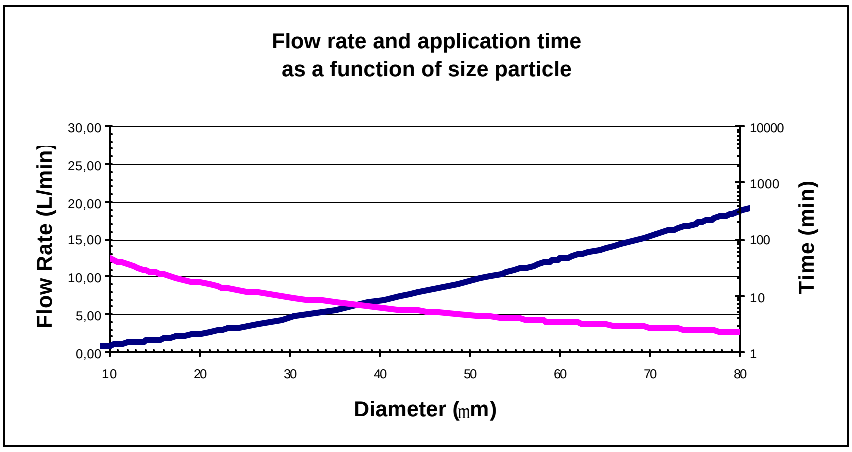

Figure 3: Graph of volume flow rate and application time, as a function of the particle diameter for ψ=1.0

After dried the sample was weighed resulting in 64.6 g of silicon particles. An aliquot of approximately 1 g was sent for particle size analysis (Figure 4).

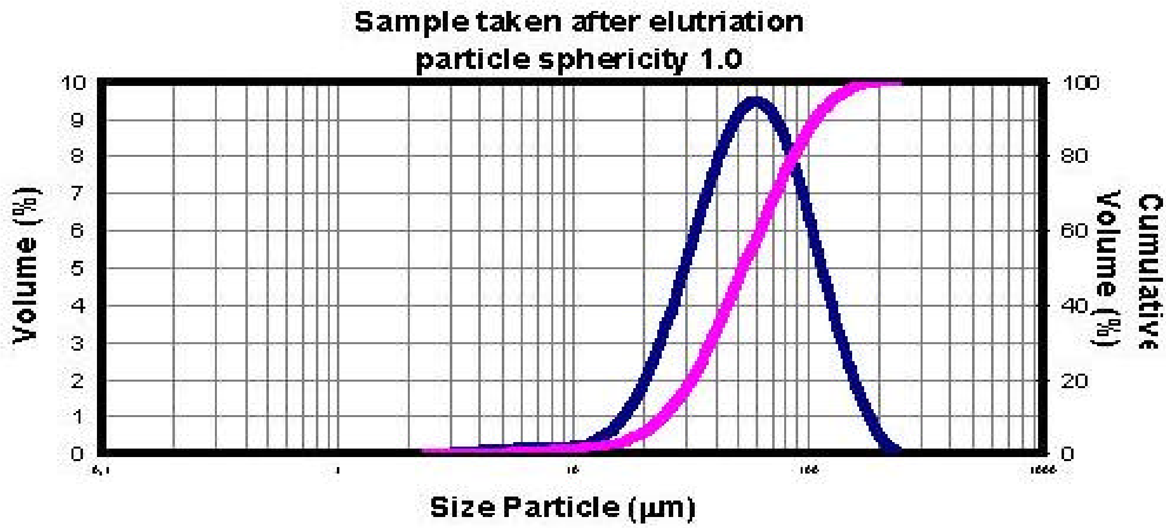

Figure 4 – Size distribution of the particles from the sample obtained by elutriation, assuming sphericity 1.0

It was observed that a more precise and symmetrical particle size classification was obtained with the elutriation when compared to the dry sieving process (Figure 2), once the elutriation process is not susceptible to the agglomeration that occurs in the sieving process and it is relatively rapid, as compared with the sieving. In a period of 40 minutes it was possible to separate the desired size fraction, while with the dry sieving it took several hours to be properly performed. Enhancing the operational method by means of fine adjustments in the cut flow rates and increasing the application times of application of the flows, thus enlarging the total separation time, it is possible to improve the elutriation process, in a way to reduce still more the presence of particles smaller than the lower size limit and particles bigger than the upper limit. Once the process is optimized for a type of material, it is possible to repeat it to obtain a larger amount of the material in consecutive lots. The developed system has the capacity to handle from five to six kilograms of solids per time and it is then possible to classify relatively big of amounts of material, taking into account that the lots of reference materials are rarely larger than 60 kg.

A characteristic point regarding elutriation is the need of a series of relatively difficult calculations and adjustments, in order to be properly implemented, that does not occur with sieving process. Aiming at solving this problem, a software was employed to support the calculations.

4. CONCLUSION

In the development of reference materials, the dry sieving is normally used to obtain the desired lot. This method is quite troublesome, requiring time and skilled manpower. Clean working conditions are also necessary, because the material to be obtained cannot be under the risk of contamination. The yield using this method is small, of approximately 30%, considering the initial amount of as received material to be preliminarily ground.

The development of a particle size classification system by elutriation was shown quite promising as an alternative to obtain more homogeneous particulate material. This method is able to handle about five kilograms of particulate material each batch. The system was shown flexible to operate with particles of different densities and shapes. This developed system can be used to establish particle size fractions for the reference material, with the goal of minimizing the experimental variability of the chemical properties.

By means of the development of the particle separation system based on the elutriation method, it can be foreseen the production of a lot within a previously specified size range displaying a more homogeneous particle size distribution.

The results insofar obtained were quite positive. The process is still in study, and the operation conditions and the software to establish the initial parameters are being improved, based on experience gathered in the experimental work.

5. ACKNOWLEDGMENTS

We thank Fundação de Amparo à Pesquisa do Estado de São Paulo – FAPESP for the support received for the development of the particle separation elutriation system presented in this work.

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