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Characterization of Crystal Size Distribution*Angelo Chianese***1.1****Introduction**

Crystalline population coming out from a crystallizer is characterized by its size distribution, which can be expressed in different ways. The crystal size distribution (“CSD”) may, in fact, be referred to the number of crystals, the volume or the mass of crystals with reference to a specific size range, or the cumulative values of number, volume or mass of crystals up to a fixed crystal size. The first approach refers to a density distribution, whereas the second one to a cumulative size distribution.

However, it is also useful to represent the CSD by means of a lumped parameter as an average size, the coefficient of variation, or other statistical parameters which may be adopted for the evaluation of a given commercial product.

In this section the more usual ways to represent both the whole CSD and the lumped CSD parameters are presented.

1.2**Particle Size Distribution**

The particle size distribution may be referred to the density distribution or cumulative distribution. Each distribution may be expressed in number, volume, or mass of crystals.

The cumulative variable, $F(L)$, expresses number, volume, or mass of crystals per unit slurry volume between zero size and the size L , whereas the density distribution function, $f(L)$, refers to number, mass, or volume of crystals per unit slurry volume in a size range, whose average size is L .

The relationship between the cumulative size variable and the density distribution size one is as follows:

$$F(L) = \int_0^L f(L) dL \quad (1.1)$$

Table 1.1 Cumulative and density variables.

Cumulative distribution			Density distribution		
Name	Symbol	Unit	Name	Symbol	Unit
Number	$N(L)$	$\#/m^3$ slurry	Population density	$n(L)$	$\#/m^3$ slurry m
Volume	$V(L)$	m^3 crystal/ m^3 slurry	Volume density	$v(L)$	m^3 slurry/ m^3 slurry m
Mass	$M(L)$	kg crystal/ m^3 slurry	Mass density	$m(L)$	kg crystal/ m^3 slurry m

or in the reverse form:

$$f(L) = \frac{dF(L)}{dL} \quad (1.2)$$

In Table 1.1 the expression of cumulative and density function variables referred to the number, volume, or mass of crystals is reported.

Examples of number and volume distributions are reported in Figure 1.1.

In spite of the three geometric dimensions of crystals, the CSD is usually referred to just one dimension, the so-called characteristic one, which is related

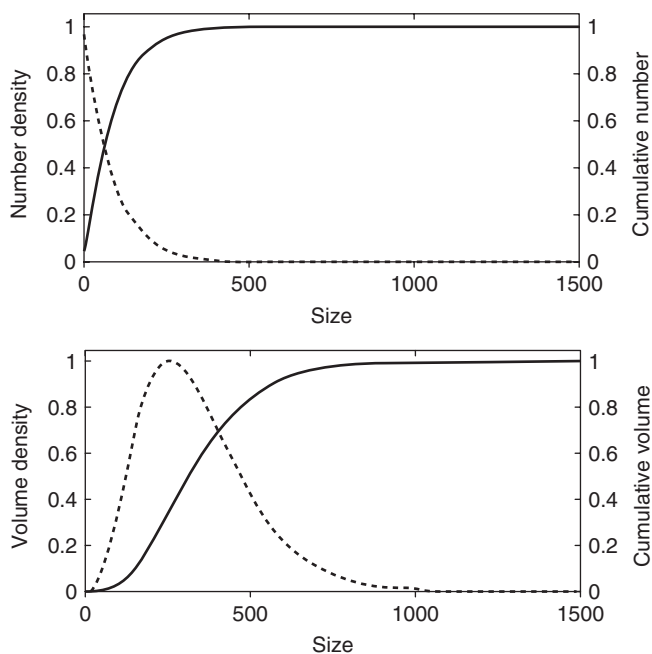


Figure 1.1 Cumulative and volume crystal size distributions (continuous line for the cumulative distribution and dotted lines for the density distribution).

to the adopted measurement technique. In the case of crystal size measurement by sieving the characteristic dimension is the second one, corresponding to the wire mesh length. Otherwise, if a laser diffraction-based analyzer is used, the characteristic dimension is the length given by the instrument, falling between the first and the second crystal dimension.

The most used density distribution variable is the crystal population density, $n(L)$. It can be used to estimate the total number, N_T , the total surface A_T , and total mass, M_T , of crystals by means of the following expressions:

$$N_T = \int_0^{\infty} n(L) dL \quad (1.3)$$

$$A_T = \int_0^L k_s(L) L^2 n(L) dL \quad (1.4)$$

$$M_T = \rho \int_0^L k_v(L) L^3 n(L) dL \quad (1.5)$$

where ρ is the crystal mass density and $k_s(L)$ and $k_v(L)$ are the surface shape factor and the volume shape factor of the crystals of size L , respectively, derived from the relationships

$$A(L) = k_s(L) L^2 \quad \text{and} \quad V(L) = k_v(L) L^3 \quad (1.6)$$

$A(L)$ and $V(L)$ are the surface and the volume of a single crystal, respectively. When the CSD is measured by sieving, the values of $k_v(L)$ can be easily determined, as follows:

$$k_v(L) = \frac{M_C(L)}{\rho L^3} \quad (1.7)$$

where $M_C(L)$ is the average mass of a single crystal retained over a sieve and L is the mean value between the dimension of two subsequent sieves, the considered and the upper ones. The surface shape factor is more difficult to calculate, since the average surface of the crystals has to be evaluated. The values of the shape factors depend on the crystal habit, which may qualitatively be defined as spherical, cubic, granular, needles, and so on. For a spherical particle the characteristic dimension is the diameter, the volume shape factor is equal to $\pi/6$, and the surface shape factor is π . Some examples of shape factors are reported in Table 1.2 (Mersmann, 2001).

In the case of size measurement by sieving, it is possible to convert the sieving data to crystal population density by means of the equation

$$n(L) = \frac{M(L)}{k_v \rho L^3 \Delta L} \quad (1.8)$$

where is $M(L)$ the overall crystal mass between two subsequent sieves, L and ΔL are the mean size and the size difference of the two subsequent sieves opening, respectively.

Table 1.2 Examples of shape factors.

Geometric shape	k_v	k_s
Sphere	0.524	3.142
Tetrahedron	0.118	1.732
Octahedron	0.471	3.464
Hexagonal prism	0.867	5.384
Cube	1.000	6.000
Needle $5 \times 1 \times 1$	0.040	0.880
Plate $10 \times 10 \times 1$	0.010	2.400

1.3

Particle Size Distribution Moments

The i th moment of a size distribution is defined as

$$m_i = \int_0^\infty L^i n(L) dL \quad (1.9)$$

The moments are useful to calculate the mean size, as follows:

$$\bar{L}_{i+1,i} = m_{i+1}/m_i \quad (1.10)$$

According to the value of i between 0 and 3 used, the mean size may be referred to the number of crystals, their length, surface, and volume, respectively. The most popular expressions of mean size, derived from the moments ratio, are the number-based and the volume-based mean size (Randolph and Larson, 1988), that is,

$$\bar{L}_{1,0} = \int_0^\infty L n(L) dL / \int_0^\infty n(L) dL \quad (1.11)$$

$$\bar{L}_{4,3} = \int_0^\infty L^4 n(L) dL / \int_0^\infty L^3 n(L) dL \quad (1.12)$$

The number, the length, the surface area, and the volume of the particles are directly related to the moments of the distribution. The relevant expression, when the shape factor is constant with size, is reported in Table 1.3.

The coefficient of variation (CV) represents the spread of the size distribution around the mean size. It is the ratio between the distribution standard deviation and the mean size.

Often, the median size, L_{median} , instead of the mean size is used. It is the mean abscissa of a graph of cumulative volume/mass fraction versus size. Note that in general the median size is different from the mean size.

Table 1.3 Properties of distribution based on the moments.

Property	Definition	Dimension
Total number	$N_T = m_0$	$(\#/m^3)$
Total length	$L_T = m_1$	(m/m^3)
Total surface area	$A_T = k_s m_2$	(m^2/m^3)
Total volume	$V_T = k_v m_3$	(m^3/m^3)
Mean (number based)	$L_{1,0} = m_1/m_0$	(m)
Mean (volume based)	$L_{4,3} = m_4/m_3$	(m)
Coefficient of variation (volume based)	$CV = \sqrt{\frac{m_3 m_5}{m_4^2}} - 1$	–

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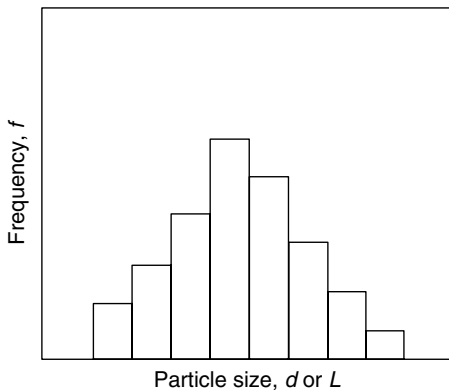
Particle Size Distribution Characterization on the Basis of Mass Distribution

The calculation of characteristic sizes of the CSD does not necessarily requires to pass through the calculations of values of the crystals population density. The most easy way is, in fact, to use directly the mass fraction of crystals measured by sieving. The mass fraction of crystals of an average size equal to L can be calculated as

$$x(L) = \frac{M(L)}{M_T} \quad (1.13)$$

Often the mass fraction is represented by means of the histogram reported in Figure 1.2, which is called the *frequency histogram*.

Sequentially adding each segment of the frequency diagram gives the cumulative distribution in terms of the mass fraction.

**Figure 1.2** Frequency histogram.

The size distribution may be characterized by two parameters: the mass mean size L_{wm} and the coefficient of variation, that is,

$$L_{wm} = \sum_i x_i(L)L \quad (1.14)$$

and

$$CV = \frac{1}{L_{wm}} \sqrt{\frac{\sum_i (L_i - L_{wm})^2}{N - 1}} \quad (1.15)$$

References

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| <p>Mersmann, A. (2001) <i>Crystallization Technology Handbook</i>, 2nd edn, Marcel Dekker, New York.</p> | <p>Randolph, A.D. and Larson, M.A. (1988) <i>Theory of Particulate Processes</i>, 2nd edn, Academic Press, San Diego, CA.</p> |
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