STEREOLOGICAL ANALYSIS OF PARTICLE CHARACTERISTICS

Ervin E. Underwood
School of Chemical Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332 USA

ABSTRACT

Important characteristics of particles are their size,
specific surface and shape. The stereological parameters
available to quantify these three attributes are discussed,
both for loose particles and particles contained within a
matrix. A shape parameter for particle roughness is deve-
loped, showing that a more complicated function expresses
the roughness characteristics better than a simple function.

INTRODUCTION

Traditionally, it appears that microscopical studies
of loose particles have tended to rely more on the projected
characteristics than on the sectioned quantities. Thus,
stereology, which leans heavily on both methods of observa-
tion, can be considered to bring another dimension to the
quantitative study of particle characteristics. Both two-
dimensional and three-dimensional quantities are considered
in discussing the choices available for size, specific surface
and shape parameters. These three topics are discussed in
turn.

SIZE

The 'size' of a particle, or of a system of particles,
can be expressed in many ways. Generally, we define some
geometrical parameter related to 'size', such as a linear,
areal or volume-based measure. Unfortunately, the 'diameter'
as a measure of size is meaningless except for circles and
spheres. For other figures or objects, the parameter
actually selected must be clearly defined. We would like

the size parameter that we select to be free of á priori
assumptions about shape, if at all possible. Moreover, it
should have a unique value that is reproducible, easily
measured, and related to the three-dimensional particle
quantities.

The most general and assumption-free choice of linear
size parameter is the mean intercept length, $\overline{L}_2$ (in the sec-
tion plane), or $\overline{L}_3$ (in three dimensions). The basic defining
equation is

$$\overline{L}_2 \equiv \overline{L}_3 = \Sigma L_i / \Sigma n_i \tag{1}$$

which states that the sum of many linear intercepts (ΣL_i)
taken along random test lines, divided by the total number
of intercepts (Σn_i), is the mean intercept length (either
two- or three-dimensional). This definition applies equally
to a single particle or a system of particles, and the
particles can have any shape (convex and/or concave).

Experimentally, we obtain the mean intercept length
from embedded particles by the relationship

$$\overline{L}_2 \equiv \overline{L}_3 = L_L / N_L = P_P / N_L \tag{2}$$

where L_L, the intercept length per unit length of test line,
is equal to P_P, the point ratio of particles hit to the total
number of grid points applied, and N_L is the number of inter-
cepts of particles per unit length of test lines. $\overline{L}_3$ is
related to other three-dimensional quantities, such as the
specific surface S/V, and is completely general and indepen-
dent of particle size, shape and complexity.

When the particles are convex in shape (or reasonably
equiaxed), additional linear size parameters are available.
For example, we can use $\overline{d}$ or $\overline{D}$, the mean tangent diameters
in the section plane or in space, respectively, or $\overline{H}'$, the
mean tangent height in the projection plane. Frequently,
the minimum or maximum values of linear size parameter are
desired. For convex bodies we have

$$D_{max} = (\overline{L}_3)_{max} = d_{max} \tag{3}$$

and for spheres of diameter D, we can write

$$D = d_{max}. \tag{4}$$

The maximum and minimum linear size parameters for particles
of known shape may be calculated. Note that the measured
values of intercept lengths vary between zero and some maximum
value, while the various measured tangent diameters vary be-
tween a non-zero minimum value and a maximum. Thus

$$\bar{L}_3 < \bar{D}. \tag{5}$$

Some of the interrelationships among these size parameters
are shown in Figure 1.
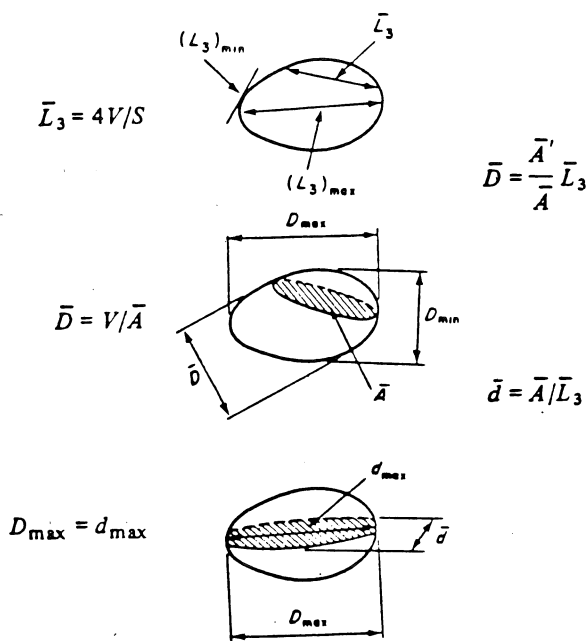
For spheres:

$$N_V = N_A/\bar{D} = N_A/d_{max} = \pi N_A/4\bar{d}$$

$$\bar{D} = \frac{3}{2}\bar{L}_3 = \frac{4}{\pi}\bar{d} = d_{max}$$

Fig. 1. Length parameters in convex bodies

In addition to linear measures of particle size, areal
and volume-based definitions of size are also possible. Areal
size definitions can employ A or A', the sectioned or pro-
jected area, or the mean or maximum values. In the two-
dimensional section plane, the mean intercept area is given by

$$\overline{A} = A_A/N_A = P_P/N_A, \tag{6}$$

and in the projection plane, the mean projected area is

$$\overline{A}' = A_A'/N_A' = P_P'/N_A'. \tag{7}$$

$\overline{A}'$ is of importance because it is related to sieve size. A spatial quantity that may be used for a size parameter is S, the surface area of the particle. For a convex particle we use the Cauchy expression

$$S = 4\overline{A}', \tag{8}$$

or a variant of Eq. 8 for a system of particles,

$$S_V = 4A_A'/t \tag{9}$$

where t is the foil thickness and A_A' is the area fraction in the projection plane. Also, the average surface area per particle, $\overline{S}$, is given simply by the defining equation

$$\overline{S} = S_V/N_V. \tag{10}$$

The definition of size in terms of volume has its staunch advocates. V, or $\overline{V}$, has the advantage of being a true three-dimensional quantity, independent of shape or orientation. For irregularly-shaped particles, the volume size parameter suffers the disadvantage of being difficult to determine directly, and requires either weakening assumptions or laborious experimental work. Moreover, it offers no information on particle shape.

In general, the mean volume is defined by

$$\overline{V} = V_V/N_V = P_P/N_V \tag{11}$$

where N_V, the number of particles per unit test volume, is difficult to obtain.

For convex particles, the three types of parameters discussed above are related by extremely general equations. Thus we have

$$\overline{d} = \overline{A}/L_3 = \overline{A}'/D = L_p/\pi \tag{12}$$

$$\overline{D} = V/\overline{A} = \overline{A}'\overline{L}_3/\overline{A} = \overline{H}' \tag{13}$$

$$\overline{A} = V/\overline{D} = \overline{L}_3\overline{L}_2' = \overline{A}'\overline{L}_3/\overline{H}' \tag{14}$$

$$\overline{A}' = V/\overline{L}_3 = \overline{H}'\overline{L}_2 = S/4 \tag{15}$$

The mean intercept length and volume relationships can also be obtained from these equations. If a specific particle shape is assumed, these general equations can be expressed explicitly in terms of a particle dimension or the other quantities. For example, for a sphere of diameter D, we have

$$D = 3\overline{L}_3/2 = 4\overline{d}/\pi \tag{16}$$

$$\overline{A} = 2\overline{A}'/3 \tag{17}$$

$$V = \pi r^2 \overline{L}_3, \tag{18}$$

etc. Moreover, only V, S and $\overline{D}$ need be known in order to calculate any of the above stereological quantities (2d or 3-d) for any convex body.

SPECIFIC SURFACE

This ubiquitous parameter appears to exert a decisive influence on the properties of many particulate systems. The three-dimensional specific surface is defined for a single particle by S/V, where the surface and volume terms refer to the particle itself. For a system of particles, we may need $\overline{S}/\overline{V}$. The specific surface is related to the mean intercept length by

$$\frac{S}{V} = \frac{4}{\overline{L}_3} \tag{19}$$

and also

$$\frac{S}{V} = \frac{S_V}{V_V} = \frac{\overline{S}}{\overline{V}} = \frac{4}{\overline{L}_3} = \frac{4N_L}{P_P} . \tag{20}$$

If loose particles are embedded and sectioned, a two dimensional analogue to S/V is obtained, i.e., $\overline{L}_p/\overline{A}$, which is the mean perimeter length divided by the mean sectioned area. These two ratios are related through

$$\frac{\overline{S}}{\overline{V}} = \frac{4}{\pi} \frac{\overline{L}_p}{\overline{A}}. \tag{21}$$

Figure 2 summarizes the equations for specific surface applicable to two important types of particulate systems. On the left is a system of particles enbedded randomly in a matrix. The other type of system, shown on the right, consists of loose, individual particles. Definitions for S and V are given for both types. It can be seen that the specific surface of sectioned particles in a matrix can be obtained by combined point and intersection counts, while for loose particles the relatively inaccessible total surface area and volume of particles are needed.

$$\bar{S} = \frac{S_V}{N_V} \qquad\qquad \bar{S} = \frac{\Sigma S_i}{N}$$

$$\bar{V} = \frac{V_V}{N_V} \qquad\qquad \bar{V} = \frac{\Sigma V_i}{N}$$

$$\frac{\bar{S}}{\bar{V}} = \frac{S_V}{V_V} = \frac{2P_L}{P_P} \qquad\qquad \frac{\bar{S}}{\bar{V}} = \frac{\Sigma S_i}{\Sigma V_i} = \frac{S_T}{V_T}$$
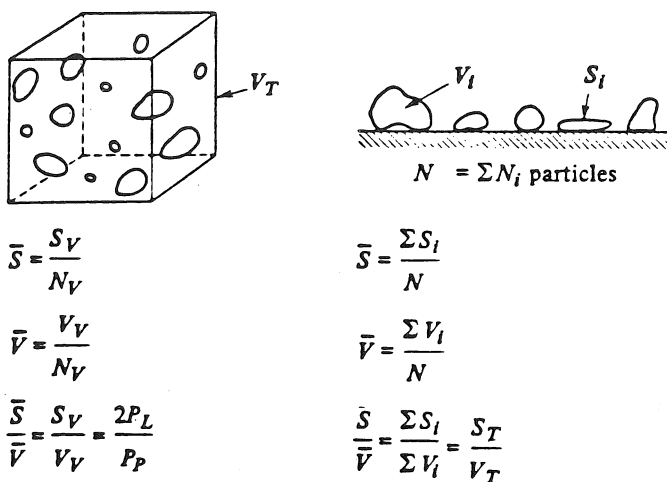
Fig. 2.   Determination of specific surface

Although the ratio S/V is determined readily enough through the basic Equations 19, 20 or 21, it is more difficult to obtain the values of S and V separately. As mentioned above, S may be determined through Equation 8, while V may possibly be calculated through Eqs. 11 or 13, although $\bar{D}$ or N_V are difficult three-dimensional quantities to obtain.

In these examples, note the difference between S_V and S/V. The first term is the standard stereological quantity ($\equiv \Sigma S_i/V_T$), equal to $2P_L$, and refers to the total surface area of the particles divided by the test volume (particles plus matrix). The second term is the specific surface, where the particle surface area is divided by the particle volume only. Accordingly, $\bar{S}/\bar{V}$ for embedded particles is equal to $2P_L/P_P$.

SHAPE

The idea of 'shape' is one of the most difficult con-
cepts to define, let alone quantify. Shape parameters are
generally used to express some particle attribute such as
roughness, roundness, acicularity, etc. Although shape
parameters are preferably dimensionless, nevertheless they
are composed largely of metric terms, e.g., length, surface
area, and/or volume. Curvature of lines and/or surfaces
and topological quantities, i.e., the <u>number</u> of corners,
edges, faces and/or bodies, may also find application in a
shape parameter.

Sometimes simple ratios or products of terms may be
combined to produce a satisfactory shape parameter. For
example, useful combinations are L/W, the length-to-width
ratio; L_p/L_2, the perimeter length divided by intercept
length; L_p^2/A, the perimeter length squared divided by the
area; or Ak_L^2, the area times the line curvature squared.
Frequently, however, more complicated parameters may be
needed to express the shape characteristics of interest.

Figure 3 defines the mean particle area $\overline{A}$ and the
equivalent circle area A_{eq}, as well as the equivalent dia-
meter d_{eq} and equivalent perimeter length $(L_p)_{eq}$. Many
shape parameters are based on ratios of the microstructural
quantity. For example, we might wish to use $(\overline{L}_p)_{meas}/(L_p)_{eq}$,
or $(\overline{L}_2)_{meas}/(L_2)_{eq}$, etc.

$$\overline{A} = \frac{A_A}{N_A}$$

mean area of   area of circle
particle sections   equals mean area

$$\boxed{A_{eq} = \overline{A}} \qquad \boxed{d_{eq} = 2\sqrt{\overline{A}/\pi}} \boxed{(L_p)_{eq} = 2\sqrt{\overline{A}\pi}}$$
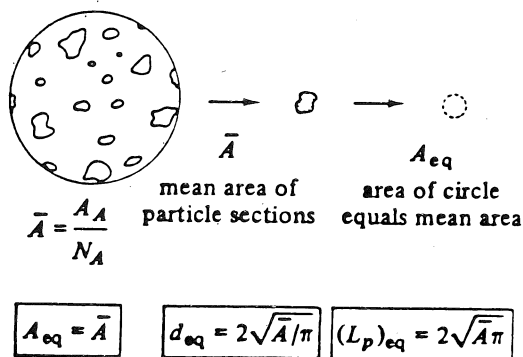
Fig. 3. Quantities calculated from equivalent area circle

Sometimes we must combine several stereological quantities in order to relate all the important shape characteristics to the particle properties or behavior. Two-dimensional quantities that are useful in shape studies are

$$\bar{A}, \ \bar{L}_2, \ \bar{L}_p, \ \bar{d} \ \text{and} \ \bar{k}_L$$

while three-dimensional quantities are

$$V, \ S, \ \bar{D}, \ \bar{L}_3, \ \text{and} \ \bar{K}_s,$$

where $\bar{k}_L$, and K_s are the mean curvature of lines in a plane and the average mean curvature of surfaces, respectively. Typical interconversions among these quantities are given in Table I. Thus, shape parameters can be expressed in terms of either two- or three-dimensional quantities.

TABLE I

BASIC RELATIONSHIPS FOR SHAPE PARAMETERS

Planar Quantities

$$\bar{A} = \frac{V}{\bar{D}} = \frac{A_A}{N_A}$$

$$\bar{L}_p = \frac{\pi S}{4\bar{D}} = \frac{L_A}{N_A}$$

$$\bar{d} = \frac{S}{4\bar{D}} = \frac{N_L}{N_A}$$

$$\bar{L}_2 = \frac{4V}{S} = \frac{L_L}{N_L}$$

$$\bar{k}_L = \frac{8\bar{D}}{S} = \frac{2N_A}{N_L}$$

Spatial Quantities

$$V = \bar{L}_2\bar{A}' = \frac{V_V}{N_V}$$

$$S = 4\bar{A}' = \frac{4N_L}{N_V}$$

$$\bar{D} = \frac{\bar{A}'}{\bar{d}} = \frac{N_A}{N_V}$$

$$\bar{L}_3 = \frac{\bar{A}}{\bar{d}} = \frac{L_L}{N_L}$$

$$\bar{K}_s = \frac{\pi}{2\bar{d}} = \frac{\pi N_A}{2N_L}$$

An example of this procedure is afforded by an analysis of a series of particle silhouettes shown in Figure 4. In order to express the particle 'roughness', we might try the ratio L_p/A. However, additional information is desirable, such as the number of projections (or 'hills') around the

perimeter. The latter is related to the number of inflec-
tion points (I) and thereby to the curvature. Additionally,
as the particles show more elongation, the aspect ratio (Q)
becomes important.

Thus the shape parameters progress from
$\dfrac{\Delta(L_p/A)}{I}$, where $\Delta(L_p/A) = (L_p/A)_{meas} - (L_p/A)_{eq}$ and I is
obtained readily by counting hills according to
$I = 2N_{hills}$; then to

$\dfrac{\Delta(L_p/A)}{(L_p/A)_{eq} \cdot I}$, which gives the relative change in L_p/A with
respect to the equivalent area circle; and
finally to

$\dfrac{\Delta(L_p/A) \cdot Q}{(L_p/A)_{eq} \cdot I}$, which introduces the aspect ratio Q.

Numerical results obtained with these three shape para-
meters are given in turn below each figure in Figure 4.
Only for the third shape parameter do the values proceed
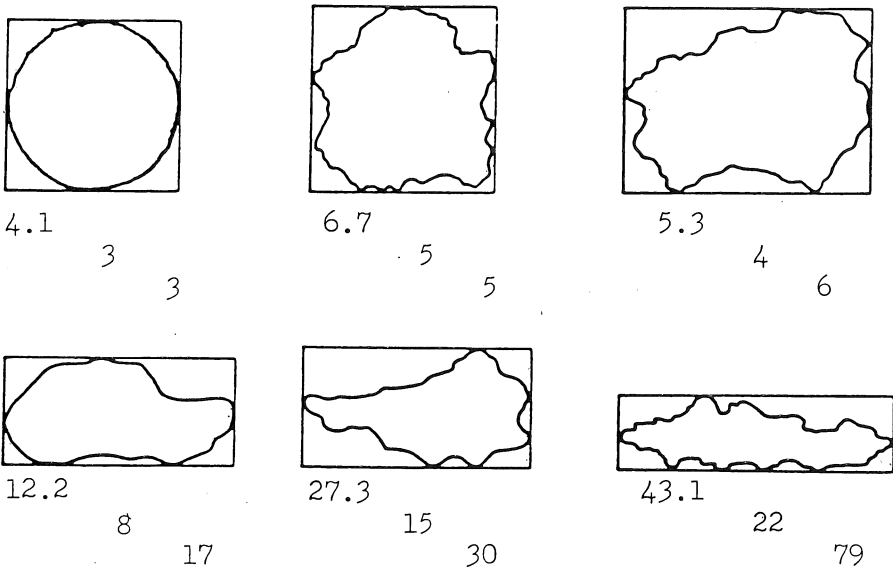systematically as the apparent roughness increases. Thus,

4.1	6.7	5.3
3	5	4
3	5	6

12.2	27.3	43.1
8	15	22
17	30	79

Fig. 4.   Variation of roughness parameters with
particle profile shape

it appears that this complexing procedure has merit. Without doubt, more refined analyses should lead to even better results.

CONCLUSIONS

Stereological quantities that express size, specific surface and shape are given for loose particles or loose particles mounted in a resin matrix. The mean intercept length, L_3, appears to represent 'size' best, while the specific surface, S/V, is given readily by a combined intersection and point count, $2P_L/P_P$. The 'shape' of particle silhouettes is expressed by a combined parameter that includes the (L_p/A) ratio; I, the number of inflection points; and Q, the aspect ratio.

ACKNOWLEDGMENT