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Particle size analysis — Image analysis methods —

Part 1:
Static image analysis methods

Analyse granulométrique — Méthodes par analyse d'images —

Partie 1: Méthodes par analyse d'images statiques

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Foreword

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The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

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ISO 13322-1 was prepared by Technical Committee ISO/TC 24, *Sieves, sieving and other sizing methods*, Subcommittee SC 4, *Sizing by methods other than sieving*.

ISO 13322 consists of the following parts, under the general title *Particle size analysis — Image analysis methods*:

— *Part 1: Static image analysis methods*

— *Part 2: Dynamic image analysis methods*

Introduction

The purpose of this part of ISO 13322 is to give guidance for a measurement description and its validation when determining particle size by image analysis.

Image analysis is a technique used in very different applications on image material with variations in material properties. Hence, it is not relevant to describe an exact standard method for determination of particle size by image analysis. The aim of this part of ISO 13322 is limited to give a standardized description of the technique used and a standardized validation.

This part of ISO 13322 includes methods of calibration verification using a certified standard graticule as a reference or by using certified standard particles. It is sensible to make some measurements on particles, or other reference objects, of known size so that the likely systematic uncertainties introduced by the equipment can be calculated.

This part of ISO 13322 gives a recommendation for a precise description of the distribution including the number of analyzed particles and an analysis window to make sure that the obtained information is valid.

Measurement of particle-size distributions by microscopy methods is apparently simple, but because only a small amount of sample is examined, considerable care has to be exercised in order to ensure that the analysis is representative of the bulk sample. This can be demonstrated by splitting the original sample and making measurements on three or more parts. Statistical analysis of the data, for example using the Student's t-test, will reveal whether the samples are truly representative of the whole.

Errors introduced at all stages of the analysis from sub-division of the sample to generation of the final result add to the total uncertainty of measurement and it is important to obtain estimates for the uncertainty arising from each stage. Indications where this is required are given at the appropriate point in the method.

Because of the diverse range of equipment and sample preparation expertise available, it is not intended to give a prescriptive procedure where use of individual methods does not jeopardize the validity of the data. However, essential operations are identified to ensure that measurements made conform to this part of ISO 13322 and are traceable.

Particle size analysis — Image analysis methods —

Part 1:
Static image analysis methods

1 Scope

This part of ISO 13322 is applicable to the analysis of images for the purpose of determining particle size distributions. The particles are appropriately dispersed and fixed on an optical or electron microscope sample stage such as glass slides, stubs, filters, etc. Image analysis can recover particle images directly from microscopes or from photomicrographs.

Even though automation of the analysis is possible, this technique is basically limited to narrow size distributions of less than an order of magnitude. A standard deviation of 1,6 of a log-normal distribution corresponds to a distribution of less than 10:1 in size. Such a narrow distribution requires that over 6 000 particles be measured in order to obtain a repeatable volume-mean diameter. If reliable values are required for percentiles, e.g. D_{90} or other percentiles, at least 61 000 particles must be measured. This is described in Annex A.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 9276-1, *Representation of results of particle size analysis — Part 1: Graphical representation*

ISO 9276-2, *Representation of results of particle size analysis — Part 2: Calculations of average particle sizes/diameters and moments from particle size distributions*

3 Terms, abbreviated terms, definitions and symbols

3.1 Terms, abbreviated terms and definitions

For the purposes of this document, the following definitions apply.

3.1.1
view field
field which is viewed by a viewing device, e.g. optical microscope or electron scanning microscope

3.1.2
measurement frame
field in a view field in which particles are counted for image analysis

NOTE The set of measurement frames composes the total measurement field.

3.1.3
binary image
digitized image consisting of an array of pixels, each of which has a value of 0 or 1, whose values are normally represented by dark and bright regions on the display screen or by the use of two distinct colours

3.1.4
edge finding
one of many edge detection methods used to detect transition between objects and background

3.1.5
Euler number
number of objects minus the number of holes inside the objects, which describes the connectedness of a region, not its shape

NOTE A connected region is one in which all pairs of points can be connected by a curve lying entirely in the region. If a complex two-dimensional object is considered to be a set of connected regions, where each one can have holes, the Euler number for such an object is defined as the number of connected regions minus the number of holes. The number of holes is one less than the connected regions in the set compliment of the object. It is important to report the Euler number together with the connectivity applied, i.e., 4-connectivity or 8-connectivity.

3.1.6
Feret diameter
distance between two parallel tangents on opposite sides of the image of a particle

3.1.7
equivalent circular diameter
ecd
diameter of a circle having the same area as the projected image of the particle

NOTE It is also known as the Haywood Diameter.

3.1.8
grey image
image in which multiple grey level values are permitted for each pixel

3.1.9
image analysis
processing and data reduction operation which yields a numerical or logical result from an image

3.1.10
numerical aperture
NA
product of the refractive index of the object space and the sine of the semi-aperture of the cone of rays entering the entrance pupil of the objective lens from the object point

3.1.11
pixel
picture element
individual sample in a digital image that has been formed by uniform sampling in both the horizontal and vertical directions

3.1.12
segmentation
⟨noun⟩ part into which something can be divided; subdivision or section

3.1.13
segmentation
⟨verb⟩ act of dividing something into segments

3.1.14
threshold
grey level value which is set to discriminate objects of interest from background

3.2 Symbols

δ	error
θ	half-angle subtended by the particle at the objective lens
λ	wavelength, expressed in micrometres
μ	refractive index of the surrounding medium
φ	shape factor
A_i	projected area of particle i
d	minimum feature length
H_{cal}	horizontal calibration factor
K	constant numerically determined by the confidence limit
N	number of particles to be measured
n_i	numbers of particles of size X_i
P	probability
P_i	probability that particle i exists in the measurement frame (also called Miles-Lantuejoul factor)
V_{cal}	vertical calibration factor
V_i	relative volume of particle i
X_{A}	diameter of spherical particle i
X_{Ai}	area equivalent diameter of particle i
X_{F1}	horizontal Feret diameter of object
X_{F2}	vertical Feret diameter of object
X_i	dimension of particle i
$X_{i\text{max}}$	longest dimension of particle i, also called maximum Feret diameter
$X_{i\text{min}}$	shortest dimension of particle i, also called minimum Feret diameter
X_{LIL}	lower limit of a class interval
X_{mean}	mean of X_i
X_{UIL}	upper limit of a class interval
X_1	horizontal dimension of object

$X_{1,m}$	horizontal dimension, expressed in micrometres
$X_{1,p}$	horizontal dimension, expressed in pixels
X_2	vertical dimension of object
$X_{2,m}$	vertical dimension, expressed in micrometres
$X_{2,p}$	vertical dimension, expressed in pixels
Z_1	horizontal side length of the rectangular measurement frame
Z_2	vertical side length of the rectangular measurement frame

4 Sample preparation demands for method description

4.1 General recommendations

4.1.1 General

The following recommendations provide a sampling of standard microscopy practices.

NOTE See References [4], [5] and [10] for additional suggestions.

4.1.2 Sample subdivision

As only a small amount is needed to prepare a sample, the whole sample shall be subdivided in a manner that ensures that the portion taken is representative of the whole.

The method used to subdivide the sample is likely to be dictated by the sample preparation method and will be decided by the laboratory performing the analysis.

Provided that the sample is well dispersed by the method and that there is no segregation of particles by size, the choice of method is left to the expertise of the laboratory, since any specialized equipment required by a particular method might not be available to all.

4.1.3 Touching particles

The number of particles touching each other should be minimized.

It is a prime requirement of the method that measurements shall be made on isolated particles. There should be as few particles as possible touching each other. Touching particles measured as one particle without a proper separation will introduce error.

4.1.4 Particle distribution

There should be an adequate distribution of particles on the sample support. The whole area of the preparation should be examined to ascertain whether there is noticeable segregation of particles (by size). Statistical comparison of the results on a frame-by-frame basis will test for uniform distribution of particles. This procedure is detailed in Clause 7.

4.1.5 Sample preparation

Electron microscope samples should be coated with a thin layer of metal (e.g. Au, Au/Pd, Pt/Pd) to reduce charging effects.

Samples should be examined as soon as possible after preparation, and should contain an expiration date.

The sample preparation method used should be fully described in the final particle size analysis report by giving quantitative details of the nominal masses, volumes and compositions of particles and products used at each stage of the preparation procedure.

4.1.6 Number of particles to be counted

The number of particles measured should be determined based on the particle-size distribution and the desired confidence limits. Assuming the particles are log-normally distributed, the required number (N) of particles with a given error (δ) and a given confidence limit is estimated in accordance with Equation (1):

$$\log N = -2 \log \delta + K \tag{1}$$

where K is numerically determined by the confidence limit, particle distribution and other parameters; see References [1] and [2].

NOTE See Annex A for detailed information.

4.2 Suggested preparation methods

Several methods can be investigated for preparing samples for measurement. The following methods may be used. They are based on the assumption that a representative sample be used to give an adequate dispersion of the particles and a sharply contrasted image.

4.2.1 Camphor-naphthalene (C-N) method

This method uses a eutectic mixture of 60 % mass fraction camphor and 40 % mass fraction naphthalene that melts at 32 °C and sublimates rapidly in a vacuum. To prepare the sample, a 1 g sample of the particles to be counted is kneaded by hand inside a plastic bag with the requisite amount of the C-N eutectic mixture. When the particles sample is fully disaggregated and well dispersed in the C-N by the heat of the hand, the plastic bag is cooled to solidify the resulting mixture. Small lumps of this solid mixture are then transferred to a microscope slide resting on a warm plate. The sample, when melted, is flattened under a cover-slip that is afterwards removed to allow the C-N eutectic to sublimate under vacuum.

This technique was found to give good dispersion of irregular quartz particles and has the advantages that the particles are viewed in air, which results in a good contrast in the refractive index, and that the slides do not age. However, tests with glass beads have been unsuccessful, as the particles segregate on the slide, do not stick well and tend to roll off, making the method unusable; see Reference [3].

4.2.2 Paste-dilution method

A sample of about 1 g of particles is mixed with a viscous liquid (gelatine, sucrose or glycerol in water, collodion in amyl acetate) on a watch glass with a spatula to give a thick paste, thus ensuring mechanical disaggregation and dispersion. A sample of the paste is then taken with the point of a spatula and diluted in the same viscous liquid to a concentration such that, after homogenization, one drop of the resulting suspension, flattened under a cover-slip, will give the required number of particles on a microscope slide, that is, about 20 particles per view frame. Depending on the choice of liquid, the slides can have only a temporary life or might be able to be stored indefinitely. Using glycerol, this method has been successful for glass beads. It gives a good uniform dispersion and a reasonably contrasted image. The use of a cover-slip aids resolution with high-magnification objectives. However, the slides tend to dry out within an hour or so and repeat counts with the same slide are not possible; see Reference [4].

4.2.3 Filtration methods

4.2.3.1 Powder or dry suspensions

A 1 g sample of particles is suspended in a suitable liquid and dispersed. A given volume of this suspension is then filtered to dryness on a suitable membrane. The concentration of the suspension and the membrane area of filtration are such that the particles are deposited in the required concentration for counting (about 20 particles per measurement frame). After air-drying, the membrane is cut into small sections which are attached by their edges to a microscope slide using an acetone-resistant glue (e.g. cyanoacrylate or “super-glue”). The gluing is to prevent the membrane from shrinking. The slide is then put in a closed container on a support over a free surface of liquid acetone, whose vapour renders the membrane transparent for viewing and particle-counting. The method has the advantage that the particles are viewed in air giving a good contrast in refractive index. Tests indicate that to avoid the membrane re-opacifying, it is preferable to perform the exposure to acetone very slowly over several hours; see Reference [5].

4.2.3.2 Liquid suspensions

A known volume of suspension, typically 100 ml, is vacuum-filtered, as described below, through a membrane of compatible material and known pore size, typically 0,8 µm cellulose nitrate for mineral oils. Particles larger than the pore size should appear well scattered across the membrane with little or no overlap. If the number of particles is too great and overlapping is excessive, the test should be repeated with a smaller known volume of suspension. Conversely, if the number of particles is too few, a greater volume of known amount should be used. The vacuum arrangement, for example a Millipore[1)] filtration system, consists of a membrane holder attached to an open flask, with a vacuum pump attached below the filter holder. A separate spray container with an integral filter attachment, typically 0,45 µm, is used with a compatible solvent to wash down the sides of the open flask to ensure that all particles are collected on the membrane for analysis, and to remove the liquid from the suspension, leaving a reasonably dry membrane for examination. The membrane should be examined as soon as possible; if there is a delay, it can be inserted between two pre-cleaned microscope slides. Appropriate glue for making the membrane transparent may be used; see Reference [6].

4.2.4 Dry deposition method

Particles may be prepared for counting by dry deposition onto a slide covered with double-sided transparent adhesive tape. Care shall be taken that all the particles in a given sample effectively stick on the slide, so as to ensure that there is no selective capture of particles by size. A microscope slide is positioned in the bottom of a vacuum chamber having a volume of about 1 l. A conical metal plug is fitted in the top of the chamber and the particles to be analyzed are placed in a groove all around the plug. When the vacuum is released by lifting the plug, the particles are sucked as a cloud into the chamber and fall on the slide. Adhesion on the slide may be enhanced by using double-sided tape or a film of grease. This method also has the advantage that particles are viewed in air, resulting in a good contrast in refractive index.

5 Image capture

5.1 General

Particle-size data can be influenced by specific parameters affecting the image formation process. It is possible to distort the reported size, particularly of the smallest particles, by using inappropriate image-capture conditions, e.g. magnification, illumination, etc. Distortion in the image might arise from a number of causes, but its presence and effect on the image can be measured by selecting a recognizable object at a number of points and orientations in a frame of view. It is important to note that the measurements made provide only two-dimensional, X and Y, information. The imaging instrument should be set up and operated in accordance with its manufacturer's recommendations considering the conditions given below.

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1) Millipore is an example of a suitable product available commercially. This information is given for the convenience of users of this part of ISO 13322 and does not constitute an endorsement by ISO of this product.

5.2 Procedures

At each operating condition used for the analysis, carry out the following steps.

a) Select a recognizable object in the image.

b) Place the feature in the centre and at the corners of the field of view in turn and measure its horizontal length (X_1).

c) Rotate the sample stage 90 degrees and repeat the measurements (X_2).

d) Record the values of X_1 and X_2 with the final result.

e) Calibrate the imaging instrument prior to the examination of samples using a certified graticule or equivalent.

f) If possible, mount the traceable calibration graticule together with the specimen in the imaging instrument.

g) Select the magnification in accordance with Annex B or Annex C and set the corresponding illumination and imaging conditions.

h) Place the calibration grating in the field of view, select a suitable area and focus it.

i) Obtain the image to be analyzed and then capture it either digitally or by use of a suitable photographic image.

j) Record a significant number of measurement frames for each sample by scanning the sample in a raster pattern as indicated in Figure 1. Once this operation is started, no changes to the operating conditions should be made.

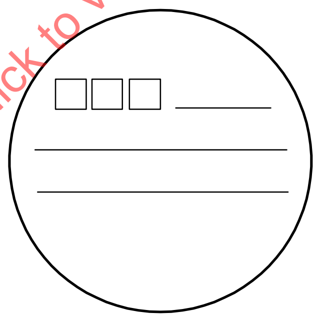


Figure 1 — Sample raster pattern

k) At the end of measurement, place the calibration graticule in the field of view and check the calibration once more. The comparison of two calibration images taken at the beginning and the end of the examination will provide a measure of the variability in instrument magnification.

l) Report the calibration constants obtained before and after the analysis together with the precise details of the microscope settings (working distance, spot size, electron microscope magnification, etc.).

5.3 Operating conditions for an image capture instrument

5.3.1 General

There are various imaging systems used for particle sizing. The setup for particle sizing using an electron or optical microscope is briefly described in 5.3.2 and 5.3.3.

5.3.2 Operating conditions for an electron microscope

The following special conditions are required for measuring particle size using an electron microscope:

a) image contrast mode: used to adjust the desired peak signal level;

b) accelerating voltage: set according to the material to be measured;

c) specimen position: The sample working distance specified by the electron microscope manufacturer for high resolution imaging should be selected. The sample should be mounted flat on the specimen holder with the stage tilt set to zero;

d) dynamic focus and tilt correction: both of these controls should be switched off.

e) Select the operating magnification after reference to Annex B. The total magnification is the product of electron microscope magnification and any image analyzer transfer magnification.

5.3.3 Operating conditions for an optical microscope

For bright-field images in the light microscope which are commonly used for particle sizing, the minimum feature length, d, expressed in micrometres, distinguishable in monochromatic light is given by Equation (2):

$$d = \frac{0{,}6\lambda}{\mu \sin\theta} \qquad (2)$$

where

λ is the wavelength, expressed in micrometres;

θ is the half angle, expressed in degrees, subtended by the particle at the objective lens;

μ is the refractive index of the surrounding medium.

The theoretical lower limit is approximately 0,2 µm, but the diffraction halo around the particle gives a gross overestimate of size. Special attention should be given to range of particle size to be measured, then to the measurement in order to obtain the required accuracy. Annex C gives the resolutions and smallest resolvable feature lengths for some typical objectives. As a rule of thumb, the smallest dimension of the smallest particle to be measured should be at least 10 times larger than this resolution limit.

6 Microscopy and image analysis

6.1 General

Modern image analyzers usually have algorithms available for enhancing the quality of the image prior to analysis and for separating touching particles. Enhancement algorithms may be used, provided that the measurements can be unambiguously associated with the particles in the original image. Irregularly shaped particles or particles with sharp corners should not be separated since this would distort the shape of the particles. All touching particles of this kind should be rejected from the measurement and a note should be made of the proportion of particles rejected from each measurement frame; see 6.3.4. Touching spherical particles may be separated, as this will give minor distortion of the area of particles. A flow-chart showing typical procedures used in carrying out measurements by image analysis is given in Annex D.

6.2 Size classes and magnification

The theoretical limit for resolution of objects by size using image analysis is one pixel and counts should be stored particle-by-particle with a maximum resolution of one pixel. Note that any compression of images might reduce the resolution. However, it is necessary to define the size classes for the final reporting of results; the desire for maximum resolution should be tempered by the necessity for precision, which is a function of the total number of particles counted, the dynamic range and the number of pixels included in the smallest objects to be considered. It is, therefore, recommended that pixels be converted to real-world dimensions prior to any reporting of size for quantitative analysis.

The magnification used should be such that the smallest particles counted have a projected area sufficient to meet the accuracy required. All particles measured should be sized and stored with a resolution of one pixel. The final results are to be reported by grouping the particles into size classes. For samples with a narrow size distribution, the grouping may be based on a linear progression and for samples with a wide distribution, the grouping may be based on a logarithmic progression. The intervals for these progressions should be based on the dynamic range and total number of particles counted. The particles assigned to a given class are those that have a diameter that is equal to or greater than the lower limit of the class interval, X_{LIL}, and less than the upper limit, X_{UIL}, as specified in Equation (3):

$$X_{\mathrm{LIL}} \leqslant X < X_{\mathrm{UIL}} \qquad (3)$$

Counts are to be checked on a frame-by-frame basis for significance using Student's t-test on the mean diameter and the F test on the variances. Counts not meeting the requirements should be rejected.

6.3 Counting procedure

6.3.1 General

The particle-size distributions should be determined by counting all particles in each measurement frame and then summing over all frames.

6.3.2 Particle edges

A suitable grey-level threshold setting should define the particle edges. Techniques for doing this depend on the sophistication of the image analysis equipment.

A half-amplitude method can be done manually, if necessary. A small region of the background located a few pixels away from the boundary of a typical particle is selected. The threshold level at which approximately half the pixels in the selected region are detected is recorded. This procedure is repeated for an area located a few pixels inside the particle boundary. The threshold level should be set at a value midway between these values; see Reference [7].

A second option is to "auto-threshold" the image, and then perform a manual check before proceeding with the measurement

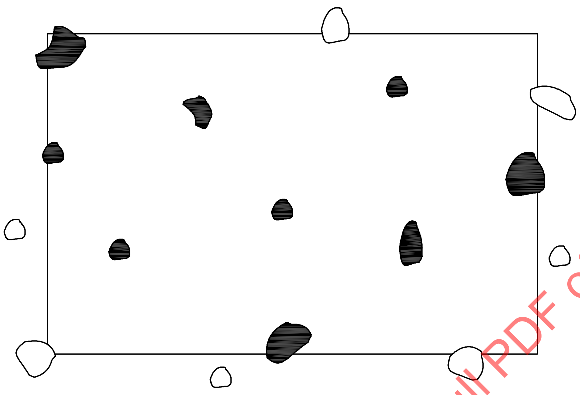
Whenever the threshold level is changed, the threshold image should be superimposed on the original image and a visual check made to determine whether all of the particles have been "thresholded" correctly. If not, the reason should be investigated and corrected before proceeding with the measurement.

6.3.3 Particles cut by the edge of the measurement frame

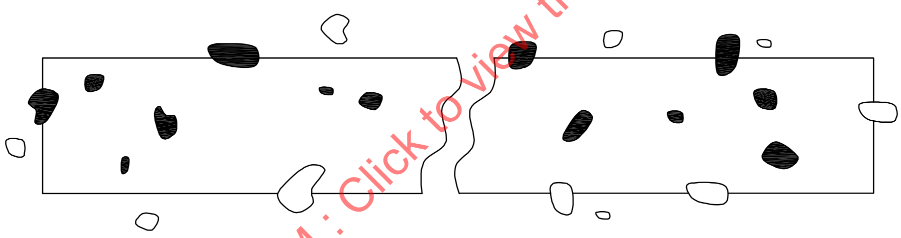
6.3.3.1 If all the objects that appear in the image frame are accepted for measurement, the accuracy of the final distribution will be impaired because some of the objects will be cut by the edge of the image frame. To overcome this, a measurement frame is defined within the image frame. The measurement frame can be used in the following two ways.

a) All the objects are allocated one pixel (e.g. the centroid) as the feature count point. Objects are accepted only when their feature count point lies within the measurement frame; see Figure 2 a). The measurement frame can be of any shape provided that there is enough space between the edges of the two frames so that no accepted particle is cut by the edge of the image frame.

b) A rectangular frame is used with the bottom and right edges defined as reject sides. Objects lying partially or wholly within the measurement frame and not touching the reject sides are accepted; see Figure 2 b). There has to be sufficient space between the top and left edges of the two frames so that no accepted object is cut by the edge of the image frame. This covers all eventualities except for particles intersecting the two opposite sides of the frame; these would either be too large to be measured at the magnification or would be so acicular as to be unsuitable for classification by area anyway. Image analysis systems that reject all particles intersecting a frame edge use an effective frame size that is different for each size class and also different for each particle shape.



a)



b)

NOTE Shaded particles are included in count; unshaded particles are excluded from count.

Figure 2 — Treatment of particles cut by the edge of the measurement frame — Counting isolated measurement frame (a); counting strip (b)

6.3.3.2 All particles entirely inside the measurement frame are accepted for counting. All particles outside, or cut by the edge, are neglected. This creates the situation where the probability for a particle to be included in the measurement frame varies inversely with the size of the particle. This, therefore, introduces a bias that is greater the larger the size of the particle considered. The probability, P_i, of particle i having a horizontal Feret, X_{F1}, and a vertical Feret, X_{F2}, in a rectangular measurement frame of size Z_1 by Z_2 is given by Equation (4):

$$P_i = \frac{(Z_1 - X_{F2})(Z_2 - X_{F2})}{Z_1 Z_2} \quad (4)$$

For spherical particles of diameter X_A this reduces to Equation (5):

$$P_i = \frac{(Z_1 - X_A)(Z_2 - X_A)}{Z_1 Z_2} \quad (5)$$

The population of particles in the measurement frame should, therefore, be divided by the probability, P_i.

EXAMPLE A square frame of size 100 units × 100 units is used for counting a population of particles of sizes ranging from 2 units to 10 units. The count of the particles wholly in the measuring frame and the correction factors are shown in Table 1:

Table 1 — Example of a corrected count

Diameter X_i arbitrary units	Raw count n_i	Probability P_i	Corrected count n_i/P_i
2	81	0,96	84
4	64	0,92	70
6	49	0,88	56
8	36	0,85	42
10	25	0,81	31

6.3.4 Touching particles

The slide-preparation method should be chosen to give a minimum number of touching particles. Nevertheless, it is inevitable that there will be touching particles in each measurement frame and some method of dealing with them is necessary.

However, the first requirement is to have an automatic method of identifying touching particles. This can be done (a) by following the number of particles "created" by numerical separation procedures, (b) by some criterion, such as the shape factor or the Euler number (the number of holes in an object) or even (c) by manual intervention. The statistical procedure for evaluating slides might also give some indications.

Numerical separation procedures are not recommended for separating particle aggregates into individual particles as they can change the size of the particles in the image and, in any case, make it difficult to ensure traceability. Such procedures for identifying aggregates can be investigated by comparing the results with the size counting performed on the original untreated image, but this would seem to be very laborious.

Identification of touching particles on the basis of shape or Euler number is not foolproof, in particular for compact overlapping agglomerates, and will not distinguish real out-of-shape or oversized particles. In cases where touching particles cannot be avoided, careful use of various techniques, e.g. fractal analysis to identify aggregates or model-based separation techniques, may be used to separate the particles.

6.3.5 Measurements

The measurement of the perimeter of particles depends strongly on the image-analysis system used. Accordingly, the primary measurement is the projected area of each particle, expressed in pixels, then the longest dimension of each particle, $X_{i\max}$, expressed in pixels.

These two determine the shortest dimension of each particle, $X_{i\min}$, thus allowing the definition of a shape factor with the greatest discrimination. It is, therefore, recommended that the primary values be

a) area of each object, A_i;

b) longest dimension of each particle, maximum Feret diameter, $X_{i\max}$;

c) shortest dimension of each particle, minimum Feret diameter, $X_{i\min}$.

These are used to calculate the area-equivalent diameter, X_{Ai}, in accordance with Equation (6), and the shape factor, φ, in accordance with Equation (7).

$$X_{Ai} = \sqrt{\frac{4A_i}{\pi}} \qquad (6)$$

$$\varphi = \frac{X_{i\max}}{X_{i\min}} \qquad (7)$$

Appropriate correction shall obviously be made if the equipment used is not based on square pixels. To aid comparison with the corresponding volumetric or mass certification method, the relative volume, V_i, of each particle i can be calculated from the projected area-equivalent diameter, X_{Ai}, of the particle weighted by the Miles-Lantoujoul factor, P_i, (see Table 1) for the contribution of the particle i to the whole population, in accordance with Equation (8):

$$V_i = \frac{(X_{Ai})^3}{P_i} \qquad (8)$$

6.3.6 Calibration and traceability

6.3.6.1 General

The equipment is first calibrated to convert pixels into SI length units, e.g., nanometres, micrometres, millimetres, etc., for the final results. The calibration procedure shall include verification of the uniformity of the field of view. An essential requirement of the calibration procedure is that all measurements shall be traceable back to the standard metre. This can be done by calibrating the image analysis equipment with a certified standard stage micrometer.

EXAMPLE The National Physical Laboratory certified chrome-on-glass reference stage graticule, National Institute of Standards Technology SRM 475 and SRM 484 or with certified spherical particles.

6.3.6.2 Recommendations and requirements

6.3.6.2.1 Touching particles

Each object in an image frame should be counted and reported in the results, together with its area, maximum and minimum Feret diameter, Euler number or a manual recognition mark indicating that the object is a group of touching particles. These data will allow the testing of criteria for detecting and rejecting touching particles.

6.3.6.2.2 Distortion

Distortion is identified as follows.

a) Select a square on a multiple-square grid feature from a reference stage graticule, e.g. the same size as the average particle. Place it at the centre and measure its width, X_1, and its height, X_2.

b) Place it at each of the four corners and measure its width, X_1, and its height, X_2, at each of the four additional positions.

c) Report the five values of X_1 and X_2 with the final results.

6.3.6.2.3 Calibration

Each setting of the microscope is calibrated as follows.

a) Determine the correspondence between image size in pixels and the size in micrometres using the multiple-square grid feature on the reference stage graticule.

b) Report the results as H_{cal}, calculated in accordance with Equation (9), and V_{cal}, calculated in accordance with Equation (10):

$$H_{cal} = \frac{X_{1,m}}{X_{1,p}} \tag{9}$$

where

$X_{1,m}$ is the horizontal dimension, expressed in micrometres;

$X_{1,p}$ is the horizontal dimension, expressed in pixels.

$$V_{cal} = \frac{X_{2,m}}{X_{2,p}} \tag{10}$$

where

$X_{2,m}$ is the vertical dimension, expressed in micrometres;

$X_{2,p}$ is the vertical dimension, expressed in pixels.

When using a matrix camera, either X_1 or X_2 and either H_{cal} or V_{cal} may be reported.

7 Calculation of the particle size results

The mean particle size, X_{mean}, and the variance, s^2, for a given number of particles, n_i, each with an associated diameter, X_i, are calculated in accordance with Equations (11) and (12), respectively:

$$X_{mean} = \frac{\sum X_i n_i}{\sum n_i} \tag{11}$$

$$s^2 = \frac{\sum n_i (X_i - X_{mean})^2}{\sum n_i - 1} \tag{12}$$

In order to ensure the homogeneity of the measurements, the mean diameter and the variance obtained in each measurement frame should to be tested by the analysis-of-variance and multiple-comparison tests (Annex E).

8 Test report

The test report shall contain as a minimum the following information:

a) identification of the test specimen;

b) reference to this part of ISO 13322 (ISO 13322-1:2004);

c) complete description of the method used for sub-sample preparation, with full quantitative details of the nominal weights, volumes and compositions of particles and products used at each stage of the sub-sample preparation procedure;

d) mean particle size, X_{mean};

e) variance, s^2;

f) full particle-by-particle results, with all dimensions in pixels, including the following:

- reference number of the particle;
- reference number of the sample;
- reference number of the sub-sample;
- reference number of the view field;
- objective used;
- frame size;
- area of particle;
- maximum and/or minimum Feret diameter;
- Euler number;
- X_1 calibration factor;
- X_2 calibration factor;
- relative volume of particle;
- any other useful information.

Usable results should be reported in tables and graphs in accordance with ISO 9276-1 and ISO 9276-2.

A micrograph should be provided of a typical field for each of the samples and for each setting of the microscope.

Annex A
(normative)

Study on the sample size required for the estimation of mean particle diameter

A.1 General

Theoretical equations had previously been developed to estimate statistical error caused by sample size (e.g. the number of sample particles). The theory gave general analytical solutions for the distribution of the sample mean diameter and the number of particles required to achieve a measurement within a certain error for a given confidential level. Computer simulation studies were carried out using a log-normal size distribution and the simulated results compared with the theoretical equation. The computer simulation results confirmed the theory. Therefore, the error caused by an inadequate number of sample particles or number of particles required in a measurement can be analytically estimated.

It was also found that the number of particles required can be reduced somewhat both by changing the evaluation basis and by increasing the assigned admissible error. However, a fairly large number of particles is required for a precise measurement that will be transformed into the mass basis.

A.2 Symbols and abbreviated terms

For the purpose of this document, the following symbols and abbreviated terms are used.

a constant

NOTE Used in Equation (A.37).

b initial value of b for the linear congruent method

$b_{i,r}$ first initial value of b

c constant equal to $\beta + \alpha / 2$

NOTE Used in Equation (A.5).

D_{CM} count median diameter

D_{MM} mass median diameter

D_{MV} mean volume diameter

D_{p} particle diameter

D_{p50} median diameter of particles

D_{p84} particle diameter which gives a cumulative undersize distribution of 84,13%

$\overline{D}_{\text{p}}$ sample-mean diameter

$\overline{D}_{\text{p}}^{*}$ population-mean diameter

d period of repeat of generated number

NOTE	From Equations (A.36) to (A.38).
e	relative error
NOTE	As defined by Equation (A.26).
$f_{\ln\kappa}$	sampling distribution of $\ln\kappa$
σ_{GSD}	geometric standard deviation
K	constant
NOTE	See Equation (A.1).
k	constant
NOTE	See Equation (A.37).
N	number of sample particles per experiment
n	number of sample particles
$n*$	number of particles required in a measurement to attain a certain level of confidence
P	probability
$P(\|e\| \leqslant \delta)$	probability that the experimental data will lie within the range of relative error $-\delta$ to $+\delta$
R	number of repeat of the random number generation
s	sample standard deviation
t_n	random number having normal probability
u	parameter of Equation (A.31) defined by Equation (A.32)
x_i	number calculated by Equation (A.37)
x_{i+1}	remainder during the calculation of random numbers
Y	experimental value
$Y*$	$Y\left(\mu^{(0)}, \sigma^2\right)$
y	process variables
$\bar{y}$	mean of y
y_i	uniform random number generated by computer
y_m	logarithmic mean diameter for a random sample of size n
z	function
NOTE	Defined in Equation (A.24).
α	constant

NOTE From Equation (A.1).

β basis number

δ relative error

κ dimensionless mean particle diameter

λ Y / Y^*

μ_0 logarithmic mean diameter for the number distribution of population

ν degrees of freedom

σ population standard deviation

σ_g population geometric standard deviation

σ_{GSD} geometric standard deviation

$\Phi(z)$ error function

$\phi(y_m, s^2)$ joint probability distribution of y_m and s^2

φ function

NOTE Defined in Equation (A.23).

ω parameter

NOTE Equation (A.34) defined by Equation (A.35).

A.3 Introduction

In general, the data obtained for a powder process scatter more widely than for other processes. Complicated phenomena, such as aggregation or adhesion, may account for some of the scatter. More fundamentally, however, the scatter can be attributed to the size distribution of the particles. Therefore, during the measurement of mean diameter of particles, the statistical error of the measured value caused by the size distribution should be included.

One of the most interesting problems relating to the error is how many particles should be sampled to achieve satisfactory results. Masuda and Iinoya [12] studied the problem theoretically and presented an equation for the number of particles, n^*, required to achieve a measurement within a certain relative error at a given confidential level. By use of this equation, the minimum number of measurements required under certain given conditions can be calculated from the particle size distribution, process variables and the basis number for the measurement.

The relationship, however, had not previously been confirmed by any experiments. We have now carried out the simulation experiments so as to examine the validity of the theoretical equation of Masuda and Iinoya [12].

A.4 Theoretical analysis for log-normal distribution

The main tenets of Masuda and Iinoya [12] are summarized here in order to define the relationships which are under examination. It is assumed that the process can be described by Equation (A.1);

$$y = KD_{\mathrm{p}}^{\alpha} \qquad \text{(A.1)}$$

where

- y is the particle property with respect to particle diameter;
- K and α are constants not equal to zero;
- D_{p} is the particle diameter.

Equation (A.1) describes one of the simplest but most important processes of particles, for example the sedimentation velocity as defined by Stokes' law. In this process, it is assumed that particle-size distribution is log-normal.

If the sample-mean particle diameter, $\bar{D}_{\mathrm{p}}$, is known, the distribution of experimental data can be calculated from Equation (A.2);

$$Y = K\bar{D}_{\mathrm{p}}^{\alpha} \qquad \text{(A.2)}$$

where

- Y is the particle property with respect to sample-mean diameter;
- $\bar{D}_{\mathrm{p}}$ is the sample-mean particle diameter;
- K and α are constants not equal to zero.

For this process, the population-mean particle diameter, $\bar{D}_{\mathrm{p}}^{*}$, adjusted by the basis number, β ($\beta = 0$ for a count basis, and $\beta = 3$ for a mass basis), is given by Equations (A.3) to (A.5); see Reference [13]:

$$\bar{D}_{\mathrm{p}}^{*} = \bar{y} / y \qquad \text{(A.3)}$$

where

- $\bar{D}_{\mathrm{p}}^{*}$ is the population-mean particle diameter;
- $\bar{y}$ is the mean of y;

$$\bar{D}_{\mathrm{p}}^{*} = \left[\frac{1}{K} \frac{K \int D_{\mathrm{p}}^{\beta} D_{\mathrm{p}}^{\alpha} f(\ln D_{\mathrm{p}}, \mu_0, \sigma^2) d \ln D_{\mathrm{p}}}{\int D_{\mathrm{p}}^{\beta} f(\ln D_{\mathrm{p}}, \mu_0, \sigma^2) d \ln D_{\mathrm{p}}} \right]^{1/\alpha} \qquad \text{(A.4)}$$

where

- c equals $\beta + \alpha / 2$ (A.5)
- μ_0 is the logarithmic mean diameter for the number distribution of the population, calculated in accordance with Equation (A.6):

$$\mu_0 = \int_{-\infty}^{\infty} \ln D_{\mathrm{p}} f(\ln D_{\mathrm{p}}, \mu^{(0)}, \sigma^2) d \ln D_{\mathrm{p}} = \ln D_{\mathrm{p50}} \qquad \text{(A.6)}$$

where

D_{p50} is the mean diameter of the particles;

σ is the population standard deviation, calculated in accordance with Equation (A.7):

$$\sigma = \ln \sigma_{\mathrm{g}} = \ln D_{\mathrm{p84}} - \ln D_{\mathrm{p50}} \qquad \text{(A.7)}$$

where

D_{p84} is mean particle diameter which gives a cumulative undersize distribution of 83.13%;

σ_{g} is the population geometric standard deviation.

Therefore,

$$\overline{D}_{\mathrm{p}}^{*} = \exp(\mu_0 + c\sigma^2) \qquad \text{(A.8)}$$

where σ^2 is the variance of the logarithmic mean diameter for the number distribution of the population.

On the other hand, the mean diameter, y_m, of the random sample of size n is calculated from Equation (A.9);

$$\overline{D}_{\mathrm{p}} = \exp(y_m + cs^2) \qquad \text{(A.9)}$$

where

$$y_m = \frac{\sum \ln D_{\mathrm{p}}}{n} \qquad \text{(A.10)}$$

$$s^2 = \frac{\sum (\ln D_{\mathrm{p}} - y_m)^2}{n} \qquad \text{(A.11)}$$

If it is assumed that the number of sample particles, n, is sufficiently large, then the distribution $\phi(y_m, s^2)$ of the combined probabilities y_m and s^2 can be calculated from Equation (A.12):

$$\phi(y_m, s^2) = \frac{n}{\sigma^3 \sqrt{2\pi}} \sqrt{\frac{n}{2(n-1)}} \exp\left\{ -\frac{n}{2\sigma^2}(y_m - \mu_0)^2 - \frac{1}{4(n-1)}\left[\frac{n}{\sigma^2}s^2 - (n-1)\right]^2 \right\} \qquad \text{(A.12)}$$

Equation (A.13) can then be derived from Equation (A.8):

$$d \ln \overline{D}_{\mathrm{p}} = dy_m + cd(s^2) \qquad \text{(A.13)}$$

Therefore,

$$d(s^2)dy_m = d(s^2)\Lambda dy_m = d(s^2)\Lambda d \ln \overline{D}_{\mathrm{p}} - cd(s^2)\Lambda d(s^2) \qquad \text{(A.14)}$$

where

$$d(s^2)dy_m = d(s^2)\Lambda d\ln\overline{D}_\mathrm{p} \tag{A.15}$$

Equation (A.15) can be rearranged in the form of Equation (A.17), the combined probability distribution:

$$1 = \int_{-\infty}^{\infty}\int_{0}^{\infty}\phi(y_m, s^2)\mathrm{d}(s^2)\mathrm{d}y_m \tag{A.16}$$

$$= \int_{-\infty}^{\infty}\left[\int_{0}^{\infty}\phi(\ln\overline{D}_\mathrm{p} - cs^2, s^2)\mathrm{d}(s^2)\right]\mathrm{d}\ln\overline{D}_\mathrm{p} \tag{A.17}$$

If a certain function $f\left(\ln\overline{D}_\mathrm{p}\right)$ is defined by Equation (A.18):

$$f(\ln\overline{D}_\mathrm{p}) \equiv \int_{0}^{\infty}\phi(\ln\overline{D}_\mathrm{p} - cs^2, s^2)\mathrm{d}(s^2) \tag{A.18}$$

it is evident that the function $f\left(\ln\overline{D}_\mathrm{p}\right)$ satisfies the condition defined in Equation (A.19):

$$\int_{-\infty}^{\infty} f(\ln\overline{D}_\mathrm{p})\mathrm{d}\ln\overline{D}_\mathrm{p} = 1 \tag{A.19}$$

Therefore, Equation (A.19) represents the distribution of $\ln\overline{D}_\mathrm{p}$. The distribution of $\ln\overline{D}_\mathrm{p}$ can be obtained by carrying out the integration of Equation (A.18) using Equation (A.12).

Then, if the dimensionless quantity $\kappa = \overline{D}_\mathrm{p} / \overline{D}_\mathrm{p}^*$ is introduced for the sake of simplicity, Equation (A.19) can be rewritten as Equation (A.20):

$$\int_{-\infty}^{\infty} f(\ln\kappa + \ln\overline{D}_\mathrm{p}^*)\mathrm{d}\ln\kappa = 1 \tag{A.20}$$

The distribution $\ln\kappa$ is denoted by $f_{\ln\kappa}$, as shown in Equation (A.21):

$$f_{\ln\kappa} \equiv f(\ln\kappa + \ln\overline{D}_\mathrm{p}^*) \tag{A.21}$$

The distribution $f_{\ln\kappa}$, as shown in Equation (A.22), can be obtained by carrying out the integration of Equation (A.18):

$$f_{\ln\kappa} = \frac{\sqrt{n}}{\sigma\sqrt{2\pi}\sqrt{2c^2\sigma^2 + 1}} e^{-\frac{\varphi^2}{2}\Phi(z)} \tag{A.22}$$

where

$$\varphi \equiv \frac{\sqrt{n}(\ln\kappa + c\sigma^2 / n)}{\sigma\sqrt{2c^2\sigma^2 + 1}} \tag{A.23}$$

$$z \equiv \sqrt{\frac{n}{2(2c^2\sigma^2 + 1)}}(2c\ln\kappa + 2c^2\sigma^2 + 1) \tag{A.24}$$

$$\Phi(z) \equiv \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{z} \mathrm{e}^{-\frac{z^2}{2}} \mathrm{d}z \tag{A.25}$$

The distribution $f_{\ln\kappa}$ is the theoretical distribution of the sample mean diameter. Thus, the probability density distribution of mean diameters obtained from simulation experiments must coincide with the distribution $f_{\ln\kappa}$ if the theoretical equation is correct.

When studying the problem of "how many particles should be sampled to get a satisfactory results", it is necessary to determine the probability, P, that the experimental data will lie within $-\delta$ to $+\delta$ of the relative error. The relative error e is defined by Equation (A.26):

$$e \equiv \frac{Y - Y^*}{Y^*} = \lambda - 1 \tag{A.26}$$

where

Y^* denotes $Y\left(\mu_0, \sigma^2\right)$

$$\lambda \equiv Y / Y^* = (\overline{D}_\mathrm{p} / \overline{D}_\mathrm{p}^*)^\alpha = \kappa^\alpha \tag{A.27}$$

Then, the probability, P, can be evaluated in accordance with Equation (A.28):

$$P(|e| \le \delta) = \left| \int_{\frac{\ln(1-\delta)}{\alpha}}^{\frac{\ln(1+\delta)}{\alpha}} f_{\ln\kappa} \mathrm{d}\ln\kappa \right| \tag{A.28}$$

The probability $P(|e| \le \delta)$, which is the probability that the mean diameter obtained for the number of sample particles, n, will be within the relative error $\pm\,\delta \times 100$ %, is calculated from the distribution $f_{\ln\kappa}$. If the distribution of $\ln\kappa$ obtained by simulation experiments coincides with the theoretical distribution of $\ln\kappa$, the probability, P, calculated from the simulation results can be expressed by Equation (A.28).

By substituting the approximation $\Phi(z) \cong 1$, Equation (A.28) can be simplified as follows:

$$P \cong \frac{\sqrt{n}}{\sigma\sqrt{2c^2\sigma^2+1}} \frac{1}{\sqrt{2\pi}} \left| \int_{-\delta/\alpha}^{\delta/\alpha} \mathrm{e}^{-\frac{\varphi^2}{2}} \mathrm{d}\ln\kappa \right| \tag{A.29}$$

$$= \frac{1}{\sqrt{2\pi}} \left(\int_{-\infty}^{\infty} \mathrm{e}^{-\frac{\varphi^2}{2}} \mathrm{d}\varphi - \int_{-\infty}^{-|u|} \mathrm{e}^{-\frac{\varphi^2}{2}} \mathrm{d}\varphi - \int_{|u|}^{\infty} \mathrm{e}^{-\frac{\varphi^2}{2}} \mathrm{d}\varphi \right) \tag{A.30}$$

$$= 1 - 2\Phi(-|u|) \tag{A.31}$$

where

$$u \equiv \frac{\sqrt{n}\delta}{\alpha\sigma\sqrt{2c^2\sigma^2+1}} \tag{A.32}$$

Equation (A.31) can be rearranged as Equation (A.33):

$$\Phi(-|u|) = \frac{1-P}{2} \quad \text{(A.33)}$$

If P, which is equal to the confidence limit, is known, u can be obtained from Equation (A.33) and the number of particles, n, from Equation (A.32). The number of particles, n, thus obtained is defined as “the number of particles required” and denoted by n^*. Equation (A.32) can then be written as Equation (A.34):

$$\log n^* = -2\log\delta + \log\omega \quad \text{(A.34)}$$

where

$$\omega \equiv u^2\alpha^2\sigma^2(2c^2\sigma^2 + 1) \quad \text{(A.35)}$$

Typical results calculated from Equation (A.33) are listed in Table A.1.

The numerical vale of parameter ω can be determined if the probability, P, is assigned and the variance, σ^2, of the particle population, the exponent, α, of the process variable and the basis number, β, of the measurement are known. Then, the number of particles, n^*, can be calculated from Equation (A.34).

Table A.1 — Parameter u calculated from Equation (A.33)

P (%)	50	75	80	90	95	97,5	99	99,5	99,8	99,9
u (–)	0,67	1,15	1,28	1,64	1,96	2,24	2,58	2,81	3,09	3,29

A.5 Simulation experiment

A.5.1 Generation of random numbers

In the simulation, 10^3 sets each containing 2×10^4 random numbers having a normal probability distribution are generated. A flowchart of the process is shown in Figure A.1. In order to generate a set of random numbers that has a normal distribution probability, it is necessary to generate random numbers having a value from 0 to 1 with a uniform probability (step 1). To generate the random numbers, y_{i+1}, the linear congruent method, based on the value of the remainder as determined from Equations (A.36) to (A.38), is employed (step 2):

$$x_{i0} = b \quad \text{(A.36)}$$

$$x_{i+1} \equiv ax_i + k \pmod d \quad \text{(A.37)}$$

$$y_{i+1} = x_{i+1} / d \quad \text{(A.38)}$$

where

x_{i+1} is equal to the remainder of $ax_i + k$ divided by d;

d is equal to the repeat period of the generated numbers;

a, k are constants selected so that the repeat period is longer than the total number of generated numbers.

Here, the values used are a = 48 828 125 (= 511) and k = 0. In this case, the repeat period d is 231 (= 2,15 × 109). The initial value of b, designated as $b_{i,r}$, is not a function of the repeat period, but is selected from the range 0 to $\sim d$. In this calculation, the first initial value $b_{i,r}$ of b is set equal to a (= 48 828 125). The χ^2 -test shows that the uniformity of the generated random numbers can be guaranteed at the 99 % confidence interval, i.e., the value χ^2 for the generated numbers is 8,89, while the theoretical χ^2 value at the 99 % confidence interval is 21,7 because the degrees of freedom v = 9 (= number of possibilities minus 1).

The first initial number, $b_{i,r}$, for the calculation of y_i is generated using the initial value b = 48 828 125 (step 1). This first initial value, $b_{i,r}$, is used as the initial value b for the generation of the $b_{i,r+1}$ in the next set (r + 1).

The random numbers, t_n , which have a normal probability distribution, can be generated from the uniform random numbers, y_i , using Equation (A.39); see Reference [14]:

$$t_\mathrm{n} = \sum_{i=1}^{12} y_i - 6 \qquad \text{(A.39)}$$

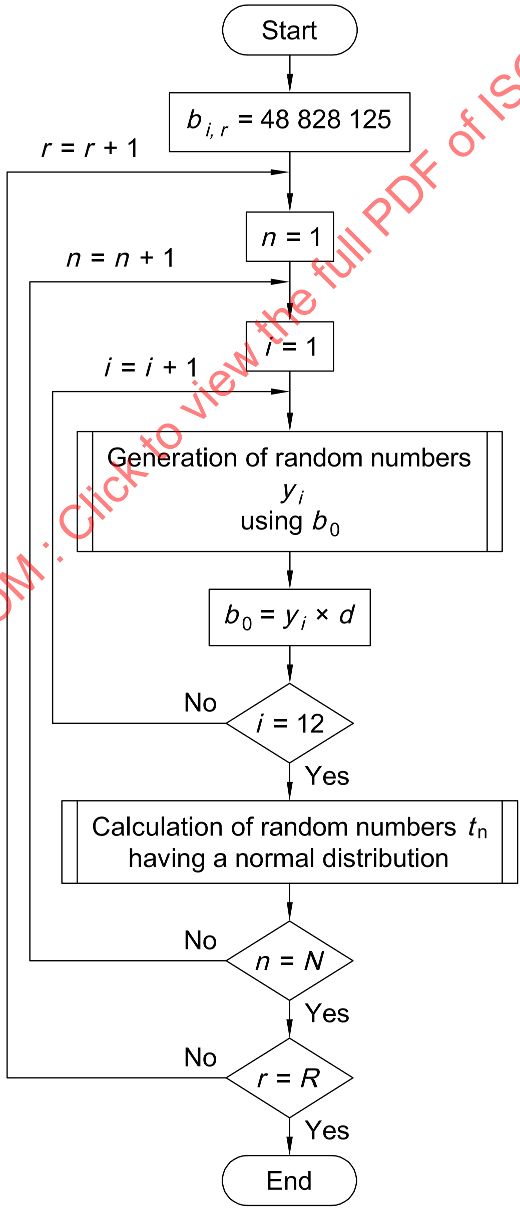


Figure A.1 — Flowchart of the generation of random numbers having a Gaussian distribution

Therefore, 12 calculations are required for y_i using the linear congruent method; see step 2 above.

In order to generate 2 × 10[4] random numbers, t_n, having a normal probability distribution, step 2 is repeated 24 × 10[4] times (step 3). Then, these 24 × 10[4] random numbers are used to generate the $N = 2 \times 10^4$ random numbers, t_n.

In this calculation, in order to generate the 10[3] sets each containing 2 × 10[4] random numbers, t_n, each having a normal probability distribution, steps 1 to 3 are repeated $R = 10^3$ times. As a result, $R = 10^3$ sets of $N = 2 \times 10^4$ random numbers, t_n, are generated using 24 × 10[7] random numbers, y_i.

A.5.2 Calculation of error introduced by sample size

By using the random numbers, t_n, having a normal distribution, the statistical error introduced by sample size (= number of sample particles) was studied. In order to transform the values t_n of the normal distribution into the values D_p of the log-normal distribution, following equation was used:

$$\ln D_p = t_n \ln \sigma_{GSD} + \ln D_{CM} \tag{A.40}$$

where

σ_{GSD} is the geometric standard deviation;

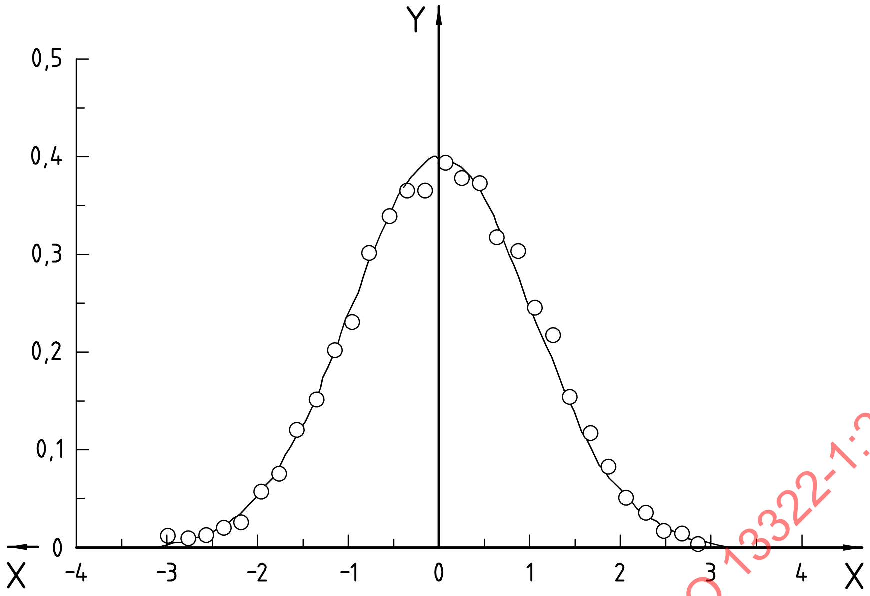
D_{CM} is the count median diameter.

Figure A.2 shows one of the distributions of D_p for the case where $N = 10^4$. It is matter of course that the distribution almost fits the log-normal distribution curve calculated from Equation (A.41):

$$f(D_p) = \frac{1}{\sqrt{2\pi} \ln \sigma_{GSD}} \exp\left[-\frac{(\ln D_p - \ln D_{CM})^2}{2\ln^2 \sigma_{GSD}}\right] \tag{A.41}$$

The mean diameter $\overline{D}_p$ of N sample particles is calculated from Equations (A.4), (A.6) and (A.8). Then, α is set equal to 2 so as to compare the results with theoretical values presented in Reference [1]. The theoretical value (for $N = \infty$) of median diameter, $\overline{D}_p^*$, is defined by Equation (A.3), which can be written as Equation (A.42):

$$\overline{D}_p^* = \exp(\ln D_{CM} + c \ln^2 \sigma_{GSD}) \tag{A.42}$$



Key

X $\ln D_p - \ln \overline{D}_p$

Y frequency $f[\ln D_p]$

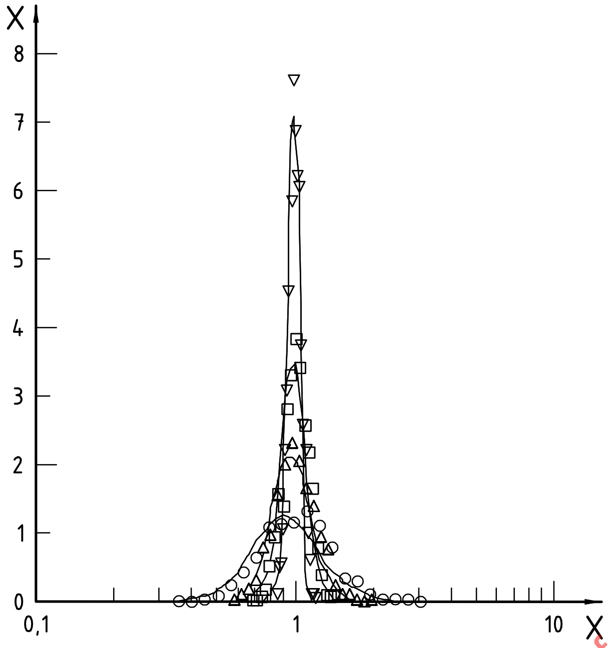
○ values from computer simulation

—— theoretical curve

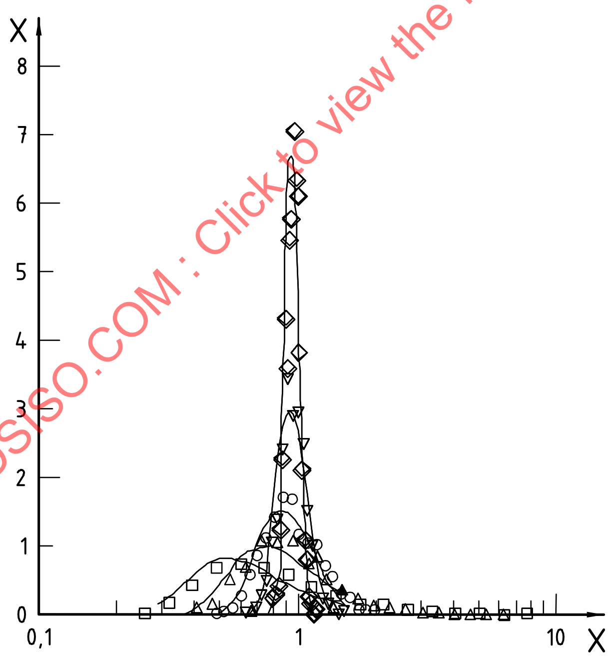
Figure A.2 — Frequency distribution of generated number

Figure A.3 shows the probability density distributions $f_{\ln\kappa}$ of the dimensionless mean particle diameters $\kappa\,(= D_p / D_p^*)$ for the count and mass bases in the case of σ_{GSD} = 1,6. When the sample number, N, is small, the distribution is broad. And when $N = 3$, κ at the peak of distribution is smaller than unity. This result shows the mean value obtained by small numbers of sample particles may include large errors and has a smaller value than the true mean diameter.

From the distribution $f_{\ln\kappa}$ shown in Figure A.3, the probability $P(|e| \leqslant \delta)$, which is the probability that the mean diameter calculated from the number, N, of sample particles will be within the relative error $\pm\delta \times$ 100%, can be calculated by the same equation as Equation (A.28). Calculated results for σ_{GSD} = 1,6 are shown in Figure A.4. Theoretical curves calculated from Equations (A.25) and (A.28) are also shown in this figure. The calculated results almost fit the theoretical curve both for count basis and for mass basis. For the count basis, the probability $P(|e| \leqslant \delta)$ is unity for the number of sample particles, N, greater than 3×10^3. Then, if the number of samples is greater than 9×10^3, the obtained mean diameter on the count basis can be recognized as the true value at the 95 % confidence interval. On the other hand, $P(|e| \leqslant \delta)$ for the mass basis is always lower than that for the count basis. This means that the required number of samples for the mass basis is larger than that for the count basis for the same level of confidence.



a) Count basis



b) Mass basis

Figure A.3 — Distribution of κ

Key

X mean diameter, dimensionless

Y probability density $f^{\ln \kappa}$ [1/ln κ]

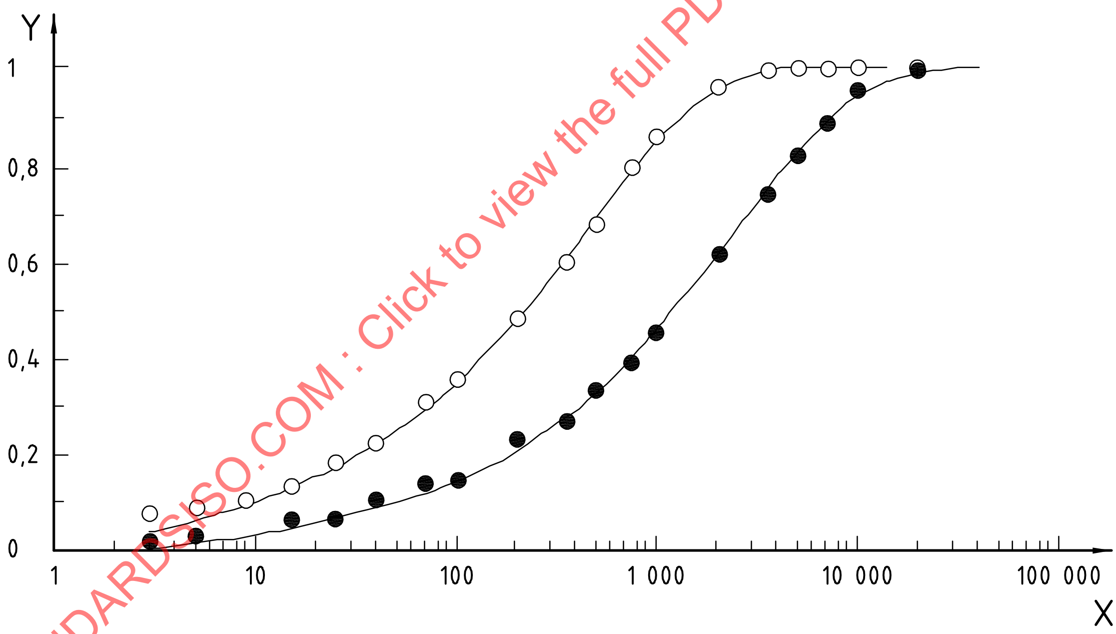
—— theoretical curve

values from computer simulation

for count basis		for mass basis	
○	N = 3	□	N = 3
△	N = 9	△	N = 9
□	N = 25	○	N = 25
▽	N = 100	▽	N = 100
		◇	N = 500

Conditions: σ_{GSD} = 1,6, R = 10^3 and α = 2.

Figure A.3 *(continued)*



Key

X sample size, N

Y P ($|e| \leqslant$ 0,05)

—— theoretical curve

values from computer simulation

○ count basis

● mass basis

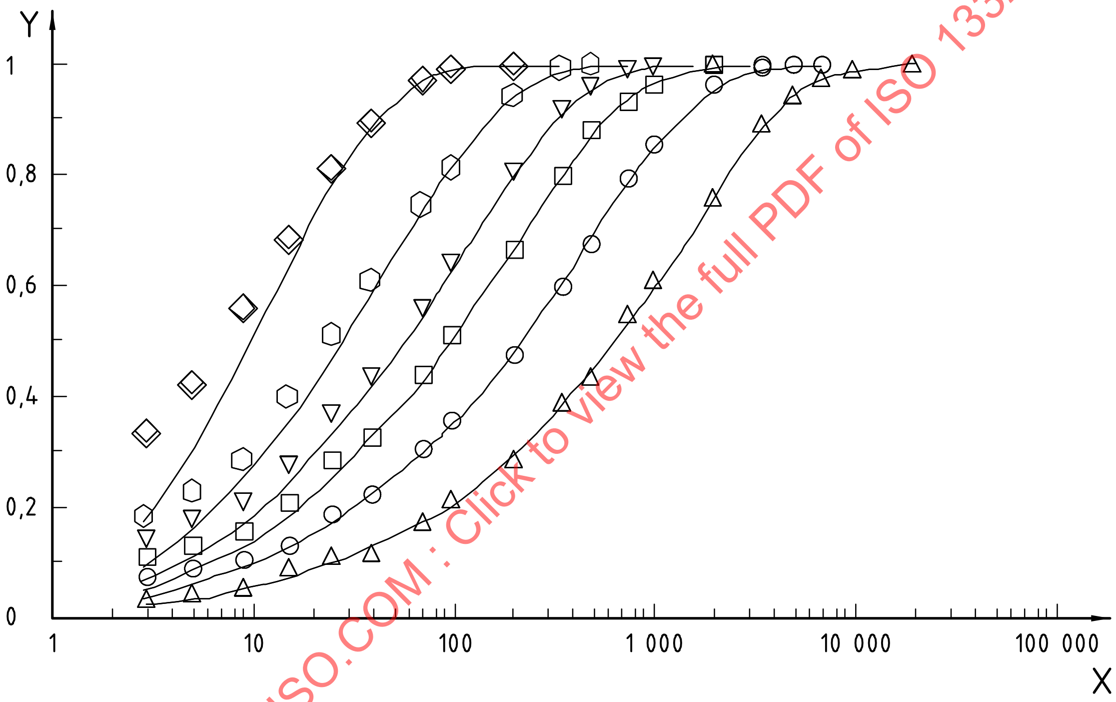
Conditions: σ_{GSD} = 1,6, R = 10^3 and α = 2.

Figure A.4 — Probability $P(|e| \leqslant 0{,}05)$ versus sample size

Figure A.5 a) shows the calculated results for the probability $P(|e| \leqslant \delta)$ for count basis, while Figure A.5 b) shows the results for the mass basis. The results obtained by the computer simulation are well represented by the theoretical lines, especially for the larger σ_{GSD}s. The observed deviation for the smaller values of n is largely due to the fact that the joint probability distribution $\phi(m, s^2)$ assumes that is the number of sample particles n is sufficiently large.

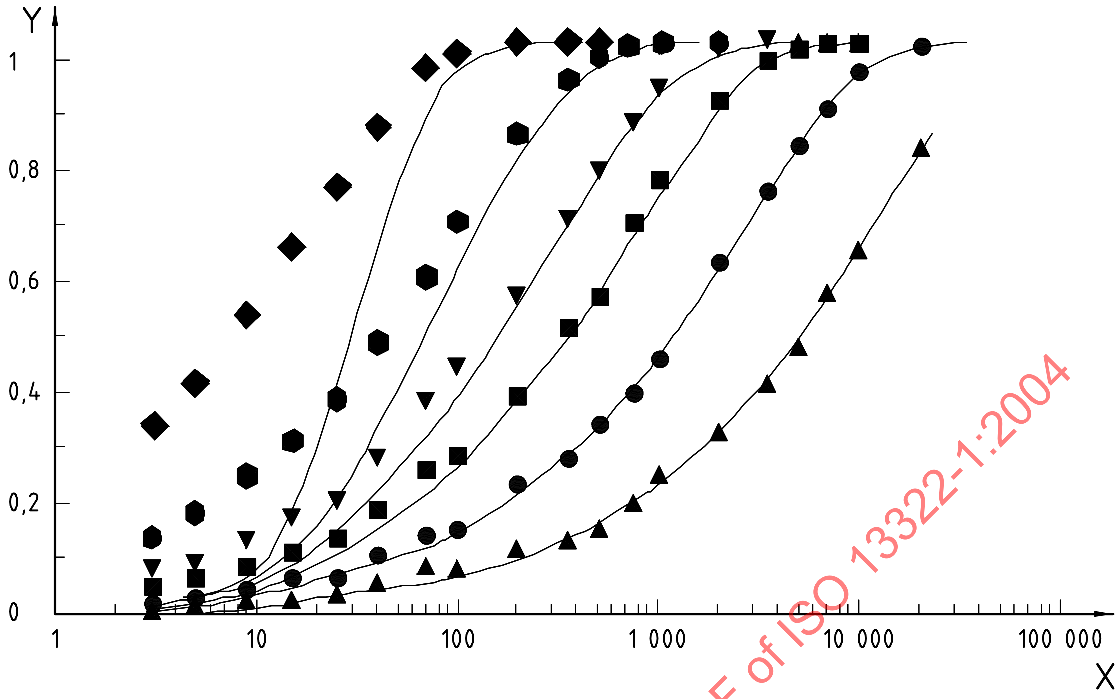
For any $P(|e| \leqslant \delta)$, the number of particles required for the measurement can be correlated to the relative error of experimental data, δ, by using parameter ω, as shown in Figure A.6. Parameter ω for the simulation experiment is calculated by substituting $\sigma = \ln \sigma_{GSD}$ into Equation (A.35). All of the calculated data coincide with the theoretical line calculated by Equation (A.34).

This result shows that parameter ω can be calculated, if the probability, P, is assigned and the σ_{GSD} of the population-particles, and if the exponent, α, of the process variable (in this study $\alpha = 2$) and the basis number, β, are known. Therefore, it is confirmed that the number of particles, n^*, required for a measurement can be obtained numerically by using Equation (A.34).



a) Count basis

Figure A.5 — Probability $P(|e| \leqslant 0{,}05)$ versus sample size, N



b) Mass basis

Key

X $P(|e| \leqslant 0{,}05)$

Y sample size, N

—— theoretical curve

values from computer simulation

◇ , ◆ σ_{GDS} = 1,1

⬡ , ⬢ σ_{GDS} = 1,2

▽ , ▼ σ_{GDS} = 1,3

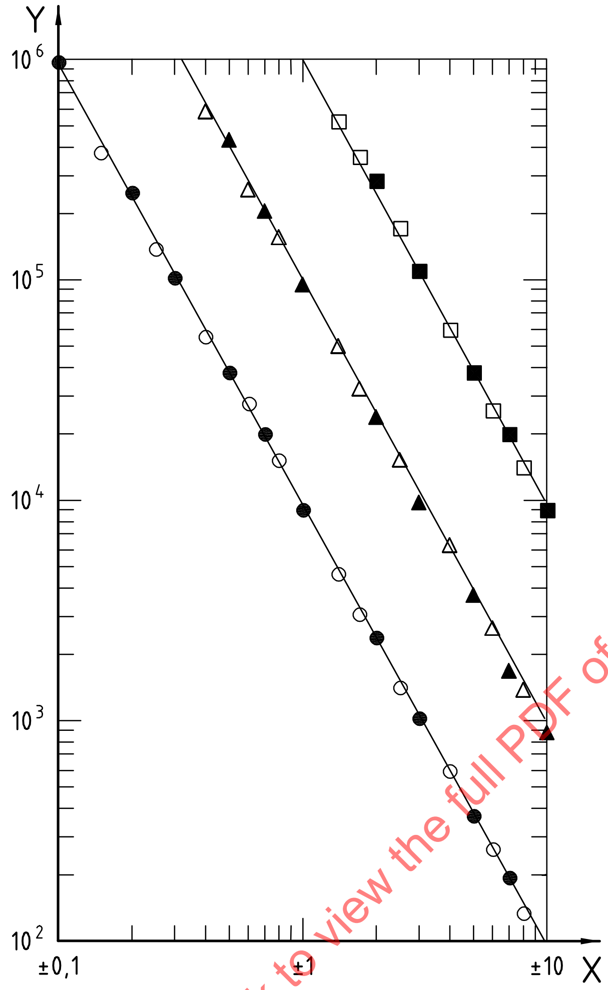
□ , ■ σ_{GDS} = 1,4

○ , ● σ_{GDS} = 1,6

△ , ▲ σ_{GDS} = 2,0

Conditions: $R = 10^3$ and $\alpha = 2$.

Figure A.5 (*continued*)



Key

X relative error of experimental data, δ, $\times$ 100, percent

Y number of particles required, n*

—— theoretical curve

values from computer simulation

○ ω = 1, count basis

● ω = 1, mass basis

△ ω = 10, count basis

▲ ω = 10, mass basis

□ ω = 100, count basis

■ ω = 100, count basis

Figure A.6 — Number of particles required, n*, versus relative error, δ