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Particle size analysis — Laser diffraction methods —

Part 1: General principles

Analyse granulométrique — Méthodes par diffraction laser — Partie 1: Principes généraux

Reference number
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Contents

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

International Standards are drafted in accordance with the rules given in the ISO/IEC Directives, Part 3.

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.

International Standard ISO 13320-1 was prepared by Technical Committee ISO/TC 24, *Sieves, sieving and other sizing methods*, Subcommittee SC 4, *Sizing by methods other than sieving*.

ISO 13320 consists of the following parts, under the general title *Particle size analysis — Laser diffraction methods*:

— *Part 1: General principles*

— *Part 2: Validation of inversion procedures*

Annexes A to E of this part of ISO 13320 are for information only.

Introduction

Laser diffraction methods are nowadays widely used for particle sizing in many different applications. The success of the technique is based on the fact that it can be applied to various kinds of particulate systems, is fast and can be automated and that a variety of commercial instruments is available. Nevertheless, the proper use of the instrument and the interpretation of the results require the necessary caution.

Therefore, there is a need for establishing an International Standard for particle size analysis by laser diffraction methods. Its purpose is to provide a methodology for adequate quality control in particle size analysis.

Historically, the laser diffraction technique started by taking only scattering at small angles into consideration and, thus, has been known by the following names:

— Fraunhofer diffraction;

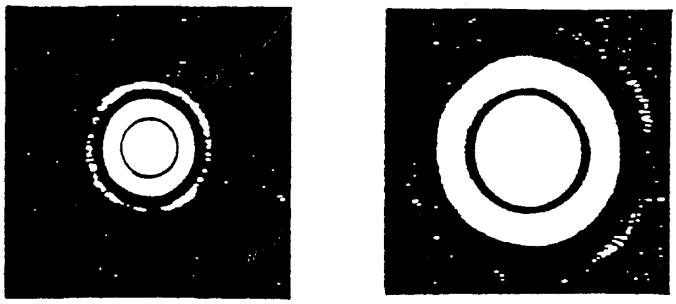
— (near-) forward light scattering;

— low-angle laser light scattering (LALLS).

However, the technique has been broadened to include light scattering in a wider angular range and application of the Mie theory in addition to approximating theories such as Fraunhofer and anomalous diffraction.

The laser diffraction technique is based on the phenomenon that particles scatter light in all directions with an intensity pattern that is dependent on particle size. All present instruments assume a spherical shape for the particles. Figure 1 illustrates the characteristics of single particle scattering patterns: alternation of high and low intensities, with patterns that extend for smaller particles to wider angles than for larger particles [2-7, 10, 15 in the bibliography].

Within certain limits the scattering pattern of an ensemble of particles is identical to the sum of the individual scattering patterns of all particles present. By using an optical model to compute scattering patterns for unit volumes of particles in selected size classes and a mathematical deconvolution procedure, a volumetric particle size distribution is calculated, the scattering pattern of which fits best with the measured pattern (see also annex A).



a) b)

Figure 1 — Scattering pattern for two spherical particles: the particle generating pattern a) is twice as large as the one generating pattern b)

A typical laser diffraction instrument consists of a light beam (usually a laser), a particulate dispersing device, a detector for measuring the scattering pattern and a computer for both control of the instrument and calculation of the particle size distribution. Note that the laser diffraction technique cannot distinguish between scattering by single particles and scattering by clusters of primary particles forming an agglomerate or an aggregate. Usually, the resulting particle size for agglomerates is related to the cluster size, but sometimes the size of the primary particles is reflected in the particle size distribution as well. As most particulate samples contain agglomerates or aggregates

and one is generally interested in the size distribution of the primary particles, the clusters are usually dispersed into primary particles before measurement.

Historically, instruments only used scattering angles smaller than 14°, which limited the application to a lower size of about 1 µm. The reason for this limitation is that smaller particles show most of their distinctive scattering at larger angles (see also annex A). Many recent instruments allow measurement at larger scattering angles, some up to about 150°, for example through application of a converging beam, more or larger lenses, a second laser beam or more detectors. Thus, smaller particles down to about 0,1 µm can be sized. Some instruments incorporate additional information from scattering intensities and intensity differences at various wavelengths and polarization planes in order to improve the characterization of particle sizes in the submicrometre range.

Particle size analysis — Laser diffraction methods —

Part 1:
General principles

1 Scope

This part of ISO 13320 provides guidance on the measurement of size distributions of particles in any two-phase system, for example powders, sprays, aerosols, suspensions, emulsions and gas bubbles in liquids, through analysis of their angular light scattering patterns. It does not address the specific requirements of particle size measurement of specific products. This part of ISO 13320 is applicable to particle sizes ranging from approximately 0,1 μm to 3 mm.

For non-spherical particles, an equivalent-sphere size distribution is obtained because the technique uses the assumption of spherical particles in its optical model. The resulting particle size distribution may be different from those obtained by methods based on other physical principles (e.g. sedimentation, sieving).

2 Normative reference

The following normative document contains provisions which, through reference in this text, constitute provisions of this part of ISO 13320. For dated references, subsequent amendments to, or revisions of, any of these publications do not apply. However, parties to agreements based on this part of ISO 13320 are encouraged to investigate the possibility of applying the most recent edition of the normative document indicated below. For undated references, the lates edition of the normative document referred to applies. Members of ISO and IEC maintain registers of currently valid International Standards.

ISO 9276-1:1990, *Representation of results of particle size analysis — Part 1: Graphical representation.*

3 Terms, definitions and symbols

For the purposes of this part of ISO 13320, the following terms, definitions and symbols apply.

3.1 Terms and definitions

3.1.1
absorption
reduction of intensity of a light beam traversing a medium through energy conversion in the medium

3.1.2
coefficient of variation
relative measure (%) for precision: standard deviation divided by mean value of population and multiplied by 100 (for normal distributions of data the median is equal to the mean)

3.1.3
complex refractive index
N_p
refractive index of a particle, consisting of a real and an imaginary (absorption) part

$$N_p = n_p - ik_p$$

3.1.4
relative refractive index
m
complex refractive index of a particle, relative to that of the medium

$$m = N_p/n_m$$

3.1.5
deconvolution
mathematical procedure whereby the size distribution of a particle ensemble is inferred from measurements of their scattering pattern

3.1.6
diffraction
spreading of light around the contour of a particle beyond the limits of its geometrical shadow with a small deviation from rectilinear propagation

3.1.7
extinction
attenuation of a light beam traversing a medium through absorption and scattering

3.1.8
model matrix
matrix containing light scattering vectors for unit volumes of different size classes, scaled to the detector's geometry, as derived from model computation

3.1.9
multiple scattering
subsequent scattering of light at more than one particle, causing a scattering pattern that is no longer the sum of the patterns from all individual particles (in contrast to single scattering)

3.1.10
obscuration
optical concentration
percentage or fraction of incident light that is attenuated due to extinction (scattering and/or absorption) by the particles

3.1.11
optical model
theoretical model used for computing the model matrix for optically homogeneous spheres with, if necessary, a specified complex refractive index, e.g. Fraunhofer diffraction, anomalous diffraction, Mie scattering

3.1.12
reflection
return of radiation by a surface, without change in wavelength

3.1.13
refraction
change of the direction of propagation of light determined by change in the velocity of propagation in passing from one medium to another; in accordance with Snell's law

$$n_m \sin \Theta_m = n_p \sin \Theta_p$$

3.1.14
scattering
general term describing the change in propagation of light at the interface of two media

3.1.15
scattering pattern
angular or spatial pattern of light intensities [$I(\theta)$ and $I(r)$ respectively] originating from scattering, or the related energy values taking into account the sensitivity and the geometry of the detector elements

3.1.16
single scattering
scattering whereby the contribution of a single member of a particle population to the scattering pattern of the entire population is independent of the other members of the population

3.1.17
width of normal size distribution
standard deviation (absolute value) or coefficient of variation (relative percentage) of the size distribution

NOTE For normal distributions about 95 % of the population falls within ± 2 standard deviations from the mean value and about 99,7 % within ± 3 standard deviations from the mean value.

3.2 Symbols

c	volumetric particulate concentration, %
f	focal length of lens, mm
$I(\theta)$	angular intensity distribution of light scattered by particles (scattering pattern)
$I(r)$	spatial intensity distribution of light scattered by particles on the detector elements (measured scattering pattern by detector)
i	indication for imaginary part of refractive index
i_n	photocurrent of detector element n, µA
k	wave number: $2\pi/\lambda$
k_p	imaginary (absorption) part of particle's refractive index
l	illuminated path length containing particles, mm
L	vector of photocurrents (i_1, i_2,..., i_n)
m	relative, complex refractive index of particle to medium
n_m	real part of refractive index of medium
n_p	real part of refractive index of particle
N_p	complex refractive index of a particle
r	radial distance from focal point in focal plane, µm
v	velocity of particles in dry disperser
x	particle diameter, µm
x_{50}	median particle diameter, µm; here used on a volumetric basis, i.e. 50 % by volume of the particles is smaller than this diameter and 50 % is larger
x_{10}	particle diameter corresponding to 10 % of the cumulative undersize distribution (here by volume), µm

x_{90} particle diameter corresponding to 90 % of the cumulative undersize distribution (here by volume), µm

α dimensionless size parameter: $\pi x/\lambda$

θ scattering angle with respect to forward direction

θ_m angle with respect to perpendicular at boundary for a light beam in medium (as used in Snell's law; see refraction)

θ_p angle with respect to perpendicular at boundary for a light beam in particle (as used in Snell's law; see refraction)

λ wavelength of illuminating light source in medium (i.e. liquid or gas/air), nm

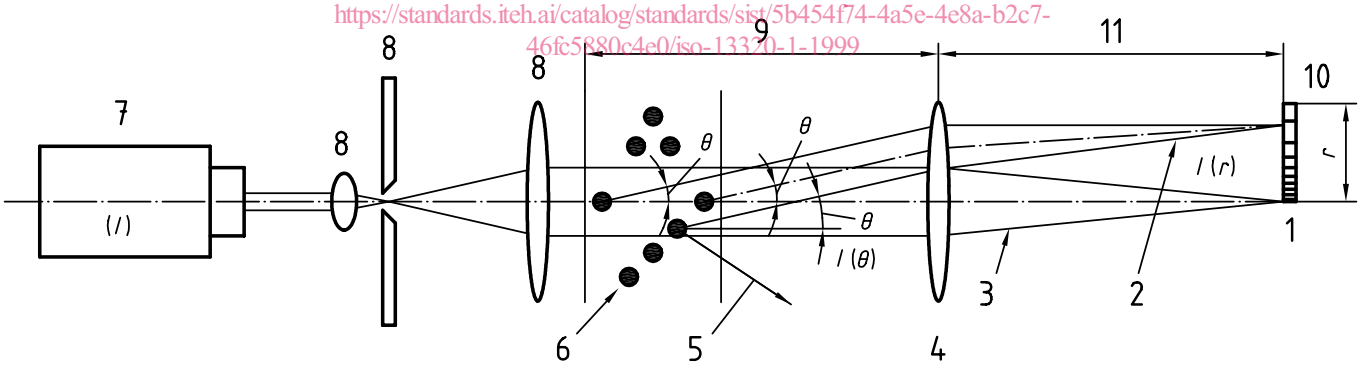
ω rotational velocity of particles in dry disperser

4 Principle

A representative sample, dispersed at an adequate concentration in a suitable liquid or gas, is passed through the beam of a monochromatic light source, usually a laser. The light scattered by the particles at various angles is measured by a multi-element detector and numerical values relating to the scattering pattern are then recorded for subsequent analysis. These numerical scattering values are then transformed, using an appropriate optical model and mathematical procedure, to yield the proportion of total volume to a discrete number of size classes forming a volumetric particle size distribution.

5 Laser diffraction instrument

A typical set-up for a laser diffraction instrument is given in figure 2.



Key

1 Obscuration detector
2 Scattered beam
3 Direct beam
4 Fourier lens
5 Scattered light not collected by lens 4
6 Particle ensemble
7 Light source laser
8 Beam processing unit
9 Working distance of lens 4
10 Multi-element detector
11 Focal distance of lens 4

Figure 2 — Example of the set-up of a laser diffraction instrument

In the conventional set-up, a light source (typically a laser) is used to generate a monochromatic, coherent, parallel beam. This is followed by a beam processing unit, usually a beam expander with integrated filter, producing an extended and nearly ideal beam to illuminate the dispersed particles.

A representative sample, dispersed at an adequate concentration is passed through the light beam in a measuring zone by a transporting medium (gas or liquid); this measuring zone should be within the working distance of the lens used. Sometimes, the particle stream in a process is illuminated directly by the laser beam for measurement, as in the case of sprays, aerosols and air bubbles in liquids. In other cases (such as emulsions, pastes and powders), representative samples can be dispersed in suitable liquids (see annex C). Often dispersants (wetting agents; stabilizers) and/or mechanical forces (agitation; ultrasonication) are applied for deagglomeration of particles and stabilization of the dispersion. For these liquid dispersions a recirculating system is most commonly used, consisting of an optical measuring cell, a dispersion bath usually equipped with stirrer and ultrasonic elements, a pump and tubing.

Dry powders can also be converted into aerosols through application of dry powder dispersers, which apply mechanical forces for deagglomeration. Here a dosing device feeds the disperser with a constant mass flow of sample. The disperser uses the energy of a compressed gas or the differential pressure to a vacuum to disperse the particles. It outputs an aerosol that is blown through the measuring zone, usually into the inlet of a vacuum pipe that collects the particles.

There are two positions in which the particles can enter the laser beam. In the conventional case the particles enter the parallel beam before and within the working distance of the collecting lens [see Figure 3 a)]. In the so-called reversed Fourier optics case the particles are entered behind the collecting lens and, thus, in a converging beam [see Figure 3 b)].

The advantage of the conventional set-up is that a reasonable path length for the sample is allowed within the working distance of the lens. The second set-up allows only small path lengths but enables measurement of scattered light at larger angles, which is useful when submicrometre particles are present.

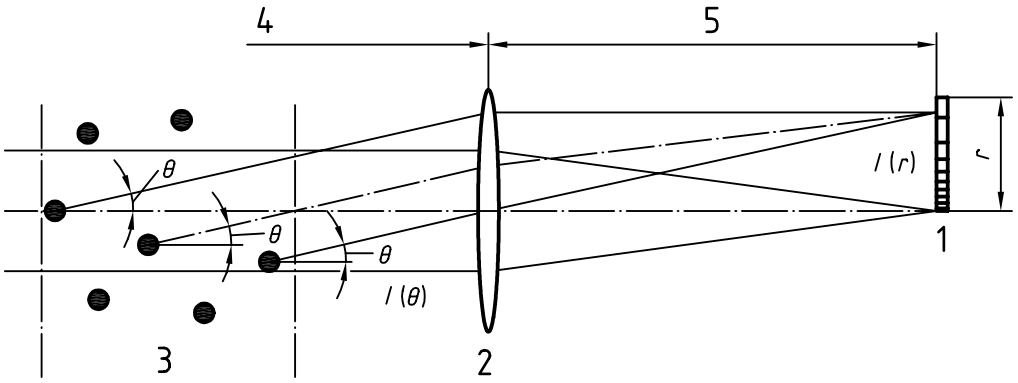
The interaction of the incident light beam and the ensemble of dispersed particles results in a scattering pattern with different light intensities at various angles (see annex A for theoretical background of laser diffraction). The total angular intensity distribution $I(\theta)$, consisting of both direct and scattered light, is then focused by a positive lens or an ensemble of lenses onto a multi-element detector. The lens(es) provide(s) for a scattering pattern which, within limits, is not dependent upon the location of the particles in the light beam. So, the continuous angular intensity distribution $I(\theta)$ is converted into a discrete spatial intensity distribution $I(r)$ on a set of detector elements.

It is assumed that the recorded scattering pattern of the particle ensemble is identical to the sum of the patterns from all individual single scattering particles presented in random relative positions. Note that only a limited angular range of scattered light is collected by the lens(es) and, thus, by the detector.

The detector generally consists of a number of photodiodes; some instruments apply one photodiode in combination with moving slits. The photodiodes convert the spatial intensity distribution $I(r)$ into a set of photocurrents i_n. Subsequent electronics then convert and digitize the photocurrents into a set of intensity or energy vectors L_n, representing the scattering pattern. A central element measures the intensity of the non-scattered light and, thus, with a calculation, provides a measure of optical concentration or obscuration. Some instruments provide special geometries of the central element in order to automatically re-centre or re-focus the detector by moving the detector or the lens. It is desirable that the detector elements are positioned so as to prevent the light reflected from the surface from re-traversing the optical system.

A computer controls the measurement and is used for storage and manipulation of the detected signals, for storage and/or calculation of a proper form of the optical model (usually as a model matrix containing light scattering vectors per unit of volume per size class, scaled to the detector's geometry and sensitivity) and calculation of the particle size distribution (see annex A for theoretical background of laser diffraction). Also it may provide automated instrument operation.

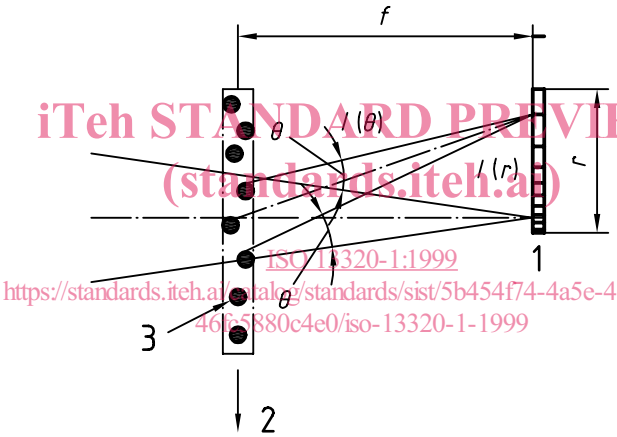
Several significant differences exist, both in hardware and software, not only between instruments from different manufacturers but also between different types from one company. The instrument specifications should give adequate information for proper judgement of these differences. In annex B recommendations are presented for the specifications of laser diffraction instruments.



Key

1 Detector
2 Fourier lens
3 Particle ensemble
4 Working distance
5 Focal distance

a) Conventional set-up: particles are in parallel beam before and within working distance of lens



Key

1 Detector
2 Flow through cuvette
3 Particle

b) Reverse Fourier set-up: particles are in converging beam between lens and detector

Figure 3 — Set-ups of laser diffraction instruments

6 Operational procedures

6.1 Requirements

6.1.1 Instrument location

The instrument should be located in a clean environment that is free from excessive electrical noise, mechanical vibration, and temperature fluctuations and is out of direct sunlight. The operating area should be well ventilated. The instrument should either contain a rigid internal optical bench or be installed on a rigid table or bench to avoid realignment of the optical system at frequent intervals.

WARNING — The radiation of instruments equipped with a low power laser can cause permanent eye damage. Never look into the direct path of the laser beam or its reflections. Avoid cutting the laser beam with reflecting surfaces. Observe the local laser radiation safety regulations.

6.1.2 Dispersion liquids

Any optically transparent liquid of known refractive index may be used. Thus, a variety of liquids is available for preparation of liquid dispersions of powders. Annex C provides requirements for the dispersion liquids.

If an organic liquid is used for dispersion, observe the local health and safety regulations. Use a cover for the ultrasonic bath when using liquids with a high vapour pressure in order to prevent the formation of hazardous vapour concentrations above the bath and/or the generation of low-temperature zones with fluctuating refractive indices in the fluid by evaporation.

6.1.3 Dispersion gases

For dry dispersion and spray applications a compressed gas is sometimes used. If used, it is essential that it is free from oil, water and particles. To achieve this, a dryer with a filter is required. Any vacuum unit should be located apart from the measurement zone, so that the output of the hot air does not reach the measuring zone. Draught should be avoided in order to avoid unstable particulate streams.

6.2 Sample inspection, preparation, dispersion and concentration

6.2.1 Sample inspection

Inspect the material to be analysed, visually or with the aid of a microscope, firstly to estimate its size range and particle shape and later to check whether the particles have been dispersed adequately.

The size distribution measured in a sample is only valid for a batch of material if the sample is representative for that batch and has been dispersed adequately.

6.2.2 Preparation

For dry powders, prepare a representative sample of suitable volume for the measurement by an adequate sample splitting technique, for instance a rotating riffler. If very small samples are required, or in the case of wet powders, it is also possible to take fractional samples out of a well-mixed sample paste. The consistency of the paste then avoids segregation errors. The pastes are formed by adding dispersant to the sample drop by drop while mixing it with a spatula. As long as the mixture forms lumps, single drops should be added while continuing the mixing after each drop. A good consistency for the paste is one like honey or toothpaste. If the paste becomes too fluid by mistake, it shall not be used, and a new preparation should be initiated.

If the maximum size exceeds the measuring range, remove the material that is too coarse, e.g. by presieving. In this case determine and report the amount/percentage removed.

Sprays, aerosols and gas bubbles in liquid should be measured directly, provided that their concentration is at an adequate level (see 6.2.3 and 6.2.4), since sampling or dilution is generally impossible without altering the particle size distribution.

6.2.3 Dispersion

6.2.3.1 Dry powders can be dispersed either in air or in liquid. The dispersion procedure shall be adjusted to the purpose of the measurement, e.g. it has to be decided whether agglomerates should be detected or broken down to the primary particles.

6.2.3.2 An adequate dry disperser should be applied; here, generally compressed air or vacuum is applied for dispersion by shear stress with the assistance of mechanical de-agglomeration by particle-particle or particle-wall collisions (see figure 4). For dry dispersion, the complete fractional sample shall be used for the measurement. Note that the use of large sample quantities can overcome the poor statistical representation of coarse particles in a wide size distribution. It is necessary to check that comminution of the particles does not occur and conversely that a good dispersion has been achieved. This is usually done by direct comparison of dry dispersion with a liquid one: ideally, the results should be the same. Another possibility for checking the degree of dispersion or comminution is by changing the dispersing energy (e.g. the primary air pressure) and monitoring the change of the size distribution. Usually upon increasing the dispersing energy the amount of fines is increased at first, due to improved dispersion, until a plateau is reached, where the size distribution is nearly constant with increasing energy. At still higher energies the amount of fines may rise again as a result of comminution. On some occasions, agglomeration has