Stream scanning methods of particle size measurement

9.1 Introduction

It is convenient to divide particle size measurement techniques involving the interaction between particles and an external field into two categories, stream scanning and field scanning. In the former, particles are examined one at a time and their interaction is taken as a measure of their size. In the latter, the interaction of an assembly of particles is interpreted in terms of the size distribution of the assembly.

The most widely used stream scanning technique employs the Coulter principle (Figure 9.1a) where the interrogating field is electrical and particle size (volume) is proportional to the change in electrical impedance as the particles pass through the field.

Particle projected area can be measured by the amount of light cut off as a particle passes through a light beam (Figure 9.1b). With a small diameter rotating or scanning beam the pulse length is a measure of a random chord length (Figure 9.1c). Scanning beams, in concert with back-scattering detectors, are also used for chord size determination (Figure 9.1g). These can be used with high concentration slurries since the beam does not have to traverse the suspension. If the incident beam is absorbed in a light trap the light scattered in the forward direction (Figure 9.1d) or at right angles (Figure 9.1e) is size dependent. The signal is greatly enhanced with the aid of an elliptical mirror (Figure 9.1f).

Interferometers (Figure 9.1h) determine particle size through the phase shift between a split laser beam, one passing through the particle and the other through the surrounding liquid. If particles are accelerated through a
Fig. 9.1 Principles of streaming systems.
nozzle, the time takes them to pass through two laser beams is a measure of aerodynamic size, (Figure 9.1i). In the phase Doppler method (Figure 9.1j) particle size is determined from the interference pattern as a particle passes through the intersection of two laser beams.

Stream scanning is generally limited to low-concentration suspensions, and is best suited to the determination of particle size distribution by number and contamination monitoring. The Hiac/Royco light blockage method is allowed in USP XXI (1985) for the evaluation of particle burden in large volume dextrose solutions. The United States Pharmaceutical method, USP XXII (1988), for determining particulates in injectable solutions is based on microscopy and suffers several disadvantages [1].

Conversion of number distribution to mass (volume) distribution can result in gross errors unless the width of the distribution is narrow. For example, if the range is 10:1 the omission of a single 10-unit particle (volume=1000) is equivalent to the omission of 1000 one-unit particles. In order to obtain accurate volume distribution data it may therefore be necessary to size millions of small particles in order to get a statistically acceptable count at the coarse end of the distribution.

9.2 The electrical sensing zone method (the Coulter principle)

9.2.1 Introduction

The Coulter technique is a method of determining the number and size distribution of particles suspended in an electrolyte by causing them to pass through a small orifice on either size of which is immersed an electrode. The changes in electrical impedance as particles pass through the orifice generate pulses whose amplitudes are proportional to the volumes of the particles. The pulses are fed to a pulse height analyzer where they are scaled and counted and, from the derived data, the size distribution of the suspended phase is determined.

The Coulter principle was patented in 1949 [2] and described in 1956 [3] as a method for counting and sizing blood cells. Kubitschek [4,5] introduced modifications which permitted the counting of bacterial cells, and pointed out that the method could be applied to the measurement of cell volumes as well as number counting. Modified instruments were soon developed with which particles could be sized as well as counted. In 1998 the company was acquired by Beckman and renamed Beckman Coulter.

Since analyses may be carried out rapidly and with good reproducibility using semi-skilled operators, the method has become very popular in a
wide range of industries. In a recent Coulter bibliography 1432 industrial references and 436 pharmaceutical references are cited [6]. The manufacturers claim more than 6000 documented references using the Coulter principle.

This type of counting device is designated in ASTM 3365-74T as a ‘Tentative method of test for concentration and size distribution of airborne particulates collected in liquid media’. It is specified that the method is suitable for particulate matter greater than 0.6 µm in diameter collected by a Greenburg-Smith or midget impinger. The original development of the method was carried out by Anderson et. al. [7] and was extended by others.

The Coulter principle is also standard for dry toners [8,9] and an accepted method for aluminum oxide powder [10], chromatography media [11], polymeric powders [12], plutonium [13], filter evaluation [14], catalytic material [15] and comparing particle size distribution using alternative types of particle counters [16]. In ASTM method C-21 it states that the experience of several laboratories indicates that the method is capable of a repeatability of 1% and a reproducibility of 3% at the 95% confidence level. Operating procedures for this technique are also covered in BS3405 [17]. The method is also the subject of an international standard [18].

9.2.2 Operating principle

The operating principle of the instrument may be followed by referring to Figure 9.2. The sample to be analyzed is dispersed in an electrolyte that is placed in a beaker. The glass sample tube, on either side of which there is a platinum electrode, is immersed in the electrolyte. A controlled vacuum initiates flow of suspension through a sapphire orifice let into the glass tube and unbalances a mercury siphon.

The instrument can then operate in one of two modes:

1 The system may be isolated from the vacuum source by closing tap A and flow continues due to the balancing action of the mercury siphon. The advancing column of mercury activates a counter by means of start and stop probes placed so that a count is carried out whilst a known volume of electrolyte (0.05 ml, 0.50 ml, 2.0 ml) passes through the aperture.
Data may be acquired in a preset count mode in which counting is initiated manually and ends when the preset count number (up to 1 million) has been reached.

In the latest versions of the instrument the mercury column is replaced by a small vacuum pump.

A current is passed through the aperture between the two electrodes. Particles drawn through the aperture cause momentary changes in the electrical impedance due to each particle displacing its own volume of electrolyte within the aperture itself. These changes in impedance are detected and presented as voltage pulses, the heights of which are proportional to the volumes of electrolyte displaced by the particles, and hence to the volumes of the particles themselves, provided they are neither unduly plate-like nor porous. The particle-generated pulses are amplified, sized and counted and, from the derived data, the particle size distribution is determined.

Particle size analysis may be performed in the overall size range of 0.4 to 1200 μm. To achieve this, a number of different sensors (aperture tubes) are required. The operating range of each sensor is from around 2% to 60% of the aperture diameter, e.g. from 2 to 60 μm for a 100 μm aperture tube. Smaller particles generate pulses that are lost in electronic noise generated within the aperture and in the electronic circuitry. Larger particles give increasingly non-linear response and tend to block the aperture if they are greater than about half the aperture diameter. For
powders with a wider size range, an extrapolation or a two-tube technique may be necessary. Alternatively the Coulter analysis can be combined with some other technique, e.g. the coarse end of the distribution can be analyzed by sieving and the two analyses combined after correction for shape factor. Care should be taken with powders having a wide size range since the uncounted fraction may form a substantial part of the distribution.

9.2.3 Theory for the electrical sensing zone method

The basic assumption underlying the Coulter principle is that the voltage pulse generated when a particle passes through the aperture is directly proportional to particle volume. The relationship between particle size and instrument response may be determined using a simplified theory. Figure 9.3a shows a particle passing through the aperture and Figure 9.3b shows an element of the particle and aperture. The resistance of an element without a particle, $\delta R_0$, is:

$$\delta R_0 = \frac{\rho_f \delta L}{a}$$

The resistance of an element with a particle included, $\delta R$, is that of two resistors $\delta R_1$, $\delta R_2$ in parallel

$$\frac{1}{\delta R} = \frac{1}{\delta R_1} + \frac{1}{\delta R_2}$$
so that:

\[
\delta R = \left( \frac{A - a}{\rho_f \delta L} + \frac{a}{\rho_s \delta L} \right)^{-1}
\]  \hspace{1cm} (9.1)

\(\rho_f, \rho_s\) are the resistivities of the fluid and particle respectively, \(A\) is the cross-sectional area of the orifice, \(a\) is the cross-sectional area of the particle and \(V\) is the volume of the particle.

Thus the change in resistance of the element, \(\delta(\Delta R)\), due to the presence of the particle is given by:

\[
\delta(\Delta R) = \delta R_0 - \delta R
\]

\[
\delta(\Delta R) = -\frac{\rho_f a \delta L}{A^2} \left( 1 - \frac{\rho_f}{\rho_s} \right) \left[ 1 - \left( 1 - \frac{\rho_f}{\rho_s} \frac{a}{A} \right)^{-1} \right]
\]  \hspace{1cm} (9.2)

The external resistance of the circuit is sufficiently high to ensure that a small change, \(\Delta R\), in the resistance of the aperture due to the presence of a particle will not affect the current \(I\); the voltage pulse generated is therefore \(I\Delta R\).

In practice, it is found that the response is independent of the resistivity of the particle. If this were not so, the whole technique would break down since a different calibration factor would be required for each electrolyte-solid system. This independency is attributed to oxide surface films and ionic inertia of the Helmholtz electrical double layer and associated solvent molecules at the surface of the particles, their electrical resistance becoming infinite [19]. The terms involving \((\rho_f/\rho_s)\) may therefore be neglected and the preceding equation becomes:

\[
\delta(\Delta R) = -\frac{\rho_f a \delta L}{A^2 \left( 1 - \frac{a}{A} \right)}
\]  \hspace{1cm} (9.3)

The response therefore, is not proportional to the volume of the particle, but is modified by the \(a/A\) term. For rod-shaped particles whose length is smaller than the aperture length, this leads to an oversizing of about 6% in
terms of the diameter at the upper limit of each aperture with distortion of the measured size distribution [20,21]. This error decreases as $a/A$ decreases.

For spherical particles of diameter $d = 2b$ the change in resistance due to an element of thickness $\delta l$ at a distance $l$ from the center of the sphere may be determined and this can be integrated to give the resistance change due to the particle [22,23].

$$\Delta R = -\frac{2\pi^2 \rho_f}{A} \int_0^b \left[ \frac{\left( b^2 - l^2 \right)}{1 - \pi \left( b^2 - l^2 \right) / A} \right] dl$$

$$\Delta R = -\frac{8 \rho_f d^3}{3\pi D^4} \left[ 1 + \frac{4}{5} \left( \frac{d}{D} \right)^2 + \frac{24}{35} \left( \frac{d}{D} \right)^4 + \frac{169}{280} \left( \frac{d}{D} \right)^6 + \ldots \right] \quad (9.4)$$

This gives a limiting value of two-thirds the Maxwellian value. Recognizing this, Gregg and Steidley multiplied their solution by three halves. This procedure has been questioned [22]. The complete solution is:

$$\Delta R = -\frac{A \rho_f}{\pi D \left[ \sin^{-1} \left( \frac{d}{D} \right) - \frac{d}{D} \right]} \quad (9.5)$$

This equation may be written:

$$\Delta R = -\frac{\rho_f V}{A^2} F_r \quad (9.6)$$

Hence, the instrument response is proportional to the volume of the sphere, $V$, modified by the function $F_r$. This equation results from a simple integration of the area available for conduction. Several approximations have been derived for $F_r$ [22,24-26] and these have been compared to find the one that agrees best with experiment [27].

The initial experimental data by de Blois and Bean were with PVC spheres, down to 0.09 $\mu$m in diameter, using a pore in a plastic sheet for an aperture. They concluded that the technique was applicable down to
0.015 μm. In a later paper [28] they reported measurements down to a diameter of 0.06 μm, using a pore in a Nuclepore filter, and they also measured the osmotic velocity of the liquid in the pore.

The error in assuming a linear relationship between resistivity change and particle volume for spherical particles is about 5.5% for \((d/D) = 0.40\). The principle may therefore be applied to higher ratios of \((d/D)\) than 0.40 provided corrections are applied and aperture blockage does not become too troublesome. For non-spherical particles, \(F\) is modified by the inclusion of a shape factor [29]. In general, as the ratio of \((d/D)\) increases, the resistance pulse generated is greater than predicted by assuming proportionality and oversizing of the larger particles occurs.

9.2.4 Effect of particle shape and orientation

It has been claimed that particle shape, roughness and the nature of the material has little effect on the analysis [30] but there is considerable evidence that the size measured is the envelope of the particle. Comparison with other techniques gives good agreement for homogeneous spherical particles; for non-spherical particles results may differ [31,32]. For porous particles the measured volume may be several times the skeletal volume, and the apparent volume for flocs is greater than the volume of the particles that make up the flocs [20].

Since flaky particles rotate as they pass through the sensor, the measured volume may be the volume swept out by the particle and this can lead to oversizing. With extreme shapes such as rods, this may cause a change in size distribution, as apertures of different sizes are used, if the whole of the rod cannot be accommodated in the sensing zone. It has been reported that silica containing large pores can be undersized by as much as 100% due to pore filling by the electrolyte [33]. This particular effect has been used to measure the amount of particulate material within a floc [34] and the porosity of porous samples [35]. Anomalous results have also been reported for fly ash [21]. Ratios of 1.31:1 have been reported for non-extreme shapes with higher values for flaky particles [36]. For these reasons, it may be worthwhile to calibrate the analyzers with the test material or carry out a mass balance routinely as recommended in BS3406 Part 5.

Model experiments have been carried out by Marshall [37], Lloyd et al. [38] and Eckhoff [39] but no firm conclusions can be drawn from them since the models used differ widely from the commercial instruments. This work was extended by Harfield et al. [40,41] using a large two-
dimensional model and they obtained a linear response for particles of diameter up to 77% of orifice diameter.

Kubitscheck [42] found that the output pulse due to the passage of a particle through the aperture was round topped not rectangular as expected theoretically (the pulse height should remain constant as long as the particle is in the aperture). This implies that the amplifier does not reach full response to the presence of each particle before it leaves the aperture and this can result in an undersizing of coarse particles. Kubitscheck found it necessary to construct apertures five times as long as they were wide, as opposed to the conventional 0.75 ratio, in order to produce flat-topped pulses. Eckhoff [39] stated that the pulses were round-topped due to the shape of the electric field and rejected the assertion that round-topped pulses were produced due to the instrument not reaching full response.

Pulses deviate from the ideal, single modal shape when they are generated from coincident particles. These pulses take on a variety of shapes always of longer duration and often having multiple peaks.

![Diagram of pulse shapes](image)

Fig. 9.4 Shape of pulses generated by particles not passing centrally through the aperture
9.2.5 Pulse shape

Grover et al. [26] conducted experimental studies of the pulses generated in the aperture and determined that the potential field was dense at the inlet and outlet edges. Particles traveling parallel to and near the walls pass regions of high potential gradient and generate M-shaped pulses. This does not affect the analysis of powders having a wide range of particle size but grossly skews narrow distributions to generate too coarse an analysis.

Thom et al. [43] used a magnified version of the Coulter Counter and, by drawing spheres through the aperture on nylon threads, mapped out the generated pulses on different streamlines (Figure 9.4). Pulse distortion was eliminated when the edges of the orifice were rounded to form a conical entrance and exit. They also made a central filament capillary for the injection of particles into the center of the orifice to eliminate pulse distortion. They called this set-up 'hydrodynamic focusing' (Figure 9.5). A secondary benefit of this arrangement is that the transit time is approximately constant for all particles. This work resulted in the development of an instrument available commercially as the Telefunken particle detector MS PD1 1105/1, which was reported on by Polke [44]. In

Fig. 9.5 Hydrodynamic focusing. Suspension is focused on the axis of the measurement opening M with a probe D. The suspension streams through the axis after dilution with particle free electrolyte E.
1973, Coulter purchased the rights to manufacture this instrument that they made available as the model TF Coulter counter.

In 1969 Coulter's patented a conical entrance and exit but in 1970 they filed another patent which indicated a preference for cylindrical apertures over the contoured variety, possibly because of plugging difficulties. They also considered channeling the particles down the center of the aperture but rejected this approach as impractical. In 1973 they were issued a patent for a rounded orifice.

In 1973 IITRI patented a trumpet-shaped orifice that included a flow straightener. Orifice plugging was eliminated by the use of a screen in the flow straightener [36,45,46]. The use of a flow directional collar protected by a micromesh sieve whose openings are equal to 40% of the orifice diameter has two advantages: By protecting the orifice it permits the use of a multi-tube system to operate in the same suspension thus eliminating the need for the conventional two-tube analysis method for wide-ranging powders. It also permits the detection and measurement of particles having different aspect ratios; this is particularly useful for the detection of fibers in the presence of other shapes.

Two types of pulse discrimination have been described; one type rejects all pulses having a rise time greater than a preset minimum; the other type accepts only those pulses generated by particles passing through the aperture on nearly central paths. A partial correction is to use an 'edit' switch so that grossly distorted pulses are not counted, but this can lead to the rejection of considerable information and is not necessarily free of bias. A better solution is to reshape the pulses electrically so as to provide information from them all. A full correction is to force the particles into a central streamline using 'hydrodynamic focusing' [49] but this complicates both the experimental set up and the analytical procedure. One effect of eliminating distorted pulses is the production of a narrower size distribution. Another is that a mass balance calibration is not possible due to count loss. The rejected pulses are oversize thus their inclusion tends to skew resulting size distribution to the right.

Elkington and Wilson [48] examined narrow size distributions of particles and resolved an additional 'artifact' peak, on the coarse side of the main or normal distribution, which was generated by particles moving non-axially through the aperture. They used a Coulter ZB, a Coulter Channelyzer C100 with 'edit' on and 'edit' off and a Coulter TF.

A comparison between the TF system, the 'edit' system and the standard system has been given by Lines [49], who also discusses the effect of using a long aperture tube.
A later development was the use of four electrodes in a tube rather than two; the outer two electrodes are used for current injection and the inner two yield voltage measurements [50]. It is claimed that this arrangement reduces errors of false counts and oversizing. The signal shape, together with signal peak, gives particle size and shape characteristics. Theoretical modeling and experiment showed that aspect ratio along with particle diameter can be measured for example for cylindrical particles.

9.2.6 Effect of coincidence

Two types of error arise due to more than one particle being in the measurement zone at any one time.

Primary Coincidence. Two or more particles in the measurement zone give rise to two or more overlapping pulses. Depending on their proximity and electrical resolution, these pulses may not be resolved, leading to loss of count.

Secondary Coincidence Two or more very close particles, which individually give rise to pulses below the threshold level, may collectively generate a pulse above the level. Thus if the concentration is too high, oversize particles begin to appear.

A coincidence correction can be applied for primary coincidence but it is preferable to use a concentration so low that this effect is negligible. Under this condition, secondary coincidence errors are also reduced.

The correction for primary coincidence is derived through Poisson probabilities of finding multiple particles in different parts of the sensing zone simultaneously. This yields the following relationship between true count \( N \) and observed count \( n \):

\[
n = \frac{v}{s} \left[ 1 - \exp\left( -\frac{s}{v} N \right) \right]
\]

If higher-order terms are neglected this gives:

\[
N = n + \left( \frac{s}{2v} \right) N^2
\]

\( v \) is the monitored volume for each count and \( s \) is the sensing zone volume which is slightly larger than the volume of the aperture.
The equation developed on the basis of transit time distribution, takes the form:

\[ n = N \exp \left( -\frac{s}{v} N \right) \]  

(9.9)

giving:

\[ N = n + \frac{s}{v} N^2 \]  

(9.10)

Note, these equations are of the form

\[ N = n + pN^2 \]  

(9.11)

The manufacturers propose the more convenient empirical equation:

\[ N = n + p n^2 \]  

(9.12)

\[ p = 2.5 \left( \frac{D}{100} \right)^3 \left( \frac{500}{v} \right) \times 10^{-6} \]  

(9.13)

\[ D \] is the orifice diameter in micrometers and \( v \) the volume of the suspension in microliters monitored for each count. The factor 2.5 was determined experimentally using a 100 \( \mu \)m diameter aperture tube 75 \( \mu \)m long with \( v = 500 \mu l \). If it is assumed that the sensing zone comprises the volume of the aperture plus a hemisphere at the entrance and exit, \( s = 1.11D^3 \) so that \( s/v = 2.22 \times 10^{-6} \) which is not greatly different to the value of \( 2.5 \times 10^{-6} \) found experimentally.

9.2.7 Multiple aperture method for powders having a wide size range

(a) General

If the size range of the powder is too wide to be covered by a single orifice, two or more aperture tubes can be used. As a general rule, if there is more than 2% of the distribution in the smallest size interval it is advisable to use a smaller orifice to determine the fine end of the distribution.
An aperture tube should first be selected to give zero count in the top size channel and a smaller aperture tube should then be selected to size the fine end of the distribution. The ratio of the orifice diameters should be less than 5:1 to ensure that the overlapping counts will match. An analysis is first carried out with the large aperture tube using a known volume of electrolyte: In order to facilitate calculation of dilution factors it is recommended that the weight of the beaker and the suspension be determined prior to the analysis. For an instrument that can provide a number count, the dilution factor can be calculated by counting the diluted suspension using the large aperture tube at two size settings at around one third of the orifice size. The ratio of this count, corrected for coincidence, to the corrected count on the original suspension, is the dilution factor.

(b) Sieving technique

The weight of the beaker and suspension is determined and the suspension is then poured through a micromesh sieve with openings approximately half the diameter of the orifice of the smaller aperture tube. The filtrate is collected in a clean, weighed beaker. The suspension beaker is next rinsed with clean electrolyte and the rinsings poured through the sieve and collected. The dilution factor is calculated from the weights of the original and final suspensions.

(c) Sedimentation technique

An analysis is carried out using the larger aperture tube as before. Using Stokes’ equation, the time is calculated for particles coarser than the upper size limit for the smaller aperture tube to settle below the orifice. After this time has elapsed, an analysis is carried out with the smaller aperture tube, after diluting if necessary. With multichannel models it is necessary to calibrate the two aperture tubes so that the channel size levels coincide. The two sets of data should coincide in the overlap region to generate the combined analysis. An application of multiple aperture technique, using wet sieving to remove oversize, has been described for fly ash in the size range 1 to 200 μm [51].

9.2.8 Calibration

Calibration is usually effected with the use of narrowly classified (monosize) spherical latex particles. Since the calibration particles are nearly monosize the pulses on the oscilloscope will be nearly uniform in
height. Rapid calibration may be made by observing the threshold level, \( t_c \), required to screen out the single height pulses displayed on the oscilloscope and to give a count \( n_c \). More accurately, a full count, \( n_f \), is taken at a visually determined threshold level of \( t_f = 0.5t_c \) and an oversize count, \( n_0 \), at threshold setting \( t_0 = 1.5t_c \). The threshold setting, to give a count equal to half the difference between these two counts \( n_c = 0.5(n_f - n_0) \) is used for calibration purposes.

An alternative procedure is to plot the number count against the instrument response and differentiate this to find the mode which is assumed to occur at \( t_c \). Calibration may also be made using powders under test provided the size range remains within the range of a single aperture tube. This procedure is a primary calibration procedure and is recommended in a recent standard as being superior to latex calibration [17,20,21,52-54]. This procedure cannot be used with the Coulter Multisizer due to count loss. One paper reported a count loss of over 30% which generated a mass loss of over 15%, which implies that the loss was preferentially of fines [55]. In a comparison between the TAII and Multisizer II weight percentage errors of ±10% were reported [56].

The volume of particles in a metered volume of suspension is:

\[
v_p = \frac{vw}{V_s \rho_s}
\]

where: \( v \) = metered suspension volume, \( V_s \) = total suspension volume, \( w \) = total weight of the powder and \( \rho_s \) = particle density.

If \( \bar{t} \) is the average threshold setting as the pulse count changes by \( \Delta n \) then:

\[
v_p = \frac{\pi}{6} k^3 \sum n \bar{t}
\]

Hence:

\[
k^3 = \frac{6 v_p}{\pi \sum n \bar{t}}
\]

\[
k^3 = \frac{6 v_p w}{\pi V_s \rho_s \sum n \bar{t}}
\]

(9.15)
If the calibration constant as determined by equation (9.15) is significantly smaller from that using monosize particles it is likely that the whole range of powder has not been examined. In that case, equation (9.15) may be used to determine the fraction undersize by comparing the experimental value of $\sum \Delta n \bar{T}$ with the expected value.

If it is significantly larger, it is possible that the particles are porous and the envelope sizes are being measured, or that the particles are flocculating. A third possibility is that the assumed powder density is incorrect.

Calibration materials in general use consist of pollens, latex spheres and glass spheres. Several investigators have discouraged the use of pollens due to non-sphericity, surface irregularities and changing size in dispersing media [57-59]. Various pollens, lattices and glass beads are available from Coulter, Dow, Duke Standards and CTI-TNO [60]. Alliet [61] describes a red bead latex available from Coulter UK, with a mean size of 18.99 $\mu$m and a standard deviation of 0.18 $\mu$m, as being a particularly promising latex. It is generally agreed that the standard deviations of the size distributions of the Dow latex particles measured by the Coulter counter are greater than those measured by microscopy and those quoted by Dow [62,63]. This may be due partly to the quality control methods used by Dow [36] and partly to the effects discussed in the section on pulse shape. Spherical hollow carbon particles (1-300) $\mu$m have also been used for calibration purposes [64].

9.2.9 Carrying out a mass balance

Although it is common practice to calibrate the Coulter Counter using a standard powder, it is possible to calibrate the instrument with the powder being examined. This is the preferred British Standard method [17]. It is reiterated that this procedure cannot be carried out with some instruments due to count loss. Essentially one balances the volume of particles passing through the measuring aperture with the known volume in the measurement sample. This serves a multiple purpose in that:

- It indicates if part of the distribution has been missed;
  (This occurs if the size range of the powder is greater than the detection range of the aperture. This is not always obvious from the appearance of the determined distribution. In particular, one mode of a bimodal distribution could easily be lost).
Particle dissolution or growth is detected;
- It checks on the accuracy of the calibration and exposes measurement errors.

This procedure should be used as routine since it indicates whether all the powder is accounted for and allows for correction for powder outside the measuring range of the aperture used. In an extreme case, it has been found that less than 5% of the total distribution was being measured and decisions were made based on these incorrect distributions. Alternatively, if the whole size range cannot be covered using a single aperture tube, a two-tube technique is required.

This is not possible if the fraction unaccounted for is below the limit of the technique and the alternative mass balance procedure, as used with BCR 66 standard quartz powder, is as follows.

Disperse $w$ gram of powder in $v_1$ cm$^3$ of liquid. Pipette out $v_2$ cm$^3$ of suspension and add it to electrolyte to make up $V_s$ cm$^3$. Determine the corrected Coulter count on a metered volume of $v$ cm$^3$.

9.2.10 Oversize counts on a mass basis using the Coulter Counter

In many powder additives, the presence of oversize particles results in faults in the finished product. A technique has been developed [65] for determining the number concentration of these oversize particles using the Coulter Counter Multisizer II.

The normal procedure for determining the number oversize is to carry out a mass balance and present the derived Coulter data graphically as counts/gram against particle size. The problem with using this procedure is that, in some cases, there are only one or two oversize particles in the presence of millions of smaller ones. It is therefore necessary to filter out many of the smaller particles and carry out a count on the oversize residue. This poses problems in that some oversize particles may be lost in the process and, even more likely, large contaminant particles may be introduced in the filtration process.

A precision transparent sieve with ±2% tolerance, introduced by Collimated Holes Inc., was tested as a simple alternative to straightforward Coulter counting and gave good, reproducible results when used in combination with a specially designed filter holder to reduce contamination. This sieve has an additional advantage in that it can be examined under a microscope to determine the nature of the oversize particles.
9.2.11 Apparatus

The original Coulter Counters have been replaced by Beckmann instruments although they are still widely used. These comprise the Coulter Counter model TAl1, Coulter Multisizer TAl1E, the Multisizer TAl1E + edit analyzer, the Coulter Counter Model D Industrial and the Coulter Counter Z series.

Beckman Coulter Z1 operates in the 1 to 120 μm size range and 1 to 60 μm using ampoule insertable aperture tubes. Metered volumes include 0.10 ml, 0.50 ml and 1.00 ml. The mercury manometer is replaced with an oil displacement pump.

Beckman Coulter Z2 uses the same technology as the Z1. It performs channelyzation of particle data into 256 channels while displaying size distribution data. The advanced user interface allows the operator to view the data in a variety of ways. Sample volumes as small as 10 ml can be handled using Accuvette II vials or less than 2 ml using ampule insertable aperture tubes.

Beckman Coulter Multisizer 3 is a new generation instrument introduced by the new owners of the company. The counter provides number, surface and volume distributions in the size range 0.4 to 1200 μm in a single run. The instrument is mercury free thus eliminating potentially hazardous spills. The calibration system is fully automated with this instrument and the parameters are stored for future reference. Advanced digital processing circuitry allows the user to increase the resolution by a factor of up to one hundred.

Malvern Sysmex SD-2000 particle counter and sizer delivers high-performance particle size analysis from 1 to 120 μm by combining electrozone sensing with hydrodynamic sheath flow focusing.

Malvern Sysmex CDA 500 employs a mercury-free vacuum pump and is designed for counting and sizing cells and particles in the size range 1 μm to 60 μm.

Micromeretics market a similar range of instruments covering a size range from 0.4 to 1200 μm, under the trade mark Elzone (previously marketed by Particle Data Inc.). Their product line is listed below with brief notes on their principal differences.

Micromeretics Elzone 5370 has one analysis station and is used primarily for particle and biocell counting and concentration analysis of particle-liquid systems. It reports particle count as a function of total liquid volume or total time. It can also provide fast low-resolution particle
sizing. It is fully self-contained featuring an internal microprocessor, control keypad and 18 cm diagonal video monitor.

*Micromeretics Elzone 5380* has one analysis station features complete sizing and counting capability and reports particle size distribution as a function of number, area or volume.

*Micromeretics Elzone 5382* has two analysis stations has complete sizing and counting capability. In addition it has automated valves that nearly eliminate the need for operator intervention after the start of an analysis.

Aperture tubes are available in 22 sizes ranging from 12 to 1900 μm in diameter. The orifice is drilled in a synthetic jewel that is permanently sealed into the wall of the tube. For diameters greater than 480 μm the holes are drilled through a ceramic insert. The effective range of particle sizes a typical orifice can cover is 3% to 70% of the diameter.

All models are available with a choice of sample dispersing options including propellor and magnetic stirrers and hydropulsers. Mercury manometers are available in eight sizes ranging from 10 to 5000 μL.

Micromeretics Elzone 5380 and 5382 are operated via a separate control module running in a Windows™ environment. Macro commands allow repetitive analyses to be scripted and executed with a few simple strokes. Collected data are presented in graphical and tabular form.

### 9.2.12 Limitations of the method

The primary limitation of this technique is the need to suspend the powder in an electrolyte. For powders insoluble in water a 0.9% saline solution is often used and dispersion effected using ultrasonics. The manufacturers also provide a list of electrolytes for use with water-soluble materials but these can cause cleaning difficulties.

The narrow size range covered using one aperture tube may necessitate the use of a two-tube procedure that can be onerous. For a powder having a wide size range, the large particles may cause troublesome aperture blockage with the smaller aperture. One procedure to cope with this is to sieve out the course particles prior to the analysis using the smaller aperture and the second procedure is to allow the coarse particles to sediment out below the aperture level. Particles, which do not pass along the axis of the cylindrical aperture, generate mis-shapen pulses that can greatly deform narrow size distributions. This effect can be reduced by editing out the mis-shapen pulses, or using a technique known as
hydrodynamic focusing, in which the particles are fed through the central streamline.

9.3 Fiber length analysis

This is of fundamental importance in the pulp and paper industry which uses a light obscuration method - the Kajaani FS-200. This instrument is also used in the chemical industry for measuring the length of man-made fibers.

The Advanced Fiber Information System (AFIS) [66] contains a mechanism for opening a hand fed ribbon of fibers so that individual fibers can be presented aerodynamically to an electro-optical system for measuring fiber length. Length measurements for 10,000 individual fibers can be obtained in 5 min.

The assumption made when the Coulter counter was first developed was that the variability in speed of the fibers, as they passed through the sensor, was small and could be assumed constant [67]. Analysis showed that this assumption was incorrect and a later instrument included a sensor to measure fiber speed [68]. Since the fibers are aligned with flow, the pulse length is a measure of fiber length and the pulse height is a measure of fiber width. The results showed excellent agreement with Suter-Webb measurements. The AFIS is widely used for cotton fiber measurement in the dry state.

Workplace exposure to asbestos fibers is usually assessed by personal sampling on a membrane filter that is subsequently examined using phase contrast or electron microscopy. Lilienfeld et. al. [69] described an instrument that monitors asbestos fiber concentration in sampled air by laser light scattering from fibers oscillating in phase with an electric field and this instrument has also been used to monitor silicon carbide dust [70]. Light scattering has also been used to detect fibers in air down to 1 μm [71] and magnetically aligned fibers on a membrane filter [72]. Rood [73] aligned fibers using a simplified Prodi instrument, then passed them through a corona discharge so that a downstream precipitator deposits the fibers on to a removable glass slide where they retain their alignment. He then used the difference in light scattered parallel to and perpendicular to flow in order to determine fiber length distribution.

The Coulter principle has also been applied to fiber length determination [74,75] in which the aperture length was made greater than the fiber length. This approach is interesting in that the pulse duration is a measure of fiber length and pulse height is a measure of fiber volume,
hence an estimate can be made of fiber thickness. An alternative approach to fiber measurement involves a flow collar upstream of the sensing zone to provide selective screening and fiber alignment with the aperture axis [36,45,48].

Elzone used a long flow tube upstream of the sensing zone to provide laminar flow and fiber alignment [76]; this flow is caused to join a clear liquid sheath that centralizes the fibers in the aperture.

**9.4 Optical particle counters**

Optical particle counters, in the stream-scanning mode, have been used for many years to determine particulate contamination levels in liquids and in aerosols.

Particle size may be determined in one of the following ways:

- by the amount of light cut off by a particle as it passes through a sensitive zone in a light beam;
- by collecting and measuring the light it scatters over a specific solid angle in the forward direction;
- by collecting and measuring the light it scatters over a specific solid angle at an angle to the incident beam (usually a right angle);
- by measuring the phase shift as a particle passes through a crossed laser beam;
- by measuring the “time of flight” between two laser beams.

Optical particle counters are available which range from simple to highly sophisticated, together with considerable design differences to cater for the wide range of applications. Instrument response depends on particle size, particle shape, particle orientation, wavelength of light, liquid flow rate and relative refractive index between the particle and its surroundings. In addition, the amount of light collected is determined by the geometry of the collecting system. The efficiency of the photo-detector will further determine the degree to which the light pulses can be converted to electronic signals that can be detected [77]. The sensitivity to particle shape is minimized with forward light scattering detectors and particles with aspect ratios of up to two-to-one can be measured [78]. Small off-axis collection angles limit the size of the sample volume thus reducing the problem of coincidence in particle flows of high number density [79]. Differentiation between particles of similar sizes is hampered by pick-up of
stray light that causes noise. Stray light is light, reflected from internal surfaces of the sensor, which falls on the detector. This phenomenon is less of a problem with properly designed off-axis detectors than with coaxial systems.

Knollenberg and Veal [80] discuss operation, design and performance of optical counters in general and a review of extinction optical particle counters has been presented by Sommer [81] (cit. 82). A comprehensive review of laser-based techniques for particle size measurement, covering both stream scanning and field scanning methods, contains 167 references [83].

![Diagram of light blocking principle](image)

**Fig. 9.6** Light blocking principle (Hiac/Royco).

### 9.4.1 Light blockage

In the light blockage technique (Figure 9.6) a narrow area of uniform illumination is established across the flow channel of the sensor so that the passage of a small particle causes an amount of light, proportional to the cross-sectional area of the particle, perpendicular to the beam, to be cut off. For a larger particle, having a diameter comparable with the width of the illuminated zone, the dependence of pulse height on particle diameter is linear. In both cases, the pulse height increases monotonically with particle diameter. Light blockage sensors are available for a variety of size ranges from 1 μm to 3 mm with a dynamic range for each sensor of 100:1 or less. Light blockage is the method of choice for sizing above a micron or so since the instrument response is less affected by variations in relative refractive index and particle morphology than light scattering whereas light scattering provides the higher sensitivity required for smaller particles.
Light obscuration particle counters determine particle size from the projected areas of particles, giving information on only two dimensions. Umhauer [84,85] determines three projected areas of each particle by measuring in three mutually orthogonal directions. An application of this system to agglomerates of two to four spheres has been reported [86]. This work was later extended to measurements on several regular figures [87]. Umhauer et al. [88] designed a 90° single particle light scattering counter to measure the size and concentration of particles in gas flows at high temperature. With this counter, in-situ measurements could be carried out in pipes with cross-sections of around 60 cm² with protection against heat and dust precipitation on the optical windows.

9.4.2 Optical disdrometer

The prototype of a novel disdrometer has been described which features low cost, easy handling, robustness and flexibility [89]. The instrument was developed from one used for classifying raindrops into 20 size classes in the field of meteorology [90]. The size and velocity of particles in the diameter range of 0.3 to 10 mm are measurable with this prototype and the lower size can be reduced to 10 μm with a second sensor. The disdrometer consists of an optical sensor, electronics and a PC. The commercially available optical sensor produces a horizontal sheet of light 160 cm long, 30 mm wide and 1 mm high. Within the receiver, the sheet is focused on a single photodiode. The transmitter and receiver are mounted in the same housing and the light sheet is folded to keep the instrument small. Particles passing through the sheet cause a decrease in received light due to light blockage and this results in a decrease in the primary voltage of 5 V. The voltage decrease is a linear function of particle cross-sectional area and the pulse duration the particle velocity. Particle size distribution was determined using a vibratory feeder at a distance of 115 cm from the disdrometer. A first analysis was made with semolina of around 0.5 mm diameter and a second was made with limestone of diameter around 1.7 mm. The data were then compared with that from a flatbed scanner (see Ch. 3). The measured distributions with the disdrometer were slightly broader with very similar medians.

9.4.3 Light scattering

The relationship between particle size and scattered light intensity at any angle may be obtained for spheres and simple shapes using Mie theory.
Particles larger than about 20 μm scatter light in proportion to the square of their diameter. For smaller particles, the amount of scattered light increases significantly to the sixth power of particle diameter for 0.2 μm particles. The scattered intensity does not necessarily increase monotonically with particle size at all angles. In order to maintain a monotonic increase, small angles or small differences in refractive index need to be used. The main disadvantage of low angle scattering is that the background scatter from the cell walls is higher than with 90° scatter and the main advantage is improved size resolution. A further advantage of low angle configurations is that the intensity is less dependent on relative refractive index than 90° scatter. The fluctuations that occur with a single well-defined angle are damped out by the use of large collection angles. These instruments cover the size range 0.05 μm to 30 μm with a dynamic range varying from 20:1 at the small end to 40:1 for particles bigger than about half a micron.

Usually, basic commercial light scattering counters pick up the light scattered in the forward direction, and great care is taken to damp out the direct beam. Greater sensitivity is obtained by incorporating a light-collecting device such as an elliptical mirror.

These instruments find a ready market in measuring low-level contamination in pharmaceutical suspensions such as intravenous liquids, and for contamination measurements in industrial liquids, and are being increasingly used for on-line process monitoring.

Often, two versions of an instrument are manufactured, one for measuring particles in suspension and the other for measuring particles in air. Simple collecting systems generate a non-monotonic response in the sub-micron size range. Figure 9.7a shows a calibration curve for a co-axial collection system that illustrates this feature. Between the sizes 0.8 μm to 1.8 μm the calibration curve changes direction so that the same output signal occurs for as many as three different particle sizes. For this reason, this type of system is limited to instruments with common size thresholds at 0.3 μm, 0.5 μm and 5 μm. Any size threshold between 0.8 μm and 1.8 μm is ambiguous. 9.7b shows the calibration curve for an off-axis collection system where the output signal for a 0.49 μm particle is the same as that for a 0.61 μm particle. Figure 9.7c shows the calibration curve for the Climet elliptical mirror system.
Fig. 9.7 Calibration curves for light scattering instruments after Chandler [91]. (a) Simple coaxial system, (b) simple off axis system, (c) Climent elliptical mirror system.

Less sophisticated systems tend to use white light illumination, others use gas lasers or solid-state laser diodes. Laser diodes are smaller and more robust than gas lasers, resulting in smaller instruments. Their spatial properties have increased the sensitivity of particle sensors by a factor of two, enabling dynamic size ranges of 700:1 to be measured. Fast analog to
digital conversion now counts every particle in semi-concentrated suspensions and provides 64,000 channels of particle size information [92].

The wavelength of white light ranges from 400 nm to 800 nm, helium-neon is 633 nm and solid state 780 nm. This variation results in differences in measured size when particles with a difference in refractive index from the calibration material are analyzed. Although the instruments are usually factory calibrated it is hardly surprising that instrument-to-instrument variability is high.

**Volumetric** sensors examine the entire stream of liquid passing through the flow cell at relatively low flowrates. **In situ** sensors focus the light on only a small portion. Focusing intensifies the illumination resulting in lower detection limits. Volumetric sensors can detect particles as small as 0.2 μm while in-situ sensors can get down to 0.05 μm. The sensitivity of volumetric sensors is limited since the amount of scattered light varies as the sixth power of particle size, hence a 0.1 μm particle scatters 1/64 of the light scattered by a 0.2 μm particle. Light scattered from the interface between the sensor wall and the fluid also reaches the detector to give a noisy base line.

Hydrodynamic focusing is found to reduce errors when measuring the size of particles in light scattering and phase Doppler instruments as well as electrical sensing zone instruments [93]. In order to increase the maximum number concentration per unit volume the size of the measuring zone has to be reduced. This is realized by using a highly focused beam. This results in a border zone error i.e. particles passing through the measurement beam at the borders generates a smaller pulse than it would be if it passed through the center. Umhauer [94] corrected for this by using a white light source and two detectors at right angles to the beam. Lindenthal and Möller [95] designed a sensing zone shaped like a T. The photomultiplier registers the pulse height for the determination of particle size and the pulse duration in combination with this to determine whether the particle is in the border zone. The Palas PCS 2000 was run with and without border correction, using filter test dusts AC fine and AC coarse. As expected the measurement without border correction was shifted towards the fines. The new instrument was evaluated using a monodisperse aerosol generated by a Sinclair-La Mer generator. The advantage of this set-up is that the correction for border zone error is no longer needed thus eliminating the need for a second PM.

An instrument employing a laser source and two photodiode arrays at 49.3° and 126.4° has been described [96]. The measurements are based on
Mie theory for horizontally and vertically polarized light. Polystyrene spheres were used for calibration.

9.5 Commercial instruments

9.5.1 Aerometrics

*Aerometrics Eclipse* is a white light blockage system designed for both liquid and gas systems. The probe head includes a light source, flow cell and photodiode. The processor can accommodate two heads covering the size ranges: 2-100 μm, 10-500 μm, 20-1000 μm and 50-2500 μm. The flow cells are compatible with most liquids and can be specially adapted for high-pressure environments.

9.5.2 Canty Vision

*Canty Vision* is an in-line system that uses lighted video images and a microprocessor-based image analysis system to visually verify particle size, length, width and distribution. The microprocessor can monitor up to eight applications, under process conditions, with a lower limit of 1 μm.

![Climet elliptical mirror optical system](image)

*Fig. 9.8 The Climet elliptical mirror optical system*

9.5.3 Climet

Climet manufacture the following range of instruments for liquid-borne and air-borne particle counting.

*CI 200 series* collect forward scattered light at angles, relative to the incident radiation, from 15° to 105°. The sample volume is located at the
Stream scanning methods

primary focal point of an elliptical mirror and the scattered light is picked up by a photomultiplier located at the secondary focal point (Figure 9.8). Its most distinct advantage over other systems is its high degree of monotonicity. Its disadvantage is that a large detector is required to collect light properly from particles at the extreme edge of the view volume and this drives up the price.

CI 220 Liquid Particle Analyzer counts particles drawn from a syringe. Four size ranges are available via panel switches: 2 to 20, 5 to 50, 8 to 80 and 20 to 200 μm.

CI 221 On-Line Monitor is designed for high purity liquids for counting particles in the size range 2 to 200 μm for flow rates of 120 to 750 ml min⁻¹. Particle concentrations up to 100 particles ml⁻¹ can be handled directly. Higher concentrations are sampled at proportionately lower flow rates down to 120 ml min⁻¹.

CI 1000 operates using light obscuration with white light and a fiber optics bundle for light collimation. Sensors are available to cover a variety of sizes from 1 to 1000 μm, each one encompassing a 50:1 dynamic range. The sampler accommodates sample sizes from 0.1 to 500 ml. The electronic module contains the electronic components for particle counting, data display and printing. Data are stored in 3000 size ranges simultaneously and can be recovered in six size ranges for display and printing. Other size ranges can be recovered by resetting the size thresholds.

9.5.4 Contamination Control Systems

Contamination Control Systems AWK analyzers measure vibratory fed dry powders in the size range of 20 μm to 10 mm and drops in the 30 μm to 3 mm range at a rate of up to 10,000 particles per second. The analysis system consists of a sensor comprising measurement optics, control electronics and a personal computer that is connected to the sensor via a parallel or serial interface and undertakes evaluation and data display. The measuring principle is based on a combination of light scatter and obscuration. Particles crossing the measurement cell produce an impulse of height proportional to size. The impulses are examined to eliminate "bad" signals and this automatically eliminates coincident pulses. The data can also be processed to give an equivalent sieve distribution. The measurement time is selectable in 0.01 s steps up to 10 min and the
generated pulses are classified into 32 size categories for final presentation in 8, 16 or 32 channels.

9.5.5 Danfoss VisionSensor

_Danfoss VisionSensorQueCheck_ system was developed for manufacturers whose products have to be checked, inspected and sorted at high speed. Using this system the lengths of components on a conveyor belt can be determined and out-of-spec components rejected.

The instrument has also been used for size analysis of sugar crystals from 40 μm to several mm in size. By means of a vibratory feeder, the sugar is fed in free fall, past a vision camera and sized every 5s. The final result of the measurement is available after 100-300 frames and documented via an interface with a database and printer. Measured data includes particle count and size or projected area.

9.5.6 Faley Status

_Faley Status 8000_ measures particles in non-corrosive liquids from 1 to 150 μm, using either of two available sensors based on the light extinction principle. Counts are collected in up to six preset particle sizes. Faley Status also manufacture several counters for airborne particulates.

9.5.7 Flowvision

_Flowvision Analyzer_ uses fiber optics to channel white light through a flowing liquid. As particles pass through the light beam they generate images that are converted to video and then analyzed using a high-speed digital computer. The computer first enhances the image and then classifies according to size. Particles are detected in the size range 2 to 1000 μm; the optimum range for sizing is 25 to 600 μm.

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**Fig. 9.9** The laser optic measuring system of the Galai Particle Analyzer.
9.5.8 Galai

*Galai CIS-1 Model 2010* (Figure 9.9) scans the sample with a shaped and focused laser beam using a rotating wedge prism [97]. The time spent by the scanning beam on a particle is interpreted as particle size. A CCD TV microscope is incorporated into the basic unit, permitting the operator to observe the sample whilst it is being measured. The images can be enhanced, processed and analyzed to obtain an independent measure of size distribution, particle shape and state of dispersion. The instrument operates in the size ranges 0.7 to 150, 2 to 300, 5 to 600 and 12 to 1200 μm. Interchangeable sample cells allow a wide variety of sample presentations; these consist of a spectrophotometer cell, a microscope slide, a liquid flow-through cell, an aerosol flow through cell and a thermoelectric cooled flow-through cell.

*Galai CIS-100* combines particle size analysis with dynamic shape characterization, covering the size range from 0.5 to 3600 μm in 300 discrete size intervals by changing a single lens. The Galai Video Microscope uses a synchronized strobe light and a black and white CCD camera to capture images at 1 or 30 times per second. The included software automatically analyses up to 30,000 particles with up to 1800 non-overlapping particles per frame. The measurement cell modules comprise systems for dry powders, aerosols, fibers in laminar flow and cells for particles in regular and opaque liquids.

*Galai Dynamic Shape Analyzer DSA-10* is a complete shape characterization system for particles in motion. All particles are classified by: maximum and minimum diameters, area and perimeter, aspect ratio, shape factor and more. A video microscope camera synchronized with a strobe light takes still pictures continuously of particles in dynamic flow, generating shape information on tens of thousands of particles, in the 1 to 6,000 μm range in minutes.

*Galai CIS-1000* is an on-line particle size analyzer. A bypass from the process line feeds the sample into the sensor unit where it is sized and either drained off or fed back to the line. Full compatibility with the laboratory instrument is maintained since it uses the identical combination of laser-based time-of-transit particle sizing using the 1001 sensor and dynamic shape analysis using the 1002 sensor. The size range covered is from 2 μm to 3600 μm with measurement of size, area, volume, shape, concentration and estimated surface area with a cycling time of 300 s.
9.5.9 Kane May

*Kane May* manufacture a series of instruments based on the light blocking principle. These consist of an on-line sampler, a small volume sampler and a large volume sampler. Each sensor operates over a size range of 75:1 using a variety of sensors and data may be processed into up to 10 size channels.

9.5.10 Kowa

*Kowa Nanolyzer™ PC-500* uses a He-Ne source and 90° light scattering to size from 0.1 μm in the size ranges: 0.1 to 0.2 μm, 0.2 to 0.5 μm and +0.5 μm at a flow rate of 20 ml min⁻¹ and a maximum pressure of 70 psi. The axis of the measuring cell is parallel to the flow path and a mask placed in front of the photomultiplier precisely defines the measuring area. The image of the mask area is centered on the laser focus, where beam intensity is constant, in order to ensure even spatial sensitivity across the measuring area. Moreover, with detection occurring along the flow path, particles flow through the mask image, so that the measuring area is not only precisely defined but is also of equal size for all particle diameters. This configuration guarantees accurate measurement of particle size and concentration in each size range. The analyzer is intended for use with clear liquids.

*Kowa Nanolyzer™ PC-30* is of similar design and intended for monitoring ultra clean water. It classifies particles in the size ranges, 0.08 to 0.1 μm, 0.1 to 0.15 μm, 0.15 to 0.2 μm and +0.2 μm, in concentrations of less than 30 particles ml⁻¹. Particles are sized according to Mie theory.

9.5.11 Kratel

*Kratel Partascope* operates using a light blocking detection system. Sensors are available, each one covering a 50:1 size range, to give an overall size range of 1 to 8000 μm. Data are presented in 4, 8, 16 or 32 channels using a multichannel analyzer module and this can be expanded to 64 channels using a computer. As with Hiac/Royco, a range of samplers is available. A sub-micron sensor is also available for the size range of 0.4 to 20 μm using near-forward laser light scattering.

*Kratel Partograph* measures size, extinction, light scattering and fluorescence of particles. Hydrodynamic focusing is used to allow single
particle centering in the beam. The optical design allows the simultaneous measurement of the particle size by forward and right angle light scattering as well as an extinction measurement. Particle size is also determined by time of flight from pulse width. Size range covered is 0.5 µm to 200 µm at concentrations up to 10^7 particles ml\(^{-1}\), at flow rates of 10-50 µL min\(^{-1}\).

![Diagram of light scattering geometry used in the Malvern Insitec PCSV instrument](image)

**Fig. 9.10** Light scattering geometry used in the Malvern Insitec PCSV instrument

9.5.12 Malvern

Malvern Autocounters [98] use light extinction sensors for particle measurement. Although designed primarily for counting particles in hydraulic and fluid power systems the ALPS 150H can be used for any liquid samples. Eight size thresholds are available in the 2 to 100 µm, 3 to 150 µm and 5 to 250 µm range with automatic verification to principal contamination standards. The flowrate can be adjusted between 1 and 30 ml min\(^{-1}\) with particle counts up to 10,000 ml\(^{-1}\). Simple, in-field calibration is an important feature of all models of this instrument.

Malvern ALPS 100 system liquid particle counter is a modular system that can be used with an autosampler for multiple samples or with an online sampler for direct measurement of flowing liquids. It can be used with sample volumes down to 0.5 ml and uses a built-in multi-channel analyzer to perform size distribution analyses of low concentration dispersions. Suitable for both aqueous liquids and solvents, it measures up to 50 size bands in the 2 to 100 µm or 3 to 150 µm size range with output from a built-in thermal printer or external dot matrix printer.

Malvern Autocounter 300A Air Particle Counter is designed for general cleanroom and environmental monitoring. It is a 0.3 µm to +5 µm, 1 cfm
Powder sampling and particle size determination

(cubic feet per minute) laser particle counter with eight preset or user defined size thresholds.

*Malvern Insitec PCSV-P* (particle counter sizer velocimeter-probe) is a non-intrusive U-shaped instrument, with the transmitter and receiver mounted on opposite sides of the tube, designed primarily for basic particle research and filter testing. The instrument is intended for *in-situ* monitoring of particle physical characteristics at medium (< 100 ppm) and very low (< 1 ppb) particle concentrations in a size range from 0.2 to 200 μm. In this instrument (Figure 9.10) an He/Ne laser beam is used to illuminate the particles and particle size is calculated from the amplitude of the scattered light measured in the near forward direction [99-101]. Employing near-forward scattering creates some degree of insensitivity for particles with aspect ratios less than 2. A separate response function is used for light absorbing and non-absorbing particles. Holve and Self [102,103] developed an intensity deconvolution algorithm that corrected for the intensity profile of the sample volume based on the assumption that trajectories of the particles through the sample volume are random. Two beam widths are used to measure the wide size range covered. Measurements on the two beam widths are made sequentially and combined to give the full distribution.

Information on the velocity of the particle is found by measuring the transit time of the particle through the sample volume [104]. The instrument has been used in large scale pulverized coal boilers [105-108] char fragmentation and fly ash formation during pulverized coal combustion [109] and for coal slurries [110,111].

**9.5.13 Pacific Scientific Hiac/Royco, Met One**

*Hiac/Royco* is a division of Pacific Scientific together with *Met One*. Hiac manufacture a wide range of counters for liquid borne systems whereas Royco and Met One manufacture counters to monitor airborne contamination. A brief description of a selection from the Hiac range is presented below.

*Hiac PC4000 portable liquid particle counter* is a contamination measurement tool, designed to run on-line analyses of hydraulic systems and fluids. The fully self-contained counter operates in the light-blocking mode using a laser diode and reports contamination levels at 4, 6, 10, 14, 21, 38 and 70 μm at a flow rate of 60 ml min⁻¹.

*Hiac 8011 liquid particle counting system* provides fast results for particle contamination analysis. Composed of a light blocking or dual
mode sensor, the system pressurizes fluid in an ABS-2 (automatic bottle sampler) for precise volumetric sampling of industrial fluids.

_Hiac 8012 liquid particle counting system_ analyzes viscous, dark, dirty fluids without dilution. The system includes the Hiac SDS (syringe driven sampler), 8000A counter and an HRLD (Hiac/Royco laser diode) sensor.

_Hiac BR-8 liquid particle counting system_ is designed for testing filter efficiency simultaneously sampling upstream and downstream. High resolution, large dynamic range laser diode sensors provide an accurate profile of the particle size distributions in the size range 0.5 to 400 μm at a concentration up to 18,000 particles per ml and a flow rate of up to 200 ml min⁻¹.

_Hiac ChemQuaP™ on line sampler_ is designed to provide a solution for the connection of a particle sensor to bulk chemical distribution systems and distribution valve boxes for pre-hookup qualification. A built-in air pump eliminates the need for an external gas supply for enclosure and line purging. For continuous monitoring applications, the sampler is also compatible with Particle Vision® Online facility monitoring software. The light source is a laser diode with count display at 4 channels 0.1, 0.2, 0.3, 0.5 μm at a flow rate of 100 ml min⁻¹.

_Hiac SDS (syringe driven sampler)_ is designed for particle contamination determination of high viscosity fluids and applications where only small amounts of sample are available. A liquid crystal display allows the user to view the distribution in tabular or histogram format.

_Hiac 3000A liquid syringe particle sampler_ incorporates precision stepper motors for sample testing, eliminating the need for pressure air­lines. The microprocessor-controlled sampler is designed to interface with the 8000A counter or with the liquid particle counting system and software. Sample testing parameters, such as volume and number of runs, are entered by the operator either at the 8000A counter or within the software. The counter and software then automatically give testing instructions to the 3000A sampler.

_Hiac ABS-2 automatic bottle sampler_ is a pressure sampling device used for batch analysis of volatile or viscous fluids. The ABS-2 delivers liquid to the sensor at flow rates of 10 to 200 ml min⁻¹. A check valve in the sample introduction line eliminates backflow and minimizes sample cross contamination, and an automatic drain mechanism allows unattended sample analysis of multiple runs. The sampler has a built-in pressure/vacuum chamber that subjects samples to pressures up to 60 psi, accommodating viscosities up to 80 centistokes. In addition, vacuum levels of 23 psi are used to degas samples which contain entrained air.
Hiac SDS syringe driven sampler is designed for high viscosity fluids and applications where only small amounts of sample are available. The sample syringe is close to the sensor, permitting particle analysis with as little as 3 ml of available sample.

Hiac 8000A eight-channel particle counter incorporates state-of-the-art electronics to process and display real-time sample data. An image of the particle size distribution data is presented using. The LCD displays the data as a histogram or in tabular format. Parameters of the analysis, alarm conditions and standards protocol are presented as text against a contrast-adjustable backlit screen. With a single keyboard entry small volume parenterals, product cleanliness levels and the cleanliness of components used in hydraulic systems can be tested. Hardcopy printout is provided through an internal 40 column thermal printer.

Hiac 2000 liquid particle counter provides an inexpensive means to transfer up to four channels of particle size information data from sensor to host computer system. Data can be viewed in real-time via a liquid crystal display. The 2000 interfaces to all Hiac liquid sensors including the MicroCount, submicron and HRLD laser sensors. Applications include point-of-use monitoring for corrosive chemical delivery systems, DI water lines, wet process tools, hydraulic oil systems and parts cleaning.

Hiac 215W on line liquid particle counter is designed for automated particle monitoring in industrial environments. The counter is rugged with a vibration isolated laser diode and clog-free' sample cell on the inside. Particle size data from 2 to 400 μm (1-300 μm optional) is presented in six channels using light blockage.

PCX interfacing Pacific Scientific liquid counter/controller interfaces Pacific Scientics complete family of liquid sensors to their Particle Vision® Online software. The iPCX is designed for online continual monitoring applications.

Hiac/Royco manufacture two counters that are compatible with a wide range of samplers. The 4100 series microprocessor based counter produces particle count data simultaneously in six channels; this is expanded to thirty-two channels with the 4300 series counter. Detection limits range from 0.25 to 25 μm for air samplers and 0.5 to 9000 μm for liquid sensors. A range of sensors is available for liquid systems, each one covering a 50:1 size range. The particles to be counted are suspended in a liquid having a refractive index different from the particles. The particles are then forced through a sensor containing a small rectangular cell with windows on opposite sides. A collimated beam of light from a high intensity, quartz halogen lamp is directed through the stream of liquid on to
a sensor. The particles pass through the sensor in random orientation and produce pulses proportional to their average projected area. These pulses are scaled and counted. The sub-micron size range is covered using near-forward laser light scattering. The shear forces in the sensing zone are small enough for the instrument to be used for sizing flocs [98].

*Hiac/Royco 4100 series* are for liquid-borne systems. Systems 4101 and 4102 are for airborne monitoring using models 1100 and 1200 airborne sensors respectively.

*Hiac/Royco Optisizer* [112] is a single particle counting system which operates with composite scattering/extinction sensors, a 64,000 channel high speed digital counter, an automatic sample dilution system and a software package. The software controls the counter/sampler and acquires, archives and reduces the data. Near forward light scattering is used to measure particles smaller than 3 µm and light extinction for particles above 3 µm. Combining the two optical sizing techniques into a single sensor allows sizing over a dynamic range exceeding 700:1. The model HPS-200 uses a He-Ne laser and covers the size range 0.2 µm to 200 µm; the model HPS-350 uses a laser diode and covers the size range 0.7 µm to 350 µm.

Hiac/Royco also offer the *Dynacount laser diode sensors* for sizing particles in liquids with a dynamic range of greater than 150:1 at number concentration levels up to 12,000 ml⁻¹. HRLD-150 covers the size range of 1 to 150 µm at particle concentrations up to 12,000 ml⁻¹ and flow rates from 10 to 75 ml min⁻¹. HRLD-400 covers the size range of 2 to 40 µm at particle concentrations up to 8,000 ml⁻¹ and flow rates between 10 and 200 ml min⁻¹.

*Hiac/Royco Model 5100 series* is a He-Ne laser-based aerosol particle counter which uses a solid-state photodiode detector to collect light scattered, at 90° to the incident beam, over an angular range from 60° to 120°. Its standard dynamic range is 40:1, from 0.25 to 10 µm at 1 cfm, with automatic collection and averaging of up to 10 sample runs. Applications include qualifying and monitoring class 100,000 to class 1 clean-rooms, downstream filter testing, pharmaceutical and semiconductor manufacture, aerospace research and inhalable particle monitoring. The sample stream is focused so that all particles in the stream are counted.

*Pacific Scientific Met One Model 211 series of sensors* are laser-based light-scattering sensors for liquid-borne particles. Their rugged design, flexibility, and low cost make them ideal for industrial day-to-day use.

*Pacific Scientific Met One Model 100 series of sensors* are laser-based light-blocking sensors for liquid by Met One. These have a simpler optics
design than light-scattering sensors and therefore cost less. They are effective when counting dark-colored particles but not as effective as light-scattering sensors when counting light-colored or small particles. They remain calibrated over a large range of flow rates, handle higher concentrations of particles and have a longer service life than light scattering sensors.

Pacific Scientific Met One 210 Liquid Particle Counter is used to measure particles in clean fluids used in electronic, pharmaceutical and other manufacturing processes. It classifies particles in six size ranges in the 0.4 to 25 μm size range using laser diode based forward light scattering. Maximum count rate is 8000 particles per minute at a fluid flow rate of 100 ml min⁻¹.

Pacific Scientific Met One 2500 batch sampling system is computer controlled for greater flexibility. Flowrate and sample volume are controlled automatically to ensure precise particle counting and sizing of liquid-borne particles. The sampler comes with either a 500 ml or 1000 ml sample capacity and uses either light scattering or light extinction sensors. The system's software displays and prints average counts per ml for up to sixteen size ranges in both graphic and numeric format. Sensors are available for various size ranges from 0.5 to 400 μm.

9.5.14 Particle Measuring Systems

Particle Measuring Systems (PMS) manufacture a range of laser-based particle sizing instruments. These are based on volumetric or in-situ sampling. The former perform particle sizing on the whole of the fluid passing through them and have relatively low flow rates. Sizing may be accomplished using either light obscuration or light scattering. Volumetric sampling may be continuous or batch; batch sampling is accomplished using a liquid sampler to deliver a sample from a vial, bottle or tank to the sensor; continuous sampling is accomplished by measuring a side-stream flow from the primary line. In situ sensors accommodate a wide range of flow rates and perform the measurements directly in-line with the main flow of fluid. Particle sizing is accomplished using light scattered from particles within an optically defined area. The ratio between this area and the flow cross-sectional area yields the fraction sampled. Particles are classified into either 8 or 16 size intervals. Their Ultra DI monitors are designed specifically for deionized water systems for contaminants as small as 0.03 μm.
Fig. 9.11 Particle Measuring Systems’ Liquid Volumetric Probe, designed for continuous monitoring of process waters, product liquids and semiconductor chemicals.

\textit{PMS CLS-700 Corrosive liquid sampler} was developed for batch sampling process chemicals at temperatures up to 150°C. Up to 15 sizing thresholds are available from 0.2 to 5 \( \mu \text{m} \). Simultaneous multipoint measurements at 0.3 \( \mu \text{m} \), 0.4 \( \mu \text{m} \), or 0.5 \( \mu \text{m} \) can be made. Sensitivity is greatly improved with the use of parabolic mirrors (Figure 9.11).

\textit{PMS CLS-900 Corrosive liquid sampler} is a compression sampler designed for integration into wet benches and process tools. The compressive sampler eliminates bubbles from process chemicals.

\textit{PMS CLS-1000 Corrosive liquid sampler} is the latest offering from PMS for monitoring the chemicals within ultra-pure processing environments.

\textit{PMS HSLIS} liquid optical counter provides continuous real-time monitoring of contamination levels in DI water and process chemicals providing size sensitivity down to 0.5 \( \mu \text{m} \) in DI water and 0.65 \( \mu \text{m} \) in process chemicals.

\textit{PMS APSS-200 Automated parenteral sampling system} for pharmaceutical fluids is designed to size and count suspended particulate matter in a wide range of liquids.

\textit{PMS APSS Automated particle sampling systems} are used to size and count suspended particles in liquids. These systems are ideal for applications where precise small volume sampling is needed.

\textit{PMS LiQuilaz\textsuperscript{®} counting spectrometer for liquids} is designed to measure particles in liquids for a wide range of applications.
Nano in-situ sensor and LS-50 liquid sampler are designed for particle measurements in process chemicals and DI water. They are ideal for sampling main process supply lines or point-of-use delivery lines where continuous monitoring of contamination levels is necessary. They can be operated either in-line or batch mode.

PMS Pressurized Automated Liquid Sampler (PALS) is designed for the fields of hydraulic oil contamination monitoring together with pharmaceutical particle size measurement of viscous and volatile liquids. This batch sampler has a self-sealing sample chamber, programmable volumetric settings from 0.1 to 100 ml per cycle, automatic flush cycle, computer controlled and monitored sample chamber pressure and corresponding liquid flow rate. The heart of the system is the Particle Sizing System AccuSizer 580 pulse height analyzer. Data are collected in 32 channels that can be expanded to 128 logarithmically spaced channels for high-resolution particle size distribution. Light extinction sensors cover the size range from 0.5 to 2500 μm.

9.5.15 Partikel Messetechnik

Partikel Messetechnik (PMT) manufacture a range of microprocessor controlled 16 and 32 channel analyzers.

PMT RBG-1000 is a dry powder dispersing system covering the size ranges 1.5 μm to 500 μm and 20 μm to 2000 μm. A dry system, with on-line capability, is also available for granules and free flowing powders covering the micron size ranges, 20-2000, 80-8000, 200-15000. The SAS sampling system covers the same size ranges as the RBG-1000 but is intended for suspensions and emulsions; this can also be used on-line.

PMT-2120 counter uses light blockage and can count up to 3,000 particles per second. It covers the size range 1-12,000 μm with selectable 16 or 32 channels.

9.5.16 Particle Sizing Systems

Particle Sizing Systems (PSS) is a designer and manufacturer of particle sizing instruments that are used for research and development, USP quality assurance, contamination and in-line monitoring.

PSS Accusizer™ 780/SPOS Single particle optical sizer consists of a patented autodilution stage, single particle optical sensor, pulse height analyzer/counter (PHA), systems computer/processor and software control. The autodiluter performs a continuous dilution of the concentrated sample.
suspension prior to its passage through the optical sensor. The PHA module continuously monitors the pulse rate during autodilution and when it falls below the coincidence level (typically 10,000 particles per second for particles smaller than 100 μm) the PHA unit starts to collect data. The resulting size distribution is displayed in real time as absolute counts versus diameter for each diameter channel (8 to 512) logarithmically displayed over the total size ranges covered by the sensors (e.g. 0.5 to 400 μm, 1.0 to 400 μm, 2.0 to 1000 μm, 3.0 to 2500 μm). Additional derived distributions are calculated from the measured number distribution. The instrument can be combined with the Nicomp 380 to extend the lower size limit to 0.003 μm.

Ancillary equipment comprises:

A syringe injection sampler, the 780/SIS, which includes 32 user-defined channels that can be converted to 512 channels to give contamination monitoring and particle size analysis in a single system.

A dry powder sampler, the 780/DPS, that has an air-flow design that minimizes cell wall contact. The flow channel has a 1 cm diameter allowing for a size range from 5 to 5000 μm. Air-flow can be controlled to change the shear forces applied to the particle stream in order to optimize particle deagglomeration. In cases where no shear forces are required the particles are fed through the sensor under gravity.

Fig. 9.12 Schematic diagram of the Particle Sizing Systems Accusizer autodilution apparatus and optical particle counter.
The autodilution system is shown in Figure 9.12. A few drops of concentrated liquid suspension containing several grams of powder are manually mixed into the mixing chamber. Filtered diluent is caused to flow into the mixing chamber, the resulting positive pressure causing some of the suspension to exit through the sensor at a rate of 25-100 ml min^-1. The PHA starts counting when the count rate falls to 20,000 s^-1 [113,114].

PSS Accusizer 780/OL is specifically designed for on-line applications. Simple customizable computer controlled liquid sampling devices are used to grab an aliquot of concentrated in-line suspension and inject it into the autodilution unit. Multi-port systems are available to monitor multiple points in an on-line process. One great virtue of the instrument is its ability to measure minute amounts of oversize particles in the tail of a distribution. The oversize fraction from a homogenizer, for example, is more pertinent for control purposes than the mean size.

9.5.17 Polytec

Polytec HC (high concentration) optical counter series is for counting and size analysis of particles, droplets and bubbles in gases or liquids in the size range 0.4 to 300 μm. The HC system uses a white light source and a relatively large aperture which smoothes out irregularities in the intensity of the transmitted beam. The near linear relationship between intensity and particle size provides sufficient resolution to allow classification of the particles into 128 size channels in a size range of 30:1 at number concentrations up to 10^5 particles ml^-1 and at a velocity of 0.1 m s^-1 to 10 m s^-1. A correction can be applied for coincidence errors.

The optical design of the HC system (Figure 9.13) defines a small rectangular measurement volume, at right angles to the flow direction, by imaging rectangular apertures with the transmitting and receiving optics. A particle passing through the illuminated measurement volume scatters light that is focused on to a photodetector. Each optical signal is converted into an electrical pulse that is electronically processed.

9.5.18 Rion

Rion Laser Based Liquid-borne Particle Counter uses an optional sampler and 'sideways' scatter to allow off-line, on-line and automatic measurement down to 0.2 μm using a range of configurations. A range of sensors is available for use with either corrosive or aqueous liquids.
Fig. 9.13 (a) Block diagram of the Polytec HC Particle Counter
(b) optics of Polytec HC white light counter

9.5.19 Spectrex

Spectrex PC 2000 Laser particle counter counts and sizes particles, from 0.5 to 100 µm in diameter, in both flowing and in-situ liquids. The PC 2000 uses near-forward scattering from a revolving laser beam (900 rpm.) for particle sizing and counting of in-situ and flowing liquids [115]. A He-Ne laser beam is focused to a small, well-defined volume (10 ml) within the liquid. Total particle count within the range 0.5 to 100 µm can be determined in less than a minute. Readout is in 1 µm steps from 0.5 to 17 µm and in five channel steps from 17 to 100 µm. Distribution may be presented on a number or a volume basis. A small vial attachment permits
inspection of vials and ampoules down to 5 ml size at concentrations up to 1000 particles ml\(^{-1}\).

Spectrex provides three sealed calibration standards containing a precise number of NIST traceable polystyrene spheres of known size in suspension. *Supercount* software is a custom interfacing electronics software to provide an easy means of analyzing and saving data generated by the PC 2000. Supercount provides instant sizing information and histogram in addition to indicating: absolute count, mean size, mass distribution, standard deviation and total suspended solids.

![Diagram](image)

**Fig. 9.14** The Spectrex SPC-510, (a) schematic of particle counter, (b) laser beam optics. 1 on-off switch, 2 digital readout, 3 count button, 4 threshold setting dial, 5 Illuminate button, 6 prism, 7 secondary lens, 8 target, 9 bottle, 10 sensitive zone, 11 scanning laser beam, 12 prism, 13 beam splitter and beam strength monitor, 14 scanner, 15 mirror, 16 lamp.
The dilution factor can be automatically computed to give absolute counts for liquids as dense as sludge. Supercount is also loaded with a special program for hydraulic fluids and phi classification.

*Spectrex SPC-510* (Figure 9.14) uses both diffuse vertical illumination for visual identification of large particles and a scanning laser for detection of small particles. The instrument is widely used for quantitative particle counts in bottles [116] including *in situ* examination of bottled beer [117].

*Spectrex ILL-1000 Particle Counter* combines the Prototron with a Particle Profile Attachment (multichannel analyzer). The instrument has been used [118] for examining volcanic ash. AC Fine Dust was used for calibration in eight 5 μm steps, which indicated that accurate data was obtained for sizes above 2 μm. It has also been shown to correlate well with the more tedious filtration and counting method for large volume parenteral liquids [119]. Although semi-transparent containers or liquids reduce the amount of transmitted light flux, the instrument gives valid data for particulates in oil [120].

*Spectrex PC 50 Partascope* viewer consists of a small, battery-operated "Black Box" which internally generates two parallel, scanning laser beams. Samples of water, oil or other liquid are introduced by beaker or bottle. Through Fraunhofer diffraction, particles in suspension, as small as 1 μm diameter, are made visible directly to the human eye. The two laser beams permit comparison of a sample with standards, or of pre- and post-filter samples.

**Fig. 9.15** Spectrex PCT-1, principle of measurement.
Fig. 9.16 The laser optic measuring system of the Brinkmann particle size analyzer

*Spectrex PCT-1 laser particle counter* is a compact liquid particle counter designed specifically to monitor real-time particle counts in ultra-pure water. The water to be monitored is fed to a rectangular cross-section Pyrex glass cell (Figure 9.15) and illuminated by a He-Ne laser at right angles to the direction of liquid flow. When a particle passes through the sensing zone, a pulse of scattered light is emitted and detected by a photomultiplier positioned at right angles to both the laser beam and liquid flow direction. Two channels of information are generated; total count coarser than 0.11 μm and counts above 0.2, 0.3, 0.5 or 0.7 μm as set in a four-position switch. Measuring time is selectable from 1 second to 1 h.

9.6 Dwell time

9.6.1. Brinkmann 201 analyzer

The Brinkman Model 2010 analyzer (figure 9.16) scans the sample with a shaped and focused laser beam using a rotating wedge prism. The time
spent by the scanning beam on a particle is interpreted as particle size. A CCd TV microscope is incorporated into the basic unit, permitting the operator to observe the sample while it is being measured. The images can be enhanced, processed and analyzed to obtain an independent measure of size distribution, particle shape and state of dispersion. The instrument operates in the size ranges 0.7 to 150, 2 to 300, 5 to 600 and 12 to 1200 μm. The measuring cells are standard, a spectrophotometer cell, a microscope slide, a liquid flow-through cell, an aerosol flow-through cell and a thermo-electric cooled flow-through cell. This instrument has been compared with the Coulter and automated image analysis for the size determination of grain based products [121]. The electrical sensing zone and the image analysis techniques gave similar mean sizes whereas the time of transition gave significantly coarser

9.6.2 Lasentec Focused Beam Reflectance Measurement (FBRM)

Lasentec's Lab-Tec 100 uses a scanning infrared laser beam to measure the particle size distribution of particles in suspension. The beam is highly focused and illuminates individual particles in its path (Figure 9.17a). The back-scattered light pulses are picked up by a non-scanning stereoscopic detection system (Figure 9.17b). The size of each particle is determined by measuring the time that the particle is in the beam hence the size is recorded as a random chord length. The laser diode and detectors are stationary while the lens, which focuses the light beam, is vibrated normal to the laser detector plane. The vibrating action causes the beam spot (focal point) to be scanned up and down normal to the direction of fluid flow. The beam amplitude is 3 mm and measurements are carried out in the central 1.5 mm where the velocity is maintained constant; since the frequency is 400 Hz the scan rate is 1.2 m s⁻¹. Since the focal point is only about a millimeter in front of the window it can operate with very high (40% by volume) concentration slurries [122].

Lasentec Labtec 1000 is a laboratory instrument that covers the size range from 0.7 to 250 μm in 28 size channels. The data are generated as 'scanned counts' an empirical frequency distribution created from classification of chords from randomly oriented particles. Software can convert these chords to a spherical equivalent distribution on the assumption that the chords were generated from an assembly of spherical particles: this software contains a filter system to reject improbable data that would tend to skew the distribution to a coarser size. A discrimination
loop sorts impulse for short rise times; only pulses from particles that pass directly through the focal spot have short rise times and will be accepted.

![Diagram of sampling configuration and measuring geometry](image)

**Fig. 9.17** (a) Sampling configuration using standard glass beakers on a magnetic stirrer plate (b) The Lasentec Lab-Tec 100 measuring geometry

*Lasentec Partec 100* was found to give reasonably accurate data for the chord length distribution of spherical particles but the accuracy deteriorated progressively as the shape became more ellipsoidal [123].

*Lasentec’s Focussed Beam Reflectance Measurement Model (FBRM M400L)* operates at a wavelength of 791 nm and the beam rotates @ 75 s\(^{-1}\) at a peripheral speed of 1.9 ms\(^{-1}\). to describe a circle of diameter 8 mm. It has been used with a 3D-model of chord length distribution for particles of a general shape as well as particles with the same shape but different sizes [124]. The focal point can be changed in the axial position but it is
recommended that the distance from the window should lie between \(d_{\text{min}}/2\) and \(d_{\text{max}}/2\) for best results [130].

**Fig. 9.18 Optical arrangement of the Lasentec Par-Tec 200/250**

Other conversions are also available, including calibration against known standards. Monnier *et al.* [125] investigated the influence of variables such as temperature, stirrer speed, focal point, concentration and the position of the focal point, on the measured size distribution and concluded that the most important, especially for small sizes, was the focal point position. They concluded that, due to this, it was not suitable for precise analyses [126].

*Lasentec Partec 200* (Figure 9.18) is an on-line version that gives a reasonably constant response over a concentration range of 5-30% by volume [127]. The Partec 200 gives much coarser data than the Lab-Tec 100, together with a lower count at similar concentrations. This can be attributed to the reflected light being collected by the same optics that are used to focus the light i.e. both the source and detector move in unison. Thus, a much smaller volume around the focal point is investigated on reflection. The beam moves in a circular path compared to the vertical vibrations of the laboratory system and different hardware is used to determine the pulses [128]. The size range of 1.9 to 1000 \(\mu\text{m}\) is covered in a \(\sqrt[3]{1.5}\) progression of sizes.
496 Powder sampling and particle size determination

The latest on-line version covers the size range 0.8 to 1000 µm in 38 channels. In an investigation of this instrument it was found that the measured chord lengths data were sensitive to the location and orientation of the probe. Normalizes chord length data were in good agreement with predictions from the measured size distributions [129].

Langston and Jones [130], starting from an assumed uniform distribution, used Bayes’ theorem to determine particle size distributions from chord distributions of non-spherical 2-dimensional images. Using numerical simulations the derived distributions are found to be quite accurate and robust for a number of different types of particle shapes [131]. With others,[132,133] they also determined particle and droplet sizes from chord size distributions. They concluded that the probability apportioning method worked well as an iterative procedure to predict particle size distributions from chord distributions. Heath et. al. [134] discuss methods of estimating average particle size by FBRM. They stated that the mode average of the square weighted chord length gave results that were comparable with other techniques for the size range 50 to 400 µm. The results were not affected by instrument focal position, suspension flow velocity and solids fraction from 0.1v to 20% w/v.

9.6.3 Messtechnik Optical Reflectance Method (ORM)

ORM system consists of a sensor probe, a signal processing unit and a personal computer running DISPAS™ software for analysis [135]. The instrument is intended for on-line analysis but can also be used off-line. Light from a 1 mW semiconductor laser is transmitted to the sensor through optical fibers. Within the sensor the beam passes an optical system, a focusing lens, a window and finally a sapphire lens before entering the particulate system. The beam is focused inside the sample and is made to rotate eccentrically. A detector picks up the light reflected back from particles and transforms it into electrical pulses. Bad pulses, selected on the criteria of symmetry and slope of the flanks, are rejected and the accepted pulses are converted into rectangular shaped signals. The dwell-times of these pulses are stored in 128 logarithmic channels that, for special requirements, can be increased to 2,000. Since the scanning speed of the laser beam is known these, effectively, give a chord length distribution. The size distribution is extracted from this chord distribution in the DISPAS™ software. The instrument measures particles in the 0.1 µm to mm size range and has been used to measure droplets from 0.1 to 125 µm [136].
Fig. 9.19 Sample chamber of the Procedyne Analyzer.

9.6.4 Procedyne

Procedyne PA-110 [137] uses a high intensity focused laser beam to scan a flowing suspension via an oscillating mirror, thus scanning the suspension horizontally and vertically (Figure 9.19). As particles interrupt the beam a pulse is generated at a photodiode, the length of which is related to particle size. The instrument is designed for the on-line analysis of process slurries at a sampling rate of 250 cm$^3$ s$^{-1}$ with a maximum mass concentration of 2% and a particle size range of 3 to 2000 μm. There are five memory registers, four calibrated for particle size and the fifth for total particle count. Hinde [138] reported on the use of the instrument for controlling mill circuits with disappointing results.

9.7 Aerodynamic time-of-flight measurement

9.7.1 Thermo Systems Incorporated

Time-of-flight aerosol beam spectrometry was first described by Dahneke in 1973 [139]. A commercial instrument, the Aerosizer, [140] was developed by Amherst Process Instruments Inc. which is now a division of Thermo Systems Inc. The TSI Model 3603 replaces the Aerosizer.
(TSI) Model 3603 aerosol time-of-flight mass spectrometer (ATOFM) is based on the precise laser based measurement of a particle’s aerodynamic time of flight (Figure 9.20). Particles to be measured are first suspended in air using a disperser. The particle laden transport air carries particles that are then surrounded by sheath air. The suspension expands through a converging nozzle into a partial vacuum and accelerates through a measurement region to near supersonic speeds with a barrel shock element; the smaller the particle, the faster the acceleration. The particle’s velocity is determined by measuring the time of flight as they cross two laser beams in the measurement region. As particles pass through the beams, they scatter light that is picked up and converted to electrical signals by two photomultipliers. One photomultiplier detects light as the particle passes through the first beam and the other detects light as it passes through the second beam. Particles are measured, with 500-channel resolution, at rates up to 100,000 per second in the size range 0.2 to 700 μm. The Aerosizer is widely used in the pharmaceutical industry since it measures the particles aerodynamic diameter. This parameter is of great importance in the design of nasal inhalers.
Fig. 9.21 Schematic of the Thermo-Systems APS showing the accelerating nozzle and the two spot laser velocimeter

This technique has been extended by inclusion of a pulsed ionization laser to vaporize the particles after their size has been determined by aerodynamic time of flight. This causes the particle to vaporize and the resulting fragments are partly ionized. Positive ions are accelerated into the flight tube of a mass spectrometer where their chemical composition is determined [141].

*TSI Model 3800 Time-of-Flight Mass Spectrometer* was developed in cooperation with the University of California as the first single airborne particle mass spectrometer to be offered commercially. It uses an aerodynamic sizing technique to size individual particles in real time. It then desorbs and ionizes the particle for chemical analysis in a bi-polar, time-of-flight mass spectrometer. It operates in the 0.3 to 3 μm with an optional disperser to extend the range to 10 μm. The instrument can save positive and negative mass spectra at a rate up to 10 particles per second [142].

*TSI Aerodynamic Particle Sizer APS 33B* counts and sizes airborne solids and non-volatile liquids at number concentrations up to 600 particles.
cm⁻³ at 0.5 μm and 45 particles cm⁻³ at 30 μm, and sorts them into 58 size channels (Figure 9.21). The aerosol is sampled into an inner tube, surrounded by a sheath of filtered air and accelerated through a nozzle. Two timers are used, a 2 ns resolution timer for sizes from 0.5 to 15 μm (small particle processor, SPP) and a 66.67 ns timer for sizes from 5 to 30 μm (large particle processor LPP). Calibration is effected using polystyrene latex spheres of known density.

The major factors influencing accuracy are particle density, particle shape, particle deformation and the particle counting process. The SPP is known to generate phantom counts and the LPP is known to reduce them. Sreenath et al. [143] provide insights on how to operate the APS to avoid counting errors and the advantages and limitations of different correction methods are discussed. A full description of the equipment is given in a paper by Blackford et al. [144]. Although designed for the size range 0.5 to 30 μm its overall efficiency falls from 95% for 3 μm particles down to less than 45% for particles smaller than 10 μm [145]. The APS Model TSI 3310 has been used to calibrate an optical particle counter and good agreement was found [146].

TSI Aerodynamic particle size spectrometer model 3321 provides two measurements, aerodynamic diameter and light-scattering intensity, in the size range 0.5 to 20 μm. The generation of paired data makes the instrument of particular interest to aerosol scientists.

9.7.2 Ancillary equipment

TSI Aerodisperser is designed to ensure that particles are properly separated from each other, even in the subμm region, by controlling the applied shear forces.

TSI Aero-Dilution model enables high concentrations of powders and aerosols to be analyzed. Since only a small sample is required (typically 0.1 g) it is particularly useful for expensive or research material. TSI Aero-breather has been developed for sampling dry powder inhalers. Barr and Cheng [147] have carried out an evaluation of this instrument.

TSI Model 3433 small-scale powder disperser breaks up agglomerated powder, by lifting particles from a turntable, and feeds them to the APS. TSI also manufacture a fluidized bed aerosol generator to generate high concentration aerosols. The instrument has been used to monitor mixing structure of cohesive dry powders [148]. Both the resolution of the Aerosizer and de-agglomeration with the Aero-disperser has been
demonstrated using albuterol [149]. In comparison tests the Aerosizer was found to agree well with image analysis, however the Malvern Mastersizer and the Coulter LS were found to give dissimilar data [150,151]. At variance with these data Kaye et. al. [152] found very good agreement between the Coulter Counter and the Aerosizer with TiO₂ coated PMMA spheres.

Naqui et. al. [153] describe a phase Doppler technique, for measuring particle velocity and statistical information about particle size of irregular particles, based on a phase shift signal. The technique works on near backscatter which leads to a robust set-up under conditions of limited optical access. Preliminary measurements in a crystallizer were presented and good agreement with TSI Aerodynamic particle sizer was found.

9.8 Laser Doppler velocimetry (LDV)

Measuring particle size from the peak amplitude of a LDV signal was first done on combustion flows [154,155]. A later paper discussed the errors associated with this method and proposed procedures to correct for them [156]. Types of error include:

1. The possibility of more than one particle being in the sample volume at the same time:
2. Particles only partly in the control volume being counted as smaller particles;
3. The dependence of the light scattering signal magnitude on the particle’s location in the sample volume;
4. The dependence of the sample volume size on the particle’s apparent diameter since larger particles have a larger effective sample volume due to the Gaussian intensity distribution of the laser beam.

The top-hat technique eliminates this problem by creating a beam with a constant intensity distribution over most of the beam width [157]. This volume is defined by the boundary where the illumination intensity falls below $1/e^2 = 0.135$ of the peak intensity. Various methods of creating this profile have been presented [158,159].

9.9 Laser phase Doppler principle

The laser phase Doppler particle analyzer (PDPA) simultaneously measures particle velocity, size and flux and may be considered an extension of laser Doppler velocimetry (LDV). It is particularly useful for
determining the local volume and mass flux within a spray cone. The basic principle is as follows. The phase Doppler method is based on the principles of light scattering interferometry. Measurements are made at a small non-intrusive optical probe defined by the intersection of two laser beams. As a particle passes through the probe volume it scatters light from the beams and creates an interference fringe pattern. A receiving lens located at an off-axis collection angle projects a portion of this fringe pattern on to several detectors. Each detector produces a Doppler burst signal with a frequency proportional to the particle velocity. The phase shift between the Doppler bursts from the different detectors is proportional to particle size. The system measures sample volume simultaneously with particle size; this enables accurate determination of particle number concentration and volume flux. No calibration is required since the particle velocity and size are dependent only on the laser wavelength and optical configuration. The instrument calibration is verified periodically using microspheres. The technique was introduced by Durst and Zare [160] and improved by others [161-163].

9.9.1 TSI Aerometrics phase Doppler particle analyzer

This was developed by Aerometrics in 1983, in collaboration with Lewis Research center, for research into pollution reduction from gas turbines. It is particularly relevant to measurements of small, spherical particles such as are found in fuel injection systems, medical nebulizers and bubbles in water. Aerometrics was later acquired by TSI who currently produce the TSI/Aerometrics PDPA 2D System. This instrument measures sizes in the 0.5 to 10,000 μm range using various optical configurations. The optical transmitter and receiver can be traversed together to move the location of the optical probe for spatial mapping of the flow field and the particle size distributions.

9.9.2 Discussion

It has been assessed that the response of PDPA is very sensitive to the roughness of conducting particles [164] (for surface irregularities equal to the wavelength of light the errors are about 23%) to inhomogeneities in droplets i.e. spray dried dairy milk powders [165] and to the periodic oscillations of droplets [166]. If these conditions do not apply, the scattered light bears irregular and ambiguous information about the phase difference Δφ that leads to incorrect particle size determination. The
various errors associated with mass flux measurements in sprays are highlighted in a paper by Dullenkopf *et. al.* [167].

Dual mode PDA combines a planar PDA with a standard PDA so that two-phase differences are measured per scattering event. These are validated against each other to see if the signal is acceptable or not. This permits the rejection of distorted events caused by multiple scattering due to poor surface quality [168]. Dual beam frequency biased LDV permits simultaneous measurement of particle size and electric charge in a two-component developer [169].

An approximate mathematical correction of a measured polydisperse size distribution has been carried out using a deconvolution technique [170]. This was verified with suspension droplets; the advantage being that it can measure such distributions; the disadvantages being that the correlation between particle size and velocity is lost and at least 5,000 data points are required for deconvolution [171].

A modified PDA has been described for on-line fiber diameter measurement [172].

Petrak [173] measured particle size and velocity using an optical probe with a fiber optical spatial technique. In order to determine particle size in a free particle-laden air stream spatial filtering was modified by fiber optical spot scanning. He found good agreement with LDV in the size range 10-1000μm at velocities from 0.01 to 20 ms⁻¹. An in-line device is available from PARSUM. Digital Particle Imaging Velocimetry (DPIV) is being studied for *in-situ* measurements of two-phase flows for mass transit [174]. A knowledge of droplet/particle size and spatial distribution is required. DPIV was found to be useful for large particles but failed for micron sized particles.

### 9.9.3 Differential phase-Doppler anemometry

The two detectors of a standard PDA system have been replaced by a charge coupled device (CCD) line scanner sensor with 256 pixels. Hence, scattered light can be measured with high spatial resolution. Instead of two signals, 256 are detected which leads to 128 phase difference values rather than one. By statistically evaluating the scattered signals even severely distorted signals can be analyzed and need not be discarded. This is called differential phase Doppler anemometry. (DPDA). It has the potential to measure the size of glass spheres with surface defects or inhomogeneous composition. The smallest particles measured were 25 μm water droplets but measurements down to 10 μm were deemed possible [175].
9.9.4 Bristol Industrial Research Association

In the phase-Doppler (PD)-Lisatec a measuring volume with a Gaussian distribution (that is without real fringes) is generated and the velocity and phase information is derived using sophisticated patented polarization techniques. The optical processing employed directly filters the signal and removes the Gaussian envelope. This allows the dynamic range for velocity measurements to be greatly increased from a usual 5:1 or 10:1 to at least 30:1. The instrument covers the size range from 1 \( \mu \text{m} \) to several mm at flow rates from 1 cm s\(^{-1}\) to 500 m s\(^{-1}\). An optional fiber link between the laser and the optics allows the laser to be remote from the experiment. This instrument was developed by the Industrial Optics Group at AEA Technology who have published over 50 papers on this subject.

Laser two-focus velocimetry is applied in the BIRAL L2F. The measurement volume is formed by focusing two laser beams to two small waists of about 10 \( \mu \text{m} \) diameter. The result is a light gate operating with concentrations orders of magnitude greater than possible with the LDV system. Particle velocity is determined by the time it takes the particle to pass from one beam waist to the other.

9.9.5 Dantec Particle Dynamic Analyzer

The Dantec Particle Dynamic Analyzer measures particle size, one to three velocity components and concentration using the phase-Doppler principle. The Doppler effect is a phase shift in light scattered from moving particles and incident illumination with the frequency shift being proportional to particle velocity. If a standard laser Doppler anemometer is combined with a second photodetector, the photodetector signals, under certain conditions, are a direct measure of particle size. A third photodetector is included to extend the dynamic range and, in two-dimensional flows, a color separator and a fourth photodetector are added to allow two velocity directions to be measured. A fifth detector is needed for measurement of three-dimensional flows. Its applications include droplet sizing, spray characterization, fuel injectors and agricultural sprays. The size range covered is 0.5 to 10,000 \( \mu \text{m} \) with a dynamic range of 40:1 with the possibility of extending this by a factor of 2.5 by varying the aperture spacing. Velocity maximum is \( >500 \text{ m s}^{-1} \); up to three components being measured using a combination of near forward scatter, near backward scatter and side scatter.
9.10 Hosokawa Mikropul E-Spart Analyzer

*Hosokawa Micron E-Spart Analyzer* carries out simultaneous measurements of the size of a particle and its electrostatic charge. Characterization of electrostatic charge and aerodynamic size of particles is of critical importance in many electrokinetic processes [176]. A number of instruments are available that can characterize aerodynamic size distribution of particles. Likewise, instruments are available to estimate the net average electrostatic charge of particles. However, the choice of instruments for real-time simultaneous measurement of aerodynamic size and electrostatic charge distribution of particles on a single particle basis is limited.

*Electrical Single Particle Aerodynamic Relaxation Time (E-SPART) Analyzer* simultaneously measures size in the range from submicron to 100 μm and particle charge distribution from zero to saturation levels [177,178].

The operating principle depends upon the phenomenon that when an airborne particle is subjected to an oscillatory external force, such as

![Diagram](attachment:figure922.png)

Fig. 9.22 (a) Schematic view of E-Spart relaxation cell (b) principle of particle measurement. Individual particles are subjected to acoustic and/or electric excitation and the resultant response is measured by LDV to determine aerodynamic size and electrostatic charge.
acoustic excitation, the resultant oscillatory motion of the particle lags behind the external driving field. The particle vibrates at the same frequency as the acoustic field but with a phase lag due to particle inertia. The phase lag increases with increasing particle size and so can be related to particle size. To determine this phase lag, the analyzer uses a differential laser Doppler velocimeter (LDV) to measure the velocities of individual particles subjected to a combination of an acoustic and a DC electric field. Simultaneously a charged particle will have its vertical position shifted by the electric field by an amount related to the charge. The maximum count rate varies from 10 to 2,000 particles per second depending on particle size that, typically, can range from 0.3 μm to 75 μm.

The particles are sampled in a laminar flow field through the LDV sensing volume. As each particle passes through the sensing volume it experiences the acoustic excitation and the superimposed DC electric field in a direction perpendicular to the direction of the laminar air flow. A typical sampling configuration is shown in Figure 9.22. A full description of the theoretical basis for the instrument has been published [179] together with some typical applications.

In a further paper, the instrument was applied to the size and charge analyses of toners [180]. The size range for toner was limited to 2 μm to 25 μm; a lower acoustic frequency is used in the improved E-Spart that increases the upper size limit, for glass beads, to 50 μm [181].

9.11 Shadow Doppler velocimetry

The SDV is based on the imaging of a conventional LDV probe volume on to a linear photodiode array and has the advantage over other sizing methods in that it is tolerant of the optical misalignment and fouling which are inevitable when passing laser beams through windows in furnaces. The technique has been used for simultaneous size and velocity measurements of irregular particles in confined reacting flows. [182].

The instrument was developed by Hardalupas et. al. [183]. The transmitting optics are identical to a conventional LDV but the receiving optics, in contrast, allow collection of the transmitting light beam so that an image of the LDV measuring volume is formed. This image is magnified by a microscope objective and projected on to a 32-element linear photodiode array. Particle size is derived by measuring the size of the shadows of the particle on the array and, because the presence or absence of the shadow is a binary phenomenon, the method is independent
of signal intensity. The analog outputs of the diode array are subsequently digitized and stored for processing and display.

The accuracy of SDV was assessed by Morikita et al. [184] who showed that the maximum difference between the arithmetic means of irregular particles by SDV and microscopic measurements was about 10%. Hishida et al. [185] recorded a maximum difference of 4% owing to beam wandering due to temperature gradients and concluded that the maximum error with increasing flame size cannot exceed 15%.

9.12 Other light scattering methods

A simple light scattering photometer was designed, to measure the angular distribution of intensity of scattered, polarized, He/Ne light, by micron and sub-micron particles [186]. The photometer used an ellipsoidal reflector and simple optical components to collect the scattered light and focus it on a 512 element photodiode array.

The intensity ratio method is based on measurements of light scattered at two angles and applies to the size range 0.1 to 10 μm [187]. Due to the possibility of large errors [188] the method has found little application.

Using Scanning Flow Cytometry the size distribution of submicron spherical particles is determined from the scattered light intensity ratios at two angles. In one example the ratio at 67° and 15° was used to determine sizes between 1 and 15μm at a flow rate of 500 particles per second [189]. This was extended to 0.5 to 14 μm using a parametric solution based on analytical approximating equations [190].

The ratio of the polarized light intensity scattered from two different coaxial beams illuminating a particle can be used to determine particle size. Azzázy and Hess [191] used two coaxial beams of different wavelengths at 30° from the forward axis polarized in different directions. The ratio of these two parameters gives a unique curve that is a function only of particle size. They found errors of a similar magnitude to those found with intensity ratio methods.

Hess [192] described an arrangement in which an LDV velocity system is positioned concentrically inside a larger sizing beam. The signal from the smaller beam is used to trigger the larger beam so that only particles passing centrally through the larger beam are counted. Wang and Henken [193] applied such a system for measuring particles in the 10 to 200 μm range.
9.13 Interferometers

9.13.1 Mach Zehnder type interferometer

Unlike light scattering instruments, interferometers do not measure the scattered light, but the phase shift in light waves. They can distinguish between gas bubbles and particles because bubbles have a lower refractive index than the surrounding liquid and therefore produces phase signals of opposite polarity to those of particles. This makes these instruments particularly useful for examining the reagents used for semiconductor cleaning since these reagents often have high vapor pressures and tend to form bubbles that can be counted as particles using light scattering or obscuration instruments.

Interferometers separate a laser beam into two beams and then recombine them to create a signal whose intensity depends on the phase difference between them. When a particle with a refractive index greater than that of the surrounding liquid passes through the beam the wave front is retarded and when a gas microbubble passes through it the wave front is advanced. The magnitude of the phase signal depends on particle size and the pulse can be calibrated with particles of known size.

![Schematic diagram of a Mach Zehnder type interferometer.](image)

**Fig. 9.23** Schematic diagram of a Mach Zehnder type interferometer.

Figure 9.23 is a schematic of a Mach Zehnder type interferometer the design of which allows for a polarized light beam to be split in two. The incoming beam with intensity $I_0$ is divided into two equally intense beam
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with a beam splitter. The beam following path 1 undergoes a 90° phase change and a second beam splitter combines beams 1 and 2 to form new beams A and B. The two beams are now 180° out of phase; when one is bright the other is dark. The intensities of the beams $I_A$ and $I_B$ are measured using separate detectors. Due to their inherent complexity, large size and susceptibility to vibration, interferometers have remained laboratory and research tools.

The variation of the polarization ratio with time has been used to determine the droplet size distribution in fuel sprays. Although polarization ratio is generally applied to an assembly of droplets it can be used for single droplets provided the incident beam is circularly polarized [194]

9.13.2 The TSI Liquitrak\textsuperscript{TM} interferometer

The \textit{TSI Liquitrak}\textsuperscript{TM} interferometer [195-197] uses a dual beam interferometer to detect slight differences in the refractive indices of particles relative to the surrounding media. It is less sensitive to vibration than the Mach Zehnder type of interferometer because it does not separate the two light beams. Instead the beams are overlapped and polarized at 90° to each other, hence the beams do not interfere and are effectively separated from each other until combined by the second beam splitter. The narrow separation of the beams reduces vibration sensitivity dramatically because any vibration interference affects both beams equally.

The dual beam interferometer also has the advantage of allowing flow-rate measurement. The instrument's flow-rate ranges from 4 to 40 ml min\textsuperscript{-1} of which 1/200 is examined. Flow-rate is measured each time a particle signal is processed hence particle concentration can be measured in a fluctuating flow.

The interferometer has several advantages over dark field scattering instruments. Because it is a bright field instrument it is less sensitive to the stray light scattered by interfaces between the instrument's capillary cell wall and the liquid medium. The instrument can also identify signals created by bubbles thus avoiding false counts. It can also measure flow-rate and it is more sensitive to particles with a refractive index near that of the liquid than dark field instruments because it can look at forward light without noise interference from the incident laser beam.

A drawback is that its inspected volume is small (0.5%) compared to the full flow stream since a highly intensive beam is required in the
viewing volume, rendering the instrument less useful for low concentration contamination measurements.

An evaluation of this procedure is available in articles by Blackford and Grant [198] and Grant [199] and the instrument is available from Thermal Systems Inc. as Model 7750.


In the flow ultramicroscope [200] dispersed particles are injected into a stream of liquid and hydrodynamically focused so as to pass through a laser beam. The scattering is detected by a photomultiplier and processed electronically as a series of pulse heights. The detector can be at right angles to the incident beam, with either a narrow or wide receiver angle, or forward angle scattering may be used. Since the scattered light intensity is highly dependent on particle size, the dynamic range of the photomultiplier can be exceeded by samples of relatively low polydispersity. The range is greatly increased by using a feedback system from the photomultiplier to the laser. The instrument measures number concentration and size distribution for spheres and particles of simple shapes in the size range 0.1 to 5 μm. Such an instrument has been used to measure particle flocculation [201].

9.14.1 ISPA image analysis system

ISPA 800 series characterizes the size and shape of 5 μm to 10 mm particles by measuring the projected areas of the particles in a strobbed image. The unit, manufactured by Greenfield, can process 15 images per second and can analyze particles moving at 30 m sec⁻¹.

9.15 Measurement of the size distribution of drops in dispersions

The most common method is direct photography [202-205]. The method is simple, easy and accurate and covers a wide size range, controlled by microscope magnification, from a lower limit of 1 μm. The technique is only suitable for low concentration systems, particularly in the case of high opacity continuous phases. The procedure requires many pictures and lengthy analysis times. Direct image analysis of the data has met with limited success.
Light attenuation is a simple, widely used method for determining interfacial area, i.e. surface-volume mean diameter if droplet concentration is known but cannot be used for size distribution determination [206-208].

Light scattering has been used for measurement of small drop sizes below 10 μm in diameter [209] and also for drop sizes below 800 μm. Although on-line measurement is possible the technique is only suitable for volume concentrations smaller than 0.05 [210].

Laser Doppler velocimetry (see section 9.6) has also been used for the measurement of a broad size range of drop sizes in solid-liquid and liquid spraying systems [211,212].

Drop size distribution in dilute suspensions of electrical conducting liquids may be determined using the Coulter principle but the need to add what may be undesirable conductive materials limits its applicability [213-215]. The use of chemical means to measure interfacial area has been used extensively for gas-liquid dispersions. Chemical reaction methods for determining the interfacial area of liquid-liquid systems involve a reaction of a relatively unchanging dispersed-phase concentration diffusing to the continuous phase. The disadvantage of this approach is that the mass transfer can affect the interfacial tension, and hence the interfacial area [216-218].

Drop stabilization methods rely on the immediate stabilization of drops by encapsulation with thin polymer films or surfactants [219-221] a photomicrographic method has been employed usually after encapsulation of drops. However this method cannot always be used due to incompatibility of the encapsulating materials with some systems. The method also has the disadvantage of the influence of the chemical treatment on drop size. A special sampling apparatus has been developed to withdraw a sample of dispersed phase from the mixing vessel to stabilize drops with a surfactant and to force the dispersed sample through a capillary with a photometer assembly to measure both droplet size and concentration [222].

The capillary method employs a fine bore capillary of the order of the drop size for sampling from the liquid dispersion. As drops pass through the capillary, they are transformed into cylindrical slugs of equivalent volume. A laser beam is split into two rays using a beam splitter and a plane mirror and the rays pass directly through the capillary precisely 0.1 mm apart. The emergent beam is collected by a ×10 microscope objective lens and focused on to a photodiode. From the measurement of the passage time of a slug at one detector and its travel time between two detectors, the velocity and diameter of the drop can be calculated. The
method can be used to obtain broad drop size distributions in the range above 50 \( \mu m \) in real time and automatically [223-227].

The scintillation method uses short-range radioactive particles for measuring interfacial area. This technique is limited by the necessity of high immiscibility between the phases as well as the availability of suitable isotopes and target materials [228].

The Lasentec particle/droplet size analyzer can be used for laboratory and in-line analysis in the +1 \( \mu m \) size range over a wide range of operating conditions.

9.16 Dupont electrolytic grain size analyzer

The EGSA provides a rapid (4 to 10 min) absolute measurement of the charge required to electrolytically reduce/oxidize AgX crystals. This electrolytic decomposition is singularly effective as a basis for measuring the particle size distribution of photographic emulsion grains since the charge is directly related to grain volume. Grains are electrolytically reduced as they are rotated under a measuring electrode and the generated pulses are sorted according to their integral size and stored in 256 logarithmically distributed channels. Three size ranges cover an overall range of 0.05 \( \mu m \) to 2 \( \mu m \) [229].

9.17 Light pressure drift velocity

The motion of individual Brownian particles is observed using a confocal tracking microscope. Particles are trapped in a strongly focused laser beam. By evaluating light-pressure-drift-velocity and the back-scattered light intensity the particle size is determined to \( \pm 2\% \). The method was demonstrated on a mixture of seven polystyrene latices between 300 and 450 nm that were divided into six size classes. A discussion of the method is presented together with a suggestion as to potential applications [230].

Tuch et. al. [231] ran a Mobile Aerosol Spectrometer (0.1 to 2.5 \( \mu m \)) and an Electrical Aerosol Spectrometer (0.5 to 10 \( \mu m \)) side by side for 6 weeks and found both to be reliable with almost identical results. Total number counts agreed with results from a Condensation Particle Counter.
9.18 Impact size monitor

Size distributions in pneumatic conveying systems are usually monitored by taking grab samples or by using the Malvern/Insitec optical diffraction particle size analyzer [232]. CSIRO has developed and patented a technique to measure particle size from measurements of the peak compression of an ultrasonic transducer subject to impact by the particles [233]. Each impact produces an independent pulse, the duration and amplitude of which conveys information about the particle size and velocity. The impact times for sub-mm particles is typically less than a μs; this short duration allows the measurement of tens of thousands of impacts per second. The instrument has been tested using several grades of ballotini from 50 to 165 μm in size and was able to differentiate between particles of size 157 and 165 μm [234]. Fluid dynamic modelling of this instrument was carried out to determine the size range of particles which could be monitored and it was determined to be applicable to the size range 50 μm to 200 μm at load rates of up to 1 kg m^-3 [235]. Impact sensors have been used previously to detect particles in streams such as sand in oil pipelines [236].

Quantitative measurements have also been carried out at low impact rates by dropping particles, in a vacuum, on to a sensor consisting of a hit plate in point contact with an ultrasonic transducer [237].

High-velocity air-laden dust has also been measured using transducers [238].

9.19 Monitek acoustic particle monitors

*Monitek Micro Pure Systems acoustic particle monitors* uses a focused acoustical beam to sense discontinuities in a flowing liquid and can detect the size and amount of suspended solids, entrained gases, fibrous material in any liquid, or oil droplets in water. The sensor mounts in-line without restricting the process flow and the acoustical beam is focused to a point approximately 0.8 to 1.5 in from the sensor tip. A piezoelectric crystal that acts both as a transmitter and receiver generates the high frequency. The transmitter emits hundreds of pulses per second and monitors the echoes; this high sampling rate makes the instrument insensitive to liquid flowrate. The amplitude of the echo is size sensitive so that a lower limit size threshold can be set; this limit can range from 0.2 μm to a few millimeters.
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9.20 Erdco Acoustical Counter

Audible sounds may be produced by particles exiting from a high-velocity laminar flow tube into a low velocity tube. The phenomenon was first reported by Langer [239] and has since been investigated by Langer [240] and others. The sensing zone of the Erdco counter is a capillary at the exit of a glass tube. As particles enter this section they interact with the boundary layer, resulting in a toroidal vortex that moves as a shock wave that is reflected back on the capillary. The pressure wave is detected by a transducer at the outlet of the capillary, whose displacement is measured by an optical probe. The displacement is proportional to particle size, which is measurable down to 4 μm.

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