LASER LIGHT DIFFRACTION TECHNIQUE
FOR PARTICLE SIZING

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ABSTRACT

This paper describes a laser diffraction particle sizing technique applications and hardware improvements. With sound theoretical background, this technique finds itself competing in the particle sizing technology market with the introduction of Malvern model series 1800, 2600 and Master Sizer of which the first two models have been tested extensively. Correction schemes to overcome some basic major problems such as multiple scattering of particles and vignetting effect also presented. Instrument calibration and comparative performance notably with the phase Doppler and other sizing techniques were made. Overall results show that by resolving some basic major problems and instrument calibration properly done then the Malvern particle sizing data is 'correctly' comparable with any other sizing technique.

1.0 INTRODUCTION

One of the instrument used widely in sizing particle is the Malvern which based on laser diffraction technique. It was commercially available in 1976 as a result of a theoretical development by Switchenbank and co workers. It is widely used in many industrial applications especially in combustion fuel spray, agriculture and chemical works. Although received well in routine sizing work, however it has many limitations and weakness as faced by any other optical techniques such as visibility, intensity or phase shift methods. Before 1984 (and seriously before 1986), its application totally overlooked the 'vignetting' effect which led to a smaller size particle distribution than the actual distribution. This article is to present the development that traces the problems
arise in its applications and make comparative performance study against other sizing techniques for the past two decades. Corrections to the vignetting effect and bias data were also highlighted in an effort to minimise instrument experimental error.

2.0 PRINCIPLE

Light that scatters from any homogeneous spherical particle appears in three basic modes of reflection, refraction and diffraction. On-axis forward scattering is dominated by the latter mode while the first two are more influential in the off-axis light scattering. Figure 1 shows a line diagram of laser diffraction set-up where a continuous laser beam is expanded by a beam expander and crosses the sample volume. Scattered light is collected by a receiving optics system and projected onto a well position detector at the receiving lens focal point. The light intensity is then said to be related to the particle size where monodisperse particles would form a pattern known as Airy pattern as in Fig. 2, while with polydisperse particles the pattern changes significantly where distribution is wider as the particles become smaller as shown by Fig. 3.

Expression for on-axis light intensity distribution due to the diffracted light by a spherical particle of radius \( a \) is given by

\[
I(\theta) = I_0 \left[ 2J_1(2\pi a\theta/\lambda)/(2\pi a\theta/\lambda) \right]^2 \tag{Eqn. 1}
\]

where \( I_0 \) and \( J_1 \) are the incident beam intensity and the first order Bessel function respectively. Particle radius is given by \( a \) and \( \lambda \) is the light wavelength.

A series of concentric alternating light and dark rings of this light distribution appear on a screen correspond to the different particle of different sizes. Summation of light distribution for polydisperse distribution can be obtained and drop size is then inferred. The exact position of the particle within the laser beam is not detrimental because of the light intensity distribution.
Fig. 1  Particle Size Analyzer Principle

Fig. 2  Diffraction Pattern due to Identical Spherical Particles

Fig. 3  Evolution of Diffraction with the Size of Particles
depends only on the scattered angles and particle size. Relation between scattering angles $\theta$ and the off-axis position, $r$, on the detector is given by

$$ r = f\theta $$  \hspace{1cm} \text{Eqn. 2}

where $f$ is the lens focal length.

Detectors sensitivities limitation is overcome by various methods and the method adopted by Malvern instrument is proposed by Swithenbank et al. (1967) where a detector contains concentric annular rings of increasing mean radius. Each ring does not suffer for the intensity variation.

From Negus and Azzopardi (1978), the energy diffracted into a ring by a single particle is given by

$$ E_y = C\pi a^2 \left[ \left( J_1^2 \left( \frac{2\pi a}{\lambda} \theta \right) + J_0^2 \left( \frac{2\pi a}{\lambda} \theta \right) \right) \right]_i \left[ \left( J_1^2 \left( \frac{2\pi a}{\lambda} \theta \right) + J_0^2 \left( \frac{2\pi a}{\lambda} \theta \right) \right) \right]_j $$  \hspace{1cm} \text{Eqn. 3}

where $i$ and $j$ are the inside and outside ring radius respectively. $J_1$ and $J_0$ refer to the Bessel function of the first kind of order 1 and 0 with $C$ is the calibration constant.

In terms of weight or volume distribution the expression for a distribution size is given by

$$ E_y = \sum_{k=1}^{M} C'' \frac{W_k}{d_k} \left[ \left( J_1^2 \left( \alpha \theta \right)+ J_0^2 \left( \alpha \theta \right) \right) \right]_a \left[ \left( J_1^2 \left( \alpha \theta \right)+ J_0^2 \left( \alpha \theta \right) \right) \right]_b $$  \hspace{1cm} \text{Eqn. 4}

where $\alpha = \frac{2\pi a}{\lambda}$ and $d_k = 2a_k$. $C''$ and $M$ are the constant and number of size increment respectively. $W_k$ is the weight or volume distribution. In processing the detected signal the system assume that the distribution an approximation of Rosin-Rammler distribution in form of

$$ R = e^{-\left( \frac{d}{x} \right)^{w}} $$  \hspace{1cm} \text{Eqn. 5}
where $R$ is the weight fraction contained in a particle of diameter greater than $d$ and $\bar{X}$ and $N$ are the characterising parameters. The initial values of $\bar{X}$ and $N$ are selected and then optimised until the difference between experimental and measured energy distribution is minimised and giving the best fit distribution.

### 3.0 INSTRUMENT APPLICATIONS

Two most commonly used Malvern instruments are the 1800 and 2600 models. Other models that have been developed outside UK come from US (model 2200) and Japan (model T-180). Continuous improvement on previous models an the need to develop a new and more competitive instrument has produce latest version of Malvern instrument called Master Sizer X (henceforth MSX).

One common feature in all previous particle sizing activities or for calibration purposes was that the laser beam diameter is less than 10 mm. More variations were found in selecting the lens focal length which ranging from 63 mm up to 1000 mm. Smaller focal length will register smaller particle size and vice versa. For example, Teixeira (1988) make use of 600 mm focal length to give sizing range from a few microns up to 1128 $\mu$m. With MSX lens focal length of 1000 mm can possibly detecting particle size up to 2000 $\mu$m. Most applications centred around finding the size distribution for either solid phase such as powder (Yamauchi and Ohyama (1982)) or liquid phase for instance fuel spray (Negus and Azzopardi (1978)).

Table 1 shows the summary of user, optical configuration and application of Malvern instrument for the past two decades. Inevitably instrument performance will be compared with other sizing techniques notably of photography and phase Doppler.

### 4.0 PERFORMANCE TEST

Comparison between two different particle sizing methods is inevitable because of the curiosity of differing results obtained at the same operating conditions by those instruments. Earliest systematic record came from photographic technique (for example Hewitt and Whalley (1969)). Assessments on instrument's performance are tabulated
Table 1  User, Optical Set-up and Application of Malvern Instrument

<table>
<thead>
<tr>
<th>Year</th>
<th>Model</th>
<th>Beam Diameter (mm)</th>
<th>Focal Length (mm)</th>
<th>Remark</th>
<th>Researcher</th>
</tr>
</thead>
<tbody>
<tr>
<td>1972</td>
<td>NA</td>
<td>30</td>
<td>250</td>
<td>Particle size analysis using diffraction technique</td>
<td>Cornillault</td>
</tr>
<tr>
<td>1980</td>
<td>1800</td>
<td>6</td>
<td>NA</td>
<td>Particle sizing in annular flow</td>
<td>Azzopardi, Freeman and King</td>
</tr>
<tr>
<td>1982</td>
<td>T-180</td>
<td>6</td>
<td>300</td>
<td>Measurement of particle size distribution</td>
<td>Yamauchi and Ohyama</td>
</tr>
<tr>
<td>1983</td>
<td>1800</td>
<td>NA</td>
<td>NA</td>
<td>Large diameter tube effect on particle size</td>
<td>Azzopardi, Taylor and Gibbon</td>
</tr>
<tr>
<td>1984</td>
<td>2600</td>
<td>NA</td>
<td>NA</td>
<td>Size and velocity measurement in annular two-phase flow</td>
<td>Haddad, Bates, Yeoman and White</td>
</tr>
<tr>
<td>1984</td>
<td>2200</td>
<td>9</td>
<td>63, 100 and 300</td>
<td>Lens size effect on particle size</td>
<td>Hirleman, Oechele and Chigier</td>
</tr>
<tr>
<td>1985</td>
<td>1800</td>
<td>6</td>
<td>NA</td>
<td>Two phase flow drop sizing</td>
<td>Azzopardi</td>
</tr>
<tr>
<td>1987</td>
<td>2600</td>
<td>NA</td>
<td>1000</td>
<td>PDA and laser diffraction: Size comparison on glass bead and spray</td>
<td>Young and Bachalo</td>
</tr>
<tr>
<td>1987</td>
<td>2600</td>
<td>6</td>
<td>NA</td>
<td>Improvement work on Malvern instrument</td>
<td>Miles, King and Sojka</td>
</tr>
<tr>
<td>1987</td>
<td>2600</td>
<td>9</td>
<td>600</td>
<td>Inserts effect on particle size</td>
<td>Teixeira, Azzopardi and Bott</td>
</tr>
<tr>
<td>1988</td>
<td>2600</td>
<td>9</td>
<td>600</td>
<td>Study of turbulence in two-phase flow</td>
<td>Teixeira</td>
</tr>
<tr>
<td>1989</td>
<td>NA</td>
<td>7</td>
<td>NA</td>
<td>Effect of gas properties on drop</td>
<td>Jeppson, Azzopardi and Whalley</td>
</tr>
<tr>
<td>1990</td>
<td>2600</td>
<td>8</td>
<td>300</td>
<td>Correction bias in particle size on spray</td>
<td>Allen and Bakker</td>
</tr>
<tr>
<td>1991</td>
<td>2600 and 3600</td>
<td>NA</td>
<td>316</td>
<td>Size comparison between photograph, laser scattering and diffraction techniques</td>
<td>Hu, Teal and Sheng</td>
</tr>
<tr>
<td>1994</td>
<td>2600 and Master Sizer X</td>
<td>NA</td>
<td>300</td>
<td>Use of Fresnel lens to study aerosol transient behaviour</td>
<td>Richer, Swithenbank and Wedd</td>
</tr>
</tbody>
</table>

NA  Not Available

which includes among other methods such as hot wire anemometer, video imaging, direct shadowing technique, laser diffraction and phase Doppler shift.

Early comparative tests by Negus and Azzopardi (1978) make use of glass spheres (ballotini) and sand (for irregular shape particle). Results in terms of Sauter
mean diameter, \( d_{32} \), were gathered from three different methods, namely, laser diffraction (of Malvern model 1800), photography (of Zeiss Ultraplot projection microscope) and sieving/weighing method. Contrary to initial believes, results from weighing/sieving method were in better agreement with the Malvern than photography with Malvern even though some difficulties came across during the sieving processes. In fact photography results showed bi-modal distribution which was not found in either sieving/weighing or laser diffraction techniques as shown in Fig. 4. The authors emphasise that a single figure \( d_{32} \) of is not likely to describe fully the size distribution since it can be found in several combinations of \( X \) and \( N \) (in the Rosin-Rammler distribution). They also indicated that particle refractive index is independent of the results which is not the case for the latest Malvern instrument model MSX today.

Two-phase annular flow database is also used in comparing some performance of particle sizer (Hadded (1986), Teixeira (1988)). With the development of two-colour particle sizing device (henceforth TCPS)(see Bates et al (1983) and Yeoman et al (1982)), Hadded measured drop size distribution using TCPS and Malvern Model ST1800. A typical histogram is given by Fig. 5 which shows that the TCPS equipment did lack in the dynamic range of droplets (140 \( \mu \)m compared to 240 \( \mu \)m). The sizing range can be extended by developing a non-linear analogue-to-digital converter and preliminary results were encouraging. Nevertheless the registered peaks from both devices are in the same region of around 90-100 \( \mu \)m. Discrepancies between the two is explained by the fact that the TCPS technique is a single particle counter instrument whereas Malvern considers the spatial array of sample distribution.

This point is being repeated by Young and Bachalo (1987) although their data presented not in form of \( d_{32} \) but in volume mean diameter, \( d_{39} \). Three sizing methods were compared here, namely diffraction (Malvern), direct shadowing (Particle Measuring System, Inc. (PMS)) and phase Doppler shift (Aerometrics Inc.). Results showed that the converted temporal sampling data result to spatial one improved the agreement among those three instruments, see Fig. 6, and this led to their conclusion that comparative data should be in an equivalent form to assess the performance correctly.

One large scale comparative study on the difference results obtained from various commercially available particle sizer was reported by Dodge (1987). Fifteen laboratories took part in that study and nine different particle size equipments were employed including PMS (direct shadowing method), KLD (hot wire anemometer), Aerometrics,
Inc. (phase Doppler), Bete (video imaging) and Malvern of model 1800 and 2600 (diffraction technique). The importance of the equivalent point data for comparison purposes was highlighted and deconvoluted procedure of Hammond (1981) is used, converting Malvern ensemble data into equivalent point data. Two types of atomiser used, Parker-Hannifin (PH) and Delavan WDB (DL) of hollow cone simplex-swirl atomiser and solid cone simplex swirl atomiser respectively. Water and calibration fluid for aircraft fuel system (MIL-C-7024 type II) were tested on each of the atomisers. He summarised the overall results by stating that the spray characteristics were reproducible to within 10-15% and iterated that the different measured size from various instruments was not due to the spray reproducibility or theoretical difference in sampling effect but postulated that it was a systematic trend in instrument response of each equipment caused the difference. For Malvern instrument group, the centre-line spray measurement with custom detector appeared to be superior to the Malvern instruments without custom factors. Spray edge variations were dependent on the experimental conditions. Calibrated Malverns were in better agreement compared to the agreement between Aerometrics instruments.

Fig. 4  Comparison of the Malvern, Weighing and Photographic Particle Distributions
Fig. 5  Typical Histogram as Compared by TCPS and Malvern Instrument

Fig. 6  Spatially Converted Data Comparison
Hu et al (1991) devised a light scattering instrument that capable measuring droplet diameter distribution in a two-phase substance. Sizing range for this instrument can be extended to about 1000 μm although their tests were done with range 5 to 200 μm (lens of 300 mm). Results showed that Malvern data were higher when compared with their instrument which was in close agreement with another method of particle sizing using photomicrographic counting technique tested on 2-90 μm glass beads (See Table 2).

Table 2  Comparison of Mean Diameter of Glass Beads by Various Methods

<table>
<thead>
<tr>
<th>Measuring method</th>
<th>Mean diameter</th>
<th>$D_{10}$</th>
<th>$D_{25}$</th>
<th>$D_{50}$</th>
<th>$D_{84}$</th>
<th>$D_{90}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Photomicrographic</td>
<td>43.47</td>
<td>41.98</td>
<td>40.20</td>
<td>40.54</td>
<td>38.66</td>
<td>36.86</td>
</tr>
<tr>
<td>2 Malvern apparatus</td>
<td>48.85</td>
<td>47.70</td>
<td>46.69</td>
<td>46.60</td>
<td>45.65</td>
<td>44.71</td>
</tr>
<tr>
<td>3 Laser scattering</td>
<td>47.28</td>
<td>44.96</td>
<td>42.71</td>
<td>42.75</td>
<td>40.59</td>
<td>38.54</td>
</tr>
<tr>
<td>Measuring I</td>
<td>42.95</td>
<td>39.59</td>
<td>36.35</td>
<td>36.49</td>
<td>34.44</td>
<td>30.65</td>
</tr>
<tr>
<td>Measuring II</td>
<td>40.94</td>
<td>38.80</td>
<td>36.61</td>
<td>36.77</td>
<td>34.62</td>
<td>32.60</td>
</tr>
<tr>
<td>Measuring III</td>
<td>43.72</td>
<td>41.12</td>
<td>38.56</td>
<td>38.67</td>
<td>36.21</td>
<td>33.93</td>
</tr>
<tr>
<td>Mean values</td>
<td>0.58</td>
<td>2.05</td>
<td>4.08</td>
<td>4.61</td>
<td>6.34</td>
<td>7.95</td>
</tr>
</tbody>
</table>

5.0  CALIBRATION AND CORRECTION METHODS

So far the discussions on results variation when using different measuring techniques focus on the data sampling effects, for example line of sight against point measurement or spatial sampling against temporal sampling. Malvern particle sizer is under scrutiny because instrument-to-instrument size measurement differs. Although the difference between experimental and theoretical data is expected in any scientific endeavour, discrepancies between instruments of the same type raise the question of how effective the calibration is and what correction method should be adopted.

Deficiencies of instrument-to-instrument variation can be minimise if the possible error sources identity are verified. For example, approximation method is one source where Hodkinson (1966) proposed that Fraunhoffer diffraction theory could be use to approximate Mie theory in measuring particle size. With this ground work, Boron and
Waldie (1978) quantified the theoretical error involved and concluded that the errors oscillate within 40%, see Fig. 7 when testing the polystyrene latex particles (size of 0.5 \( \mu \)m to 3.2 \( \mu \)m) suspended in air and water using forward scattered intensity ratio technique. Response curve oscillation in the small particle region of the particle distribution is expected if using Mie theory.

![Efficiency of Rings 29 & 30](image)

**Fig. 7** Possible Error due to Approximation

Dodge (1984) briefly reviewed a calibration procedure as a result of Malvern instrument-to-instrument variation which he reckon due to the variations in responsivities of the detector assemblies. Each instrument has a detector assembly consisting of 30 annular detectors arranged coaxially. With none particle in the sample volumes all the incident light fall onto the central detectors. When particles exist in the sample volume, some fraction of incident light will be scattered onto the annular detectors surrounding the central detector. A systematic increase or decreases in detector responsivity in progressing from the inner to outer detectors causes the computed size distribution to be smaller or larger, respectively, than the actual distribution. To correct this situation is to calibrate each detector responsivity and then correct the responsivities feed of the computer model (that come with the instrument). To do this the detector assembly needs to be removed and placed in a position so that it can be illuminated with uniform light. The responsivity of each 30 detectors may be calculated by using several illumination levels and correcting the detector areas.
The calibrated results were verified using two standard reticles (of RR-50-2.0-0.03-102-CF-#74 and RR-50-3.0-0.08-102-CF-#115). The reticles do not follow a Rosin-Rammler distribution exactly and have discrete sizes. Thus, there is some discrepancy between the actual $d_{15}$ for the distribution and the $d_{15}$ that is calculated from the best fit $\bar{X}$ and $N$ for a Rosin-Rammler distribution. Before calibration the $\bar{X}$ were high by 14 to 24% and $d_{15}$ by 13 to 22%. After calibration the instrument accuracy is within the calibration reticle accuracy.

The second main problem that appeared during Malvern instrument application was vignetting effect. Vignetting is the effect of exceeding maximum allowable distance between the light scattering medium and the receiver lens. It is a function of lens diameter and its focal length. Any measurement that experiences vignetting effect will have a bias result skewing to the larger particles. Hirleman et al (1984) did study the lens and optical sample volume position effect on the response characteristics of Malvern instrument. Using a geometrical relation and assuming the scattering angle is small, an expression is derived to predict the onset of the vignetting point and given by

$$z_p = \frac{f(d_l - 2r_p)}{2r_{oj}} \quad \text{Eqn. 6}$$

where $z_p$ is the sample volume from the lens, $r_p$ is the sample volume from the optical axis. The focal length of the denoted by $f$ and $d_l$ is the receiving lens diameter and $r_{oj}$ is the outer radii of the $j^{th}$ annular ring detector. The vignetting effect is negligible and the above equation fails when there is not enough scattered energy on the detector element.

Dodge (1984) developed a calibration technique for Malvern instrument because no method is available then to check the calibration or recalibrate the instrument if the data is in error. He used a calibration reticle developed by Hirleman (1984) to verify his hypothesis. The equation for maximum allowable distance is the same as given by Hirleman, see Eqn. 6. With 300 mm lens he reported that the total sample volume should be 336 mm of the lens. He also mentioned about other work on vignetting effect (References 7 and 8 of Dodge (1984)) but both analyses lacking the important laser beam diameter. Wild and Switchenbank (1986) proposed placing a beam stop to avoid multiple scattering and vignetting effect. By defining efficiency as the ratio of energy collected on a ring at point P to the energy collected on a ring at point P from a particle field of the
same extent but using infinite lens aperture, they compared experimental data with prediction of Hamidi and Swithinbank (1985) and the result is in good agreement. See Fig. 8. The latest development on resolving the vignetting effect is reported by Richer et al (1994) where they reported that by using Fresnel lens (to the MSX and HSD2600 Malvern model) maximum allowable distance can be extended further than before.

![Efficiency of Rings 29 & 30](image)

**Fig. 8  Effect of Vignetting (Comparison between Experiment and Theory)**

The multiple scattering of particles is another problem that has been under constant examination. Early Malvern model could not response to this problem which occurred in a dense spray where the scattering angle is higher caused the appearance of smaller particle diameter than the actual one. This error will gives a broader size distribution than it was. Dodge (1984) looked into this problem and suggesting a correction scheme experimentally. With seven nozzles (of Delavan and Hugo), the procedure started with first nozzle operating alone and then two at a time and so on until all the nozzles operating simultaneously. This means the laser beam obscuration is at its highest or laser beam transmission at its lowest with all the nozzles operating. A correction factor is introduced as defined as the measured value divided by the dilute spray value (transmission =1). The observed data were fitted to some equation for all data available which was given by
where \( d_{320} \) is \( d_{32} \) for dilute spray, \( T \) is the transmission factor and \( a \), \( b \) and \( c \) are the constants. A table of best fit parameter applied to \( N = 2 \) and \( d_{32} \) ranging from 20 to 60 \( \mu \)m is also given and he proposed that the correction factor should be only used if the transmission unscattered light is less than about 50\% (see Table 3).

Before that Gomi (1986) adopted a numerical solution to correct multiple scattering problem using ray tracing method and resulting in quite early multiple scattering onset at 90\% transmission. He showed that Malvern measurement at 30\% transmission with water/gas spray would give \( d_{32} \) of 38 \( \mu \)m and when considering his multiple scattering model increases \( d_{32} \) to 47 \( \mu \)m. Hamidi and Swithinbank (1987) continued to work on multiple scattering problem and resolved it mathematically by considering both Fraunhoffer and anomalous diffraction theories. Using data obtained by Dodge (1984), they produced Fig. 9 which shows the effect of particle size (\( d_{32} \)) with and without scattering multiple in place. Tests on the bi-modal distribution were conducted to illustrate the method general application and typical result is shown in Fig. 10.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>a</th>
<th>b</th>
<th>c</th>
</tr>
</thead>
<tbody>
<tr>
<td>SMD</td>
<td>0.9456</td>
<td>3.811</td>
<td>0.0204</td>
</tr>
<tr>
<td>N</td>
<td>0.4264</td>
<td>3.672</td>
<td>0.0130</td>
</tr>
</tbody>
</table>
Fig. 9  Results With and Without Correction for Multiple Scattering (Data of Dodge (1984))

Fig. 10  Size Distribution Comparison 87% Obscuration
REFERENCES


