Investigations on non-Spherical Reference Material Using Laser Diffraction and Dynamic Image Analysis

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ABSTRACT

For shape and size characterisation Sympatec has introduced the QICPIC dynamic image analysis system. The dispersers for dry powders and suspensions are modular and interchangeable between QICPIC and HELOS laser diffraction systems. QICPIC can handle particle numbers of more than 10^8 particles per measurement and its results can reach a statistical relevance comparable to that of laser diffraction.

The direct comparison of image analysis and laser diffraction measurements under identical particle dispersing conditions is now possible. For arbitrarily shaped particles an equivalent HELOS laser diffraction signal is calculated from the images of the QICPIC measurement. This approach solves the fundamental problem of how to quantify the influence of the infinite number of appearances of particle shapes on laser diffraction results. With this method the absolute scale of both systems may now be verified by direct signal comparison, even without relying on evaluation modes or inversion procedures.

Keywords: Laser Diffraction, High Speed Image Analysis, Reference Material, Particle Size

1 Introduction

In all laser diffraction (LD) systems a radial variation of the intensity pattern of the diffracted light is recorded. It is generally difficult to interpret the relationship between the diffraction pattern obtained and the equivalent size distribution of spherical in the case of non-spherical particles. Average azimuthal variations of the intensity pattern only exist if non-spherical particles are aligned with the medium flow in the cuvette. The azimuthal dependency can be detected with the help of wedge shaped photo elements. Some extensions of LD systems are available which calculate a size distribution and some shape characteristics of particles from this data (Heffels 1995). But the signal depends strongly on the flow condition of the dispersing system.

Our strategy is to stay with the classical concept of LD and to accept the influence of the particle shape on the result. The orientation dependence within the projection plane should be eliminated with a semi-circular detector. If shape information should be detected, an image analysis (IA) system shall be used. Until now the comparison of IA with LD has been limited because of differences in dispersion and average particle orientation. In many IA systems the particles are statically observed on glass slides, whereas free flowing particles are used in an LD system. Because particle detection had been time consuming in the past and because of low frame rates of standard imaging devices only a few thousand particles had been measured.

At PARTEC 2004 a new concept of digital IA was presented (Witt 2004), combining for the first time high-speed image analysis with powerful dry dispersion in a table-top instrument. The short exposure time of 1 ns makes it possible to apply the same dispersing devices which have been developed for the standard laser diffraction line of instruments (figure 1). The high frame rate of up to 500 fps at full resolution of 1024x1024 pixels and the fast handling of large particle numbers per measurement ($>10^8$) fundamentally overcomes the weakness of typical image analysis systems – low particle numbers resulting in large statistical errors (Witt 2005). Now for the first time image analysis data is available with the same statistical significance as of LD results. The results of both instruments can be obtained under identical dispersion conditions. A direct comparison of both methods is possible.

Figure 1: OASIS wet (SUCELL) and dry (RODOS) dispersing system. Left: Set-up in the QICPIC image analysis sensor. Right: Set-up in the HELOS/BF laser diffraction sensor.

2 LINK BETWEEN THE TWO METHODS

2.1 Comparison of detection principle

The optical set-up of the QICPIC image analysis system and the HELOS laser diffraction system is in principle very similar (figure 2). In both systems a parallel beam of light is created by an adaptable beam expansion unit. This beam of light is directed at the measuring zone of the dispersing system. In the LD system a lens transforms the diffracted light to a diffraction pattern which is recorded by a multi-element photo detector. In an IA system the full amplitude and phase distribution of the diffraction pattern is back-transformed to a real image, which is recorded by the image sensor.

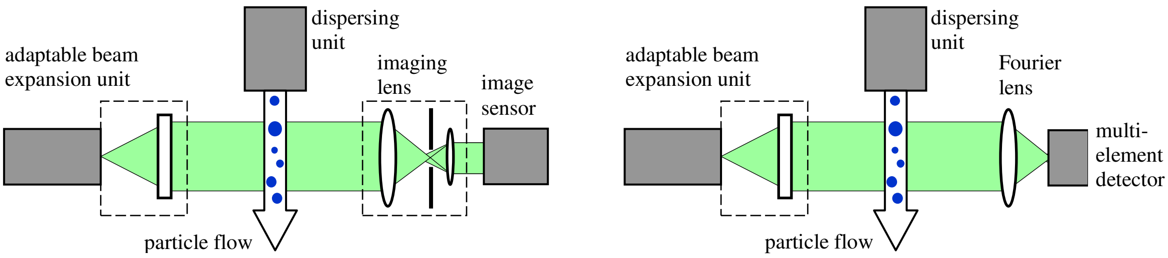

Figure 2: Optical set-up of the QICPIC image analysis sensor (left) and of the HELOS laser diffraction sensor (right).

To measure the particle size in SI length units, the IA system is calibrated with a certified standard scale. The effective magnification of the imaging lens and the size of the sensor are measured and are thus traceable back to the standard metre. In practice the calibration is a small correction to the nominal magnification of the optical design. Like an IA system a laser diffraction system is based on first principles. To measure the particle size in SI length units, the wavelength of the light, the scale of the detector and the exact focal length of the system must be known.

With IA and LD Systems it is still necessary to qualify the whole system (sensor and disperser) with the help of reference materials (RM). This will confirm the correct scale of the instrument and reveal misalignment, optical defects or malfunction of the dispersing or feeding system. Since 1992 we have introduced a variety of non-spherical RMs in the size range from 0.1 µm to above 1 mm. For the stable silicon carbides (SiC) relative standard deviations σ have been determined to $\sigma < 0.01\%$ for the same sample, about 0.3% for different samples and it was possible to improve the system-to-system comparability of the HELOS LD PSA systems to below 1% (including sampling errors) by introducing these RMs as the final check in our systems integration.

2.2 Comparison of the evaluation modes

The LD technique assumes a spherical particle shape in its optical model. For non-spherical particles a size distribution is reported in which the predicted diffraction pattern for the volumetric sum of spherical particles matches the measured diffraction pattern. Many different diameters and evaluation modes may be selected as an evaluation mode within the software of IA systems. The equivalent projection area of a circle (EQPC) is assumed to give the best agreement with LD. In theory both evaluation modes will give the same results if the particles are spherical. If the particles are non-spherical a bias may be observed. For lager particles it is possible to quantify this bias because there is a direct relation between the diffraction pattern and the so called “object function” of the particles which is recorded as an image.

Fraunhofer diffraction is a well known model for describing forward light scattering by opaque particles at a large distance compared to the size of the particle. Analytical solutions exist for the Fraunhofer diffraction integral at circular or rectangular shaped objects. For general shapes we write the diffraction integral as a Fourier integral in terms of (x, y)-coordinates in the object plane and $(\tilde{x}, \tilde{y})$-coordinates in the focal plane of a lens with a focal length of f.

$$U\left(\frac{\tilde{x}}{\lambda f}, \frac{\tilde{y}}{\lambda f}\right) = C \cdot \iint G(x, y) e^{-2\pi i\left(\frac{\tilde{x}}{\lambda f}x + \frac{\tilde{y}}{\lambda f}y\right)} dxdy \qquad (1)$$

The term $\tilde{x}/f$ is an approximation of the direction cosine of the wave vector, λ is the wavelength of the light and C is constant. The object function $G(x, y)$ represents the spatial transmission function of a two-dimensional object, like a slide with a distribution of particles on it. If an opaque particle is present at (x, y) then $G(x, y) = 0$, and $G(x, y) = 1$ in the case of no particle. The result U of the transform represents the scalar complex field at the detector. The diffraction pattern of the intensity is the square of the field’s amplitude.

A binary digital image represents a sampled and discretised form of the object function which is scaled to the object plane with the optical magnification M. We can now simulate diffraction for a large amount of real particles in different orientations recorded by an imaging system with the help of the Discrete Fourier Transform. An equivalent LD signal can then be calculated by applying a digital representation of the detector geometry. The scales of both systems (M and f) which have been set-up with absolute methods can now be compared by direct signal comparison without even relying on evaluation modes.

2.3 Instrument set-up and experimental conditions

The QICPIC IA system was calibrated with a certified standard scale. In previous investigations (Koehler 2007) we have aligned the laser diffraction instrument with the help of spherical material to the IA results to obtain best agreement of the results. Now we use a single lens for which it was possible to calculate the location of its principle plane from optical design data. The laser diffraction sensor was placed at the minimum focus diameter. The distance from the principal plane to the detector surface was determined with the help of a large calliper gauge. The tolerance is less than 0.5 mm at a nominal focal length of 1000 mm.

The same dispersing systems and conditions were used for both IA and LD. In IA the depth of focus must be considered carefully. We used a cuvette for IA with an optical path length of 1 mm, and the same cuvette for the LD measurements. In LD, contamination of liquid is subtracted as a background signal with the help of a reference measurement. In IA a particle filter on shape and size may be applied to recognize and eliminate air bubbles but such a filter was not used in the experiment.

It was not possible to use the same optical concentration. LD requires a strong and reliable signal and therefore relatively high particle concentrations. In contrast, IA requires that overlapping particles must be strictly avoided. In IA the optical concentration is defined as a geometrical obscuration. In LD systems it is defined as the extinction of the laser beam in focus. According to the Fraunhofer diffraction theory this value is twice the geometrical obscuration of an IA system.

3 RESULTS

3.1 Comparison with spherical particles

Even in the case of a monodisperse sample the LD result will be a distribution of sizes because smoothing constraints are commonly required in the inversion procedure. Thus it is advisable to compare polydisperse particle samples having size distributions within at least three size classes of the LD system. Opaque spherical glassy carbon particles having a narrow size distribution have been selected for the first comparison between QICPIC and HELOS sensors. The result of the laser diffraction analysis is obtained with the advanced HRLD iterative method. A standard Phillips-Twomey inversion method will show a broader distribution. The IA evaluation is based on the equivalent projection area of a circle (EQPC).

The particle size distributions are presented in figure 3. For spherical particles the results of both evaluation methods are nearly identical. They show a very good correspondence between the image scale and the focal length of the LD lens. The correspondence of both techniques can also be cross-checked by direct comparison of the measured diffraction pattern with the simulated pattern from IA data. A difference in scale of the focal length of the HELOS compared to the magnification of the QICPIC is then observed as a relative shift between both patterns.

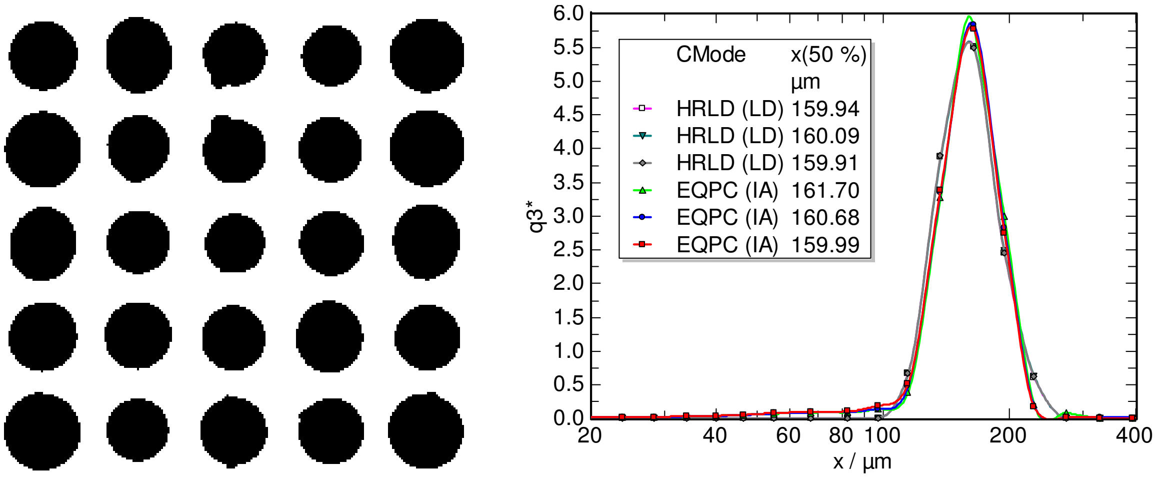

Figure 3: Size distribution of opaque spherical glassy carbon particles. Sample images are shown above. The HELOS (LD) result is obtained with the HRLD evaluation mode. The QICPIC (IA) result is obtained with the EQPC evaluation mode.

3.2 Comparison with irregular particles

Irregular silicon carbide particles are used by Sympatec as an internal RM to certify and recertify IA and LD sensors according to Sympatec's own specifications. The main advantage compared to spherical material is more realistic wet and dry particle feeding characteristics, resulting in a better overall system test. A huge database of results is available at Sympatec and the long-term availability is guaranteed. With the introduction of the QICPIC small but noticeable differences in the results of LD compared with IA have been observed. A very small amount of these differences may be attributed to differences in feeding and particle orientation, but even if exactly the same dispersing conditions are used the results are not identical (figure 4, left). This is an experimental demonstration that for non-spherical particles the EQPC and the equivalent spherical diameter measured by LD are not the same.

To overcome this fundamental problem more than 10000 images of the same QICPIC measurement are converted by a Fast Fourier Transform after removal of particles which touch the border. The results are accumulated to an equivalent laser diffraction signal. These values are passed through the LD inversion algorithm. This procedure leads to a very good agreement of both results (figure 4, right).

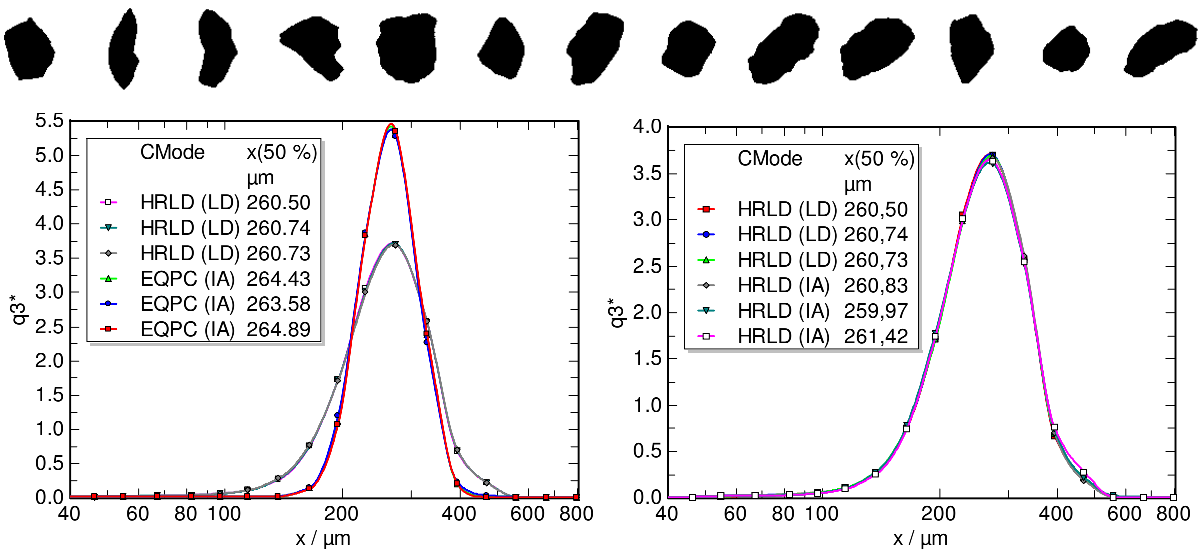

Figure 4: Size distribution of irregular SiC-P80 particles. Sample images are shown above. In the left diagram the HELOS (LD) results are obtained with the HRLD evaluation mode. The QICPIC (IA) results are obtained with the EQPC evaluation mode. On the right diagram the QICPIC (IA) results are obtained after calculating an equivalent diffraction pattern by Fast Fourier Transform and using the HRLD laser diffraction evaluation mode.

4 CONCLUSION

A non spherical material has been characterised as an equivalent laser diffraction result through use of transformed image analysis data. This approach has finally solved the fundamental problem of how to handle the influence of the infinite number of appearances of particle shapes on LD results. The results show that the observed main differences between LD and IA arise primarily from the effect of the particle shape on the evaluation procedure.

This data has been obtained by the QICPIC image analysis sensor, where the image scale is calibrated by a certified standard. It is a common opinion that a laser diffraction instrument can only be traced back correctly with spherical reference particles. But for these experiments the LD system has been set-up by a traceable method. With respect to the presented comparison it is obvious that LD as well as IA are suitable for the characterization of RM as a primary method. It is also completely valid to prove the performance of individual HELOS LD instruments with irregular shaped particles since the properties of that material can be characterized for each combination of sensor and dispersing system by an absolute method.

5 REFERENCES

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