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PARTICLE SIZE DISTRIBUTION MEASUREMENT BY STATISTICAL EVALUATION OF LIGHT EXTINCTION SIGNALS

Common extinction photometers deliver the product of particle concentration and particle size. Small measuring zones allow us to measure additionally the standard deviation of the fluctuating extinction signal. For monodisperse particles the particle size and particle concentration can be calculated from the measured mean value and standard deviation of the extinction signal. The determination of particle size distribution is not possible.

A variation in the measuring conditions allows us also to detect the effects of the polydispersity of a particle system using this technique. The variation in the size of the measuring light beam influences the measurable standard deviation of the extinction signal. A reduction of the beam size leads to an increased standard deviation and the particle size-depending amount of particles in the border zone of the light beam that is not completely illuminated. These particle size-depending effects allow the measurement of a particle size distribution.

SYMBOLS

A_{meas} – cross-section of the light beam,
B_{meas} – width of the light beam,
E – extinction,
K_{ext} – extinction coefficient,
m – relative refractive index,
q – density distribution of particle size,
x – particle size,
λ – wavelength,
A_p – geometric particle projection area,
C_{ext} – extinction cross-section,
H_{meas} – height of the light beam,
$\overline{N}$ – expected particle number in the measuring volume,
P_B – presence probability of a particle in the border zone,
T – transmission,
σ_T – standard deviation of extinction signals.

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1. INTRODUCTION

Optical principles of particle characteristics are suitable for many industrial and scientifical applications because the measurement is carried out without mechanical contact or any other physical influence on the particle system. For particle characteristics or monitoring on an industrial scale, the extinction principle is a useful, simple and reliable method.

Ordinary extinction photometers deliver the product of the particle concentration and particle size being known. Small measuring zones additionally allow us to measure the standard deviation of the fluctuating extinction signal. For monodisperse particles, the particle size and particle concentration can be calculated from the mean value measured and standard deviation of the extinction signal [1], [2]. The determination of the particle size distribution is not possible.

In order to detect additionally the effects of the polydispersity of a particle system, using this technique, the measuring conditions have to be varied. The method presented in this paper consists in using a defined variation of the size of the measuring light beam. This size variation influences the measurable standard deviation of the extinction signal and principally allows the determination of the particle size distribution. This paper presents a model enabling us to calculate the expected value and standard deviation of fluctuating transmission signals for the given particle systems, particle concentrations and sensor parameters, e.g. the beam size of the illumination system. Furthermore, a measuring setup and the first experimental results will be presented.

2. MODELLING THE EXPECTED VALUE AND STANDARD DEVIATION OF FLUCTUATING, COINCIDENT TRANSMISSION SIGNALS

2.1. EXPECTED VALUE OF TRANSMISSION

The Lambert–Beer's law may be used to describe the light transmission T in dispersed systems:

$$T(x, N, \lambda, m) = \frac{I}{I_0} = e^{-\frac{\overline{N} K_{\text{ext}}(x,\lambda,m) A_p(x)}{A_{\text{meas}}}}, \tag{1}$$

where:

I – transmitted light intensity,

I_0 – incident light intensity.

Equation (1) is valid for many practical applications. Its correction is necessary if the signal is influenced by multiply scattered light or particle–particle interactions.

2.2. STANDARD DEVIATION OF TRANSMISSION

2.2.1. STANDARD DEVIATION OF TRANSMISSION BASED ON PARTICLE NUMBER FLUCTUATIONS

A simple model of the standard deviation of transmission signals is based on the effect of particle fluctuations in a volume element in a time element. The presence probability of N particles in the light beam can be described by a binominal distribution. Very important is the fact that the standard deviation of the particle number in a volume element is equal to the square root of the expected particle number. A combination of that fact with the Lambert–Beer's law gives the following equation of the standard deviation of transmission [1], [2]:

$$\sigma_T = \frac{T\left(N-\sqrt{N}\right)-T\left(N+\sqrt{N}\right)}{2} = \dots = e^{-\frac{\overline{N} K_{\text{ext}} A_p}{A_{\text{meas}}}} \cdot \sinh\left(\frac{\sqrt{N} K_{\text{ext}} A_p}{A_{\text{meas}}}\right). \tag{2}$$

Principally, the standard deviation σ_T and the expected value of transmission signals allow us to detect a mean particle size and concentration simultaneously. Figure 1 shows a characteristic field for different particle sizes and concentrations. In practice, this simple model is erroneous in the range from 5 to 25%. However, measurements in dispersed and flocculated particle systems have shown interesting results and a profitable application to industrial processes [3].

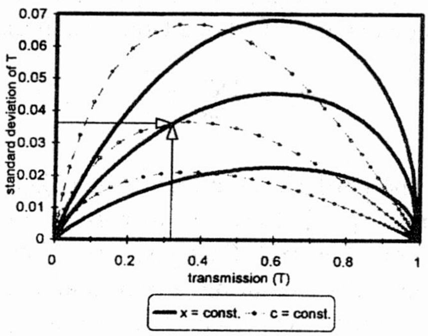

Fig. 1. T and σ_T for different particle sizes and concentrations
c – particle concentration

2.2.2. CONSIDERATION OF FURTHER EFFECTS

To reduce the errors of the model of σ_T, the most important signal effects have to be considered. Besides the effect of the particle number fluctuation described above, the following effects influence the standard deviation T (see also figure 2):

- border zone effects,
- effects of particle overlapping,
- effects of polydispersity.

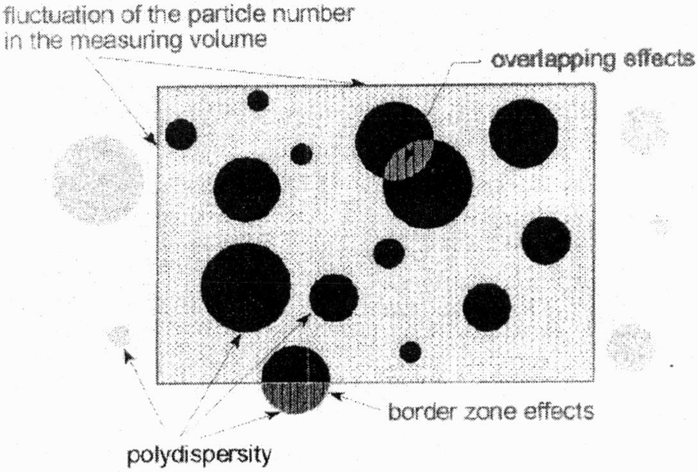

Fig. 2. Effects of transmission fluctuations in disperse systems

The model presented here is rating the signal effects individually. As the influence of each effect on the transmission signal is independent of each other, the standard deviation of the transmission can be calculated from the single effect as follows:

$$\sigma_T = \sqrt{\sum_i \sigma_{T;i}^2} \; . \tag{3}$$

Border zone effects. The border zone effect, as it is known due to single particle counters or image processing [4], [5], decreases the particle projection area in the border zone of the measuring volume. Figure 3 illustrates this effect. A reduced mean projection area of a single particle in the measuring volume and an apparent polydispersity of the particles are caused by border zone effects. The presence probability of a particle in the border zone is equal to the ratio of the border zone area to the whole cross-section, where the particles may be in contact with the light beam (eq. (4)). Equation 4 allows us to calculate the mean particle projection area in the measuring volume:

$$\overline{A}_p = [GP_B + (1 - P_B)]\, A_p \, . \tag{4}$$

The coefficient G describes the part of a particle projection area that is illuminated in the border zone. Its value is approximately 0.5. The mean particle number, which may be inside measuring volume which includes the border zone, is also important:

$$\overline{N}_{\text{eff}} = -\ln\left(\overline{T}\right)\frac{\left(H_{\text{meas}}B_{\text{meas}}\right)}{A_p} \approx \frac{\left(H_{\text{meas}}+x\right)\left(B_{\text{meas}}+x\right)}{H_{\text{meas}}B_{\text{meas}}}\overline{N}, \tag{5}$$

where N_{eff} denotes the effective particle number inside the measuring volume.

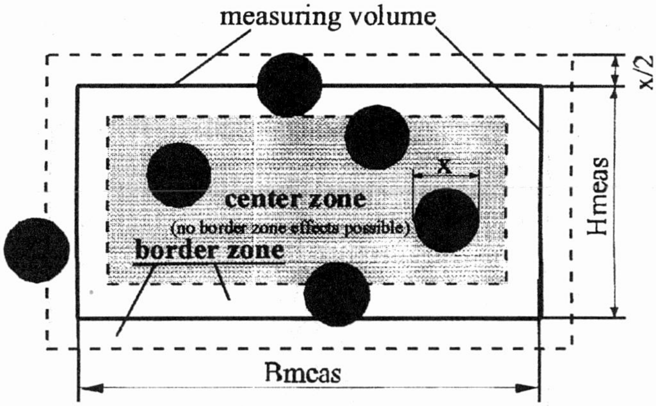

Fig. 3. Border zone effects of particles in a light beam

Now the standard deviation of transmission caused by the reduced projection area of a single particle in the measuring volume can be formulated by eq. (6):

$$\sigma_{T;B} = \overline{T}\sinh\left(\sqrt{P_B\overline{N}_{\text{eff}}}\;\frac{GA_b}{A_{\text{meas}}}\right), \tag{6}$$

where $\sigma_{T;B}$ is the standard deviation of transmission including border zone effects.

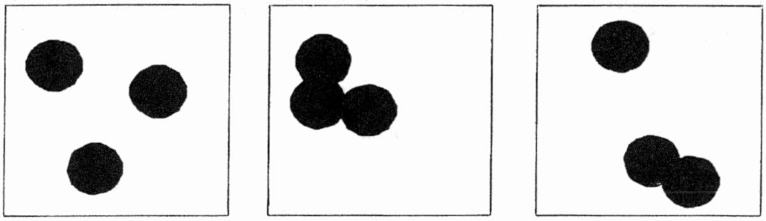

Fig. 4. Particles arranged randomly

The effect of the apparent polydispersity of the particles will not be described here, but it is considered in all calculations presented in this paper.

Overlapping effects. Figure 4 gives an impression of a random arrangement of three particles in a measuring volume. A change in the arrangement of a fixed particle number in a measuring volume also causes a change of the transmission. This effect cannot directly be detected by measurements. Therefore, computer simulations were necessary to investigate the overlapping effects. In figure 5, a typical result of a computer simulation for a given particle size and concentration is shown. The density distribution of the transmission is plotted for a simulation of overlapping effects (triangle), the complete transmission signal (rectangle) and the discrete value of the Lambert–Beer's law.

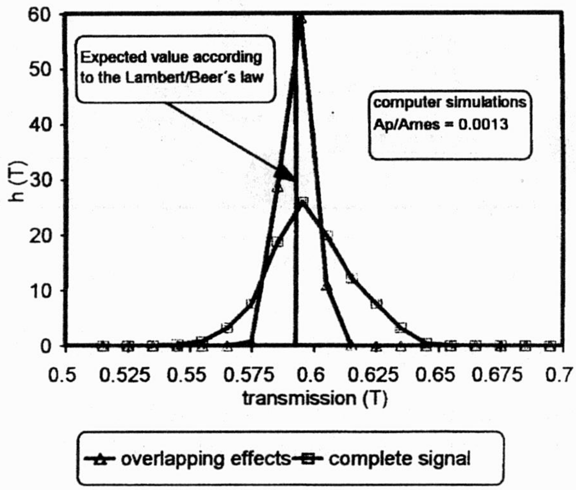

Fig. 5. Density distribution of transmission of overlapping effects, the complete signal and the Lambert–Beer's law

An empirical relationship between the standard deviation of the transmission signal caused by overlapping effects and the complete signal (all effects) was found by computer simulations:

$$\sigma_{T;\text{ovl}} \approx \sigma_{T;\text{means}} \frac{\sqrt{E}}{2}, \tag{7}$$

where:

$\sigma_{T;\text{ovl}}$ – standard deviation of transmission caused by overlapping effects,

$\sigma_{T;\text{meas}}$ – measured standard deviation of transmission.

An influence of the particle size was not observed with the simulation.

Polydispersity. The transmission signal in polydisperse systems is composed of the quantities of all particle size fractions which are present in the measuring volume. In case of spherical, polydisperse particles, the Lambert–Beer's law has the following form:

$$\ln\left(\frac{1}{T}\right) = E = \frac{\pi}{4} c_N L \int_{x_{\min}}^{x_{\max}} K_{\text{ext}}(x) x^2 q_0(x) dx\,, \tag{8}$$

where:

c_N – particle number concentration,

L – cuvette path length,

q_0 – density distribution by number.

The integral in eq. (8) describes the mean extinction cross-section of the polydisperse system. If a small measuring volume is used to obtain detectable transmission fluctuations, the number of particles illuminated becomes smaller. The polydispersity of the illuminated "particle sample" is changing for each measuring value. This effect principally allows a determination of the particle size distribution. It is possible to estimate the standard deviation of the transmission that is caused by polydispersity. Here, the polydispersity is simplified to the standard deviation of the particle cross-section:

$$\sigma_{T;A_p} = \overline{T} \sinh\left(\sqrt{N_{\text{eff}}} \frac{\sigma_{A_p}}{A_{\text{meas}}}\right), \tag{9}$$

where:

$\sigma_{T;A}$ – standard deviation of transmission caused by polydispersity,

σ_{A_p} – polydispersity of the particle system (standard deviation of the particle projection area).

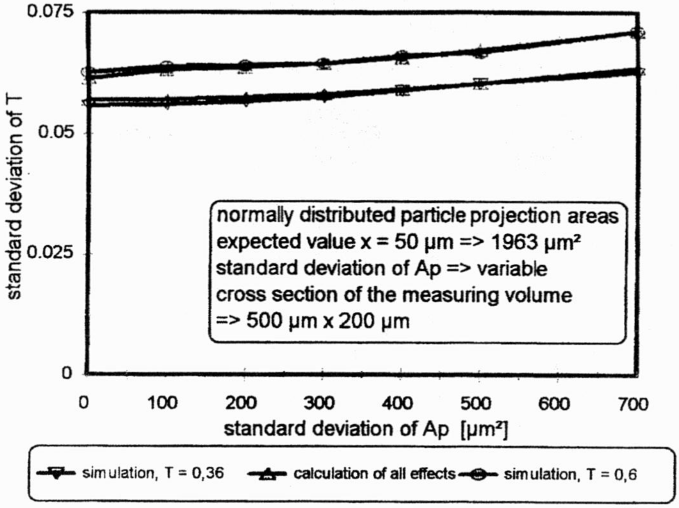

Fig. 6. Comparison of computer simulated and calculated (eq. (2)) standard deviations of transmission for different concentrations and polydispersities

A comparison of computer simulated (circle) and calculated (triangle) standard deviations of transmission is shown in figure 6 for different values of concentration and polydispersity. There was found a good agreement between the computer simulations and the modelling of important causes of transmission fluctuations.

3. A METHOD FOR THE MEASUREMENT OF PARTICLE SIZE DISTRIBUTION

3.1. PRINCIPLE

As mentioned before, the mean extinction cross-section depends on the dimensions of the measuring volume. This effect increases if the particles are larger than the height or the width of the measuring volume.

Transmission measurement with variable sizes of the illuminating light beam allows us to detect a characteristic extinction cross-section of a particle system for each size of the measuring light beam. This effect is shown in figure 7 for a variable beam height.

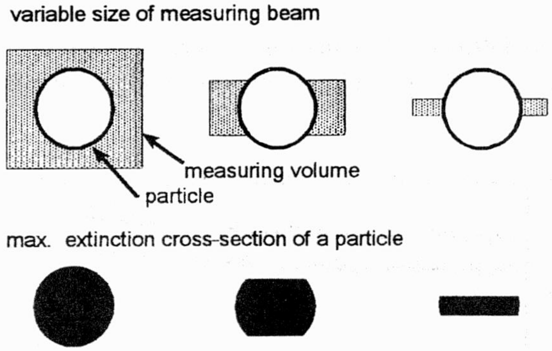

Fig. 7. Maximum particle projection area for different sizes of the illuminating light beam

In figure 8, the maximum particle projection area of three particle sizes is plotted versus the height of the light beam according to figure 7.

The mean extinction cross-section of a polydisperse particle system can be calculated, depending on the number density distribution $q_0(x)$ of the particles and the cross-section of the light beam (A_{meas}):

$$C_{ext}(A_{meas}) = \int_{x_{min}}^{x_{max}} q_0(x) C_{ext}(A_{meas}, x) dx . \quad (10)$$

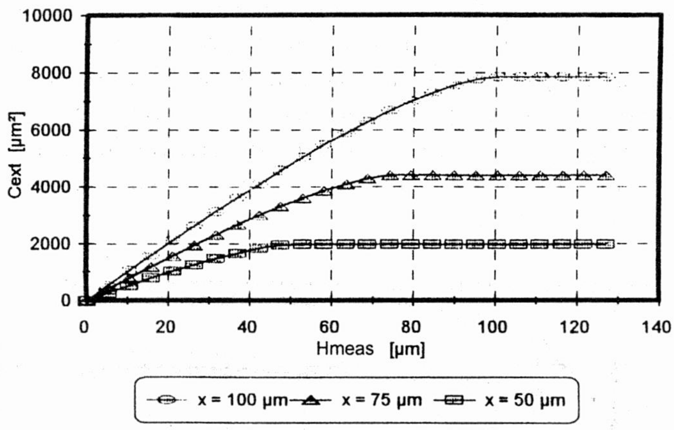

Fig. 8. Maximum extinction cross-sections of different particle sizes versus the height of the illuminating light beam (B_{meas} = 610 µm = const.)

The relationship shown in equation (10) is a Fredholm integral equation of the first kind. This kind of equation can be solved for different boundary conditions, e.g. non-negative or non-oscillating solutions, or solutions of a given type. Therefore all kinds of methods are available.

3.2. EXPERIMENTAL RESULTS

For practical investigations, a small-angle photometer was used. The setup is shown in figure 9. The variation of the size of the light beam was realised by an axially shiftable flow cell (K) in the combination with a convergent laser beam (D-photodetector).

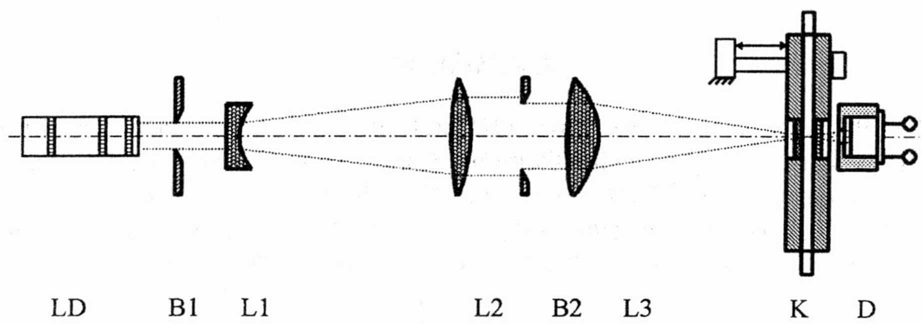

Fig. 9. Optical setup for extinction measurements with a variable beam size
LD – laserdiode 680 nm, B1 – aperture 1 mm, L1 – lens f = –10 mm,
L2 – lens f = 150 mm, B2 – aperture 8 mm, L3 – lens f = 80 mm

The measurements of $\overline{T}$ and σ_T were carried out with several quasi-monodisperse particle fractions and different beam sizes. In figure 10, the measured extinction cross-sections versus the particle size and the axial flow cell position in the light beam are plotted. Several mixtures of quasi-monodisperse particles were tested. A mixture of 1.09 μm and 43 μm particles shows distinct differences in comparison to quasi-monodisperse particles in the characteristic of the detected extinction cross-section for variable measuring volumes.

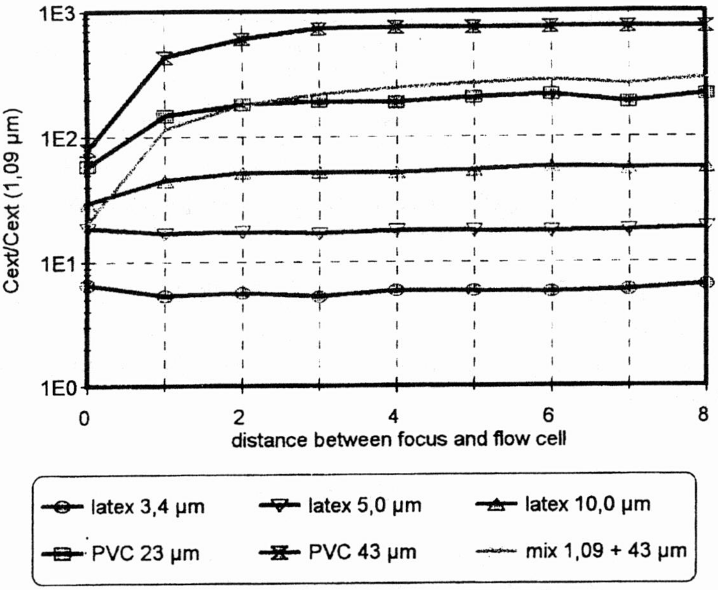

Fig. 10. Results of measurements with quasi-monodisperse particles compared to a mixture of 1.09 μm and 43 μm particles

4. SUMMARY

A principle of optical particle size measurement has been presented, which uses a statistical evaluation of fluctuating light transmission signals. In an extended model for the measurable standard deviation of transmission, the border zone effects, the overlapping effects and the polydispersity of a particle system are taken into account. The determination of a particle size distribution from the measuring data seems possible.

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OKREŚLENIE ROZKŁADU WIELKOŚCI CZĄSTEK METODĄ STATYSTYCZNEJ OCENY SYGNAŁÓW EKSTYNKCJI ŚWIETLNEJ

Standardowe fotometry ekstynkcyjne pozwalają określić stężenia cząstek i ich rozmiary. Wąskie przedziały pomiarowe umożliwiają dodatkowo wyznaczenie odchylenia standardowego zmiennego sygnału ekstynkcji. Gdy cząstki mają charakter monodyspersyjny, ich rozmiar i stężenie można obliczyć na podstawie zmierzonej wartości średniej i odchylenia standardowego sygnału ekstynkcji. Określenie rozkładu wielkości cząstek nie jest możliwe.

Modyfikacja warunków pomiarowych pozwala uwzględnić efekty polidyspersyjności systemu. Zmiana wielkości mierzonej wiązki promieniowania świetlnego wpływa na wielkość odchylenia standardowego sygnału ekstynkcji. Gdy zmniejsza się rozmiary wiązki promieniowania, wtedy zwiększa się wartość odchylenia standardowego i liczba cząstek w strefie granicznej promieniowania świetlnego, która nie jest całkowicie oświetlona. Te zależne od rozmiarów cząstek efekty pozwalają określić rozkład wielkości cząstek.