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The inclusion-concentration measurement of suspension droplets based on Monte Carlo ray tracing

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Abstract
In this paper we analyse suspension droplet light scattering with respect to measuring the size of the host particle and the inclusion concentration. The light-scattering simulations show significant changes in the scattering distribution of suspension droplets with different inclusion concentrations. The evaluation reduces to only two parameters, namely angular fringe spacing and the slope in the scattering domain 30–70°. This method relies on the simulation of scattering with parameters such as the refractive indices of the host and the inclusions and the size of the inclusions.

Keywords: particle size, inclusion concentration, optical characterization

1. Introduction

Optical techniques for particle characterization are of great interest in a variety of engineering applications. They are non-intrusive and can be used for on-line process control. Emulsions and suspensions are utilized in several industrial applications. In chemical engineering unit operations such as spray drying or flue-gas scrubbing inhomogeneous droplets are generated so as to increase the area which is involved in the heat or mass transfer. The inclusion concentration, which is a quantity of enrichment, and the droplet size are of interest.

In this paper we present a method for the evaluation of the inclusion concentration of inhomogeneous particles. This method relies on knowledge of other important particle parameters such as the size and the refractive index of both the host and the inclusions. Therefore, each measurement has to be accompanied by light-scattering simulations.

2. Simulations

For the simulation of the light scattering of the inclusion droplets we use a conventional Monte Carlo method [1, 2]. The position of the incident photon is generated randomly. The beam profile can be modelled as either a Gaussian beam profile or as a plane wave. At the point where the photon hits the surface of the host particle the Fresnel coefficients r_s and r_p are computed. In the interval [0, 1] a random number ξ is generated. If $\frac{1}{2}\left(r_s^2+r_p^2\right) > \xi$ the photon is reflected. Otherwise it is transmitted. After a photon is refracted into the host sphere it is allowed to travel a free path length $l = -\bar{l}\log\xi$, where $\bar{l}$ is the mean free path length between two subsequent scattering events. If the photon has not reached the outer boundary of the host sphere, a new random number ξ is generated and two subcases arise. If $\xi < Q_{\mathrm{abs}}^{\mathrm{incl}}/Q_{\mathrm{ext}}^{\mathrm{incl}}$, the photon is absorbed. Otherwise the photon is scattered. $Q_{\mathrm{abs}}^{\mathrm{incl}}$ and $Q_{\mathrm{ext}}^{\mathrm{incl}}$ denote the absorption and the extinction efficiencies of the inclusions, respectively. If the photon is scattered, the new direction is randomly determined according to the phase function of the inclusion. This procedure is repeated until the photon reaches the host boundary surface, where it is again subjected to reflection or refraction events.

Important parameters for the simulation are the volume concentration c_{v} of inclusions in the host particle, the size parameters α and α_i and the refractive indices n, n_i of the host and inclusions, respectively. The size parameter is defined by $\alpha = \pi d/\lambda$, where d is the particle diameter and λ is the wavelength.

In our simulations we focus on model suspensions of water with polystyrene (PS) particles as inclusions. The size of the host droplets varied from $\alpha = 280$ to 1670. The size of the inclusion particles varied from $\alpha_i = 2.75$ to 21.4. The described programme is able to simulate the scattering distribution for complex refractive indices n, n_i. For comparison with the experimental results we used the refractive index $n = 1.334$ for water and $n_i = 1.6$ for the inclusions.

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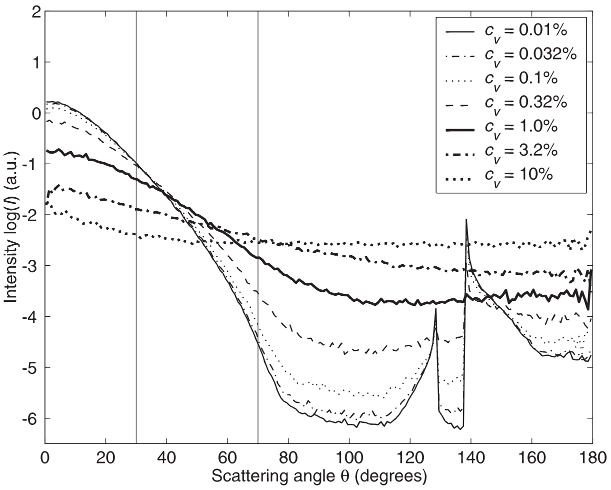

Figure 1. Phase functions computed with the described method for different inclusion concentrations c_v. The size parameters for the host and the inclusions are $\alpha = 460$ and $\alpha_i = 2.75$, respectively. The refractive indices are $n = 1.334$ and $n_i = 1.6$.

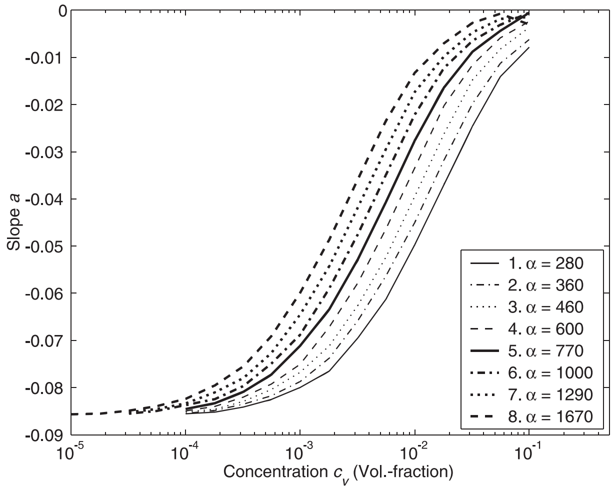

Figure 2. The approximated slope a as a function of the inclusion concentration c_v for different sizes of the host particle α. The size of the inclusions is $\alpha_i = 2.75$.

An example of a simulation result is shown in figure 1 for $\alpha = 460$ and $\alpha_i = 2.75$. The volume concentration of inclusions c_v vary from 0.01 to 10%. The range of the scattering angle, θ, of 30–70° is analysed with regard to changes in the slope a of the scattered intensity with the inclusion concentration.

The scattered intensity distribution in the range of 30–70° is approximated by the relation $\log(I) = a\theta + b$, where a is the slope and $b = \log(I_{\theta=0})$. It can be seen that the slope of the scattered intensity varies with the inclusion concentration in the scattering domain of 30–70°.

In figure 2 we plot the slope a as a function of the particle concentration c_v for different α values and a given value of $\alpha_i = 2.75$.

In order to obtain a one-to-one dependence between the inclusion concentration and an auxiliary quantity we analyse the dependence of the slope on the parameter αc_v for different α_i values.

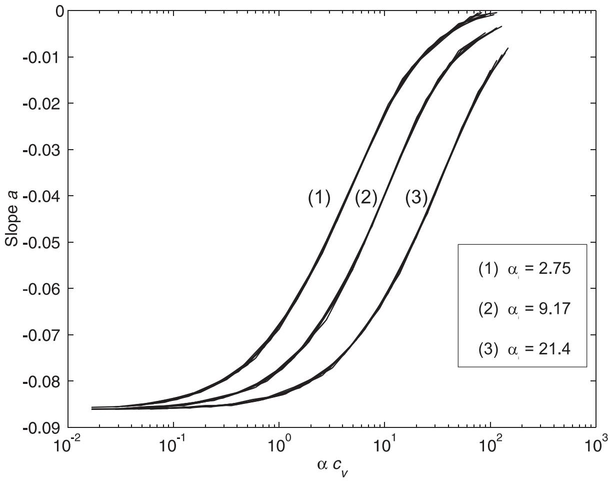

Figure 3. The approximated slope a as a function of the auxiliary quantity αc_v for different sizes of the inclusions, α_i. Each given value of α_i is represented by eight graphs with host particle sizes in the range $280 < \alpha < 1670$.

From figure 3 we see that for a given value of α_i the dependence of the slope on the parameter αc_v is a monotonically increasing function in a certain domain of αc_v.

This suggests that for a known value of α_i we can obtain the parameter αc_v by measuring the slope of the scattered intensity. In addition if we have information about the size α of the host particle we have found a method for measuring the inclusion concentration.

For concentrations below 1% we can use the method of König *et al* [3] to obtain information about the particle size. This method is based on relating the particle size to the angular fringe spacing in the measured intensity. The advantage of the CCD linescan is the fast and detailed measurement of the scattering distribution. For reasons of simplicity we use this setup to obtain the size together with the inclusion concentration. For concentrations above 1% it is recommended to use reliable particle sizing methods based on small-angle diffraction.

Hesselbacher *et al* [4] introduced an advanced formula (1) which gives a relation between the measured angular fringe spacing $\Delta\theta$ and the size α of the particle:

$$\Delta\theta = \frac{2\pi}{\alpha}\left(\cos\left(\frac{\theta}{2}\right) + \frac{m\sin(\theta/2)}{\sqrt{1+m^2-2m\cos(\theta/2)}}\right)^{-1}. \tag{1}$$

This relation shows that the angular fringe spacing $\Delta\theta$ is not only dependent on the size α of the particle but also on the real part of the refractive index m and the scattering angle θ. Since suspension droplets consist of media with different refractive indices, the dependence on the angular fringe spacing needs further investigation.

Increasing the concentration of inclusions results in a greater effective refractive index. The effective refractive index of inhomogeneous particles can be computed with the appropriate mixing rules of Maxwell–Garnett or Bruggemann [5]. These mixing rules require dipole scatterers as the inclusions. Instead of the effective refractive index it is possible to use the equivalent refractive index as described in [2]. The equivalent refractive index of the examined

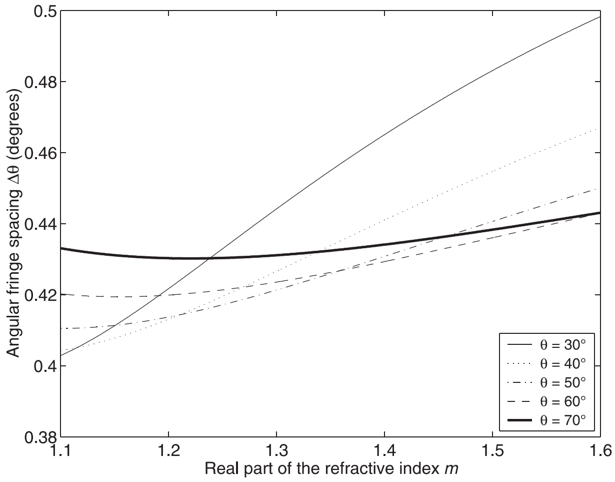

Figure 4. The angular fringe spacing $\Delta\theta$ as a function of the real part of the refractive index m of the droplet for different values of the scattering angles θ. The size of the droplet is $\alpha = 460$.

inhomogeneous particles lies within a close range near the refractive index of the host particle. Therefore, it can be stated that the influence of the inclusion concentration on the angular fringe spacing can be neglected. In figure 4 we plot the angular fringe spacing $\Delta\theta$ for different angles θ as a function of the real part of the refractive index m. The size of the droplet is $\alpha = 460$. It can be clearly seen that the dependence of m on the angular fringe spacing is weak. Since $\Delta\theta \propto \frac{1}{\alpha}$, this is also valid for other size parameters α.

3. Experimental setup

The examined model suspension is composed of water and monodisperse spherical PS particles. The PS particles have been produced in our laboratories at the Department of Chemical Engineering. The particles used for the experiments are verified through scanning electron microscope images to be monodisperse with a diameter of $d = 420 \pm 10$ nm. The concentration c_v of the inclusions varies from 0 up to 1.0% volume fraction. With a piezoelectric droplet generator [6] we are not able to produce droplets of suspension with higher concentrations. The generated droplets are monodisperse and single droplets can be emitted on demand. The size of the droplets can be verified with a microscope CCD-camera with an accuracy of about 10%.

For the setup we use an argon ion laser ($\lambda = 514.5$ nm) with an output power of 800 mW. The laser beam is focused by a lens. This causes a higher intensity at the point where the droplet passes the beam. The light scattered by a suspension droplet is collected by a lens system with a small focal length. The layout of the experimental setup is shown in figure 5.

The scattered light is detected by a high-speed linescan camera which is able to sample 100 000 lines s^{-1}. Each line contains 256 pixels with a depth of 8 bit.

In figure 6 we depict the measured intensity distribution of a water droplet without inclusions. The oscillations are well modulated and evaluation of the angular fringe spacing leads to a droplet diameter of 90 ± 4 μm. The measured slope is $a = -0.09$ and shows good agreement with the simulated value of -0.086.

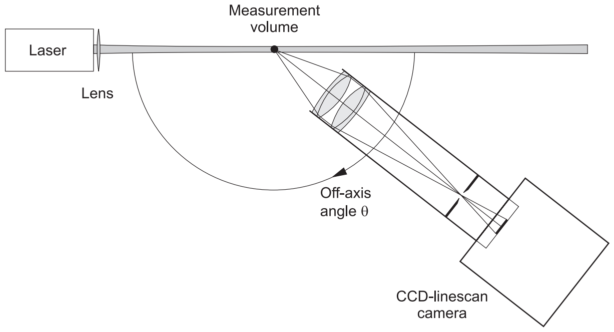

Figure 5. Experimental setup.

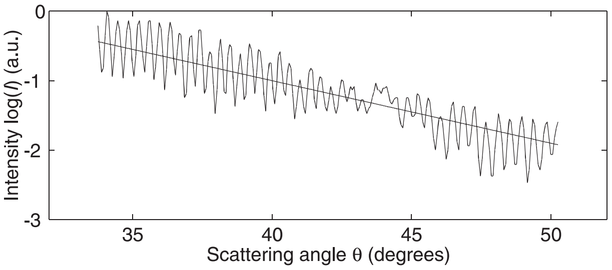

Figure 6. Measured scattering distribution of a single water droplet with a size of 90 ± 4 μm with slope $a = -0.09$ (line shown).

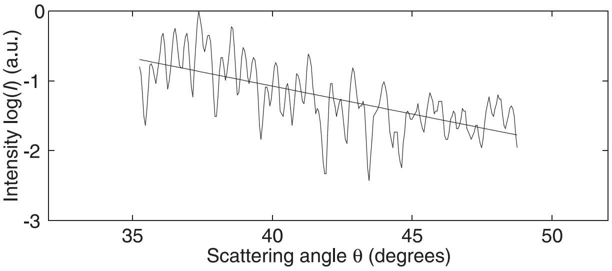

Figure 7. Measured scattering distribution of a single water droplet with a size of 81 ± 4 μm and slope $a = -0.078$ (line shown). The concentration of the inclusions is $c_v = 0.06\%$.

Figure 7 shows another example of a measured intensity distribution. Here, the concentration of the PS inclusions amounts to $c_v = 0.06\%$. The oscillations are less regularly modulated. The size of the droplet is 81 ± 4 μm. The measured slope of $a = -0.078$ lies within the expected range.

Finally, in figure 8 we would like to show the good agreement between the simulated and the experimentally obtained results. The accuracy of the obtained results obviously varies with the value of αc_v. The best accuracy in obtaining the concentration c_v is given for the slope values $-0.07 < a < -0.02$. Small uncertainties in the determined values of the slope a lead to significant errors beyond this area.

4. Conclusion

In this paper we presented a method for the determination of the inclusion concentration by the slope of the intensity distribution in a selected scattering domain. The method implies measurement of the particle size and knowledge of other particle parameters such as inclusion size and the refractive indices

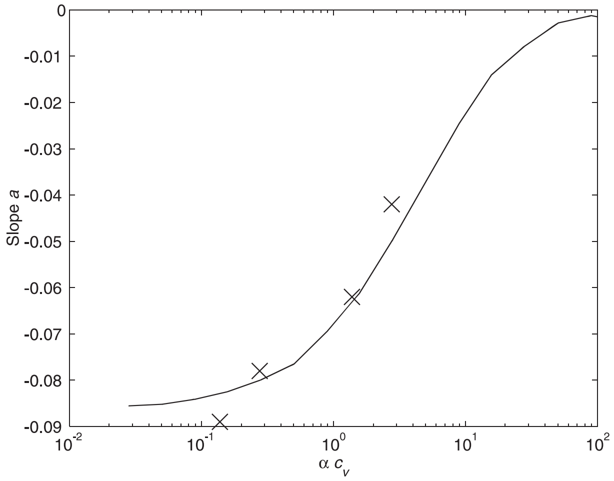

Figure 8. Measurement results from suspension droplets ($\alpha_i = 2.75$) with different concentrations c_v. The slope a as a function of the quantity αc_v. The solid curve represents the simulated slope. The crosses show the measured data. Each point is generated by an average of 100 measured droplets.

of the host and the inclusions. Exemplary measurements confirmed the applicability of the method described.

The limitations of this method are still under further investigation. The simulation method covers a wide range of the essential parameters involved. Considering different droplets of suspension, each composition needs its own set of simulations. Due to the variety of parameters involved (α, α_i, n, n_i, c_v) the limitation of the method needs to be evaluated for each individual configuration.

Acknowledgment

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