Analysis of particle distribution according to sizes with improved resolution

Vladimir Rysakov, Feliks Rejmund

Institute of Fundamental Technological Research, Polish Academy of Sciences, Świętokrzyska 21, 00-049 Warsaw, Poland
tel.: +48-22-826-12-81 ext.273, fax: +48-22-826-98-15, e-mails: rysakow@ippt.gov.pl, freymund@ippt.gov.pl

Abstract

A new method is proposed of analysis of emulsions with the aid of light scattering, in which it is possible to separate in the detecting signal the contributions of the particles of different sizes and thus to determine the particle distribution according to the sizes with improved accuracy. The effect is achieved by introducing into the emulsion additionally an ultrasonic beam which, as a result of the Doppler effect, modulates the scattered light in frequency. Application of a special frequency-phase detector, sensitive to the amplitude of the frequency deviation, makes it possible to separate the signals from the particles of different sizes.

1. Introduction

The light, scattered by an ensemble of particles, contains important information about these particles. However, extraction of this information, i.e., solution of the inverse problem of diffraction, even for the case of homogeneous emulsion, presents (without additional assumptions) in many cases practically insurmountable difficulties. But if the ensemble of particles in the emulsion is multicomponent, then with the aid of scattering of light it is difficult to determine even the total particle concentration, since scattering cross-section depends on the particle size. Since this dependence changes from a^4 to a^0, without the knowledge of the particle distribution on sizes the final concentration cannot be determined [1, 2]. At the same time, the determination of particle distribution according to the sizes with the aid of scattering of light with reasonable accuracy is also impossible, since the scattered light from the particles of different types and sizes overlaps at any angles. At present, there is no procedure for separation of the contribution of individual components to the recorded signal.

In the presented paper is proposed a method of separation of these contributions, which gives the possibility to carry out the analysis of particle distribution of sizes with the average resolution better than $a/\Delta a \approx 150$. In the existing procedures it is usually less than $a/\Delta a \approx 2$, which is insufficient for practical purposes in the region of high technologies.

2. Schematic diagram of instrument

The essence of method consists in the following (Fig.1). The beam of ultrasound is introduced into the cuvette with the emulsion. The particle of emulsion performs oscillations under the action of the ultrasound. The amplitude of oscillations depends on the size (and density) of the particle. Due of the Doppler effect the scattered light is shifted in the frequency. Signal with the difference frequency is separated by heterodyning the scattered and incident light in the photodetector. The deviation of the frequency of scattered light (and, correspondingly, of the signal from the photodetector) is determined by the amplitude of oscillations (i.e. of the particle size), while the modulation frequency is identical for all particles and is equal to the doubled ultrasonic frequency. The use of a special frequency-phase detector, sensitive to the maximum deviation, makes it possible to divide the contributions to the scattered signal from the particles of different sizes and thus to determine the particle distribution

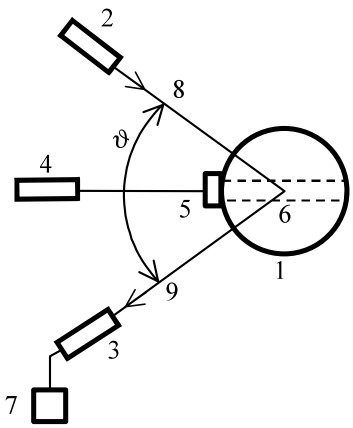

Fig. 1. Schematic idea of instrument. 1 - cuvette; 2 - laser; 3 - photodetector; 4 - generator of ultrasound; 5 - piezoradiator of ultrasound; 6 - beam of ultrasound;7 - recorder; 8 and 9 - incident and scattered light beams.

according to the sizes. It is natural that with the interpretation of results it is necessary to consider the dependence of differential scattering cross-section on the particle size. The resolution of the method is determined first of all by the filter pass-band of the frequency-phase detector and therefore it can be made high.

3. Light scattering by oscillating particle

Consider the process of light scattering by the particle, which oscillates in the travelling acoustic wave. If the harmonic travelling wave has the intensity I_0, then the amplitude of oscillation velocity of the medium will be $v_0 = \sqrt{2I_0 / \rho c_0}$, where ρ is the density of liquid, c_0 is the velocity of sound in the liquid. Stokes's force acts on a particle as a result of fluid friction against the particle, which we consider to be spherical:

$$F = 6\pi\eta a\,(v - v_c)\,, \qquad (1)$$

where v and v_c are the instantaneous velocities of particle and liquid, a is the radius of the particle, η is viscosity. Solution of this equation, together with the equation of motion, gives the possibility to determine the relation between the velocity amplitude of the particle and the velocity amplitude of the medium, which is conveniently expressed by off-beat complex transmission factor on the velocity K:

$$|K| = v_c/v = \frac{1}{\sqrt{1+\left(\dfrac{a}{a_0}\right)^4}} \quad , \quad \varphi = -\,\mathrm{arctg}\left(\frac{a}{a_0}\right)^2 , \qquad (2)$$

where $a_0 = 3\sqrt{\dfrac{\eta}{2\rho_1\Omega}}$ is the characteristic radius of the particle, ρ_1 is the density of the particle, Ω is the frequency of ultrasound (Fig.2). It is evident that the oscillation velocity of small particles actually approaches the oscillation velocity of the liquid, while large particles are practically motionless. In the range (0.5 – 5)a_0 the particle velocity strongly depends on the particle size. Accordingly, in this range it is possible to reach high resolution. If the incident light of the laser and direction of scattering are symmetrical with respect to the beam of sound and the angle between them is ϑ (Fig.1), then the frequency shift of the scattered light Δν will be:

$$\Delta\nu/\nu = 2(v_c/c)n \sin(\theta/2)\ , \qquad (3)$$

where ν is the frequency of the laser emission, n is the refractive index of the liquid.

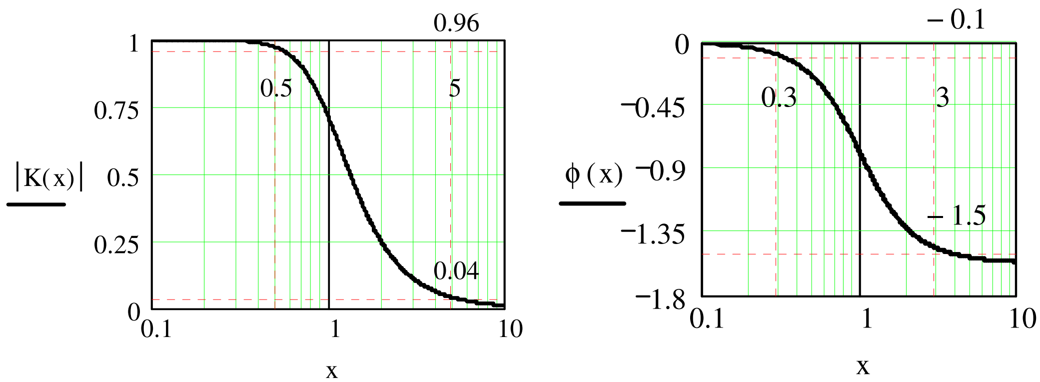

Fig. 2. Dependence of the transmission factor of the velocity on the normalised size of the particle $x = a/a_0$.

4. Resolution of the instrument

The resolution of the method is determined by the slope of curve K(x) and by the ratio of the passband of the recorder ΔF with maximum deviation of the frequency of the scattered light $\Delta\nu_0$:

$$a/\Delta a = (\Delta\nu_0/\Delta F)\,(dK/dx)\,. \tag{4}$$

Fig.3 presents the dependence of dK/dx on x. It is evident that the maximum resolution will be for x = 1, i.e. when $a = a_0$, it is equal to $a/\Delta a = (\Delta\nu_0/\Delta F)/\sqrt{2}$. On the boundaries of range $(0.2 – 5)a_0$ it decreases approximately 35 times. The resolution rapidly decreases outside the boundaries of the range. In the effect, in the range $a_{min} = 0.1a_0$ - $a_{max} = 10a_0$ at one side of the boundary, total concentration of all particles smaller than a_{min} will be recorded, and at the other larger than a_{max}.

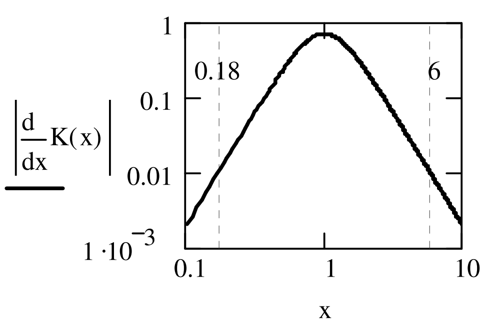

Fig. 3. Dependence of the slope of curve K(x) on the normalised size of the particle $x = a/a_0$. Curve describes a change the resolution with the deviation of a particle radius from optimum a_0.

5. Numerical estimations

Numerical estimations lead to the following results. For the water and the intensity of ultrasound $I=1W/cm^2$, the amplitude of oscillation velocity equals approximately $v_0 \approx 11$ cm/s. Thus, for the ultrasonic frequency of the order of 20 kHz the optimal radius is $a_0 \approx 1.5$ μm. This gives the range of the measured radii of particles approximately from 0.7 to 7 μm, i.e. exactly the range which is most difficult for the measurement by other methods. By a change in the frequency and the power of ultrasound, the measured range can be slightly displaced. For the laser working in the green domain, the maximum frequency shift of the scattered light $\Delta\nu_0$ will be of the order of 700 kHz. Using the recording device with the frequency passband of the order of 1 kHz, the maximum resolution will be approximately 500, and it decreases on the boundaries of the operating range to ~15 and reaches one when $a = 0.1a_0$ or when $a = 10a_0$. In many practical cases large particles are few in the emulsion (they are sediments), on the other hand the influence of small particles, as a rule, does not depend on their size. Therefore decrease of the resolution on the boundaries of the range and information only about the total concentration of both small and large particles seems to be not a substantial limitation.

6. Construction of instrument

Technical instrument, which operates on the basis of the principle described above, can be designed analogously with the usual spectrum analyzer, showing the particle distribution on sizes in the emulsion in real time directly on the display screen. The proposed procedure and the technical solution of the instrument for determining the particle sizes distribution in the emulsions with improved resolution are protected by the patent A1(21)320900 (22) 97 06 30 6(51) G01N 15/02.

7. Conclusion

The article describes the method of determination of particle distribution on the sizes in the emulsions with the maximal resolution equal to $a/\Delta a \approx 500$, on the average over the range to 150. Such a high resolution is reached due to the fact that in the receiving equipment, the contributions to the scattered signal from the particles of different sizes are separated. For this purpose, an additional ultrasonic wave into the emulsion is introduced, in which the particles perform oscillatory motions. The frequency of oscillation is equal to the ultrasonic frequency; however, the amplitude of oscillations depends on the size (and density) of the particle: the amplitude of large particles approaches zero, the amplitude of small particles is in effect equal to the amplitude of oscillations of the liquid. The scattered light due to the Doppler effect will be shifted in frequency, the maximum deviation of frequency depending on the particle size. The use of a special frequency-phase detector with the narrow-band filter makes it possible to separate from the obtained signal the part, which corresponds to the light scattered by the particles of the certain size (with the possible resolution). Scanning the frequency of filter allows in the real time to obtain on the display screen the particle distribution according to the sizes.

The paper gives the theoretical analysis of operation of the proposed instrument and the numerical estimations, which show that at the reasonable requirements for the elements of the instrument (the power and the ultrasonic frequency etc.) it is possible to reach a high resolution.

Acknowledgement

The paper was financially supported by the State Committee for Scientific Research, Grant No. 4T08A 056 23

References

[1] M.Kerker, The Scattering of light. (Academic Press, NY, 1969).

[2] V.Rysakov, M.Stoń, „ Light Scattering by a Suspension of Particles: IV. The Determination Limits of Parametrs of Suspensions by Light Scattering”,Optica and Spectroscopy, 82, 280-284 (1997).