

Method for measuring optical parameters in weakly absorbing turbid media

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This article develops a simple, yet effective technique of measuring optical parameters in weakly absorbing turbid samples. Although based on diffusion theory, this technique largely relaxes its strict non-boundary and spot source requirements by choosing a suitable source-detector distance. Moreover, the technique is applicable not only to liquid samples, but also to solids or on-line measurements. Experimental results demonstrate that, measured by this method, the reduced scattering coefficients of intralipid suspensions are in good agreement with those obtained by other authors. The paper also reports on the application of this technique to scattering measurements in pulp.

Keywords: scattering coefficient, optical diffusion, scattering in weakly absorption medium.

1. Introduction

Techniques based on time-of-flight, time-resolved photoacoustics and diffusion theory have been developed to measure the optical parameters of various media [1]–[4]. Although these techniques provide accurate results, their use in particle applications has some disadvantages. For example, the instrumentation required by the time-of-flight technique, including an ultrashort pulse laser and a single-photon avalanche photodiode, is very expensive. The same is true of the time-resolved photoacoustic technique, which depends on a high power pulse laser and a wideband ultrasonic transducer. Further, these techniques impose strict requirements for the position of the light source and the detector. Diffusion theory, on the other hand, requires that both the light source and the receiver are placed either in an infinite medium or at least far away from the boundaries of a sufficiently large medium.

However, for most practical applications, accuracy requirements are less strict than for laboratory measurements. Instead, they call for instrumentation that is cheap, convenient and accessible on-line. To this end, this article presents a simple, but effective technique that employs a cheap pulse laser and a photodiode, to measure

optical parameters in weakly absorbing turbid samples. Although this technique is based on diffusion theory, it disposes of the strict non-boundary and spot source requirements. Experimental results demonstrate that the reduced scattering coefficients of intralipid suspensions measured by this method are closely identical to those obtained by other authors. Finally, the paper presents an application of this technique to measurement of the scattering properties of pulp.

2. Theoretical background

When a light beam is incident on a weakly absorbing, highly turbid medium, photons in the beam are scattered in all directions. The intensity and direction of this scattering is determined by the medium's optical parameters, such as absorption coefficient, scattering coefficient and phase function. At any observation point located several reduced mean free path (mfp) lengths away from the incident light source, photons appear to migrate in accordance with diffusion theory. If the laser beam hitting the sample's surface is focused into a small spot and the illuminated sample is large enough to be thought of as semi-infinite, the fluence rate of light in the medium can be described as

$$F(r) = \frac{P_0}{4\pi Dr} \exp\left(-r \sqrt{\frac{\mu_a}{D}}\right). \quad (1)$$

In this equation, D is the diffusion constant, r – the distance from the light source to the observation point and μ_a – the absorption coefficient of the turbid medium. Because the sample can be regarded as semi-infinite, part of the incident optical power will be reflected from its surface. Thus, the light spot power going into the sample decreases to

$$P_0 = (1 - R_{d\infty})P_l \quad (2)$$

where P_l is the laser output power and $R_{d\infty}$ – the total diffuse reflectance calculated as [5]

$$R_{d\infty} = \exp\left[-\frac{7}{\sqrt{3(1 + \mu'_s/\mu_a)}}\right] \quad (3)$$

where μ'_s is reduced scattering coefficient. For a highly turbid sample where $\mu'_s \gg \mu_a$

$$D = \frac{1}{3\mu'_s + \mu_a} \approx \frac{1}{3\mu'_s}. \quad (4)$$

The amplitude of the detection signal from a pin photodiode is $V = R_T R_S F(r) S$ (R_T is the transimpedance of the receiving circuit, while R_S and S represent the response and the active area of the pin photodiode, respectively). By substituting (1)–(4) into V , multiplying with the distance and then taking the logarithm, we get

$$\ln[rV(r)] = -\sqrt{3\mu_a\mu'_s} r + \ln\left\{\frac{3\mu'_s}{4\pi} R_T R_S P_l S \left[1 - \exp\left(-7\sqrt{\frac{\mu_a}{3\mu'_s}}\right)\right]\right\}. \quad (5)$$

Hence, a linear fitting of the measurement points in Eq. (5) allows the absorption coefficient and reduced scattering coefficient to be deduced from the slope and intercept of the straight fitting line, provided that R_T , R_S , P_l and S are known. In the case of an aqueous suspension or a solution in which the absorption coefficient of the scattering particles is almost equal to or smaller than that of water, the reduced scattering coefficient can be calculated directly from the slope of the line, since the absorption spectroscopy of water is well known.

3. Experiment and results

Figure 1 displays a schematic drawing of the setup used in these experiments. As the excited source, the setup uses a pulsed laser with an output energy of $1.52 \mu\text{J}$ at the 1064 nm wavelength. The duration of each pulse is 10 ns and the pulse repetition rate is 200 Hz. The laser beam is projected on the incident window of a cuvette and the diameter of the light spot is estimated at 0.5 mm. Scattered light is detected by a photodiode (Telefunken Electronic, model BPW 86) which is placed at a distance of 3 mm from the incident window, in close vicinity of the cuvette's side wall. The feet of the photodiode are directly connected with a transimpedance amplifier (Avtech, model AV-149-BW1-2k-PIN-SP1). During the experiment, the light spot was moved transversely by a translation stage, while the cuvette and the photodiode were stationary.

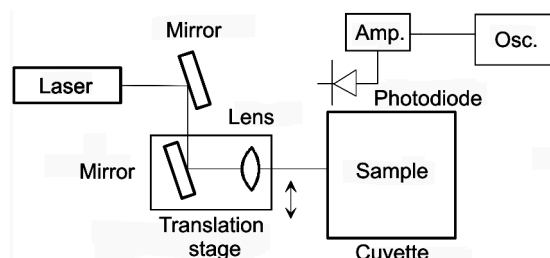


Fig. 1. Experimental setup for measurements of scattering fluence rate.

3.1. Intralipid suspensions

This preliminary study used 10% Intralipid® (Fresenius Kab. AB., Uppsala, Sweden) and its water dilution as samples, with Intralipid concentrations of 0.2%, 0.3%, 0.5%, 1%, 2%, 5% and 10%. These samples were loaded successively in a cuvette with a thickness of 25 mm, width of 25 mm and height of 40 mm. A small hole and a valve at the bottom of the cuvette were used to replace the samples without moving the cuvette. The measurement results are drawn into the relationship between $\ln(r \times V)$ and r , as shown in Fig. 2. The estimated error at any measurement point is smaller than the marks. The left hand graph of Fig. 2 shows that the relationship between $\ln(r \times V)$ and r is not linear throughout the r range. Thus, as r decreases below a definite value, the increase in the amount of $\ln(r \times V)$ gradually decreases to a point where it starts to deviate from the linear relationship. This is probably due to two reasons. Firstly, the laser beam incident on the samples cannot be regarded as a spot source, when the beam is too near the detector. Secondly, when the light source is too close to the cuvette's side wall, a lot of photons will leak from it. Together these two factors affect the light distribution in the cuvette such that it apparently deviates from the solution given by the diffusion equation (1). For samples with a higher intralipid concentration, the deviation in linear relationship occurs at smaller values of r .

On the other hand, the linear relationship does not hold for dense samples either, such as 2%, 5% and 10% intralipid, at larger r distances (e.g., 16 mm). In such samples, the reduced mean free path length is so short that the samples actually absorb a great number of photons, before they arrive at the photodiode. Consequently, the received signal is lower than that of a diluted sample. If the distance between the source and

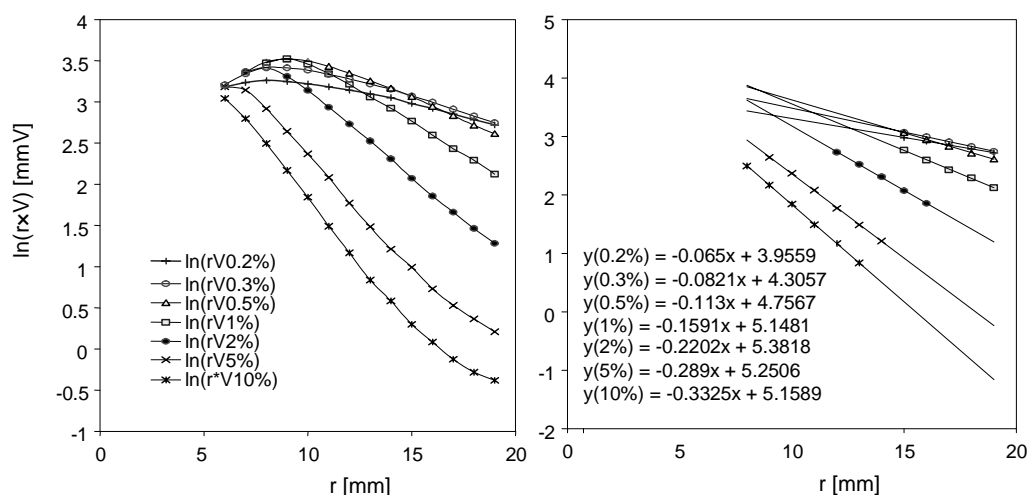


Fig. 2. Relationship between $\ln(r \times V)$ and r in the measurement of different intralipid samples.

Table 1. Slopes of fitting lines and reduced scattering coefficients of intralipid suspensions with different concentrations at the 1064 nm wavelength.

	0.2%	0.3%	0.5%	1%	2%	5%	10%
Slope of line μ_{eff} [mm^{-1}]	0.065	0.0821	0.113	0.1591	0.2202	0.289	0.3325
μ'_s [mm^{-1}] deduced by slope of line	0.12	0.20	0.37	0.73	1.4	2.4	3.2
μ'_s [mm^{-1}] after VAN STAVEREN <i>et al.</i> [1]	0.13	0.20	0.34	0.67	(1.34 ?)	(3.35 ?)	(6.7 ?)

the photodiode is sufficiently large, the incident boundary will reflect enough backscattering photons to produce an apparent increase in the signal received by the photodiode. This probably explains why the $\ln(r \times V)$ values of dense intralipid samples deviate from the linear relationship at larger values of r .

On the basis of these findings, we may conclude that the linear relationship of the $\ln(r \times V) - r$ curve is approximately true within a suitable source – detector range. A linear fitting of the data in this range, as in the right hand graph of Fig. 2, allows us to deduce optical parameters of the samples. Because the absorption coefficient of a low concentration intralipid suspension is nearly equal to that of water (0.0115 mm^{-1} at the 1064 nm wavelength), the reduced scattering coefficient can be calculated from the slopes of the fitting lines listed in Tab. 1. For comparison, the table also lists van Staveren's results [1].

At low concentrations, the reduced scattering coefficients measured by this technique are practically identical with van Staveren's results. Because his measurement was carried out in a very low concentration of intralipid suspension (1 ml of 10% intralipid diluted in 999 ml of water), a linear forecast for high concentrations (5% and 10% intralipid suspensions) is not possible. ZACCANTI *et al.* [2] showed experimentally and GIUSTO *et al.* [6] demonstrated theoretically that the reduced scattering coefficient of intralipid suspensions is not linear with concentration, but approximates quadratic behaviour from 1% to 20%. Hence, our results is reasonable in that the tendency of the reduced scattering coefficient to increase at high concentrations gradually diminished.

3.2. Paper pulp samples

The pulp samples used in this study were made of the long fiber fraction of thermo-mechanical pulp (TMP) and the fines fraction of groundwood pulp (GW). The long fiber fraction is formed by filtering the original pulp through a 0.297 mm screen, while the fines fraction consists of particles that pass a 74 μm screen, while being rejected by a 37 μm screen. Such fibers have a diameter of a few tens of micrometers and measure a few millimeters in length. This study included six samples with different mixtures of long fibers and fines. These mixtures have two different total consistencies,

Table 2. Reduced scattering coefficients of five measurements and their average values and standard deviations for six pulp samples at the 1064 nm wavelength.

Sample	0.5% fiber	0.5% fines	0.5%(10)	0.5%(40)	0.9%(10)	0.9%(40)
μ'_s [mm^{-1}]	0.05	0.19	0.03	0.15	0.32	0.22
	0.27	0.20	0.14	0.11	0.13	0.27
	0.09	0.20	0.02	0.05	0.14	0.28
	0.04	0.19	0.13	0.08	0.14	0.30
	0.09	0.20	0.16	0.10	0.09	0.29
Average μ'_s	0.11	0.20	0.10	0.10	0.16	0.27
St. dev.	0.09	0.00	0.07	0.04	0.09	0.03

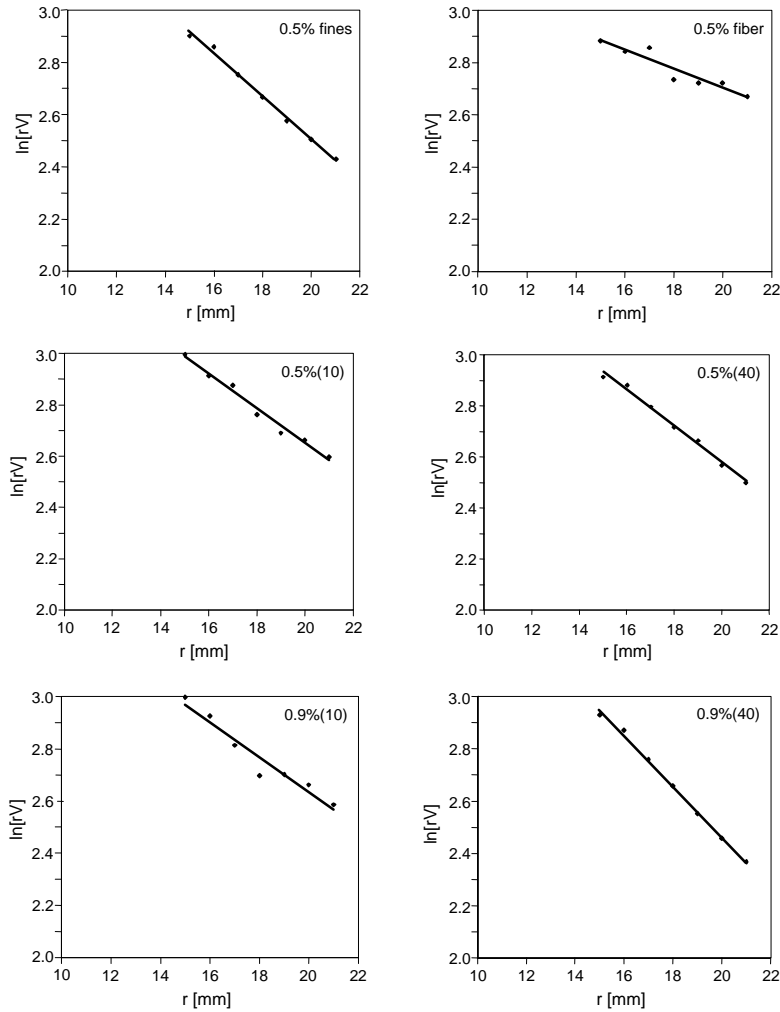


Fig. 3. Typical $\ln(r \times V) - r$ relationships in measurements of different pulp samples.

such as 0.5% fines, 0.5% fiber, 0.5% (10), 0.5% (40), 0.9% (10) and 0.9% (40) (note that 0.5% and 0.9% refer to the dry consistency of the samples, while (10) and (40) refer to the percentage of fine particles relative to the sample's total dry weight). Naked eye observations suffice to establish that some of the samples are clearly inhomogeneous. Thus, the scattering coefficient will differ in the various parts of the samples. Further, as the samples consist of a liquid-fiber/fine mixture, their local scattering coefficients can be changed by stirring.

Nevertheless, it is still possible to apply this technique to evaluation of the scattering properties of these samples, provided that they are stirred as homogeneously as possible and also measured a few times to obtain an average value and its standard deviation. Consequently, every sample was measured five times and stirred homogeneously before each measurement.

The results of the experiments are shown in Tab. 2. It can be seen that μ'_s should be smaller than 0.3 mm^{-1} in all samples. As pulp consistency increases, μ'_s tends to increase. At the same dry consistency level, fines samples show larger scattering than the corresponding fiber samples. It is also interesting to note that although fibers introduce non-homogeneity into the samples, fines improve the samples' homogeneity and increase their scattering property. In more detail, the samples of 0.5% fiber and 0.5% (10) are apparently non-homogeneous; hence, the measured μ'_s shows great variation between different measurements. This can be seen more clearly in Fig. 3, which presents a typical $\ln(r \times V) - r$ relationship for one measurement in every pulp sample.

Many measurement spots in the left column of Fig. 3 appear to deviate from the fitting lines. This demonstrates that these samples are still inhomogeneous, even after stirring, implying that the samples contain a number of local scattering coefficients. Hence, the μ'_s deduced from these measurement data represents an average local reduced scattering coefficient. As shown by the last row of Tab. 2, some samples, such as 0.5% fiber, 0.5% (10) and 0.9% (10), have a larger standard deviation of μ'_s . On the other hand, measured data for 0.5% fines and 0.9% (40) samples show good linear fitting (right column of Fig. 3), meaning that these samples are sufficiently homogeneous for deducing their μ'_s by this technique.

4. Discussion and conclusions

In diffusion theory, the fluence rate calculated by formula (1) is the solution to the steady-state diffusion equation for a point source located in an infinite turbid medium. An exact measurement of optical parameters requires that these requirements are satisfied. To this end, laboratory measurements are usually implemented by inserting two optical fibres into the centre of a sufficiently large liquid vessel [1], [2]. A small diffusive sphere is then attached to the ends of the fibers to simulate a point-like isotropic source and a point-like all-direction detector. This arrangement, however, is not convenient for on-line measurements and is impossible for solid media applications. The method presented here can solve these problems.

Moreover, experimental results show that the method described here is effective, provided that the distance between the source and the detector is appropriate. This can be explained by two approximations. First, when the medium studied is weakly absorbing and highly scattering, the incident light spot is scattered such that the contour of the fluence rate becomes spherical in shape at a definite distance sufficiently far from the source. Hence, seen from that observation point, it approximately resembles an isotropic point-like source. Second, the fluence rate at any one point can be approximately measured by a detector with a small receiving surface, on condition that it is vertical to the propagation direction of the fluence.

However, when the incident light is too close to the photodiode, it cannot be seen as a point-like source by the diode. Moreover, many photons leak out from the side wall of the cuvette, apparently changing the fluence rate at the detection point. On the other hand, when the light spot is too far from the photodiode, the fluence rate at the measurement point becomes so weak that it is easily interfered with by the reflection of backscattering photons from the incident boundary. Consequently, a careful selection of source–detector distance for different turbid media serves to diminish these problems and results in a good approximation of the measured parameters.

Equation (1) is valid when $\mu'_s \gg \mu_a$. At the 1064 nm wavelength, the absorption coefficient of intralipid samples is equal to 0.0115 mm^{-1} . As a result, the measurement becomes invalid for samples with a lower concentration than 0.2%, since their $\mu'_s < 0.13 \text{ mm}^{-1}$. However, the absorption coefficient becomes much smaller at red and short NIR wavelengths, such as 0.0018 mm^{-1} at 800 nm and 0.00026 mm^{-1} at 633 nm. At these wavelengths, then, this method is applicable to measuring intralipid suspensions with concentration lower than 0.1%.

Contrastingly, as shown by Tab. 1 and Fig. 3, when the concentration exceeds 2%, the reduced scattering coefficient no longer increases linearly with concentration. This phenomenon occurs when the inter-particle distance becomes comparable with the wavelength, with the result that the correlation of the electromagnetic fields between the particles should be taken into account. The independent scattering approximation, which is satisfied in the case of low concentrations, does not hold for denser concentrations. However, the reason behind quadratic behavior between scattering property and particle concentration remains unclear [2].

For a high concentration intralipid suspension, the deduced μ'_s may be smaller than the true value. Because the boundary of the cuvette tends to diminish the linear fitting slope, especially in dense samples (see left hand graph of Fig. 2). It can be seen that quadratic behaviour will be better satisfied, if the μ'_s of 5% and 10% intralipid suspension is slightly larger than the values calculated in Tab. 1. Another reason is that the absorption coefficients of dense samples deviate from the water absorption value, due to the replacing effect of intralipid drops.

For the pulp measurement described above, the measured μ'_s is close to the true value only for 0.5% fines and 0.9%(40) samples. For other pulp samples, owing to their poor homogeneity, the deduced μ'_s represents only the average value of the local

Table 3. Evaluating scattering properties of pulp samples by the amplitude of the scattering signal.

	0.5% fiber	0.5% fines	0.5%(10)	0.5%(40)	0.9%(10)	0.9%(40)
	1.1	1.46	1.184	1.416	1.66	1.6
	1.08	1.46	1.132	1.36	1.6	1.48
	1.05	1.46	1.136	1.344	1.51	1.52
	1.34	1.456	1.192	1.412	1.47	1.46
	1.12	1.456	1.24	1.348	1.47	1.53
	1.14	1.476	1.224	1.384	1.47	1.53
	0.98	1.464	1.364	1.428	1.29	1.59
	1.28	1.456	1.412	1.428	1.53	1.65
	1.12	1.472	1.596	1.504	1.56	1.5
	1.06	1.468	1.132	1.364	1.42	1.58
Average (V)	1.127	1.463	1.261	1.399	1.498	1.544
St. dev. (V)	0.108	0.007	0.152	0.049	0.102	0.059

scattering coefficient, which makes it difficult to make a reliable comparison of the scattering properties of various samples. To solve this problem, another method was used to study the scattering property of these pulp samples. Since the inhomogeneity of the samples is caused by fibres with a length in the few millimetre range, we expanded the diameter of the incident laser beam to 10 mm. The laser beam itself was located at a distance of 15 mm from the detector. All samples were measured ten times and stirred after every measurement. As shown by Tab. 3, the samples can be easily distinguished from one another by the scattering signal voltage. When consistency increases, scattering increases as well. The table also shows that fines produce larger scattering than fibres, which is identical with the result in Section 3.2. However, the amplitude of the scattering signals is not only dependent on the scattering coefficient, but also on the laser–detector distance, absorption coefficient of the samples and specific measuring instrument. As a consequence, it is better to evaluate the scattering property of the samples on the basis of their scattering coefficient.

This experiment and analysis demonstrate that our technique offers a convenient method for measurements in highly scattering, weakly absorbing media. Concentrations of intralipid suspension ranging from 0.2% to 10% were studied at 1064 nm. Measurement values of lower concentration (< 2%) samples were found to be in good agreement with previously published data (although data concerning higher concentrations studied at this wavelength are not yet available). Further, this study also demonstrated quadratic behaviour at high concentrations. Hence, selection of a suitable measurement distance enables the scattering property of weakly absorbing, highly turbid media to be measured reliably. Applying this technique to pulp measurements shows that it offers an efficient, yet convenient way of deducing the scattering coefficient. The result is identical with evaluating the scattering property by amplitude of scattering signal.

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