

Subscripts

- b = refers to bottom plate
 t = refers to top plate

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Interfacial Areas of Liquid-Liquid Dispersions from Light Transmission Measurements

A mathematical model simulating light transmission through liquid-liquid dispersions has been developed. Numerical solutions of the model for various drop size distributions show that the fraction of parallel light which passes through a dispersion is a unique function of a dimensionless group, here named the Transmission Number, regardless of the drop size distribution. The results, which were verified by actual light transmission experiments, show that the interfacial area of a liquid-liquid dispersion can be calculated from a light reading provided that the light detector receives only parallel light. Application of the results to other two-phase dispersions is indicated.

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SCOPE

When two immiscible liquids are brought into contact to form a dispersion, knowledge of the interfacial area is frequently important. For example, in the analysis of heat or mass transfer between two phases, the interfacial area must be known in order to determine the corresponding transfer coefficient. If the interfacial area can be measured, then the area-free coefficient can be found, giving information on the independent effects of area and resistance. In this way, direct measurement of interfacial area can contribute to a better understanding of heat and mass transfer and their industrial applications.

There are cases where drop size itself is an important consideration, particularly in emulsion and other types of polymerization. The same data from interfacial area measurements will also give average drop size in terms of Sauter mean diameter.

Several methods have been used to measure the inter-

facial area or drop size of dispersions, but none to date seem to combine simplicity with accuracy. Photographic techniques are probably the most accurate method of measuring interfacial area, but the procedure usually requires many pictures and lengthy times for analysis. Furthermore, in a concentrated dispersion, only the drops nearest the camera can be measured. Light transmission through dispersions has proven to be the simplest method, but the theory, until now, has often been limited to dilute dispersions or to a specific detector system, as in the works of Bailey (1946), Langlois et al. (1954), Vermeulen et al. (1955), and Trice and Rodger (1956).

The most widely used theory for light transmission through liquid-liquid dispersions has been developed by Calderbank (1958) where a uniform drop size was assumed. Excellent summaries of the previous work in this field are given by Trice and Rodger (1956) and Calderbank (1958).

In this investigation, light transmission theory has been applied to nonuniform dispersions to determine the rela-

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tionship between the transmitted light and the characteristics of the dispersion. A mathematical model was developed to simulate a dispersion of spherical drops and the light which would pass through this dispersion. The model was solved numerically on a digital computer for various concentrations and drop size distributions, and

the results were used to correlate, theoretically, the fraction of light transmitted as a function of the volume fraction, drop size distribution, Sauter mean diameter, and interfacial area. Finally, the accuracy of the theoretical results was tested by actual light transmission measurements for glass bead suspensions in a mixing tank.

CONCLUSIONS AND SIGNIFICANCE

A mathematical model based on probability theory has allowed the application of light transmission theory to concentrated polydisperse systems. In the model, the assumptions are kept to a minimum so that the results may be applied to the greatest possible number of cases. Although the model was developed specifically for liquid-liquid dispersions, it may be used for any two-phase dispersion as long as the fundamental assumptions are satisfied. No requirements are made on the refractive index of the dispersed phase; it may be transparent or opaque; solid, liquid, or gas.

Numerical solution of the model has shown that the fraction of parallel light transmitted f through a dispersion is a unique fraction of the dimensionless Transmission Number, $N_t = \phi l/\bar{d} = a l/6$. This fraction, expressed by the exponential relationship $f = \exp [-1.5 N_t]$, is independent of the drop size distribution for all

the cases studied.

Experimental work has verified the accuracy of the above relationship and showed that the assumptions of the mathematical model can be adequately satisfied in the real case.

Application of these results should facilitate the study of two-phase interfacial areas. With a properly constructed light probe, the interfacial area per unit volume of total dispersion can be calculated from the fraction of light transmitted and the optical path length. The drop (or particle) size distribution and volume fraction need not be known. To calculate the Sauter mean diameter of the drops (or particles), or the interfacial area per unit volume of dispersed phase, values of the volume fraction, fraction of light transmitted, and optical path length are sufficient.

MATHEMATICAL MODEL OF LIGHT TRANSMISSION THROUGH A DISPERSION

When a parallel light beam is passed through a dispersion, the light is attenuated by diffraction, reflection, refraction, and absorption. If one drop is considered, the angular scattering of the parallel rays can be pictured as in Figure 1. As illustrated, each of the four scattering mechanisms prevents the rays from maintaining their original direction

To determine the net result of the four scattering mechanisms, the scattering coefficient of the drop must be considered. The scattering coefficient is defined by

$$a_s = K_s a_p$$

In this case, the scattering coefficient of the drop is primarily a function of two variables: the size of the drop

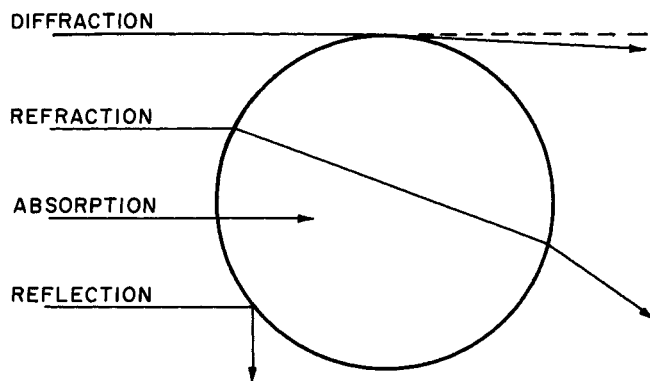


Fig. 1. The four mechanisms of light attenuation.

relative to the wave length of the incident light and the refractive index ratio of the two phases (van de Hulst, 1957). If all of the drops are larger than 0.1 mm in diameter, which is true for many dispersions, then K_s becomes a function of the refractive index ratio alone (Lothian and Chappel, 1951). Furthermore, if the light detector is placed at a large distance from the dispersion so that it receives only parallel light, the refractive index will have little effect on the amount of light received, making the effective value of K_s equal to 1.0^* (Diermendingian, 1969).

The above development is accurate except for the phenomenon of multiple scattering. Multiple scattering occurs when light which has been scattered by one drop is scattered again by another drop. This causes the light detector to receive an excess amount of light. However, by the same reasoning as that given for forward scattering, the error introduced by multiple scattering will be minimized if the light detector has a high angular resolution (Boll and Sliepcevich, 1956; Sheppard, 1958). Thus, with proper construction of the light detector, the effects of multiple scattering and forward scattering will not be significant, and the scattering cross section of the drops will equal the geometric cross section.

To consider the dispersion geometry, Figure 2 shows a parallel light beam incident on a rectangular volume of dispersion with dimensions w , h , and l . If all of the drops are assumed to be spherical,[†] which is a valid assumption for small drops, the volume fraction of dispersed phase is given by

* Actually, the light detector will receive light which has been scattered at very small angles no matter how far it is from the dispersion. However, this effect of forward scattering can be made negligible for typical liquid-liquid dispersions by using a light detector of high angular resolution (Walstra, 1965; Dobbins and Jizmagian, 1966).

† See application of the Results for a discussion of this assumption.

$$\phi = \frac{\pi \sum_{i=1}^n d_i^3}{6 whl} \quad (2)$$

The interfacial area per unit volume of dispersion is related to the volume fraction by

$$a = \frac{6\phi}{\bar{d}}$$

where \bar{d} is the Sauter mean diameter of the drops

$$\bar{d} = \frac{\sum_{i=1}^n d_i^3}{\sum_{i=1}^n d_i^2} \quad (4)$$

To simulate the amount of light transmitted through this dispersion, the geometry may be reduced from three to two dimensions, and Figure 2 reduces to Figure 3. This figure illustrates the two-dimensional characteristics of the light transmission process. The area not covered by the circles is the area through which parallel light would pass without hindrance, and thus is a measure of transmitted light.

Use of Probability Theory

To calculate the free space of this model, probability theory was found to be a most useful tool. In Figure 3, the probability that an arbitrary ray of light does not strike the i th drop is

$$P = 1 - \frac{\pi d_i^2}{4wh} \quad (5)$$

provided the drops are randomly located.

The probability that the ray does not strike the i th or the j th drop is

$$P = \left(1 - \frac{\pi d_i^2}{4wh}\right) \left(1 - \frac{\pi d_j^2}{4wh}\right) \quad (6)$$

It follows that the probability of the light ray not striking any of the drops is

$$P = \prod_{i=1}^n \left(1 - \frac{\pi d_i^2}{4wh}\right) \quad (7a)$$

Since this probability is equal to the fraction of light transmitted, Equation (7a) may be rewritten as

$$f = \prod_{i=1}^n \left(1 - \frac{\pi d_i^2}{4wh}\right) \quad (7b)$$

Equations (2) to (4), and (7b) relate the theoretical

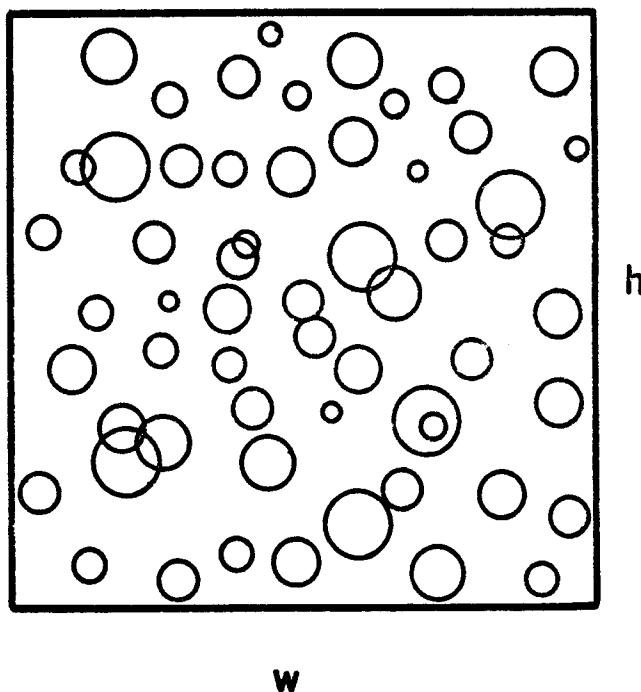


Fig. 3. The two-dimensional aspect of the light transmission process.

value of f to the dispersion characteristics for a given case. These characteristics are the drop size distribution, the Sauter mean diameter, the volume fraction, and the interfacial area.

CALCULATION PROCEDURE

Calculations using these equations were carried out on the Purdue University CDC 6500 computer. For each case study, 10,000 drop radii were generated to give the desired drop size distribution, and the total volume of the drops was calculated. The desired volume fraction and optical path length were specified, allowing calculation of the light beam cross-sectional area through Equation (2). Equations (3) and (4) gave the interfacial area and the Sauter mean diameter for the dispersion under study. Finally, Equation (7b) was used to calculate the fraction of light transmitted for the corresponding values of the dispersion parameters. Repetitive calculations gave data on the behavior of f as a function of the drop size distribution, interfacial area, Sauter mean diameter, volume fraction, and optical path length.

Three different types of drop size distributions were investigated: log-normal, volume-normal, and Schwarz-Bezemer. Of these three distributions, the log-normal is most often found (Rushton and Roy, 1955; Nagata and Yamaguchi, 1960; Yamaguchi and Yabuta, 1963; Keey and Glen, 1969; McLaughlin, 1970), with volume-normal (Chen and Middleman, 1967) and Schwarz-Bezemer (Schwarz and Bezemer, 1956; Sprow, 1967) less often reported.

The parameters in each distribution equation were varied to give drop size distributions with different Sauter mean diameters. For instance, the radius of a drop under a log-normal density function is given by

$$r_i = e^{(\mu + \sigma N_i)} \quad (8)$$

where μ (the mean of the natural logarithm of the radius) and σ (the standard deviation of the natural logarithm of the radius) are parameters of the distribution function. Experimental investigations have shown that the value of σ in Equation (8) generally lies in the range from zero to 2.0 for liquid-liquid dispersions. A value of zero

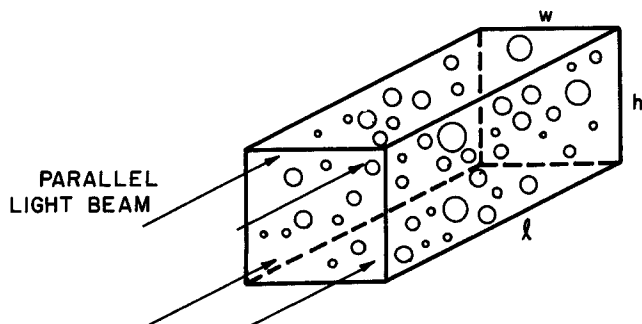


Fig. 2. Light transmission through a dispersion.

corresponds to a uniform drop size, while larger values gradually increase the range of drop sizes for that dispersion. The value of μ in Equation (8) largely determines the Sauter mean diameter of the dispersion. These two parameters were varied (σ from zero to 2.0 and μ from -2.3 to -9.0) along with the dispersion parameters to give the following range of variables studied for the log-normal distribution:

$$\bar{d}: 0.025 \text{ to } 0.50 \text{ cm}$$

$$a: 0.70 \text{ to } 7.8 \text{ cm}^{-1}$$

$$l: 1.0 \text{ to } 10.0 \text{ cm}$$

$$\phi: 0.01 \text{ to } 0.60$$

$$f: 0.007 \text{ to } 1.0$$

A similar range of variables was investigated for the volume-normal and Schwarz-Bezemer drop size distributions.

RESULTS

Log-Normal Distribution

The results of the calculations for a log-normal drop size distribution are summarized by Figure 4 where the fraction of light transmitted f is plotted against a dimensionless grouping of the variables ϕ , l , and \bar{d} . The plot shows 62 representative points picked at random from the 249 cases calculated for this distribution to illustrate the range of the results. The dimensionless group, defined here as the Transmission Number N_T , is

$$N_T = \frac{\phi l}{\bar{d}} \quad (9a)$$

If Equation (3) for interfacial area is combined with this definition, the transmission number can also be expressed as

$$N_T = \frac{a l}{6} \quad (9b)$$

Figure 4 shows that the transmitted light is a unique function of the transmission number for all values of σ studied. This is equivalent to saying that the fraction of light transmitted through a log-normal dispersion is a unique function of N_T regardless of the range of drop sizes. The data and curve are summarized by

$$f = \exp[-1.5 N_T] \quad (10)$$

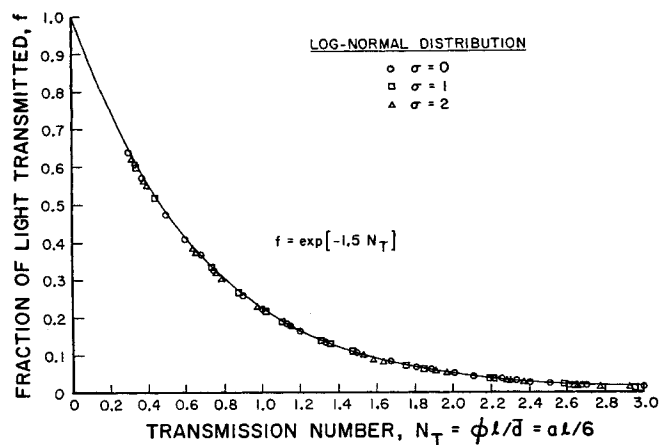


Fig. 4. Effect of transmission number on light transmitted; log-normal drop size distribution.

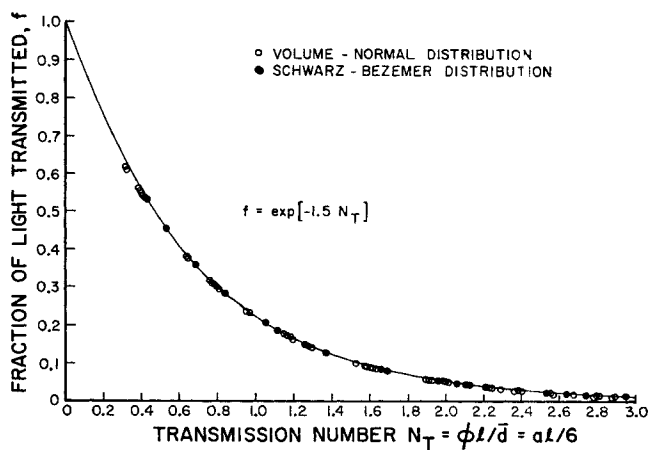


Fig. 5. Effect of transmission number on light transmitted; volume-normal and Schwarz-Bezemer distributions.

Volume-Normal and Schwarz-Bezemer Distributions

Figure 5 shows the relationship between the transmitted light and N_T for the volume-normal and Schwarz-Bezemer drop size distributions. Once more, the transmitted light is a unique function of N_T , regardless of the range of drop sizes. These data also fit Equation (10) and the curves for all three types of distributions coincide exactly, showing that the light transmission characteristics of a dispersion are independent of the type of drop size distribution as well as the range of drop sizes, and the coefficient of 1.5 is exact.

Uniform Drop Size

Equation (10) in a different form has been proposed by Calderbank (1958) for uniform drop size.

$$\log(1/f) = \frac{a l}{9.210} \quad (11)$$

We now find that this equation is applicable to nonuniform distributions as well. Or, conversely, that Equation (10) covers uniform size as well as different size distributions. It is interesting to note that Equation (7b), when applied to a uniform drop size, approaches Equation (10) in the limit as the number of drops studied goes to infinity.

Sauter Mean Diameter

With regard to nonuniform drop size distributions, Dobbins and Jizmagian (1966) concluded that the mean scattering cross section of a nonuniform dispersion is primarily a function of the Sauter mean diameter, and this is in agreement with these results.

LIGHT TRANSMISSION EXPERIMENTS

To investigate the accuracy of the simulation results in an actual application, experiments were carried out in which light transmission measurements were made for glass bead suspensions in water. The glass beads were transparent, spherical, and ranged from 0.10 to 0.84 mm in diameter. Division of the beads into six size ranges allowed the study of four different uniform sizes and three different log-normal size distributions.

The glass beads were suspended in a continuous water phase by a 13 cm six flat-blade turbine impeller in a 30 cm baffled mixing tank. The mixing tank geometry and impeller design were the same as those recommended by Rushton (1950), where the cylindrical mixing tank was fitted with four equally spaced wall baffles, each one-tenth of the tank diameter in width. The liquid depth in each experiment was equal to the tank diameter, and the impeller was placed at one-third off bottom.

A light probe, shown schematically in Figure 6, was immersed into the suspensions to measure the fraction of light

transmitted for each suspension. This light probe incorporates a parallel light beam and a small phototube receiving angle (less than 1°), as required by the assumptions of the mathematical model. First-surface mirrors and optical black paint were used to help ensure that the phototube receives only parallel light. The horizontal section of tubing adjacent to the optical gap was threaded so that different tubing lengths could be inserted to give optical path lengths from 10 to 43 mm. Calibration of the light probe was accomplished with neutral density filters relating the electrical output of the phototube to the fraction of light transmitted across the gap.

For each experiment, a known volume of glass beads (of known size distribution) was placed into the mixing tank, and water was added to give a total depth of 30 cm. The impeller was then rotated at the maximum speed for which no air was drawn below the surface, which suspended the glass beads as uniformly as possible. After a homogeneous suspension was achieved, the light probe was placed into the mixing tank so that the optical gap was in the discharge stream of the impeller, perpendicular to the flow at that point. Finally, the fraction of light transmitted was measured for the particular volume fraction, bead size distribution, and optical path length under study.

The results are illustrated in Figure 7 where the transmission number calculated from the light reading [Equation (10)] is plotted against the known transmission number of the suspension [Equation (9a)]. The plot shows that Equation (10) adequately describes the light transmission characteristics of all the dispersions studied regardless of the size distribution, volume fraction, or optical path length used. Thus, the results of the mathematical model were verified, and it was shown that the assumptions can be satisfied in the real case.

APPLICATION OF THE RESULTS

The results from the computer study, verified by actual light transmission experiments, indicate that measurement of the amount of parallel light transmitted through any liquid-liquid dispersion will allow calculation of the interfacial area (and drop size) even though the drop size distribution is unknown. It must be emphasized, however, that this relationship will hold only if the fundamental assumptions of the mathematical model are satis-

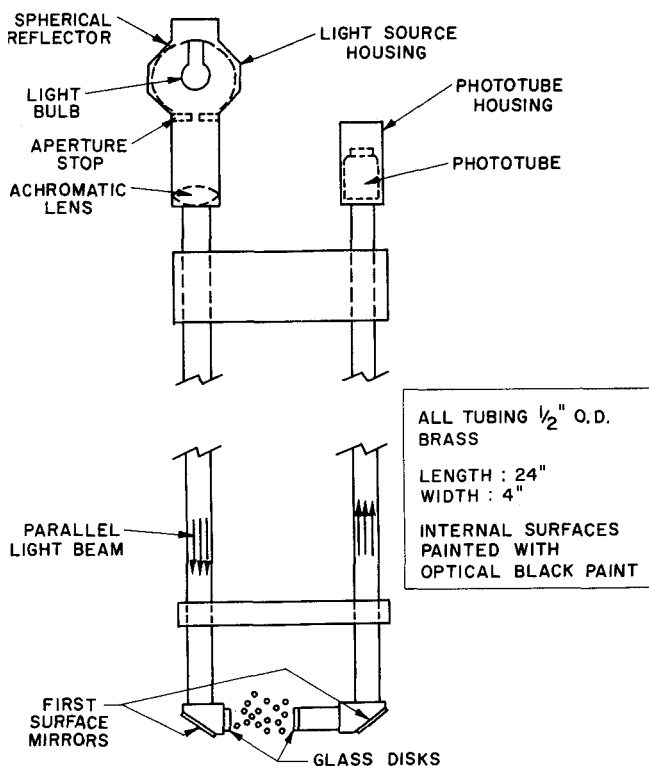


Fig. 6. The experimental light probe.

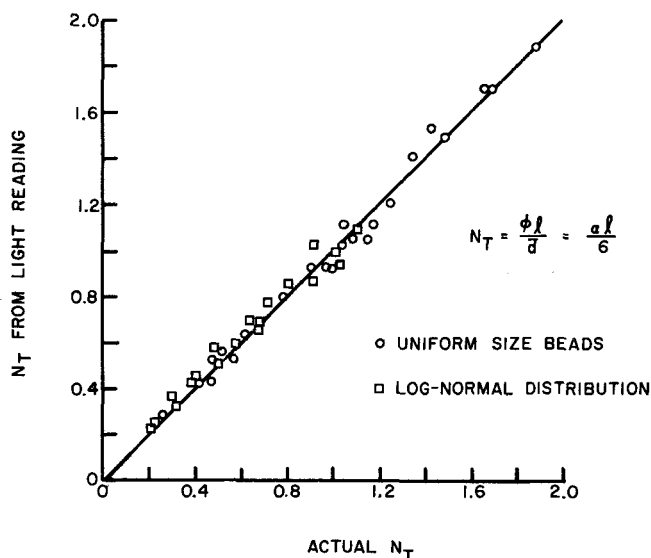


Fig. 7. Calculated versus actual transmission numbers.

fied in the application of these results. The necessary conditions are that:

1. There be a transparent continuous phase;
2. There be random drop locations;
3. The drops are greater than 0.10 mm in diameter (that is, the minimum value of α is 450);
4. The drops are subject to random orientation in the light beam;
5. The drops have no concave surfaces;
6. The light source emits an incoherent parallel light beam; and
7. The light detector receives only parallel light.

It should be noted that the restriction of spherical drops has been replaced by assumptions 4 and 5. Actually, the assumption of spherical drops was made earlier as a matter of convenience [for Equations (2), (4) and (5)] rather than necessity. It has been shown by Cauchy (1841) that the average projection area of any particle is one-fourth of its surface area, provided it has no concave surfaces and is subject to random orientation in the light beam. Thus, Equation (10) applies to spheres, ellipsoids, cubes, cylinders, etc., as long as the equations for volume fraction and Sauter mean diameter are modified accordingly.

As mentioned previously, the refractive indices of the two phases are an additional consideration in the practical application of these results. If the refractive index ratio of the two phases is near 1.0, then a light detector of high angular resolution will be required to prevent errors due to forward scattering. On the other hand, an opaque dispersed phase will not require as high an angular resolution because forward scattering would be negligible in this case. Lothian and Chappel (1951) and Walstra (1965) discuss these considerations in detail.

To give an example of the application of Equation (10), it can be transformed to

$$a = \frac{-4 \ln f}{l} \quad (12)$$

If a dispersion of drops transmits 32% of the incident light over an optical path length of 2.10 cm, the interfacial area per unit volume of dispersion is

$$a = \frac{-4 \ln (0.32)}{2.10} \text{ cm}^2/\text{cm}^3$$

$$a = 2.17 \text{ cm}^2/\text{cm}^3$$

Furthermore, if the volume fraction is 0.06, the Sauter mean diameter of the dispersion is

$$\bar{d} = \frac{6\phi}{a}$$

$$\bar{d} = \frac{6(0.06)}{2.17} \text{ cm}$$

$$\bar{d} = 0.166 \text{ cm}$$

Note that the volume fraction of dispersed phase is not needed to calculate the interfacial area per unit volume of dispersion. The volume fraction must be known only to calculate the Sauter mean diameter or the interfacial area per unit volume of dispersed phase ($a' = a/\phi$).

Whatever the application, it is recommended that the fraction of transmitted light be kept above 0.10. Below this value the errors due to multiple and forward scattering increase sharply. Figures 4 and 5 show that the fraction of light transmitted will be greater than 0.10 if the value of N_T is less than 1.53. Thus, the maximum recommended optical path length for a dispersion defined by ϕ , \bar{d} and a is given by

$$N_T \text{ max} = 1.53 = \frac{\phi l \text{ max}}{\bar{d}} = \frac{a l \text{ max}}{6}$$

or

$$l \text{ max} = \frac{1.53 \bar{d}}{\phi} = \frac{9.18}{a} \quad (13)$$

Equation (13) emphasizes that the volume fraction alone does not determine the light transmission characteristics of a dispersion.

ACKNOWLEDGMENT

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NOTATION

- a = interfacial area per unit volume of dispersion
- a' = interfacial area per unit volume of dispersed phase
- a_p = geometric cross section of a drop
- a_s = scattering cross section of a drop
- \bar{d} = Sauter mean diameter of the drops, otherwise called the area average diameter
- d_i = the diameter of the i th drop
- f = the fraction of light transmitted through a dispersion
- h = the height of a lightbeam cross section
- K_s = the scattering coefficient of a drop, defined by $\alpha l/6$
- l = optical path length
- n = the total number of drops in the dispersion
- N_i = a random deviate under a standard normal density function
- N_T = the Transmission Number, defined as $\phi l/\bar{d}$ or $a l/6$
- P = the probability of an event
- r_i = radius of the i th drop
- w = the width of a lightbeam cross section

Greek Letters

- α = size parameter for a dispersion, equal to $\pi d/\lambda$

- λ = wavelength of the incident light
- μ = mean of $\ln(r)$ for a log-normal drop size distribution
- π = numerical constant approximated by 3.14159
- σ = standard deviation of $\ln(r)$ for a log-normal drop size distribution: a measure of the range of drop sizes
- ϕ = volume fraction of dispersed phase

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