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Theoretical and experimental study of the diffuse transmission of light through highly concentrated absorbing and scattering materials Part I: Monte-Carlo simulations

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ABSTRACT

In many technical materials and commercial products like sunscreen or paint high particle and absorber concentrations are present. An important parameter for slabs of these materials is the diffuse transmission of light, which quantifies the total amount of directly and diffusely transmitted light. Due to the high content of scattering particles not only multiple scattering but also additional dependent scattering occurs. Hence, simple analytical models cannot be applied to calculate the diffuse transmission. In this work a Monte-Carlo program for the calculation of the diffuse transmission of light through dispersions in slab-like geometry containing high concentrations of scattering particles and absorbers is presented and discussed in detail. Mie theory is applied for the calculation of the scattering properties of the samples. Additionally, dependent scattering is considered in two different models, the well-known hard sphere model in the Percus–Yevick approximation (HSPYA) and the Yukawa model in the Mean Spherical Approximation (YMSA). Comparative experiments will show the accurateness of the program as well as its applicability to real samples [1].

1. Introduction

The diffuse transmission of light through thin slabs of scattering and absorbing materials is an interesting and important topic as it has wide applications in daily life, e.g. for cosmetics, paints or solar cells. A deep understanding of the light transport in these layers and a quantification of the diffusely transmitted or reflected light is therefore of utmost importance for the design of the above-mentioned materials.

Due to high particle concentrations, typical for such systems, multiple scattering and in case of particle interaction even dependent scattering occurs. Additionally, in the presence of absorbers absorption influences scattering and vice versa. The diffuse transmission of these materials is therefore not a simple function of its scattering and absorption properties (namely the scattering and absorption coefficients μ_s and μ_a, respectively). Hence, different approaches have been developed to model the light transport through this kind of systems like the Analytical Theory (AT) [2, Chapter 14] or Radiative Transfer Theory [3]. Whereas in the first one the problem of light transport is treated by solving Maxwell's Equations which can be mathematically complex, the second one treats light transport as a transport of power in a heuristic way. The solution of the Radiative Transfer Equations (RTE) is a complex problem depending on many aspects like the optical properties of the considered material, the beam shape of incident light or the geometry of the sample.

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There are several approximate solutions of the RTE like Kubelka–Munk Theory [4], Four-Flux Theory [2,5] or the Diffusion Approximation (DA) [2, Chapter 9, 6]. Whereas the first ones lack the ability to introduce lateral sample boundaries and in case of Kubelka–Munk Theory to consider collimated incident light, the DA is restricted to samples where $\mu_a \ll \mu_s$ and the results obtained with the DA can vary depending on the applied boundary conditions. On the contrary, Monte-Carlo (MC) simulations provide an excellent alternative to model the light transport in such systems without making any rigorous assumptions. MC simulations can even be applied to highly structured samples consisting of various materials with different optical properties.

MC simulations are widely used to model light transport in scattering and absorbing media [7–11]. In most cases the Henyey–Greenstein phase function is used to calculate polar scattering angles. However, to our knowledge there is no work including the effect of dependent scattering. In this study a MC program was developed to calculate the diffuse transmission of a liquid dispersion containing scattering particles and absorbers with arbitrary optical properties. The program is designed to match a real experiment and includes all experimental parameters like sample boundaries, refractive indices of the materials used or the detector geometry. In contrast to other work Mie theory was applied providing the exact phase function for arbitrary particle sizes. Additionally, dependent scattering is considered either applying the well-known hard sphere model in the Percus–Yevick approximation (HSPYA) or the Yukawa model in the Mean Spherical Approximation (YMSA) for additional electrostatic particle interactions. In this paper the details of the program will be presented as well as results obtained for different parameters. In a second study [1] the MC simulations will be compared with experimental results.

2. Theory and Monte-Carlo program

In the following the optical parameters and the considered geometry used in this work are described as well as the MC program.

2.1. Intensity and attenuance

In the following a slab-like sample with thickness L_z [m], consisting of spherical particles of radius a [m] embedded in a liquid medium, is considered. In Fig. 1 the scheme of the sample is shown. Collimated light of intensity I_0 [J m^{-2} s^{-1}], incident on the slab can be either scattered by the particles or absorbed by the particles or the liquid medium. Here, μ_s [m^{-1}] and μ_a [m^{-1}] are the scattering and absorption coefficients of the sample, respectively. Provided that the light is only scattered once during its path through the sample (low particle concentrations, low slab thickness L_z, small μ_s) a detector with a small acceptance angle placed at a large distance behind the sample would measure the directly transmitted intensity

$$I_T = I_0 \exp(-[\mu_a + \mu_s] L_z). \tag{1}$$

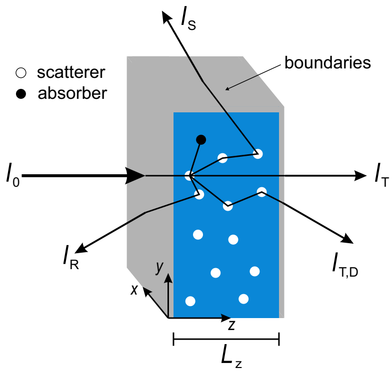

Fig. 1. Schematic slab-like sample consisting of scattering and absorbing particles embedded in a liquid absorbing medium.

The attenuance of the transmitted light is defined as

$$D = \ln\left(\frac{I_0}{I_T}\right) = [\mu_a + \mu_s] L_z. \tag{2}$$

The total transmitted intensity also including diffusely transmitted light $I_{T,D}$ has the intensity $I_T + I_{T,D}$. Here, the diffuse attenuance D_D is defined as

$$D_D = \ln\left(\frac{I_0}{I_T + I_{T,D}}\right). \tag{3}$$

D_D is not proportional to μ_a and μ_s anymore. Due to surface reflections or scattering light of intensity I_R and I_S can leave the sample on the front or the side (for finite extensions in x or y), respectively.

2.2. Absorption and scattering

As long as no interaction between the absorbers takes place the absorption coefficient μ_a is proportional to the molar concentration [*Abs*] [mol L^{-1}] of any absorbing species in the sample like the particles or the surrounding liquid medium. In case of a pure homogeneous medium of a complex refractive index $\tilde{n} = n + i\kappa$, μ_a can be described by the imaginary part κ

$$\mu_a = \varepsilon_a [Abs] = \frac{4\pi\kappa}{\lambda}, \tag{4}$$

where ε_a [m^{-1} L mol^{-1}] is the molar absorption coefficient and λ [m] is the wavelength of light. The scattering coefficient can be calculated by

$$\mu_s = \frac{3\phi Q_s}{4a}, \tag{5}$$

where $\phi = N_p V_p / V$ is the volume fraction of the dispersed particles in the sample ($V_p = 4\pi a^3/3$: volume of one particle [m^3], V: volume of the sample [m^3], N_p: number of particles within the sample). Q_s is the scattering efficiency for a particle of radius a and is calculated by

$$Q_s = \frac{1}{x^2} \int_{-1}^{1} i_{12}(\theta_s)\, d\cos(\theta_s), \tag{6}$$

where i_{12} is the angular scattering intensity, $x = 2\pi a/\lambda$ is the size parameter and θ_s is the polar scattering angle. In this study Mie theory is applied for the calculation of i_{12}.