Anisotropic reflectance from turbid media. II. Measurements

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The anisotropic reflectance from turbid media predicted using the radiative transfer based DORT2002 model is experimentally verified through goniophotometric measurements. A set of paper samples with varying amounts of dye and thickness is prepared, and their angle resolved reflectance is measured. An alleged perfect diffusor is also included. The corresponding simulations are performed. A complete agreement between the measurements and model predictions is seen regarding the characteristics of the anisotropy. They show that relatively more light is reflected at large polar angles when the absorption or illumination angle is increased or when the medium thickness is decreased. This is due to the relative amount of near-surface bulk scattering increasing in these cases. This affects the application of the Kubelka–Munk model as well as standards for reflectance measurements and calibration routines. © 2010 Optical Society of America

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1. INTRODUCTION

Radiative transfer theory has been studied throughout the last century to describe the propagation of light in turbid media. Different models of varying degree of complexity have been applied in several scientific and industrial fields, ranging from astrophysics and neutron diffusion to color predictions in plastics, textile, and paper. The first theoretical attempts were made by Schuster [1], while the general radiative transfer theory is usually ascribed to Chandrasekhar [2]. The most widespread industrial model is the one presented by Kubelka and Munk [3–5] (hereafter denoted KM), but now efficient general solution methods exist, such as DISORT [6] and DORT2002 [7].

Schuster acknowledged the importance of angular variations in the light distribution within a medium, but found the matter too complex for further investigation. Other authors have observed angular variations, or anisotropy, and have discussed its potential importance [8–10]. One important difference between two-flux models like KM and more general solution methods like DORT2002 is that the latter can describe and handle the angular resolution of light. The KM model assumes an isotropic single scattering process, and is further based on the implicit assumption that this, together with a perfectly diffuse (Lambertian) illumination, will give isotropic light intensity throughout the medium. This is not at all true, although several industrial applications rely on this assumption. The disagreement between KM and general radiative transfer theory has been investigated by Edström [11], who noted that the error is larger for optically thin and highly absorbing media.

Part I of this work [12] (appearing consecutively in this issue) theoretically investigates anisotropy, explains how it arises in plane-parallel turbid media and shows that anisotropy is present in all real situations. It is shown that the anisotropy depends on the relative contributions of different scattering depths in the medium, and that *near-surface bulk scattering* increases the relative reflectance in large polar angles. Increasing absorption, transmittance, or the average angle of incidence of the illumination causes the amount of near-surface bulk scattering to increase, and consequently also the relative reflectance in large polar angles increases. In this sense the anisotropy increases as the amount of near-surface bulk scattering increases. These results hold even for an isotropic single scattering process, and asymmetric single scattering will obviously introduce further anisotropy. It is then concluded that there can be no perfect bulk-scattering diffusor, which has practical impact on, e.g., calibration routines. It is further shown that anisotropy makes instruments measuring only in one direction overestimate or underestimate the total reflectance, depending on the particular instrument geometry and medium properties. Part I also derives validity conditions for the KM model, and shows that errors can be 20–40% in a large region of medium parameters. But it is also shown that an optimal detection angle can be determined that minimizes the KM model error for a given medium or experimental setup, which could be exploited for designing reflectance measurement setups.

The purpose of part II (this paper) is to experimentally confirm the theoretical findings in part I and to discuss measurement related issues concerning anisotropy and consequences for industrial applications. A specific aim is thus to use measurements to verify the theoretical predictions from part I regarding increased anisotropy—in the form of increased relative reflectance in large polar angles—for highly absorbing and optically thin media as well as oblique illumination. Another aim is to verify the predicted anisotropic reflectance of an alleged perfect diffusor. One additional aim is to discuss consequences for some of these findings on practical measurements and industrial applications, e.g., for standardized diffuse reflectance measurements, their calibration, and their interpretation using the KM model. One final aim is to discuss the possibility to design a measurement setup with a corresponding data interpretation model that is in some respect optimal.

This part of the paper is more applied, and the reader is referred to part I [12] for a theoretical treatment, including definitions and equations.

2. METHOD

In order to experimentally investigate angle resolved reflectance and how it depends on medium properties, a series of paper handsheets was made and their reflectance measured with a goniophotometer. The handsheets contain different amounts of dye and vary in grammage. Increasing the amount of dye is equivalent to decreasing the single scattering albedo in the wavelengths corresponding to the region of absorption of the dye. The grammage can be treated as equivalent to the paper thickness [13], and is proportional to the paper optical thickness. The handsheets thus allow for an investigation of how the deviation from an isotropic intensity distribution depends on albedo and medium optical thickness. A corresponding investigation was also made for a “perfect reflecting diffusor” using a sample that is provided by instrument suppliers for calibration purposes. In this case the illumination was varied to study its influence on anisotropy. To compare measurements with the theoretical predictions in part I of this work, simulations were made with the angle resolved DORT2002 model. (The DORT2002 software is freely available from the authors.)

A. Material

The paper handsheets used in this work were prepared with a Formette Dynamique. All handsheets were made from a mix of equal amounts of kraft and birch pulps having Shopper/Riegler numbers of 18° and 23° SR, respectively. Sheets of two approximate grammages, 30 and 60 $\mathrm{g/m^2}$, were prepared. All sheets contain 0.02% PAM, 0.2% Bentonit, 0.35% Cerestar, and 22% filler (PCC Syncarb FO 474-MJ). The amount of blue dye was changed from 0 to 1% of the fiber weight in four steps. This gives eight sheets in total and is summarized in Table 1. The dye used was Levacell Fast Blue KS-6GLL Liquid, manufactured by Lanxess. This is a cationic direct dye, and it does not affect the structure of the paper. The sheets were dried in a cylinder dryer for 5 min at a temperature of 105 °C and a pressure of 1.50 bars.

Table 1. Overview of the Paper Handsheets Used in This Work

Sheet No.	Grammage ($\mathrm{g/m^2}$)	Dye (% of fiber weight)
1	31.80	0.00
2	31.69	0.25
3	32.54	0.50
4	32.27	1.00
5	64.97	0.00
6	65.73	0.25
7	66.91	0.50
8	65.87	1.00

This process assures that the paper sheets have minimal surface effects, such as, e.g., gloss. In this way they can be considered to be purely bulk scattering turbid media, and their interaction with light is thus accurately described by radiative transfer theory.

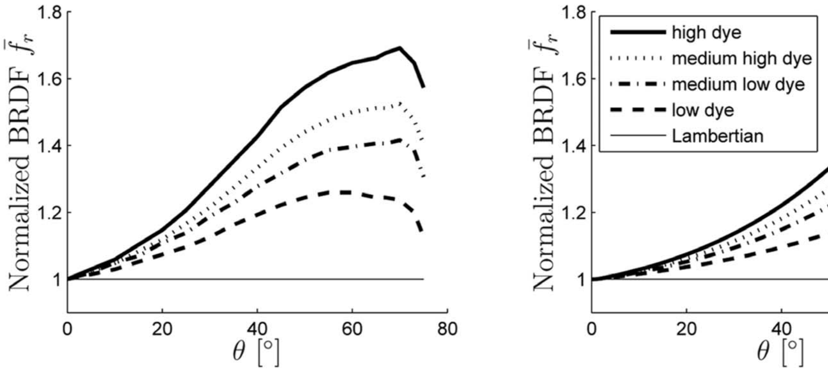
B. Measurements

The angle resolved measurements were made with a Zeiss GP3 goniophotometer. This instrument uses collimated illumination from an incandescent lamp, and the detector angle can be adjusted in an interval 0°–75° from the sample normal on the side opposite to the lamp. The illumination angle was kept constant at 45° when illuminating the paper samples. An area much larger than the measurement spot is illuminated. The instrument is calibrated by adjusting the detector reading from a glass plate with known specular reflectance. The detector aperture has a radius of 2.5 mm, and the reflected light is filtered through a Y-filter giving the Y tristimulus value [14]. Measurements were made with this instrument on single paper sheets and on opaque pads of paper sheets. A thick black glossless paper was used as background. This instrument was also used to assess the angle resolved reflectance from an alleged perfect diffusor. The barium sulphate diffusor delivered with the instrument was used. The angle of incidence of the illumination was varied in this case in order to study its influence on the reflectance from the diffusor.

C. Simulations

The DORT2002 model [7] is adapted to light scattering simulations in paper and print, has been successfully applied to real paper industry problems [11,15,16], and has been thoroughly evaluated [17,18]. DORT2002, being angle resolved, is in this work used to predict the paper samples’ deviation from isotropic reflectance and the corresponding dependence on albedo and optical thickness. The same model is also used to predict the anisotropic reflectance from the “perfect diffusor.” DORT2002 is adjusted to simulate the specific illumination and detection geometry of the measurement setup, and a numerical Y-filter [14] is implemented. The experimental results can thus be compared with the theoretical results.

In order to obtain values for the paper samples’ albedo and optical thickness, the samples were measured with a Datacolor SF450 Elrepho complying with ISO2469 [19]. This instrument has d/0 geometry, meaning that the illumination is diffuse and the detector is centered around 0°, i.e., the normal direction of the paper. Using these reflectance data, calculations were performed with DORT2002 adjusted to this instrument geometry (more details are given by Edström [16]). This procedure gives numerical values for the paper samples’ albedo and optical thickness that can be used when simulating the goniophotometer



(a) Measured BRDFs (b) Simulated BRDFs

Fig. 1. (a) Measured and (b) predicted BRDFs for increasing absorption. Absorption is increased in four steps corresponding to increasing amounts of dye. The BRDFs are scaled to coincide at polar angle $\theta=0°$. Lambertian (perfectly diffuse) reflectance is included for reference. It can be seen that the anisotropy increases with increasing absorption in both measurements and simulations in the same characteristic way where relatively more light is reflected at large polar angles.

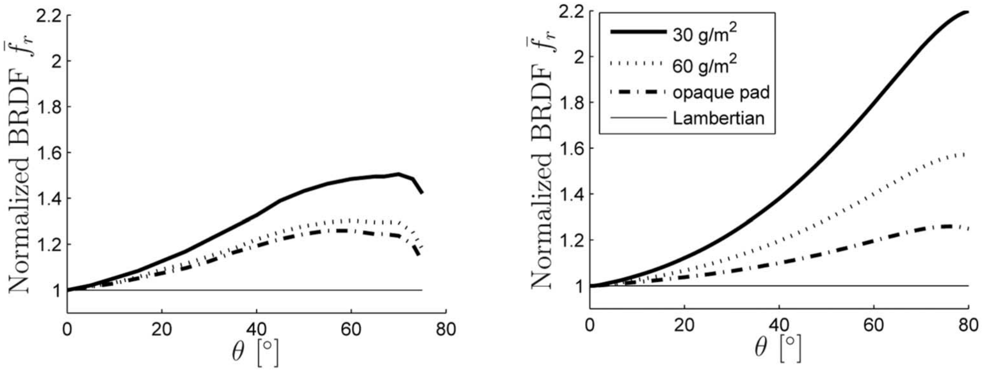
described in 2.BSubsection 2.B. The asymmetry factor of the Henyey–Greenstein phase function [20] describing the anisotropy of each single scattering, usually denoted g, is set equal to 0.8, which is the only reported value for g in a paper application [21]. This corresponds to forward scattering dominating the single scattering process, since $g=1$ is complete forward scattering and $g=0$ is isotropic single scattering.

3. RESULTS

A. Dependence of Anisotropy on Medium Absorption and Thickness

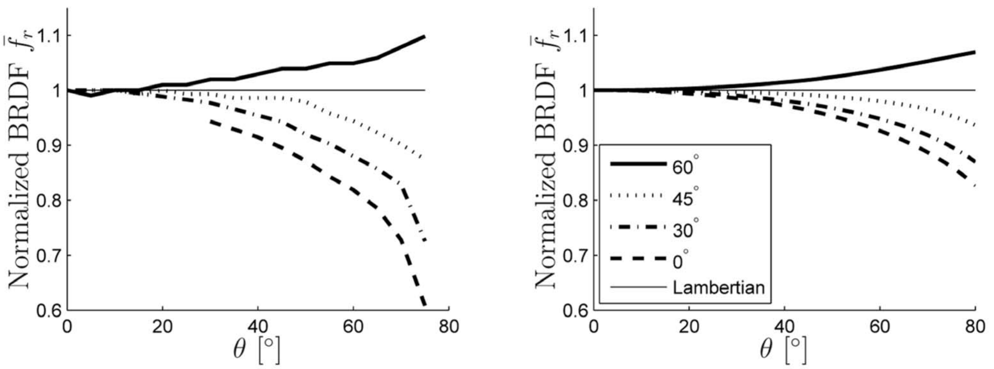
To study the dependence of anisotropy on absorption, opaque pads of paper sheets 1–4, with varying amounts of dye, are measured and simulated. In this way the effect of absorption is isolated, since there is no transmittance. Goniophotometer measurement data and the corresponding simulations are shown in Fig. 1. The data are presented in terms of the bidirectional scattering-distribution function (BRDF, f_r), which is proportional to the intensity I and the reflectance factor R [22]. The BRDFs are normalized to coincide at polar angle $\theta=0°$ in order to highlight the dependence of the anisotropy on absorption. That is, if the BRDF of sample i is denoted f_r^i, the normalized BRDF $\bar{f}_r^i$ is $\bar{f}_r^i(\theta)=f_r^i(\theta)/f_r^i(0°)$, where the dependence on polar angle θ is included. It can be seen in Fig. 1 that the anisotropy increases as the absorption increases since relatively more light is reflected at large polar angles. This characteristic behavior is common to both measurements and simulations, and is in agreement with the findings of part I of this work. The decrease of the measured reflectance at grazing angles is unexpected and can probably be attributed to instrument specific limitations in these angles.

The dependence of anisotropy on medium optical thickness can be studied by using measurement data from paper sheets of varying grammage and an opaque pad of paper sheets. Sheets 1 and 5 are used since they do not contain dye, thus isolating the effect of transmittance. In Fig. 2 these data together with the corresponding model predictions are shown. It can be seen that the anisotropy increases in the same characteristic way in both measurements and simulations as the medium optical thickness decreases. Relatively more light is reflected at large polar angles for thin media, i.e., when transmittance is significant, and the anisotropy is larger in this sense for thin media. The reflectance closest to isotropic (Lambertian) is



(a) Measured BRDFs (b) Simulated BRDFs

Fig. 2. (a) Measured and (b) predicted BRDFs of a 30 $\mathrm{g/m^2}$ paper, a 60 $\mathrm{g/m^2}$ paper, and an opaque pad of paper sheets. No papers contain dye. The BRDFs are scaled to coincide at polar angle $\theta=0°$. Lambertian reflectance is included for reference. It is seen that the relative reflectance in large polar angles increases as the medium thickness decreases. Thus the anisotropy is larger in this sense for thin media.



(a) Measured BRDFs (b) Simulated BRDFs

Fig. 3. Measured BRDFs of (a) a barium sulphate diffusor and predicted BRDFs of (b) a non-absorbing and non-transmitting medium with varying illumination angle. Angles 0°, 30°, 45°, and 60° are used. It is seen that the relative reflectance at large polar angles increases as the illumination angle increases. The reflectance can thus be anisotropic both from an alleged perfect diffusor (a) and an ideal bulk scattering diffusor (b). Lambertian reflectance is included for reference.

as expected obtained from the opaque pad of paper sheets. These results are in complete agreement with the findings of part I of this work.

B. Dependence of Anisotropy on Illumination: The Non-Diffuse Reflectance of the Perfect Diffusor

The dependence of anisotropy on illumination can be investigated by measuring and simulating the reflectance from a highly scattering and non-transmitting medium, i.e., what is normally believed to be a perfect diffusor. This eliminates effects of absorption and transmittance. The barium sulphate diffusor delivered with the Zeiss GP3 goniophotometer is used here, and in the DORT2002 model the albedo is set equal to one, and the optical thickness is increased until the point of zero transmittance. Isotropic single scattering, i.e., $g=0$, is assumed in the simulations. Assuming $g\neq 0$ would introduce further anisotropy, and isotropic single scattering is a necessary condition for isotropic reflectance in this case. In both measurements and simulations the illumination angle is varied, and incident angles 0°, 30°, 45°, and 60° are used. When illuminating in 0° the reflectance cannot be measured for polar angles $\theta < 30°$ because of physical constraints of the instrument. Figure 3 shows the measured and predicted BRDFs with varying illumination angle. Also in this case the normalized BRDF $\bar{f}_r$ is plotted. It can be seen that the relative reflectance at large polar angles increases as the illumination angle is increased, and that measurements and simulations show the same behavior. The relative reflectance is larger in the normal direction if the illumination is incident around the normal direction. These results are also in agreement with the results from part I of this work.

4. DISCUSSION AND CONCLUSIONS

It has been seen that the measured and model predicted reflectance from various turbid media, including an alleged perfect diffusor, show the same type of dependence between anisotropy and medium absorption, optical thickness, and illumination. Increasing absorption and illumination angle or reducing the thickness has the same characteristic effect on the anisotropy, since this increases the relative reflectance at large polar angles. This is in complete agreement with the conclusions of part I of this work. The correlation between measurements and simulations shows that an explanation of the observed phenomenon can indeed be found within the framework of radiative transfer theory. Using the conclusions from part I, the results can be understood by considering the amount of near-surface bulk scattering. Increasing absorption or illumination angle, or reducing the optical thickness increases the amount of near-surface bulk scattering, which leads to higher reflectance at large polar angles. This holds since light deeper inside a highly absorbing or transmitting medium has a high probability of being absorbed or transmitted, and since the light has to travel a larger distance to reach a certain depth if the illumination angle is increased. The angular distribution of the reflected light is thus determined by the relative contributions to the reflectance from different depths in the medium, and the relative contributions are determined by the absorption, optical thickness, and illumination.

A. Consequences for the Kubelka–Munk model

It has been seen in this work that the light reflected from common paper samples can be highly anisotropic. The samples studied are comparable to, e.g., common office paper or newsprint, so the phenomenon is present in everyday situations. Because of this, the KM model is invalid in practically all situations, since it is based on the false assumption that the light is isotropic throughout the medium. The amount of error introduced when assuming this was indicated in part I of this work to be 20–40% in a large region of parameters, but this holds under the assumption of isotropic single scattering. Anisotropic single scattering will increase the error further. To reduce this error, angle resolved models are necessary, but it is also possible to customize a measurement setup for a particular set of samples as described in part I. The determination of the KM scattering and absorption coefficients is a standardized procedure [23], but it is important that anisotropy be taken into account in these calculations.

B. Effect of Anisotropy on Established Instruments

If reflectance measurements are performed with instruments having different illumination and detection geometries, anisotropy has to be taken into account to be able to compare the measurements. If assuming isotropically reflected light the inter-instrument difference will vary depending on the measurement setup and the sample properties. This is further investigated for the common 45/0 [24] and d/0 [19] geometries by the authors [25].

Calibration routines involving perfect diffusors are affected by the anisotropy of the light reflected from the diffusor if the anisotropy is not taken into account. Judging from Fig. 3, the barium sulphate diffusor, which is a commonly used material, probably has anisotropic single scattering, since in the measurements the light tends to be scattered more in the forward direction compared to the simulations. This further increases the effect of anisotropy, and it is more severe if directed illumination is used since illuminating diffusely to some extent reduces this effect. Though, calibration can in many situations improve the results if the total reflectance is overestimated or underestimated in the same way in both calibration and actual measurements. But without taking angular variations into account the measurements will contain errors due to the anisotropy, and standards for reflectance measurements should be modified accordingly.

C. Optimal Instrument Geometry for Reflectance Measurements

As described in part I of this work an optimal instrument in the sense that it minimizes the error in the KM model can be readily constructed for a set of samples. The instrument detection geometry would have to be modified if changing the error tolerance or the set of samples, and the modification would require angle resolved calculations. In order to construct a generic instrument without need for recalibration it is necessary with several detection angles. Given that the reflectance is fairly monotone with respect to polar angle, it could be enough with two or three detection angles. Combining accurate measurements in several directions with an angle resolved model such as DORT2002 would give access to the objective and physical scattering and absorption coefficients together with the asymmetry factor of the medium. This would significantly improve the model-measurements interaction and open up possibilities for further investigations. However, other phenomena such as surface roughness or gloss can affect the measurements, and to stay optimal these have to be included in the model.

5. FUTURE WORK

Measuring the spectral reflectance in several directions allows for an investigation of the spectral dependence of the asymmetry factor. This work is planned by the authors and will be published shortly. Furthermore, fluorescence is present in many situations. By including this in an angle resolved model it can be understood more thoroughly. Also, effects of surface roughness, gloss, or the refractive index have to be taken into account to develop a more comprehensive model. The authors believe that better instruments for reflectance measurements can be constructed, and that they can be combined with angle resolved models such as DORT2002. This would allow for a better characterization of the light scattering properties of the medium and open up possibilities to optimize, for example, the optical appearance.

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