Optical calculations for multilayer papers

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ABSTRACT: The experimental results of this study showed that the one-dimensional Stokes equation cannot always predict the reflectance of multilayer papers. The optical properties of a layer depend on the angular distribution of the incident light. Multilayer calculations based on different assumptions showed that the change in the angular distribution of light may explain observed deviations in experimental testing. Because all particles modify the angular distribution of light to a degree, the phenomenon interferes with most multilayer calculations.

KEYWORDS: Computation, equations, errors, experimental design, multilayers, optical properties, paper grades, reflectance.

To obtain the desired mechanical, optical, and printing properties, most paper and board grades consist of several layers. In practice, many experiments are often necessary to obtain the best physical properties of multilayer papers at a reasonable cost. To reduce this number of experiments, predictions of estimates for the optical properties of multilayer papers have used the Stokes equation. A theoretical problem with that equation, however, is its treatment of the propagation of light in one dimension. The actual light-scattering phenomenon is three-dimensional. It is therefore quite possible that the equation does not incorporate all the optical phenomena involved.

Kubelka *(1)* introduced the Stokes equation to paper science. **Figure 1** illustrates the treatment for a system having a top layer and a background. Diffuse incident light hits the top layer. As light travels in the layers, there is an assumption that the angular and wavelength distributions of the light remain unchanged. This simplifies the analysis so that the treatment of fractions scattered forward and backward use single numbers instead of matrices to express the angular dependence. An additional assumption is that it is possible to analyze the optical effects by considering the interaction at the layer interfaces. If the background is so thick that no light will transmit through it, summing the geometrical series in Fig. 1 provides the following expression of the reflected fraction:

$$R_S = R_U + R_G T^2 + R_D (R_G)^2 T^2 + \ldots =$$

$$R_U + (T^2 R_G)/(1 - R_D R_G) \qquad (1)$$

where

- R_S = reflectance of the two-layer system
- R_U = reflectance of the top layer measured on the top side
- R_G = measured reflectance of the background
- R_D = reflectance of the top layer measured on the bottom side
- T = transmittance.

The literature contains only a few theoretical studies comparing the results of one-dimensional calculations to those of the exact three-dimensional solution. Using the Kubelka-Munk theory as an example for one-dimensional approximations, van de Hulst *(2)* found deviations greater than 10% from the exact result. Blewin and Brown *(3)* showed that normalizing the Kubelka-Munk curve to coincide with the exact solution at 0.50 reflectance produces an error less than 0.002. Both these studies deal with the continuous case—the reflectance is a continuous function of the layer thickness. Although there has been no systematic study of the discrete case of Eq. 1, there are some weaknesses in the one-dimensional calculations.

Clark and Ramsay *(4)* and Ramsay *(5)* measured the optical properties of a coated paper at one coating weight and the properties of the respective basestock. Then they used the results to predict the optical properties of coated paper at

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other coating weights. By measuring the actual optical properties of those coated papers, they found that the brightness from Eq. 1 agreed well. The opacity deviated up to 0.007 from the measured value, however. One can always speculate that the error was because the structure of the coating layer changes with coating weight. Comparing different techniques to determine the Kubelka-Munk light scattering coefficient of a coating, however, Starr and Young *(6)* found that the light-scattering coefficient depended on the coating weight and base paper reflectance.

Karipidis *(7)* tried to evaluate the optical properties of different top layers or sheets using the Stokes equation—the two-background method. He found that measurements with different pairs of backgrounds deviated from each other. This is consistent with the findings of Starr and Young *(6)*. Karipidis also studied different factors to determine the greatest influence on the two-background method. Because he could not identify a clear explanation for the deviations, however, he concluded that the top layer changes the geometrical relationship between the background and the light falling on the background. As a result, the measured reflectance of the background, R_G, is not valid for the calculation of the optical properties of the top layer.

Jordan *et al.* *(8)* gave two other examples that show that the one-dimensional equation is not always realistic. They showed that the reflectance of a mirror measured in a commercial instrument is near zero, because the gloss trap largely removes the last specular reflection before the light enters the detector. According to Eq. 1, the system with the mirror as a background for a pulp sheet should give a reflectance close to the same pulp sheet with black cavity as a background. In the actual measurement, however, they found that the reflectance with the mirror as a background was greater than estimated from Eq. 1. The explanation is that the black cavity absorbs the light transmitted through the sheet. It returns by a mirror so that part of the intensity goes to the detector. Jordan *et al.* also found the same phenomenon by using a matte and a glossy opal glass as a background. The reflectance of the matte glass was higher than that of the glossy one. With the same pulp sheet, however, the pile having the matte glass background had a lower reflectance than the pile having the glossy glass background. Jordan *et al.* reasoned that part of this opacity sidedness was due to surface reflectance effects both in the top layer and in the background. They also suggested that the Kubelka-Munk theory might be erroneous. They did not, however, give physical explanation for this suggestion.

To explain the findings of Starr and Young, Karipidis, and Jordan *et al.*, one can use the theoretical data of van de Hulst *(2)*. These show that the optical properties of a layer with similar single scattering properties depend on the angular distribution of the incident light. This has a particular implication for multilayer papers. If the top layer modifies the angular distribution of the incident light, the reflectance of the background may appear different with different top layers.

There is very little experimental data on the effects of angular distribution. It is most likely that the angular distribution of transmitted light will not be similarly diffuse, if perpendicular light enters different sheets. Norman and Wahren *(9)* have shown that the sideways diffusion distance of the transmitted light depends on the type of pulp and the grammage of the sheet.

This study will measure the R_S of different sheets to investigate the possible size of the deviations between the measured reflectances and those calculated from Eq. 1. Multilayer calculations based on the theoretical data of van de Hulst *(2)* will show that the observed deviations in the experimental testing can result from different angular distributions. One motivation for this study was to show that the systematic deviations between the calculated and measured reflectances found in a previous paper *(10)* are due to changes in the angular distribution of light.

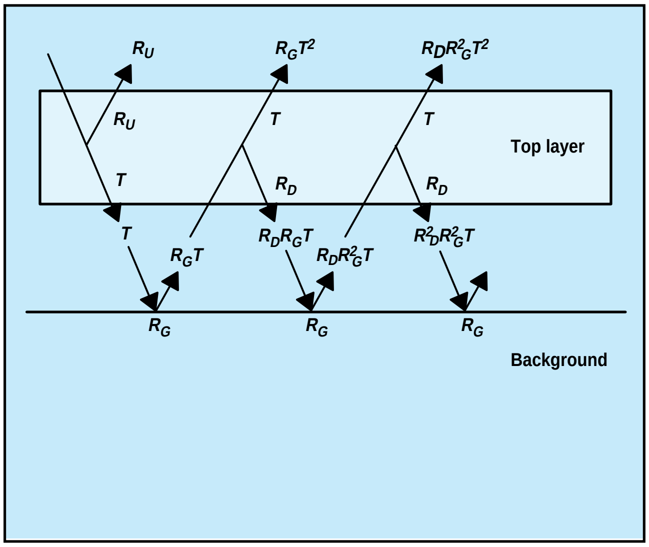
Materials and methods

Experimental testing

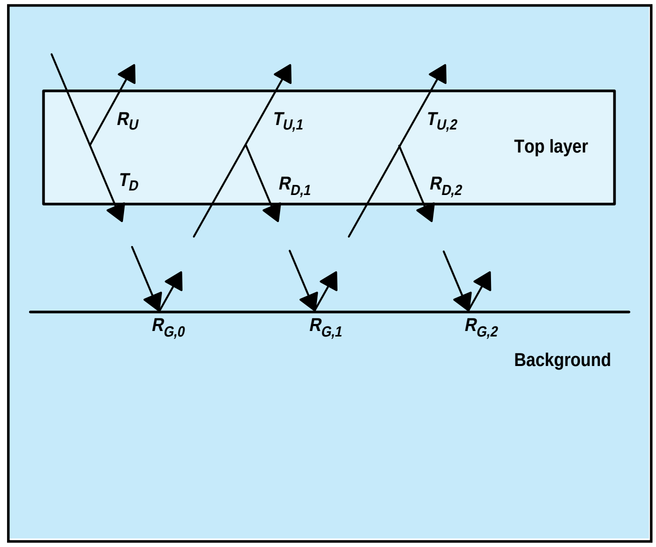
To determine R_S with Eq. 1, one first measures the reflectances, R_U and R_D, of the top layer in Fig. 1 and the reflectance, R_G, of the background. The customary way to determine the transmittance, T, of the top layer is by calculation using Eq. 1 with the measured reflectance, R_∞, for a thick pile of the top layer as R_G. If the equation then provides a realistic description of light propagation in the system of Fig. 1 with the measurement conditions used in the instrument, it should be possible to calculate R_S of the system with any background R_G.

To test this hypothesis, this work investigated the piles of different top layers and backgrounds shown in **Table I**. To increase the interaction between the top layer and the background, selection of the top layers used low opacity. The grammage of the top layers was 40–52 g/m^2. Some top layers and the backgrounds were machine-made. Since none had heavy calendering, they were not glossy. A commercially available instrument measured the reflectances with brightness wavelength 457 nm. In the instrument, the incident light hits the sample from the whole upper hemisphere. Detection of the reflected light is at the angle that is perpendicular to the surface of the sample. Setting the UV-filter at zero removed UV light. Using R_U, R_D, T, and R_G first allowed calculation of the reflectance, $R_{S,c}$. Then there was measurement of the respective $R_{S,m}$. The small differences between the

1. System comprising a top layer and a background



2. Reflectances and transmittances of the spreadsheet calculations



reflectances R_U and R_D had no effect in the calculations. The calculations used the average values denoted as R_0 in Table I of those two reflectances.

Theoretical comparison

The results of van de Hulst *(2)* explained the observed findings. He had tabulated results of theoretical three-dimensional calculations for different single-scattering parameters and angular distributions. His results were for perpendicular, lambertian, or isotropic incident light. Reflected intensity collection was from the whole hemisphere. His results therefore reverse the incident and reflected light compared to the commercial instrument geometry. The Helmholtz Reciprocal Principle *(2)* says that it is possible to reverse the incident and reflected light for both direction and angular flux. If the Helmholtz Reciprocal Principle is valid, the conclusions drawn from the data of van de Hulst would apply also for the commercial instrument geometry. In this paper, however, one does not have to assume that the Helmholtz Reciprocal Principle is strictly valid. There is only qualitative use of the data of van de Hulst to suggest possible mechanisms.

The layer calculations were spreadsheet calculations that considered separately the reflectances R_U or $R_{D,i}$, $R_{G,0}$, and $R_{G,i}$. The transmittances T_D and $T_{U,i}$ came from each interface i where i = 1, 2, . . . as **Fig. 2** shows. The method was the same as the one van de Hulst used to calculate the fate of scattering energy in planetary atmospheres *(2)*.

Using the data of van de Hulst, this work investigated three idealistic systems: reference, nondiffusing top layer, and diffusing top layer. The light hitting the top layer in all these cases is perpendicular. Depending on the properties of the top layer, the angular distribution of light can then either change or not change. In the calculations, the single scattering albedo of the top layer was 0.99. That of the background was either 0.99 or 0.60. The respective background reflectances for perpendicular incident light were 0.752 and 0.155. The single scattering asymmetry parameter was always 0. The forward scattered and the backward scattered fractions were therefore equal. There were similar calculations for values of the optical thickness of the top layer, b, of 1 and 5.

Reference. There was an assumption that the angular and wavelength distribution of the incident light remained unchanged. It was perpendicular as light traveled in the layers. The reflectances of the top layer and the background and the transmittance of the top layer therefore remain constant at each interface.

Nondiffusing top layer. This alternative represents a pile that has a nondiffusing top layer with diffusing background. The incident light reflects from the first surface similar to the reference case. The angular distribution of the fraction transmitted through the top layer is still perpendicular. In the background, however, the angular distribution of the light changes and the light returning to the top layer is either lambertian or isotropic. The angular distribution of light then remains either lambertian or isotropic, respectively. $R_{D,i}$ $T_{U,i}$, and $R_{G,i}$ are different from R_U, T_D, and $R_{G,0}$, respectively.

Diffusing top layer. The incident light again reflects from the first surface like the reference case. The essential feature here is that the top layer diffuses light. An assumption is that the angular distribution of the transmitted light becomes either lambertian or isotropic. A second assumption is that the angular distribution remains either lambertian or isotropic, respectively. Compared to the nondiffusing case, $R_{G,0}$ and $R_{G,i}$ are now equal.

Results

Experimental testing

The experimental results of **Table II** show that the deviations were clearly larger than the measurement accuracy of the instrument for approximately half the cases. This suggests that Eq. 1 is not realistic enough to describe light propagation in those piles. The reflectance R_G for KK or the reflectance R_∞ for E and F did not exceed the value of 0.90 from Table I. The measured reflectance R_S was nevertheless 0.924 for the background KK with the top layer E or F. This is not possible according to Eq. 1.

The deviations in Table II show trends that seem to correlate with the filler content shown in Table I. The deviations increase and become positive with greater filler content in the top layer. All the large negative deviations occur when the background is either LL or MM. These backgrounds contain about 10% PCC particles or 15% spherical ground calcium carbonate particles, respectively. One would therefore expect them to change the angular distribution of light effectively.

I. Properties of the samples

Sample	*Raw materials*	R_0	R_∞
Top layer A	Greaseproof: well beaten BSK	0.396	0.688
Top layer B	Handsheet: BSK	0.567	0.819
Top layer C	Handsheet: BHK and BSK, 9% talc	0.648	0.839
Top layer D	Handsheet: BHK, 7% calcined clay	0.687	0.828
Top layer E	Finepaper: BHK and BSK, 10% PCC	0.742	0.898
Top layer F	Finepaper: BHK and BSK, 15% calcium carbonate	0.702	0.889
Background GG	Greaseproof: well beaten UBSK		0.158
Background HH	Handsheet: groundwood		0.646
Background II	Uncalendered magazine paper: mechanical pulp and BSK, 27% clay		0.719
Background JJ	Handsheet: BHK, 7% calcined clay		0.828
Background KK	Blotting paper: bleached softwood kraft		0.895
Background LL	Finepaper: BHK and BSK, 10% PCC		0.898
Background MM	Finepaper: BHK and BSK, 15% calcium carbonate		0.889

BSK = bleached softwood kraft
BHK = bleached hardwood kraft
UBSK = unbleached softwood kraft
PCC = precipitated calcium carbonate

II. The deviations $R_{S,m}$ - $R_{S,c}$ in percentage

	Top layer					
Background	*A*	*B*	*C*	*D*	*E*	*F*
GG	0.1	0.0	0.0	0.1	0.3	0.6
HH	0.1	-0.2	-0.1	0.1	1.4	2.2
II	-0.3	-0.5	0.0	0.0	1.9	2.5
JJ	-0.1	-0.2	0.3	0.3	2.6	3.2
KK	-0.3	-0.3	0.4	0.2	2.8	3.3
LL	-1.9	-2.4	-1.9	-1.7	0.0	0.0
MM	-1.9	-2.1	-1.8	-1.5	0.4	0.4

Theoretical comparison

If one assumes that the top layer is nondiffusing and the background is diffusing, **Table III** shows there are smaller reflectances obtained than in the reference case. This suggests an explanation for the negative deviations found in the experimental testing of Table II, if the reference case represents the customary way of using the Stokes equation. The top layer with zero or low filler content does not change the angular distribution of the incident light effectively. The calcium carbonate or PCC containing backgrounds do change the angular distribution, however. The light returning to the top layer is therefore diffuse. As a result, R_S is smaller than in the reference case, or $R_{S,m}$ is smaller than the respective calculated $R_{S,c}$ in the experimental testing.

The predicted deviations appear surprisingly similar to those found in the experimental testing. One must remember, however, that the reference case cannot fully represent the customary way of using the Stokes equation. The problem is that all the measured reflectances involve effects of three-dimensional optical phenomena. They may therefore compensate for some deviations arising from strictly one-dimensional measurements and calculations.

If one assumes that the top layer is diffusing and the background is diffusing, Table III shows there are greater reflectances compared to the reference case. This also agrees with the results of experimental testing. The positive deviations increase with greater filler content of the top layer, i.e., with increasing ability of the top layer to change the angular distribution of light. The theoretical calculations here did not predict deviations as large as those seen in the experimental testing.

A qualitative explanation for both the negative and positive deviations observed in the experimental testing is possible. It is therefore probable that the erroneous predictions

III. Multilayer calculations with L denoting the lambertian and I the isotropic angular distributions, respectively

Top layer	*Angular*	$R_{G,0}$	$R_{D,i}$	$T_{U,i}$	$R_{G,i}$	R_S
b = 5	Reference	0.155	0.680	0.221	0.155	0.689
R_U = 0.680,	Nondiffusing L	0.155	0.738	0.173	0.195	0.687
T_D = 0.221	Diffusing L	0.195	0.738	0.173	0.195	0.689
	Nondiffusing I	0.155	0.769	0.149	0.225	0.686
	Diffusing I	0.225	0.769	0.149	0.225	0.689
	Reference	0.752	0.680	0.221	0.752	0.755
	Nondiffusing L	0.752	0.738	0.173	0.795	0.749
	Diffusing L	0.795	0.738	0.173	0.795	0.753
	Nondiffusing I	0.752	0.769	0.149	0.818	0.747
	Diffusing I	0.818	0.769	0.149	0.818	0.752
b = 1	Reference	0.155	0.333	0.651	0.155	0.402
R_U = 0.333,	Nondiffusing L	0.155	0.436	0.544	0.195	0.393
T_D = 0.651	Diffusing L	0.195	0.436	0.544	0.195	0.408
	Nondiffusing I	0.155	0.504	0.474	0.225	0.387
	Diffusing I	0.225	0.504	0.474	0.225	0.411
	Reference	0.752	0.333	0.651	0.752	0.758
	Nondiffusing L	0.752	0.436	0.544	0.795	0.741
	Diffusing L	0.795	0.436	0.544	0.795	0.764
	Nondiffusing I	0.752	0.504	0.474	0.818	0.729
	Diffusing I	0.818	0.504	0.474	0.818	0.763

given by Eq. 1 may be due to the changes in the angular distribution of light.

The theoretical calculations also give two additional results that are very logical:

- The deviations between the reference case and the other two cases decrease with increasing opacity of the top layer. The effect of the background on the optical properties of the pile decreases with increasing opacity of the top layer.
- The deviations from the reference case are larger for the isotropic calculation than for the lambertian one. This apparently results because the isotropic angular distribution is more diffuse than the lambertian one.

Discussion

The experimental testing showed that Eq. 1 cannot always predict R_S of the pile by measuring the optical properties of the top layer and the background. The tables of van de Hulst show that the optical properties of a layer with similar single scattering properties depend on the angular distribution of the incident light. The theoretical results based on that data indicated that the change in the angular distribution of light may explain the observed deviations in the experimental testing. This conclusion agrees with the speculation of Karipidis *(7)*.

Jordan *et al. (8)* considered that the surface reflections caused the spurious observations in their experiments. Although different surface reflections play an important role, the results of this paper suggest that the scattering properties of the particles alone may cause the change in the angular distribution of light. All the fibers and the mineral pigment particles used in papermaking change the angular distribution of light to some extent. One would therefore expect the change in the angular distribution of light to be a very general phenomenon in multilayer papers.

Windle and Gate *(11)* have shown that the changes in the angular distribution of light affect the brightness measurement. By testing a series of clays having different brightness, they found an increase in the brightness difference between two brightness testers as a function of the sample brightness. They concluded that a geometrical factor resulting from the different modes of illumination and light collection in the systems caused the differences. Clays with different brightness also have different goniophotometric characteristics. This means that any theory deduced for reflectance values obtained with one optical geometry causes a systematic error in the application of values measured with another geometry. As this study showed, the samples may be different enough to result in different measurement geometry. In the commercial instrument geometry in this study, this means that the instrument will rank two samples differently, if their ability to reflect to the perpendicular direction is different, although they may reflect equally well to the whole upper hemisphere.

The fact that one cannot always predict R_S of the pile from Eq. 1 by measuring the optical properties of the top layer and the background has other consequences for using the equation. These include two examples that both involve the determination of the optical properties of the top layer by measuring R_s with two different backgrounds (two-background method).

A determination of the light-scattering coefficient of a film *(12)* often describes the structure of coating films. Coatings having different light diffusing properties—they modify the angular distribution of the transmitted light differently—may well show different behavior depending on the background used.

Equation 1 can determine the yellowing tendency of the pulp by measuring the absorption coefficient of the pulp sheet after treatments in different atmospheres *(13)*. One would expect the changes in the angular distribution of light here to be smaller than with coatings. This is because the optical changes of the pulp sheets occur only in the light absorption ability of the pulp. The light-scattering and the light-diffusing properties remain almost constant.

An impetus for this study was to explain the systematic deviations between the calculated and measured reflectances found previously *(10)*. In that study, use of the Stokes equation assumed that the properties of a layer corresponded to the properties of a single particle. According to the results of this paper, the changes in the angular distribution of light result in systematic deviations from the predictions of the Stokes equation. It therefore most likely caused the deviations in the previous study.

Conclusion

To find better agreement with the measured and calculated reflectances, it is necessary to modify the calculations or the measurement standards. Improved calculations should clearly use matrices expressing the intensity of light using both the incident angle and the scattering angle. The literature *(14)* contains examples of such calculations. The experimental determination of the optical properties of papers using scattering angle would be much more tedious than the present standard. Some improvement in measuring standards might occur if the reflectometers prevented any light passing directly to the detector *(15)*. It is not clear whether this modification would result in better agreement between the measured reflectances and those calculated from Eq. 1, however. If one also changes the measuring standards, he needs to examine carefully which measurement correlates best with the subjective opacity and brightness judgment.

The simple Stokes equation may occasionally be accurate enough for practical purposes. Sometimes the different sources of error may counterbalance each other. One can often extend it beyond its physical limits, however, because the angular distribution of light changes. In multilayer optical calculations, a paper physicist must therefore know what really happens in the sample and interpret his results accordingly. TJ

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