

Quantification of the Intrinsic Error of the Kubelka–Munk Model Caused by Strong Light Absorption

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The Kubelka–Munk (KM) model is widely used within the paper industry to interpret diffuse reflectance factor measurements of paper and its components. It has been found in the literature that the addition of a dye colourant to a paper sheet not only increases its KM light absorption coefficient but, for strong absorption, also decreases its KM light-scattering coefficient. This effect has been attributed previously to the intrinsic error of the KM model induced by light absorption that tends to orient the light fluxes perpendicular to the plane of the sheet. In this paper, we have mapped the intrinsic error of the KM model by comparing light-scattering calculations from the KM model with the more accurate discrete ordinate radiative transfer model DORT2002. We found that the models agree within 2.3% in reflectance and that the intrinsic error in the KM model explains ~20% of the previously observed interdependence of the KM coefficients for heavily dyed sheets.

Le modèle Kubelka–Munk (KM) est largement utilisé dans l'industrie du papier pour interpréter les mesures des facteurs de réflectance diffuse du papier et de ses éléments. Nous avons trouvé dans la documentation que l'ajout d'un colorant à une feuille de papier non seulement accroît son coefficient KM d'absorption de la lumière, mais que, lors d'une forte absorption, il réduit aussi son coefficient de KM de diffusion de la lumière. Cet effet avait auparavant été attribué à l'erreur intrinsèque du modèle KM induite par l'absorption de la lumière qui tend à orienter le flux lumineux perpendiculairement à la feuille. Au cours du présent travail, nous avons appliqué l'erreur intrinsèque du modèle KM en comparant les calculs de diffusion de la lumière du modèle KM avec le modèle DORT2002 plus précis du transfert radiatif des ordonnées discrètes. Nous avons trouvé que les modèles concordent à ± 2.3 % de la réflectance, et que l'erreur intrinsèque du modèle KM permet d'expliquer environ 20 % de l'interdépendance observée des coefficients KM pour les feuilles très colorées.

INTRODUCTION

The Kubelka–Munk (KM) model [1–3] is used widely within the paper industry to interpret diffuse reflectance factor measurements of paper [4]. This model considers the interaction of two diffuse light fluxes (reflection and transmission direction) with the bulk of a plane parallel light-scattering and absorbing medium. The strength of these fluxes depends on the light-scattering and absorption coefficients (s and k), which represent the ability of the medium structure to scatter and absorb light. Paper properties such as the micro-topographical surface structure, the free surface area, and the size, shape, and chemical composition of pig-

ments and fibres are known to influence the optical response. However, the interpretation of the KM coefficients in terms of such structural features is not always obvious. One example that has attracted some interest in the literature is the behaviour and interpretation of the KM coefficients for a paper with strong light absorption.

The discussion started with the experimental findings of Foote in 1939 [5]. He studied the KM coefficients of dyed sheets and found that both the k value and the k/s ratio increased nonlinearly with increasing dye concentration. The results were unexpected, since the light absorption of a dye is strong, while the light scattering is weak. Similar results were also obtained by Nordman et al. [6]. Later, Rundlöf and Bristow [7] proposed an experimental method to determine the limiting values of k at which an acceptable determination of s is no longer possible. They observed that the apparent scattering coefficient was significantly reduced after dyeing at wavelengths corresponding only to the light absorption peaks of

the dye colourant. Based on this observation, they argued that the dyeing process did not alter the paper structure and the corresponding true scattering coefficient. Hence, the decreased apparent scattering coefficient at strong light absorption was regarded as physically incorrect. By using the wavelength dependence of the apparent scattering coefficient of the uncoloured paper as a reference, they were able to estimate quantitatively the decrease of the apparent s with increasing apparent k . The results showed an approximately linear decrease of the apparent s with increasing apparent k value with the approximate slope

$$\frac{ds}{dk} = -\frac{5}{8} \quad (1)$$

as seen in Fig. 6 of their article [7]. Another important result was that the apparent k values for unbleached thermomechanical pulp in the blue wavelengths of the spectrum are so large that they are likely to decrease the s values.

There are basically three explanations in the literature for the observed behaviour of the

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KM coefficients for paper samples with strong light absorption. One attributes the behaviour to spatial variations in the sheet structure, which are not considered with the one-layer KM model [8]. This explanation was discarded by Koukoulas and Jordan [9] by experimental results from a series of dyed sheets with variable formation. Instead, they attributed the decrease of s at strong absorption to the mutual dependence between the real, n , and the imaginary, κ , part of the complex refractive index of the fibre wall. This mutual dependence (the Kramers–Kronig relation) is fundamental and relies only on causality and a linear optical response in the fibre wall medium [10]. Koukoulas and Jordan argued that, while κ of the fibre wall affects only the light-absorption properties of the sheet, both n and κ affect its light-scattering properties. With this interpretation, the decreasing s with increasing k becomes a natural consequence of the physical properties of the paper, rather than an artificial relation. The third explanation attributes the behaviour to an intrinsic error of the KM model which occurs for a medium with a large k/s ratio [11]. This error depends on the fact that oblique rays suffer greater absorption than rays propagating along the normal in a plane parallel medium. The slight orientation of the light fluxes perpendicular to the sheet induced by light absorption alters the optical response of the medium. This orientation effect introduces an intrinsic error because the KM model does not consider any change in the angular distribution of light fluxes.

The size of the intrinsic error has been studied in the literature by comparing the KM model to more accurate models. These studies have resulted in validity conditions of the KM model but also in approximate relationships between the medium parameters used in the KM model and those in so-called radiative transfer models [12–15]. In the present work, we apply these results to estimate the size of the intrinsic error of the KM model for paper applications. We do that by simulating diffuse reflectances with the discrete ordinate radiative transfer (DORT) model DORT2002 [16,17] and compare these to results from KM model calculations. Finally, we analyze how much of the experimental interdependence of the KM coefficients found by Rundlöf and Bristow [7] that can be explained by the intrinsic error in the KM model.

MODELS

The DORT2002 model version 2.0 [16,17] can be used to calculate the angle-dependent diffuse reflectance ρ and transmittance τ from a bulk medium. The optical response of the medium depends on the illumination distribution and the medium parameters σ_s (scattering coefficient), σ_a (absorption coefficient), g (asymmetry parameter) and t_{phy} (physical thickness). All medium parameters but the physical thickness depend on the wavelength λ of the incident radiation. The scattering coefficient represents the fraction of the incident radiation scattered in all directions per unit propagation length along the path of the beam.

The absorption coefficient σ_a analogously represents the fraction of the incident radiation absorbed per unit propagation length along the path of the beam. It is meaningful to separate the optical thickness τ_{opt} which is an absolute measure of the light-scattering and absorption sites in the medium, and the density parameter t_{phy} , which relates the number of scattering and absorption sites to a measurable quantity such as the physical thickness of the medium. Depending on preference the physical thickness can be substituted for concentration or grammage. Naturally, this will change the units of the scattering and absorption coefficients accordingly. The optical thickness is dimensionless and is defined as

$$\tau_{opt} = (\sigma_s + \sigma_a) \cdot t_{phy} \quad (2)$$

Another optically meaningful quantity is the coalbedo ϖ given by

$$\varpi = \frac{\sigma_a}{\sigma_s + \sigma_a} \quad (3)$$

A ϖ value near 1 corresponds to a medium where the absorption processes dominate over the scattering process, while a value close to 0 corresponds to dominantly scattering processes. A medium already is considered strongly absorbing at small values of ϖ .

The discrete ordinate method used in DORT2002 refers to the division of light fluxes into multiple ordinates, or channels, that describe different propagation directions. Throughout the present work, we used 30 channels in the DORT2002 calculations. This is sufficient to guarantee that the simulations of the total reflectance are fully accurate, i.e. more channels would not yield more accurate results using this model, nor would any other radiative transfer method based on a discrete ordinate model geometry.

The KM model can be used to calculate the hemispherical reflectance and transmittance from a bulk medium illuminated by a completely diffuse light source, given the KM scattering coefficient s , the KM absorption coefficient k , and the density parameter t_{phy} of the medium. The KM equations are given in closed form and the coefficients can be calculated readily given reflectance or transmittance values at different t_{phy} values. The most common approach used in the pulp and paper industry is to use the measured diffuse reflectance factor R_0 for one sheet and R_∞ for a semi-infinite stack of sheets, and to simply assume that these correspond to the diffuse bulk reflectances ρ_0 and ρ_∞ , respectively [4]. The grammage is commonly used to represent t_{phy} of the sheet instead of the physical thickness and, consequently, the KM coefficients get the unit 1/grammage. The KM equations are [4]

$$s = \frac{\rho_\infty}{t_{phy}(1-\rho_\infty^2)} \ln \left(\frac{\rho_\infty - \rho_0 \rho_\infty^2}{\rho_\infty - \rho_0} \right) \quad (4)$$

$$k = \frac{s(1-\rho_\infty)^2}{2\rho_\infty} \quad (5)$$

Since the light-scattering and absorption coefficients within the KM and the DORT2002

model are deduced under different conditions, they are not equal.

However, so-called similarity relations have paved the way for a relevant comparison between different parameter sets. One such similarity relation [18] makes it possible to substitute nonisotropic scattering in a bulk medium with isotropic scattering by using a reduced scattering coefficient σ'_s to approximate the total diffuse bulk reflectance and transmittance. In mathematical terms, this is

$$\sigma'_s = \sigma_s(1-g) \quad (6)$$

Based on this similarity relation, Mudgett and Richards [13,14] found that it was possible to approximately relate the KM parameters and parameters from any DORT model, given a completely diffuse illumination and hemispherical detection conditions.

$$s = \frac{3}{4} \sigma'_s \quad (7)$$

$$k = 2\sigma_a \quad (8)$$

The proportional relation between the DORT and the KM model parameters indicate that the s and k parameters represent reasonably well the physical processes σ'_s and σ_a . Billmeyer and Richards [12] gave two validity conditions for the times Eqs. (7) and (8) can be applied in practice. These are

$$\rho > 50\% \quad (9)$$

$$\tau < 20\% \quad (10)$$

A modification of Eq. (7) to extend its usefulness to slightly more absorption was given by Star et al. [15]

$$s = \frac{3}{4} \sigma'_s - \frac{1}{4} \sigma_a \quad (11)$$

This equation was deduced within the diffusion approximation that is valid for a strongly light scattering medium with weak light absorption. The diffusion approximation is a two-channel DORT model where the influence of g is approximate. For the conditions in the present article using $g=0$ and no collimated light source, the KM model and the diffusion approximation coincide. The last term in Eq. (11) reveals that at least part of the intrinsic error in the KM model will show up as a linear decrease in the KM scattering coefficient when the light absorption increases.

METHOD

In the present work, we estimate the intrinsic error of the KM model and analyze how this error will show up in paper applications. We do this by comparing results from the DORT2002 model with results from the KM model and by using Eqs. (7) and (8) to convert the model coefficients. To mimic the way the KM coefficients are obtained for paper applications in practice, we consider the reflectance of a single-plane parallel medium ρ_0 and the reflectance of an infinitely thick medium ρ_∞ . While we realize that the actual reflectance dis-

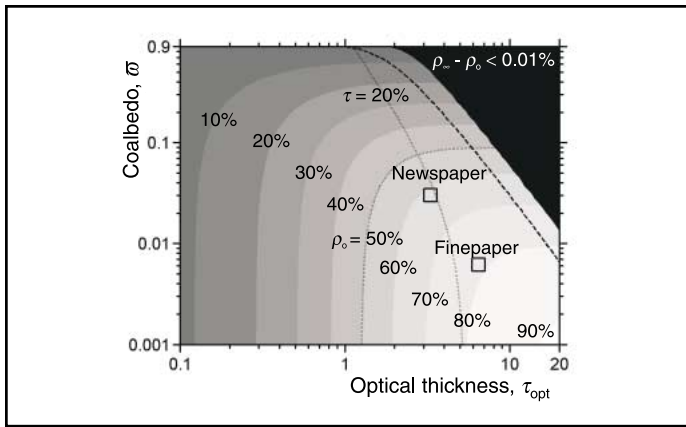


Fig. 1. The diffuse reflectance simulated with DORT2002 as a function of the optical thickness, τ_{opt} , and the coalbedo, ω . The reflectance is plotted in grayscale. The two dotted lines represent 50% reflectance and 20% transmittance. The dashed line represents $\rho_{\infty} - \rho_0 = 0.1\%$ and the black area in the upper right corner represents $\rho_{\infty} - \rho_0 < 0.01\%$.

tribution affects the diffuse reflectance factor response of real paper samples, the current study is limited to the total bulk reflectance from a medium illuminated by a completely diffuse light source. This approach makes it possible to reduce the DORT2002 simulations for any nonisotropically scattering medium ($g \neq 0$) to the isotropic case ($g = 0$) by using Eq. (6).

In the first model comparison (method 1), we used the DORT2002 model to calculate $\rho_{0,DORT2002}$ values for a parameter space spanned by different ω and τ_{opt} values. Then we used Eqs. (2) and (3) and Eqs. (7) and (8) to calculate the corresponding KM parameters $s_{MR} \cdot t_{phy}$ and $k_{MR} \cdot t_{phy}$. Here the subscript *MR* stands for the authors Mudgett and Richards. From these parameters we calculated $\rho_{0,KM}$ values with the KM model by inverting Eqs. (4) and (5). The difference between the $\rho_{0,DORT2002}$ (i.e. correct values) and the $\rho_{0,KM}$ values gave the intrinsic error of the KM model as it showed up in the reflectance domain.

In the second comparison (method 2), which perhaps is more relevant for the papermaker, we mapped how the intrinsic error of the KM model shows up in the KM coefficients. First, we used the DORT2002 model to calculate $\rho_{0,DORT2002}$ and $\rho_{\infty,DORT2002}$ values for a parameter space spanned by different ω and τ_{opt} values. Then we used Eqs. (4) and (5) to calculate the apparent KM coefficients $s_{app} \cdot t_{phy}$ and $k_{app} \cdot t_{phy}$ that corresponded to these $\rho_{0,DORT2002}$ and $\rho_{\infty,DORT2002}$ values. By this method, the apparent KM coefficients now contained the intrinsic error of the KM model. Finally, we used Eqs. (2) and (3) and Eqs. (7) and (8) to calculate the corresponding KM coefficients $s_{MR} \cdot t_{phy}$ and $k_{MR} \cdot t_{phy}$, which we consider to be the true medium parameters. The relative error in the KM coefficients due to the intrinsic error of the model is given by

$$\frac{\Delta s}{s} = \frac{s_{app} \cdot t_{phy} - s_{MR} \cdot t_{phy}}{s_{app} \cdot t_{phy}} = \frac{s_{app} - s_{MR}}{s_{app}} \quad (12)$$

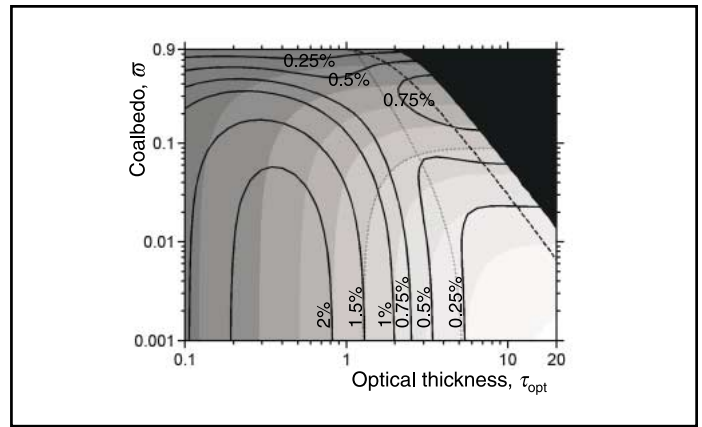


Fig. 2. A contour diagram of the difference between the reflectance calculated with the DORT2002 and the KM model superimposed onto the reflectance grayscale levels. See also caption for Fig. 1.

$$\frac{\Delta k}{k} = \frac{k_{app} \cdot t_{phy} - k_{MR} \cdot t_{phy}}{k_{app} \cdot t_{phy}} = \frac{k_{app} - k_{MR}}{k_{app}} \quad (13)$$

Figure 2 in Rundlöf and Bristow's paper [7] shows what happened to the apparent s and k values of an uncoloured wood-free paper when it was dyed with a yellow colourant. To be able to compare directly those experimental findings with the theoretical analysis in the present work, we applied method 2 to the spectral behaviour of the dyed sheet. We started by using Eqs. (7) and (8) to calculate parameters of the uncoloured paper ($\sigma'_{s,uncol}$, $\sigma_{a,uncol}$) and the yellow paper ($\sigma'_{s,uncol}$, $\sigma_{a,yellow}$) from the measured apparent $s_{MR,uncol}$, $k_{MR,uncol}$, $k_{MR,yellow}$ values. The latter values are considered to be the true medium parameters. The $\sigma'_{s,uncol}$ values were used in both datasets because we assume they are unaffected by the dye, hence we are not considering the explanation proposed by Koukoulas and Jordan [9]. Rundlöf and Bristow used sheets having a grammage of 80 g/m². * Given these input parameters, we readily calculated the spectral ρ_{∞} and ρ_0 values for both the uncoloured and the yellow paper sheet with the DORT2002 model. From these ρ_{∞} and ρ_0 values, we used Eqs. (4) and (5) to calculate the KM coefficients of the uncoloured ($s_{app,uncol}$, $k_{app,uncol}$) and the yellow paper ($s_{app,yellow}$, $k_{app,yellow}$).

RESULTS

Quantification of the Intrinsic Error in the KM Model

Figure 1 shows a contour diagram of the diffuse reflectance as grayscales simulated with DORT2002. The diffuse reflectance is plotted against both the optical thickness (τ_{opt}) and the coalbedo (ω). A doubling of τ_{opt} represents a doubling of scattering and absorption sites in the medium, while a doubling of ω represents a

doubling of the relative amount of absorption sites in the medium. The dotted lines in the figure show the medium parameters when 50% of the incoming light is reflected or 20% is transmitted, respectively. The area to the right of the line $\tau = 20\%$, but where $\rho_0 > 50\%$ gives the validity domain of the KM model as specified by Billmeyer and Richards (Eqs. 9,10). To familiarize the papermaker with this type of diagram, we include typical coalbedo and optical thickness values for a 40 g/m² newspaper and a 80 g/m² filled fine paper. These were obtained by applying Eqs. (7) and (8) to typical KM coefficients of mechanical pulp ($s = 60$ g/m², $k = 5$ g/m²) and filled fine paper ($s = 60$ g/m², $k = 1$ g/m²) [19]. However, the KM coefficients of a paper can vary strongly over the visual spectrum and depend strongly on the papermaking processes.

The standardized method to obtain KM coefficients relies on ρ_0 and ρ_{∞} values (Eqs. 4,5). When the difference between these values has a size similar to the measurement uncertainties, the KM coefficients obtained become irrelevant in practice. The black area to the upper right in Fig. 1 gives the parameter space when $\rho_{\infty} - \rho_0 < 0.01\%$, and the dashed line represents the case when $\rho_{\infty} - \rho_0 = 0.1\%$.

Figure 2 shows $\rho_{0,DORT2002} - \rho_{0,KM}$, i.e. the difference between the reflectance calculated with the DORT2002 and with the KM model (method 1). From the figure, it is evident the DORT2002 model always gives the largest reflectance over the whole parameter space. The difference is small both for an intensely scattering medium (small ω , large τ_{opt}) and for a medium with large light absorption dominance (very large ω). The largest difference (2.3%) was obtained for small ω values at $\sim 25\%$ reflectance. The calculations also show that the validity domain proposed by Billmeyer and Richards (Eq. 9) corresponds to a difference less than 0.6%.

Figures 3 and 4 show the relative error in the KM coefficients as calculated from Eqs. (12) and (13). These figures show that the relative error is largest near the black area for large ω values but, also, when the τ_{opt} value is small. The relative error of s within the validity do-

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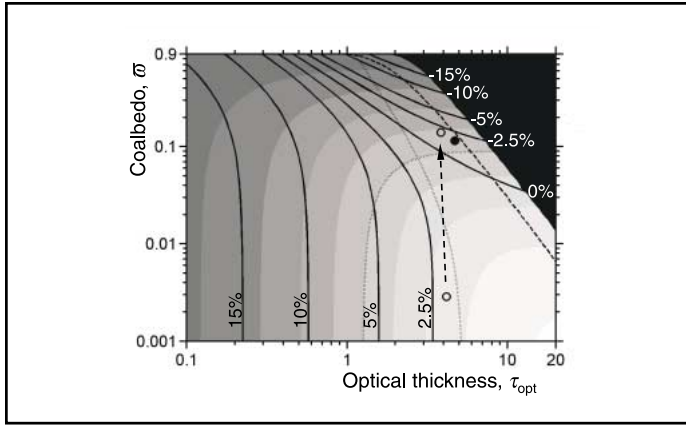


Fig. 3. The relative error in s due to the intrinsic error in the KM model. A contour diagram of the difference between the reflectance calculated with the DORT2002 and the KM model superimposed onto the reflectance grayscale levels. The unfilled circles represent the parameters of an uncoloured sheet (lower) and a sheet dyed with a yellow colourant (upper). The filled circle represents $s_{app, uncol}$ and $K_{app, yellow}$. See also caption for Fig. 1.

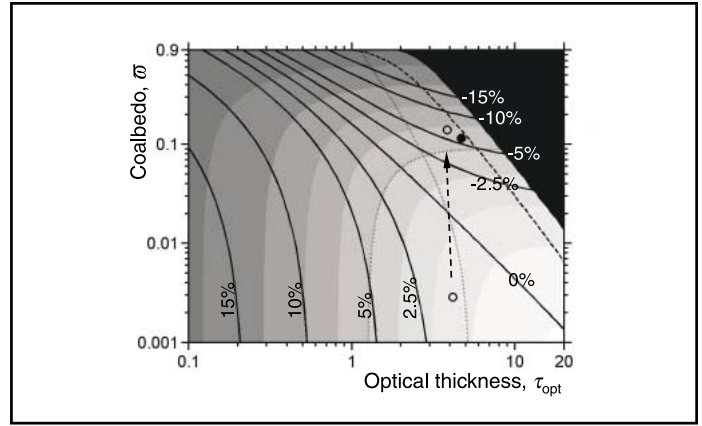


Fig. 4. The relative error in k due to the intrinsic error in the KM model. A contour diagram of the difference between the reflectance calculated with the DORT2002 and the KM model superimposed onto the reflectance grayscale levels. The unfilled circles represent the parameters of an uncoloured sheet (lower) and a sheet dyed with a yellow colourant (upper). The filled circle represents $s_{app, uncol}$ and $K_{app, yellow}$. See also caption for Fig. 1.

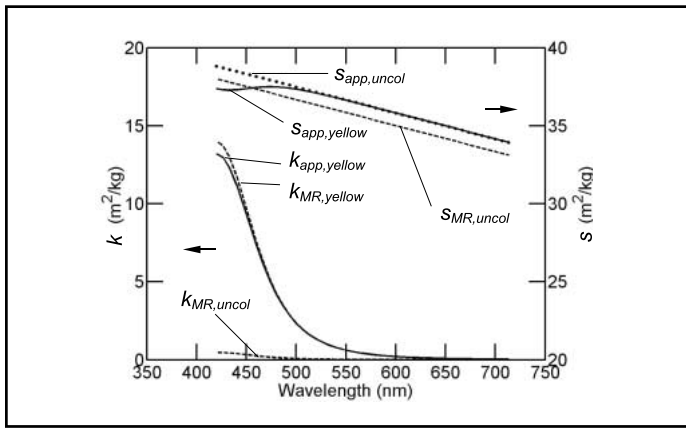


Fig. 5. The spectral dependence of the true (MR) and the apparent (app) s and k values of an uncoloured sheet and a sheet dyed with a yellow colourant.

main given by Eqs. (9) and (10) is less than $\pm 2\%$ and the relative error in k is between -6 and $+2\%$. If the apparent s had decreased linearly, as in Eq. (11), this would have shown up in Fig. 3 as contour lines of equal relative error, parallel to the horizontal axis. Such behaviour can be approximated close to the black area where both coalbedo and optical thickness values are high.

Comparison with the Experimental Findings of Rundlöf and Bristow

In this section, we compare our theoretical results of the intrinsic error of the KM model with the findings in Figs. 2 and 6 in the work of Rundlöf and Bristow [7]. In Figs. 3 and 4 of the present work, we have plotted the apparent KM coefficients obtained for the yellow-dyed sheet at the wavelength $\lambda = 420$ nm, which corresponds to the absorption peak of the yellow dye. The unfilled circles represent the medium parameters of the uncoloured paper and the dyed paper. The arrows in Figs. 3 and 4 show how the medium parameters change when the sheet is dyed. The coordinates were calculated by applying Eqs. (7) and (8) to the

apparent KM coefficients obtained for a sheet grammage of 80 g/m². We note that the upper unfilled circle is close to the line representing 0.1% difference between ρ_∞ and ρ_0 . In this region, small systematic deviations in the reflectance values can have large implications on the KM coefficients obtained.

The apparent optical thickness of the medium decreased after the yellow dye was added, because the upper unfilled circle is positioned slightly to the left of the lower unfilled circle in Figs. 3 and 4. This was not expected, based on the assumption that the true scattering coefficient was not affected by the addition of the dye. The filled circle shows the hypothetical parameter coordinate of the medium if the scattering coefficient had remained constant after the addition of the dye. In Fig. 3, one can see that the decreasing s with increasing k can be explained, at least partly, by the intrinsic error in the KM model. Here, the relative error in s is $+2.5\%$ for the uncoloured paper and -2.5% for the dyed paper.

To illustrate further the experimental observations, we applied method 2 to the spectral behaviour of the yellow-dyed sheet. The true (subscript *MR*) and apparent coefficients of the uncoloured and the dyed sheet are presented in Fig. 5. The data show that the apparent s is larger than the true s at weak absorption, and that both the apparent s and the apparent k underestimate the real coefficients at strong absorption (subscript *yellow*). The difference between the $s_{app, uncol}$ and the $s_{app, yellow}$ curve clearly shows that the apparent s decreases for wavelengths with considerable light absorp-

tion. This difference corresponds to $\sim 1/8$ of the k_{yellow} values. This is equal to the correction term proposed by Star et al. when substituting σ_a in Eq. (11) by $k/2$ from Eq. (8)

$$s = \frac{3}{4} \sigma'_s - \frac{k}{8} \quad (14)$$

$$\frac{ds}{dk} = -\frac{1}{8} \quad (15)$$

This means that about 20% of the decrease of the apparent s with increasing apparent k , which was observed by Rundlöf and Bristow (Eq. 1), can be explained by the intrinsic error of the KM model. In addition, our results confirm the correction term by Star et al., which was deduced within the diffusion approximation. However, our results also show that this correction term does not give the whole picture at smaller optical depths, since the contour lines in Fig. 3 are not parallel to the horizontal axis.

DISCUSSION

We have observed that the decrease in s due to large k happens in a region where the difference between ρ_∞ and ρ_0 is small. This implies that even a small systematic deviation of either ρ_∞ or ρ_0 can strongly influence the KM coefficients obtained. In our analysis to explain 20% of the decrease in apparent s with increasing k , we considered the total reflectance from an isotropically scattering bulk medium without boundary surfaces. We realize that this is only a simplified description of diffuse reflectance factor measurements of paper. Both the top surface boundary and the asymmetry parameter g will influence the actual reflectance distribution and, hence, the signal to the optical instrument [15,20–22]. It is also well known that the internal reflectance at the top surface is quite sensitive to the angular distribution of light that is incident on it [14]. Based on this, we find it likely that the combination of a top surface and strong light absorption in the bulk can explain a larger part of the effect than 20%. To be able to test this sugges-

tion, we therefore call for models that account for both the geometry of the optical instrument and the light scattering from rough surfaces. It would also be interesting to study how much of the effect that can be explained by the Kramer–Kronig relation. To be able to do that, one would most likely need models such as P3D [23] or *Grace* [24], which resolves the paper structure into its individual fibres.

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