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A partial explanation of the dependence between light scattering and light absorption in the Kubelka-Munk model

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KEYWORDS: Kubelka-Munk, Paper optics, Parameter dependence, Reflectance measurements, Anisotropic reflectance.

SUMMARY: The Kubelka-Munk scattering and absorption coefficients of a set of paper samples are assessed using reflectance measurements in d/0 geometry. The coefficients display the widely studied dependence between light scattering and light absorption, since the light scattering coefficient decreases in regions of high absorption. It is shown using general radiative transfer theory that part of this dependence can be explained and eliminated by taking into account the geometry of the d/0 instrument and the single scattering anisotropy, thus capturing the angular variations of the light reflected from the samples. These findings allow the papermaker to better predict the reflectance from mixtures of pulps, fillers, dye, and FWA, and to better evaluate bleaching efforts.

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To predict the visual appearance of scattering media such as paper, and to quantify the light scattering properties of these media, mathematical models are often used (Billmeyer, Abrams 1973; Chandrasekhar 1960; Edström 2010; Glassner 1995; Hutchings 1999; Moussa et al. 2009; Pauler 2002; Schmitt 1999; Thomas, Stamnes 1999). The paper industry has since long been using the widespread Kubelka-Munk (KM) (Kubelka, Munk 1931) model together with reflectance measurements in d/0 geometry (ISO 2469) to calculate the light scattering and absorption coefficients s and k (ISO 9416). The KM model is appealing due to its simplicity, but it is well known that it is over simplified and inaccurate in many cases (Neuman, Edström 2010a; Nobbs 1985). A widely investigated problem is the decrease in scattering coefficient s as the absorption coefficient k increases. This can be observed both over a spectral range where k increases (Nordman-Aaltonen-Makkonen anomaly (Nordman et al. 1966)) and at a specific wavelength when dye is added to the paper (Foote effect (Foote 1939)). This has been studied and discussed by many authors (Van den Akker 1949, 1990; Foote 1939; Granberg, Edström 2003; Koukolas, Jordan 1997; Moldenius 1983; Nordman et al. 1966; Olf 1989, 1990; Rundlöf, Bristow 1997), but still lacks a satisfactory explanation. For papermakers, the KM model has proven useful for example to predict the reflectance from mixtures of

pulps, fillers and dyes. These predictions assume that s and k are independent. For instance, sheet dyeing is believed to affect only k , whereas calendering is expected to affect s and not k . Understanding the dependence between s and k is therefore relevant for the papermaker since this can make KM based predictions of the reflectance from mixtures of different pulps, fillers and dyes more reliable. Furthermore, accurate k -values are important when evaluating pulp bleaching efforts (Moldenius 1983; Karlsson et al. 2012). In the present work we show that using models more comprehensive than KM leads to a reduced dependence between light scattering and absorption.

Another approach to modeling of light scattering and absorption in paper is general radiative transfer (RT) theory (Chandrasekhar 1960). An advantage of general RT is that it takes the d/0 geometry into account when calculating the scattering and absorption coefficients, thereby capturing all angular variations of the light intensity in the d/0 instrument. Neglecting these variations introduces a geometry induced error in all practical situations (Edström et al. 2010; Neuman, Edström 2010a,b). A further advantage of general RT theory is that it includes the single scattering anisotropy. That is, it takes into account if light is on average scattered more in the forward direction at each scattering site. This single scattering anisotropy is often quantified using the asymmetry factor g . If g equals zero light is scattered isotropically at each scattering site and if g approaches unity light is mostly scattered in the forward direction. Previous work has shown that light is indeed scattered more in the forward direction in organic materials and in paper (Cheong et al. 1990; Granberg, Béland 2004; Joshi et al. 2006; Prahel et al. 1993).

General RT theory follows from Maxwell's equations under specific assumptions (Mishchenko 2010), rendering the theory legitimacy based on fundamental physical principles. KM theory is a limited case of general RT theory and has been shown to be exactly valid only when a non-absorbing opaque medium is diffusely illuminated (Neuman, Edström 2010a,b). In all other situations angular variations (anisotropy) arises, which is neglected by the KM model.

This work shows that using general RT, thus taking the instrument geometry and the single scattering anisotropy into account, we can partly explain the anomalous dependence between scattering and absorption in the KM model. This allows the papermaker to assess light scattering properties that are more reliable since they are not contaminated by a geometry induced error, which for example will give better predictions of the reflectance at different amount of dye or FWA, and when evaluating pulp bleaching. Furthermore, we quantify the deviation between general RT theory and KM theory. In this way

we can indicate for what types of pulp and paper samples that the model error is significant.

Method

We use a set of paper samples with both heavily dyed Formette papers and office paper. The samples are measured with a d/0 instrument, using both an opaque pad and a single sheet, thus giving the R_0 and R_∞ reflectance factors. We introduce and calculate the local model deviation δ and study this deviation for the samples. If $\delta = 0$, then the RT and KM models will respond in the same way to changes in R_0 and R_∞ , and if $\delta \neq 0$ they will not. Furthermore, we calculate the scattering and absorption coefficients from d/0 reflectance factor data using general RT, thereby eliminating the geometry induced error. We then compare these to the scattering and absorption coefficients calculated using the KM model. This will reveal how much of the dependence between scattering and absorption in the KM model that can be explained by the geometry induced error. A detailed description of the method follows below.

Material and measurements

The Formette samples are prepared with a Formette Dynamique from a mix of equal amounts of soft and hardwood kraft pulps, having Shopper/Riegler numbers 18° and 23° SR respectively. The samples have grammage 30 g/m^2 and contain 22% fillers. One undyed sheet and one sheet with blue shading dye are prepared. The amount of blue dye is 1 % of the dry fiber weight and the dye used is Levacell Fast Blue KS-6GLL Liquid, manufactured by Lanxess. This dye is a cationic direct dye and is assumed not to affect the structure of the paper. The sheets are dried in a cylinder dryer during 5 min at a temperature of 105°C and a pressure of 1.50 bars. Special care is taken when preparing the samples to minimize gloss. Gloss measurements were made with a gloss meter (Zehntner) to ensure low gloss levels, and the gloss was found to be low with an average of 1.0 for 20° and 2.9 for 60° and 75° . The Formette samples do not contain fluorescent whitening agents. The office paper that we use has a grammage of 80 g/m^2 .

All samples are measured using a Lorentzen & Wettre Elrepho d/0 instrument complying with ISO2469 in UVX illumination, thus excluding wavelengths shorter than 420 nm. The R_0 and R_∞ reflectance factors are obtained by measuring both a single sheet over a black background and an opaque pad of paper sheets. Fig 1 shows these reflectance factors and the KM scattering and absorption coefficients s and k . We can see that the scattering coefficient of the dyed sample decreases in the absorption band of the dye, both when considering a spectral range of the dyed sample (solid blue line) and when comparing the dyed and undyed samples at a specific wavelength (compare solid blue and solid black lines). We thus observe both the NAM anomaly and the Foote effect.

Simulations using general RT theory

In contrast to the equations of KM theory, the equation of general RT theory cannot be solved analytically, but there exists a vast amount of solution methods and

implementations of numerical solutions to the RT equation (see e.g. Thomas and Stamnes (1999)). In this work we solve the general RT equation numerically using the implementation contained in the DORT2002 model developed by Edström (Edström 2005), which is freely available. The DORT2002 model is adapted to paper industry applications and contains predefined and well tested settings for simulations of geometries such as d/0 (Edström 2008).

Deviation between general RT and KM theory

We quantify the deviation between general RT and KM theory by introducing a measure that captures how well the two models coincide in describing the changes in scattering coefficient as the R_0 and R_∞ reflectance factors vary. This will reveal in what regions of the $R_0 R_\infty$ space that the geometry induced error is significant.

The KM scattering coefficient s is given by KM theory using the equation (Pauler 2002)

$$s = \left[w \left(\frac{1}{R_\infty} - R_\infty \right) \right]^{-1} \log \left[\frac{(1-R_0 R_\infty) R_\infty}{(R_\infty - R_0)} \right], \quad [1]$$

where the grammage is denoted w . The general RT scattering coefficient σ_s is determined by optimization using the DORT2002 model since there is no closed form expression relating this scattering coefficient to the

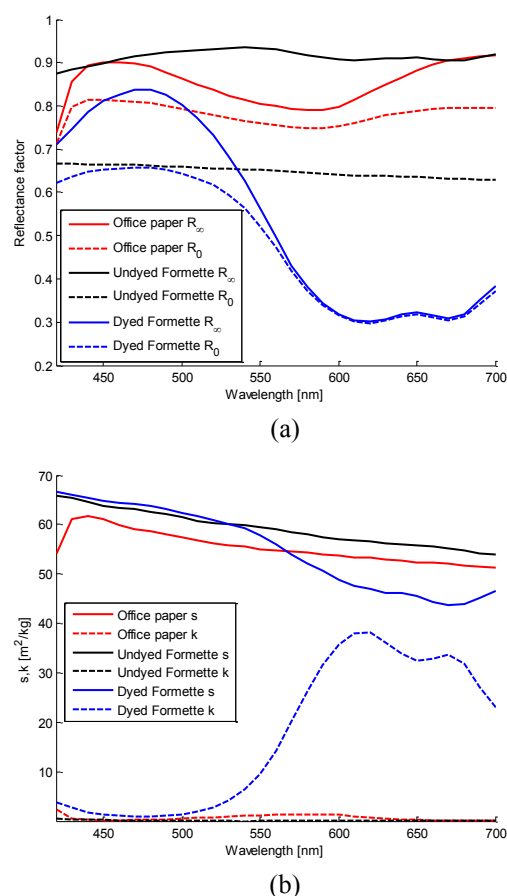


Fig 1. The R_∞ and R_0 reflectance factors measured in d/0 geometry (a) and the Kubelka-Munk scattering and absorption coefficients (b) of the samples used in this work. We can see that the scattering coefficient of the dyed sample decreases in the region of absorption of the dye.

reflectance factors. We calculate the respective scattering coefficients for all values of $\{R_\infty, R_0\}$ such that $0.16 \leq R_\infty \leq 0.92$, $0.15 \leq R_0 \leq 0.8$ and $R_\infty - R_0 > 0.01$, thus studying the phase portrait of the models in this space containing the most interesting variations. The model deviation is quantified by calculating the angle between the gradients of the respective scattering coefficients. Denoting this angle δ it can be calculated as

$$\delta = \arccos[\nabla\sigma_s(R_\infty, R_0) \cdot \nabla s(R_\infty, R_0)] \frac{180}{\pi}, \quad [2]$$

where we have taken the inverse cosine of the scalar product of the gradients. If the respective model gradients point in the same direction (i.e. if $\delta = 0$), then the RT and KM models will respond in the same way to changes in R_0 and R_∞ . If the gradients do not point in the same direction (i.e. if $\delta \neq 0$), then the models will respond differently to changes in R_0 and R_∞ and give different changes in light scattering. This measure thus shows in what regions of the R_0R_∞ space that the behavior of the general RT and KM models differ.

Light scattering and light absorption with general RT theory

The measured R_0 and R_∞ reflectance factors are used to calculate the RT scattering and absorption coefficients σ_s and σ_a by employing the pre-defined d/0 settings in the DORT2002 RT model. This simple procedure gives us scattering and absorption coefficients that are unaffected by the geometry induced error since the anisotropy of the reflected light is taken into account by the DORT2002 model. Necessary input to the model includes values of the grammage and of the asymmetry factor g . It has been shown that forward scattering dominates in paper and other organic materials (Cheong et al. 1990; Granberg, Béland 2004; Joshi et al. 2006; Prahl et al. 1993), and $g = 0.8$ is therefore a reasonable value to use when simulating paper media. The spectral dependence of g in paper is however an open issue that is currently being studied by the authors. We use the so-called similarity relations (Mudgett, Richards 1971, 1972; van de Hulst 1980) to scale the RT scattering and absorption coefficients that we obtain to the same order of magnitude as the KM scattering and absorption coefficients. The similarity relations are $s' = 3/4(1 - g)\sigma_s$ and $k' = 2\sigma_s$, where s' and k' denote the scaled coefficients, and the scaling is thus constant over the entire spectrum. This is done only for the sake of convenience, and there exists no exact translation of this kind (Neuman, Edström 2010a,b).

We use a modified version of the method introduced by Rundlöf and Bristow (1997) to compare how light scattering changes with absorption in the two models. Rundlöf and Bristow calculated Δs as a function of k in order to identify a critical value of k where the decrease in s becomes significant. However, the KM s and k are inherently model dependent and therefore cannot be meaningfully compared to the physically objective scattering and absorption coefficients of general RT. We therefore introduce the relative light scattering deviation as $\Delta s = (s_d - s_w)/s_w$ for KM and $\Delta\sigma_s = (\sigma_{s,d} - \sigma_{s,w})/\sigma_{s,w}$ for general RT. Here subscript d denotes the

dyed paper and subscript w denotes the undyed paper. This is a dimensionless measure of the relative change in light scattering of the two models.

Results

Deviation between general RT and KM theory

Fig 2 shows the local model deviation δ as a function of R_0 and R_∞ together with reflectance values of the office paper and of the blue Formette sample. We see that the model deviation is small when the reflectance is high, as for the office paper. But even for this paper there is a model deviation for shorter wavelengths, where the absorption increases (see Fig 1(b)). For wavelengths less than 420 nm it would be even more significant due to absorption of FWAs. The local model deviation increases as the reflectance factors decrease, i.e. as the absorption increases, and for wavelengths in the region of absorption of the dyed Formette paper the deviation is significant. It is interesting to note that for more translucent papers the deviation would be even more significant, and that there seems to exist a paper thickness where the model deviation reaches a maximum. Judging from these simulations, the geometry induced error is small when $R_0 > 0.75$ and $R_\infty > 0.8$ approximately. It should be noted that δ is a local measure of the deviation and therefore shows how the two models responds to changes around a particular point. When for example moving from weak to strong absorption in the R_0R_∞ space, as for the dyed Formette paper, the model deviation increases, which means that the model responses diverge. That is, the angle between the model gradients increases in this case.

Light scattering and light absorption with general RT theory

Fig 3 shows the relative deviation in light scattering as a function of absorption for the dyed Formette sample compared to the undyed Formette sample when using the KM model and general RT respectively. We can see that when using general RT the deviation in light scattering when the absorption increases is less than when using KM. This is a consequence of the model deviation

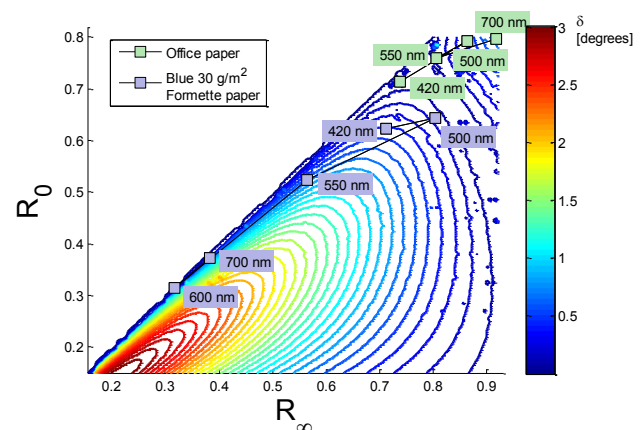


Fig 2. The local deviation between the KM model and general RT theory. The model deviation is small for the office paper but significant for the dyed Formette sample. For more translucent papers the model deviation would be even more significant.

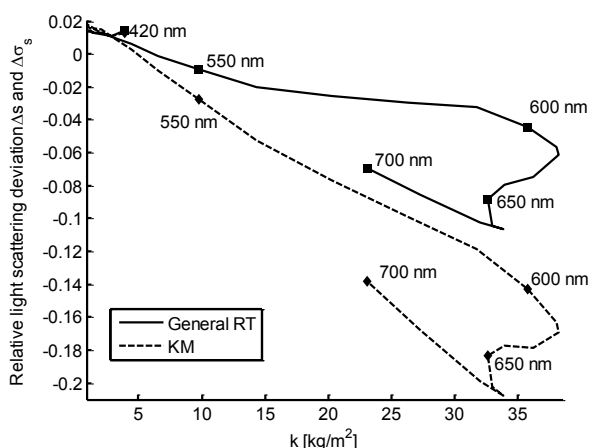
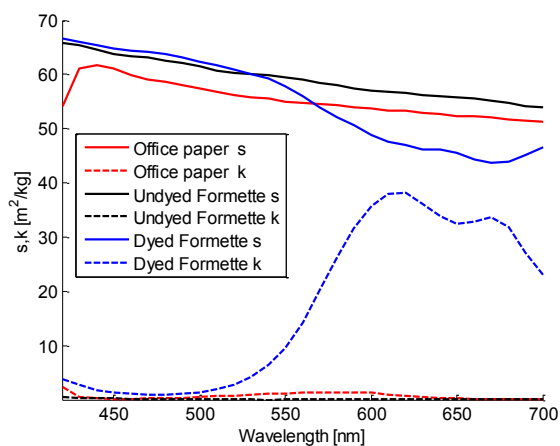
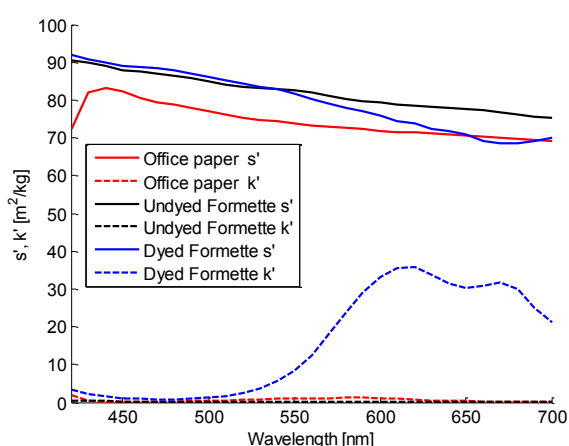


Fig 3. The relative deviation in light scattering as a function of absorption for the dyed Formette paper using general RT (solid) and KM (dashed). The decrease in light scattering is less when using general RT, which is a direct consequence of eliminating the geometry induced error.



(a)



(b)

Fig 4. The scattering and absorption coefficients calculated with the KM model (a) and with general RT theory (b). The decrease in scattering as the absorption increases is less when using general RT (compare solid blue in (a) and (b)). This is entirely due to the KM model neglecting angular variations of the light reflected in the d/0 instrument. General RT describes these variations accurately.

represented in Fig 2, which in turn is a consequence of the angular variations of the reflected light. These results thus show that neglecting the instrument geometry and the single scattering anisotropy indeed causes part of the dependence between light scattering and absorption in the KM model, and that general RT can relieve this situation. Fig 4(a) shows light scattering and absorption coefficients for all samples when using the KM model, and Fig 4(b) shows the coefficients when general RT is used. We can see also here that the decrease in scattering as the absorption increases is less when using general RT than when using KM theory (compare solid blue in (a) and (b)). This is entirely due to the KM model not capturing the geometric characteristics of the d/0 instrument and the single scattering anisotropy. The angular variations of the light reflected in the d/0 instrument are therefore neglected when using the KM model, but general RT describes these variations accurately, which reduces the dependence between light scattering and absorption.

Discussion and Conclusions

We have seen that the since long observed and debated dependence between light scattering and light absorption can be partly explained by taking the geometry of the d/0 instrument and the single scattering anisotropy into account when calculating the light scattering and absorption coefficients. This means that more detailed models, which include the instrument geometry, are more appropriate when assessing the light scattering characteristics of paper samples since otherwise we get a geometry induced error. When the samples are strongly absorbing, the error is considerable. Using general RT instead of KM can thus allow the papermaker to better predict the reflectance from mixtures of pulps, fillers and dye, at different dye or FWA amount and to better evaluate bleaching efforts.

The remaining dependence between light scattering and absorption, that we were unable to explain here, can potentially be explained completely by the spectral dependence of the single scattering anisotropy, i.e. the asymmetry factor g . Assessing the spectral asymmetry factor requires sophisticated measurements and optimization algorithms. Research on this subject is currently being conducted by the authors and will be reported shortly.

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Literature

Billmeyer Jr., F. W. and Abrams, R. L. (1973): Predicting reflectance and color of paint films by Kubelka-Munk analysis, I. Turbid medium theory, J. Paint Technol. 45, 23.

- Chandrasekhar, S.** (1960): Radiative Transfer, Dover.
- Cheong, W. F., Prah, S. A. and Welch, A. J.** (1990): A review of the optical properties of biological tissues, IEEE J. Quantum Electron. 26, 2166.
- Edström, P.** (2005): A Fast and Stable Solution Method for the Radiative Transfer Problem, SIAM Rev. 47, 447.
- Edström, P.** (2008): A Two-Phase Parameter Estimation Method for Radiative Transfer Problems in Paper Industry Applications, Inverse Probl. Sci. Eng. 16, 927.
- Edström, P.** (2010): Simulation and modeling of light scattering in paper and print applications, in Kokhanovsky, A. A. (ed.), Light Scattering Reviews 5, Springer, p. 451.
- Edström, P., Neuman, M., Avramidis, S. and Andersson, M.** (2010): Geometry Related Inter-Instrument Differences in Spectrophotometric Measurements, Nord. Pulp Pap. Res. J. 25(2), 221.
- Foote, W. J.** (1939): An investigation of the fundamental scattering and absorption coefficients of dyed handsheets, Pap. Trade J. 109, 397.
- Glassner, A. S.** (1995): Principles of Digital Image Synthesis, Volume Two, Morgan Kaufman.
- Granberg, H., Béland, M.-C.** (2004): Modelling the angle-dependent light scattering from sheets of pulp fibre fragments, Nord. Pulp Pap. Res. J. 19(3), 354.
- Granberg, H., Edström, P.** (2003): Quantification of the intrinsic error of the Kubelka-Munk model caused by strong light absorption, J. Pulp Pap. Sci., 29(11), 386.
- van de Hulst, H. C.** (1980): Multiple Light Scattering. Tables, Formulas and Applications. Vol. 2., Academic Press, New York.
- Hutchings, J. B.** (1999): Food Color and Appearance, Aspen Publishers.
- ISO 2469** (1994): Paper, board and pulps – Measurement of diffuse reflectance factor, International Organization for Standardization: Geneva, Switzerland.
- ISO 9416** (1998): Paper - Determination of Light Scattering and Absorption Coefficients (Using Kubelka-Munk Theory), International Organization for Standardization: Geneva, Switzerland.
- Joshi, N., Donner, C. and Jensen, H.W.** (2006): Noninvasive measurement of scattering anisotropy in turbid materials by nonnormal incident illumination, Opt. Lett. 31, 936.
- Karlsson, A., Enberg, S., Rundlöf, M., Paulsson, M. and Edström P.** (2012): Determining optical properties of mechanical pulps: sheet forming procedure and investigation of different ways to evaluate light absorption, accepted for publication in Nord. Pulp Pap. Res. J.
- Koukoulas, A. A. and Jordan, B. D.** (1997): Effect of Strong Absorption on the Kubelka- Munk Scattering Coefficient, J. Pulp Pap. Sci., 23(5), 224.
- Kubelka, P. and Munk, F.** (1931): Ein Beitrag zur Optik der Farbanstriche, Z.Tech. Phys. 11a, 593.
- Mishchenko, M. I.** (2010): Poynting-Stokes tensor and radiative transfer in discrete random media: The microphysical paradigm, Opt. Express 18(19), 19770.
- Moldenius, S.** (1983): Light absorption coefficient spectra of hydrogen peroxide bleached mechanical pulp, Paperi Puu 65, 747.
- Moussa, A., Dupont, D., Steen, D. and Zeng, X.** (2009): Multiangle study on color of textile structures, Col. Res. Appl. 34, 274.
- Mudgett, P., Richards, L.W.** (1971): Multiple Scattering Calculations for Technology, Appl. Optics 10(9), 1485.
- Mudgett, P., Richards, L.W.** (1972): Multiple Scattering Calculations for Technology II, J. Colloid Interface Sci. 39(3), 551.
- Neuman, M. and Edström, P.** (2010a): Anisotropic reflectance from turbid media. I. Theory, J. Opt. Soc. Am. A. 27(5), 1032.
- Neuman, M. and Edström, P.** (2010b): Anisotropic reflectance from turbid media. II. Measurements, J. Opt. Soc. Am. A. 27(5), 1040.
- Nobbs, J. H.** (1985): Kubelka-Munk theory and the prediction of reflectance, Rev. Prog. Coloration 16, 66.
- Nordman, L., Aaltonen, P. and Makkonen, T.** (1966): Relationships between mechanical and optical properties of paper affected by web consolidation, Trans. 3rd Fundamental Res. Symp., Cambridge, UK.
- Olf, H. G.** (1989): Correspondence between the Kubelka-Munk and the Stokes model of strongly light-scattering materials, Part II: implications, Tappi J. 72(7), 159
- Olf, H. G.** (1990): Reply to Johannes A. Van den Akker's response, Tappi J. 73(2), 13.
- Pauler, N.** (2002): Paper Optics, AB Lorentzen & Wettre, Kista.
- Prah, S. A., van Gemert, M. J. C. and Welch, A. J.** (1993): Determining the optical properties of turbid media by using the adding-doubling method, Appl. Opt. 32, 559.
- Rundlöf, M. and Bristow, J. A.** (1997): A note concerning the interaction between light scattering and light absorption in the application of the Kubelka-Munk equations, J. Pulp Pap. Sci., 23(5), 220.
- Schmitt, J. M.** (1999): Optical coherence tomography (OCT): A review, IEEE J. Sel. Top. Quant. 5, 1205.
- Thomas, G. E. and Stamnes, K.** (1999): Radiative Transfer in the Atmosphere and Ocean, Cambridge University Press.
- Van den Akker, J. A.** (1949): Scattering and absorption of light in paper and other diffusing media, Tappi J. 32(11), 498.
- Van den Akker, J. A.** (1990): Comments on the correspondence between the Kubelka-Munk and Stokes model, Tappi J. 73(2), 12.