

## KUBELKA-MUNK THEORY IN DESCRIBING OPTICAL PROPERTIES OF PAPER (II)

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Subject review

Kubelka-Munk theory was quickly adapted for use by the papermaking industry and has now been in widespread use for decades in the measurement and prediction of colour, brightness and opacity of paper sheets. Though it remains the most used in practice, it has some disadvantages and the second part of the article will cover a system that is less applicable to the theory itself (an ink-dyed sheet of paper or strongly absorbing media) as well as numerical methods (simulations of individual photon paths) which are used to improve the results obtained by the K-M theory. The most commonly used are Monte Carlo simulation model, GRACE and DORT. Most of the information reported here is taken from the references cited at the end of the article which should be consulted for a more in-depth study.

**Keywords:** *absorption, Kubelka-Munk, paper, scattering*

### Kubelka-Munk teorija u opisivanju optičkih svojstava papira (II)

Pregledni članak

Kubelka i Munk dali su sažeti oblik i pristup s jasnim ciljem ka praktičnim metodama mjerenja. Njihova metoda je bila brzo prihvaćena u papirnoj industriji i danas ima široku primjenu u mjerenju i procjeni obojenja, svjetline i opaciteta pri proizvodnji papira. Unatoč tome navedena metoda posjeduje neke nedostatke i drugi dio članka će pokriti sustave na koje se ne može tako kvalitetno primijeniti (otisnuti i reciklirani papir). Također će biti riječi i o numeričkim modelima (simulacije putanja pojedinačnih fotona) koji se koriste kao nadopuna rezultatima dobivenima K-M metodom. Najčešće korišteni su Monte Carlo simulacijski modeli, GRACE i DORT. Većina informacija u ovom članku preuzeta je iz referenca čiji se popis nalazi na kraju članka i koje je potrebno konzultirati za detaljniju analizu.

**Cljučne riječi:** *apsorpcija, Kubelka-Munk, papir, raspršenje*

## 1

### Introduction

Knowing the optical properties of paper is of a great importance in all papermaking industry. One of the most successful and simplest models of describing them is that of Kubelka and Munk (K-M). With this model, optical properties of paper under diffuse illumination can be predicted from effective absorption and scattering coefficients of the material. Though the Kubelka-Munk theory remains the most used in practice, it has some disadvantages, like imprecision in some cases (fluorescence problems for instance). To improve the results obtained by the use of the K-M theory, numerical methods are common nowadays which simulate individual photon paths (Monte Carlo simulation model, GRACE, DORT).

## 2

### Interactions of light with strongly absorbing media

While the KM theory enjoyed great success in a range of scientific and industrial applications [1-3] its shortcomings prevented the model from being applied to specific systems with media containing absorptive components. If we have any absorption at all, oblique rays will be attenuated more than vertical rays when passing between layers, and this will invalidate the assumption that we have diffuse light everywhere in the medium. In particular, the incident and the reflected light will have different angular distributions and will not be properly described by a symmetric pair of differential equations like (1) and (2) (Eq. 1 and 2, Part 1) [60]. One important example of a system that is less applicable to the KM theory is an ink-dyed sheet of paper or strongly absorbing media [4-10] and attempts have been made to attribute some of this behavior to intrinsic errors of the KM mode [11-15]. Understanding the origin of the theory's shortcomings has attracted

continued interest from the researchers worldwide [16-21].

### 2.1

#### The coefficients $K$ and $S$

Researchers seem to agree that the problem lies essentially with the inherent nonlinear relationship between the K-M proportionality constants (the K-M coefficients),  $K$  and  $S$ , and the physical, intrinsic optical properties of the materials  $\sigma_a$  and  $\sigma_s$ , represented by the absorption and scattering probabilities, i.e. by the mean free path in a medium.

K-M theory assumes that light propagates in just two opposing directions, with the flux variation at any point in the medium being linearly proportional to the two local opposing fluxes. The proportionality constants,  $K$  and  $S$ , are assumed dependent on the absorption and scattering properties of the medium. Approximate relations between the KM coefficients and physically objective parameters scattering cross sections  $\sigma_a$  and  $\sigma_s$ , have been suggested.  $K$  is obviously closely related to  $\sigma_a$ , but since the incident light is diffuse, the average path length for a light ray passing through a layer of thickness  $dx$  is  $2dx$ . This means that

$$K = 2\sigma_a. \quad (1)$$

$S$  is subject to the same change as  $K$  due to the average path length, but additionally, in the formulation of equations (1) and (2) (Eq. 1 and 2, Part 1) [60],  $S$  addresses only the part of the scattered light that changes direction from upwards to downwards, or vice versa. For isotropic scattering, this is exactly half of the actually scattered light. Thus, the two effects compensate, and

$$S = \sigma_s. \quad (2)$$

When comparing the predictions of K-M theory (Eqs. 1 and 2) with proper analytical or numerical calculations, it has been reported by Nobbs [15] and Mudgett and Richards [22, 23] that the relation between  $S$  and  $\sigma_s$  is actually better described by the modified equation:

$$S = \frac{3}{4} \sigma_s. \quad (3)$$

These relations are approximate, since dependencies between  $S$  and  $K$  have been reported [4-6, 10, 24], while  $\sigma_a$  and  $\sigma_s$  are considered to be independent.

Other relations have been suggested in different fields of application to explain the apparent dependence between  $S$  and  $K$ . These relations must all be approximate because KM model is fundamentally simpler and a translation to higher-order models could never be complete. They should be regarded as the first term of some series expansion.

A recent contribution in this matter is from Yang and co-workers [25-28], who in their work propose a rederivation to correct an oversight of the original KM theory.

L. Yang and B. Kruse [25] analyzed the shortcomings of the Kubelka–Munk theory from a purely theoretical-physics viewpoint, i.e., independent of any specific application, and proposed a revision that would include more accurately in a general way the effects of light scattering in a turbid medium. The fundamental idea [28] was to take into account the effect of internal scattering of light on the total path traversed by a photon within the medium. The essential point is that the scattering that takes place has an influence on the final probability of a photon being absorbed. This feature was overlooked in the original KM theory. This revised description led first to both KM coefficients of absorption and scattering,  $K$  and  $S$ , becoming dependent on both of the physically meaningful coefficients of absorption and scattering,  $\sigma_a$  and  $\sigma_s$ . Second it led to a nonlinear relationship between the two sets of coefficients. These relationships can be summarized by the set of equations:

$$K = \alpha \mu \sigma_a \quad (4)$$

$$S = \frac{\alpha}{2} \mu \sigma_s \quad (5)$$

where the quantity  $\mu = \mu(\sigma_s, \sigma_a)$  is a scattering-induced path variation (SIPV) factor describing the influence of light scattering on the total path length and is nonlinearly dependent on both the absorption and scattering properties of the medium. Mathematically  $\mu$  is defined as the ratio of the true path length  $L$  and the corresponding straight-line displacement  $r$ .

In the original K-M theory the coefficients of absorption and scattering,  $K$  and  $S$ , were assumed linear functions of the corresponding intrinsic physical parameters  $\sigma_a$  and  $\sigma_s$ ; (Eqs. 1 and 2) and hence correspond to  $\mu = 1$  (no effect of scattering on path length) and  $\alpha = 2$  (diffuse light distribution).

On the basis of a physical model assuming strong light absorption the expression is [28]:

$$\mu = \sqrt{\frac{\sigma_s}{\sigma_a}} \quad (6)$$

which clearly articulates the mutual dependence of absorption and scattering. This fairly simple description can explain the experimental finding that  $S$  decreases significantly when the absorption  $\sigma_a$  increases, while  $\sigma_s$  stays essentially constant. For a less absorptive medium, however, the influence of absorption is overestimated by this expression. Consequently, there is a need for a more general expression for the SIPV factor  $\mu$  capable of accommodating a much wider range of absorption influences.

Yang and co-workers derive their expressions for  $S$  and  $K$  for a layer of finite thickness, but they inadequately combine this with KM theory which is a differential equation and thereby implicitly use infinitesimal layers. To get adequate results, the limiting process should be explicitly carried out to obtain expressions for  $S$  and  $K$  for infinitesimal layers. This limiting process is omitted in their work as P. Edstrom pointed out. More about that problem you can find in [29].

Another situation where K-M theory should be used with caution is when the material under consideration is highly translucent, i.e., when a significant amount of light passes straight through the medium without scattering. In that case the thickness of the medium needs to be considerably larger than the mean free path in order for the assumptions to hold.

However, if it is irrelevant what the exact relations between the model parameters and the physical parameters are, the K-M theory is in fact applicable to a wide range of physical materials. The parameters  $K$  and  $S$  may be determined entirely from indirect measurements, e.g. from measurements of the diffuse reflectance over two or more backgrounds with different reflectance. So, KM method gives information about the bulk of materials and must be determined for each substrate individually. As such an empirical model, treating the parameters as measurement data without any explicit relation to the physical scattering and absorption cross sections, the K-M theory has been successfully used in many applications.

## 2.2

### Deinked paper (recycled paper and handsheets)

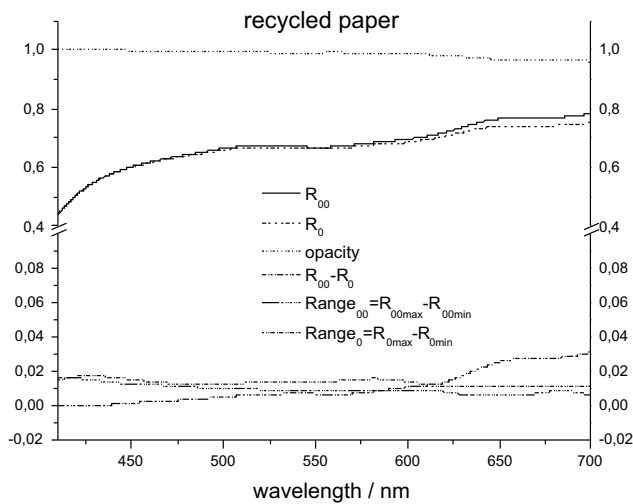
Ink removal is one of the most important steps in recycling of mixed recovered office paper, old magazines and old newspapers. Ink removal efficiency of a recycling operation is characterized by the brightness ( $R_{457}$ ) increment of the final paper over that of feed stock. The brightness ( $R_{457}$ ) method was developed to monitor the bleaching of pulp, because the reflectivity is changed the most at these short wavelengths (400-500 nm) during the bleaching of the pulp. With the introduction of modern spectrophotometers it has been convenient to use reflectance at 460 nm [30]. It is practically the same whether the reflectance at a wavelength of 457 or 460 nm is used.

The measurement of effective residual ink concentration (ERIC) in recycled papers depends on their opacity. Opacity, the degree of non-transparency, is one of the fundamental optical properties of the paper. For the paper to be acceptable, opacity has to exceed 80 %. The opacity should be so high that text and pictures from the underlying side are not interfering. For opacity lesser than 97 %, the method is based on the application of the Kubelka-Munk theory to diffuse reflection from papers measured once with a black backing  $R_0$  and again with a thick backing of the same papers  $R_\infty$  (TAPPI 519) [31]:

$$O = \frac{R_0}{R_\infty} 100\% \tag{7}$$

Low-grammage paper has a low  $R_0$  value (low opacity value) and the  $R_0$  value approaches  $R_\infty$  when grammage increases or/and number of ink particles after recycling process increases. So for deinked paper we expect high opacity value because of the remainder of ink particles after the recycling process. At opacities above 97 %, the two reflection values tend to become statistically indistinguishable.

As we can see from Fig. 1 [32] in the blue part of the reflectivity and reflectance spectra of recycled paper there are no differences between  $R_0$  and  $R_\infty$ . In that part of spectrum opacity is  $\approx 1$  and two measured reflectance values are statistically indistinguishable. In the red and infrared part of spectra, the absorption from lignin and dyes can be ignored. While the ink is the predominant absorber,  $R_\infty > R_0$  and they are not any more indistinguishable. So, towards the red part of spectrum, opacity is  $\approx 0,97$  and the difference of mean values of reflectance and reflectivity become higher than dissipation values of measured reflectance and reflectivity spectra (Fig. 1).



**Figure 1** Recycled paper mean values of reflectance  $\bar{R}_0$  and reflectivity  $\bar{R}_\infty$  spectra, their differences  $\bar{R}_\infty - \bar{R}_0$  compared with their ranges,  $\text{Range}_0$  and  $\text{Range}_g$ , and opacity.

Jordan and Popson [33] developed a near-infrared reflectance technique to measure effective residual ink concentration (ERIC) in paper made of deinked pulp using the K-M theory. The technique measures reflectance at an infrared wavelength ( $\approx 950$  nm) from a paper sample over black backing,  $R_0$ , and reflectance from a thick stack of paper from the sample,  $R_g$ :

$$ERIC = \frac{k_{\text{sample}}}{k_{\text{ink}}} 10^6 \tag{8}$$

Black inks have been shown [34] to have an absorption coefficient  $k_{\text{ink}} = 10000 \text{ m}^2/\text{kg}$ , while the specific absorption coefficient of the sample  $k_{\text{sample}}$  can be calculated from the two measured reflectance values,  $R_0$  and  $R_g$  (Eq.12, Part 1) [60]. It is directly related to the residual ink concentration in the paper sample when measured at a near-infrared wavelength, because the ink is the predominant absorber of infrared

light. The technique has been adopted by TAPPI as a provisional test method T 567 pm-97 [35] to measure effective residual ink concentration (ERIC) of deinked pulp. The estimation of residual ink by optical measurements in the infrared area is more accurate and much less time consuming for submicron ink particles [36] (below  $10 \mu\text{m}$ ) than determination by image analysis method. The technique works fairly well except for the large standard deviations encountered in measuring papers of high opacity resulting from high grammage, ash content, or ink concentration (high ERIC values). These large standard deviations are magnified through the use of several different kinds of spectrometers to measure ERIC in practice, some of which (like most UV-Vis spectrometers) do not extend into the infrared. Low measurement accuracy for high-ERIC papers occurs primarily because the denominator of the logarithmic function in the Eq. (12) (Part 1) approaches zero as  $R_0/R_g$  approaches unity. The condition  $R_g = R_0$  defines a singular point. In tests of opaque papers, measurements of  $R_0$  and  $R_g$  become statistically indistinguishable, resulting in an indeterminate value for  $S$  in Eq. (12) (Part 1) and forcing the use of an approximate value.

Many authors have used measurements of  $R$  and  $T$  to solve the inverse variables of absorption  $K$  and scattering  $S$  [37-42].

A new approach proposed by D.W. Vahey and co-workers [43] measures the reflectance at the incident surface,  $R$ , and transmission at the back surface,  $T$ , from the same paper sample without back reflectance or remounting. The inverse equations expressing  $s$  and  $k$  in terms of  $R$  and  $T$  are:

$$s = \frac{2R}{w\sqrt{(1-T^2 + R^2)^2 - 4R^2}} \sinh^{-1} \left[ \frac{1}{2T} \sqrt{(1-T^2 + R^2)^2 - 4R^2} \right] \tag{9}$$

$$k = \frac{(1-R)^2 - T^2}{w\sqrt{(1-T^2 + R^2)^2 - 4R^2}} \sinh^{-1} \left[ \frac{1}{2T} \sqrt{(1-T^2 + R^2)^2 - 4R^2} \right] \tag{10}$$

These results agree with Eqs. (12) and (13) (Part 1) and with those of Knox and Wahren [44]. A singularity occurs when  $T = 0$ , which is consistent with that of K-M Eq. (12)(Part 1) since a paper for which  $T=0$  must also have  $R_0 = R_g$ . However, when  $T$  is small but finite, differences between  $R_0$  and  $R_g$  will be on the order of  $T^2$ , and therefore much harder to measure accurately than  $T$  itself. This translates into greater ERIC measurement accuracy using Eq. (10) in place of Eqs. (12) and (13)(Part 1), [60].

For opacities above 97 %, corresponding to  $T$  about 3 %, Eq. (13) (Part 1) is approximated as

$$k = \frac{\bar{s} \cdot (1 - R_\infty)^2}{2R_\infty} \tag{11}$$

where  $\bar{s}$  is an average value of scattering coefficient.

D. W. Vahey and co-workers [43] compare three methods for determining ERIC:

- a) RT method, Eq. (10)
- b)  $R_\infty R_0$  method, Eqs. (12) and (13)(Part 1) [60]
- c)  $\bar{s} R_\infty$  method, Eq. (11).

The main advantage of the RT method is that  $T$  can usually be measured more accurately in absorbing papers

than differences between  $R_0$  and  $R_\infty$ . A secondary advantage is that it only requires measurement of a paper's reflectance at the incident surface and transmittance at the back surface. No sample remounting is required to measure the two properties, while the  $R_\infty R_0$  method requires measurement of a paper twice to know its properties: first, as a single sheet; second, as part of an opaque stack of paper from the same sample. Because of the non-homogeneity of ink and fiber distribution in paper, the two measurements introduce variability unless perfectly repeated alignment and contact between papers in the stack are maintained from test to test. At high opacities, the precision of the  $R_\infty R_0$  method is significantly compromised. A disadvantage of the RT calculation is that it becomes error prone at low ERIC values.

Therefore, the RT method suffers in measuring low-opacity samples just as the  $R_\infty R_0$  method suffers in measuring high-opacity samples. The two techniques are complementary to each other. The superiority of the  $R_\infty R_0$  method is not demonstrated until ERIC values are below 150 ppm, a value seldom achieved in commercial practice, while RT method works well from the first stages of ink removal to the commercial range. The  $s R_\infty$  method is found to be intermediate to the RT and  $R_\infty R_0$  methods.

D. W. Vahey and co-workers [43] suggest that refinement of the RT method deserves incorporation into standard methods for ERIC measurements such as T567 pm-97. It may prove useful whenever diffuse reflection geometries are limited because of high sample opacities.

### 3 Models for simulating light scattering in paper

Recently, the paper industry has shown an increased interest in radiative transfer theory, in order to simulate and predict light scattering in paper. Interactions between photons and paper are very complex, and the Kubelka-Munk model oversimplifies the problem by considering radiation in only two directions – upwards and downwards. Although a simple model, it has been commonly used since it is fast and easy to use, i. e. the measured quantities (reflectance factors) are directly related to the parameters of the model ( $S$  and  $K$ ). However, the model is crude and computational savings are not an argument for not considering more sophisticated models.

#### 3.1 Discrete Ordinate Radiative Transfer model

The Kubelka-Munk model cannot provide information about the angular distribution of the transmitted and scattered light, nor does it include any surface contributions to scattering. To include angular distribution, Mudgett and Richards [45], building up on Schuster's ideas [46], developed a discrete ordinate model with several channels. Their main interest lay in the optics of paint films. Later, Berglind [47] proposed a straightforward implementation of this model, referred to as DORT (Discrete Ordinate Radiative Transfer model) [48].

The paper industry has adopted DORT for use in optical design, due to its ability to simulate angle-resolved scattering, and the scattering of light in multilayered structures with different index of refraction in different layers, such as coated paper.

DORT2002 is adapted to light scattering simulations in paper and print [49, 50], while DISORT is mostly applied to atmospheric research [51]. Both of these models were designed for much more challenging tasks, so they fully include the KM situation as a simple special case [52] where the illumination and scattering is completely isotropic and where only two channels are considered.

P. Edstrom and co-workers [48] propose a Stable Multilayer Discrete Ordinate Radiative Transfer model (SM-DORT) to simulate the scattering of light in coated paper and similar structures.

#### 3.2 Monte Carlo simulation methods

Since most of the detailed mechanisms of scattering are known and are simple to model, it is possible to successfully simulate individual photon paths by pseudo-random Monte Carlo methods. Using this approach, even very general problems with arbitrary geometry, inhomogeneous media and anisotropic scattering can be simulated. The drawback is that it is extremely time-consuming, but with the recent advances in computer technology, Monte Carlo simulation of multiple scattering has become quite tractable. A few authors have reported some success in calculating the reflectance properties of paper by making a three-dimensional model of the paper constituents and performing Monte Carlo simulations on the model structure [53, 54].

Hainzl [55] recently proposed a Monte Carlo simulation model, GRACE. This model is a modern simulation tool for light scattering in paper [56]. It takes into account the three-dimensional nature of the paper describing light interaction with individual component in the paper structure. The model does not require any restrictions for the media and can be used, after some improvements, to model real paper in a real environment. The ability to model "the real world" is the greatest disadvantage of GRACE, since it involves substantially more parameters, many of which are unknown or difficult to measure. The calculation time increases, since the paper structure is modeled as a statistical distribution. Therefore, hundreds of thousands or millions of wave packets need to interact with the paper structure in a simulation.

KM should be used where its accuracy is sufficient, and a DORT tool should be used where higher accuracy is needed. The reported problems are largely due to the low resolution of the KM two-flux model, and can be resolved with a radiative transfer model of higher resolution.

In the future, the Kubelka-Munk inverse solutions for  $k$  and  $s$  may be replaced by software such as DORT2002 [57-59].

### 4 Conclusion

Kubelka-Munk theory, though it remains the most used in practice, has some disadvantages, like imprecision in some cases (fluorescence problems for instance), initial assumptions that are too much simplified. Real paper never completely satisfies all that assumptions, but researchers are interested in finding a model to upgrade that theory probably due to its explicit form, simple use and its acceptable prediction accuracy in many cases useful in paper, paint and colorant industry.

To improve the results obtained by the use of the K-M

theory, numerical methods are common nowadays which simulate individual photon paths (Monte Carlo simulation model, GRACE, DORT).

## 5

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