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This is a published version of a paper published in *Nordic Pulp & Paper Research Journal*.

Citation for the published paper:

Hägglund, H., Norberg, O., Neuman, M., Edström, P. (2012)

"Dependence between paper properties and spectral optical response of uncoated paper"

Nordic Pulp & Paper Research Journal, 27(2): 440-444

URL: <http://dx.doi.org/10.3183/NPPRJ-2012-27-02-p440-444>

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Dependence between paper properties and spectral optical response of uncoated paper

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KEYWORDS: Paper properties, Variations, Optical, Spectral, Grammage, Filler content, Uncoated, PLS, Light scattering

SUMMARY: This paper presents a method to describe, with good accuracy, the relation between variations in paper properties and variations of the spectral optical response of an uncoated paper. The dependence between density, filler content, grammage, and the spectral optical response is characterized by a multivariate model. The model is based on large-scale measurements data on a set of paper samples that have been produced with different values of grammage, density and filler content, representing the variations within a normal 80 g/m² uncoated paper. From the optical measurements the light scattering (s) and light absorption (k) coefficients have been estimated according to the Kubelka-Munk theory.

The results from this study will give valuable input to optical modeling activities, where the optical variations are predicted from measured small-scale variations in underlying paper properties.

The variations in the paper properties can be used to model the light scattering coefficient, s , but there were too small variations in the light absorption coefficient, k , to find any significant dependence to the paper properties for the samples studied in this work. Furthermore, linear models give sufficient accuracy in the intervals studied. Additional findings from this study are the different effects of wet-pressing and calendering on the light scattering coefficient.

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The optical characteristics of paper such as whiteness, opacity and brightness are in general derived from large-scale measurements. However, the visual impression of a paper is not only affected by the average value of the optical properties but also by small-scale variations that might impair the visual appearance. These variations are also often amplified in print. The optical variations are due to spatial variations of paper properties, such as filler content, grammage, and density. Furthermore, paper-making in most cases implies compromising between different paper properties and also production cost. Therefore, it is relevant to find out what properties should be given priority when designing the paper. This will be

helped by a good understanding of the relations between paper properties and the optical response of the paper.

Related research

The qualitative relations between paper properties and optical properties like whiteness, brightness and opacity are generally well known (Pauler 2002, Holik 2006, Alava 2006, Fellers et al., 1998). Formation and print mottle are commonly used measures for evaluations of the small-scale variations in paper and print. There are several reported works that focus on the comparison of the interrelation between different paper properties and the optical response at high resolution. To evaluate the combined influence of local paper properties on the final product, a common method is to use image registration methods to combine 2D paper property measurements. Kajanto (1989) investigated the interrelation between offset print mottle and formation. Mettänen et al. (2008) used image registration to relate paper formation, local surface topography and local print density. The relationship of local basis weight and offset fiber picking was studied by Chinga, Syverud (2007) from 2D paper measurement maps. Hirn et al. (2008) presented a robust image registration method to combine 2D paper measurement maps with arbitrary resolution.

However, in the present study, efforts have been put on describing the dependence between small-scale variations in paper properties and the spectral changes of the optical response that occur in a paper.

Aim of the present study

This study attempts to present a method for high-accuracy characterization of the interrelations between variations in paper properties and variations of the spectral optical response of an uncoated paper. Previous studies have indicated a non-linear relationship between filler content and the light scattering coefficient, s , at high filler loadings (Middleton et al., 1994). Therefore, possible non-linear relations between paper properties and optical response are also evaluated.

The results from this study will increase the understanding of the complex interaction between light and paper and may in a later work be combined with image registration methods to evaluate the influence of measurement scale on the interrelations between paper properties, optical response and appearance of the paper.

Materials and Methods

Determination of variation span

Local variations of paper properties in paper will affect the optical response and give rise to inhomogeneity in print. When regarding relatively large surfaces, the optical variations between two measurements will be small but with increased measurement resolution the variation spans will become larger. Furthermore, at a

spatial scale in the order of 1 mm², the variations can be considered as perceptual disturbing at normal viewing distance; variations at an even smaller scale are less disturbing for the human eye (Fairchild, 1998).

In order to assess the intervals of variations of paper properties that are representative for this study, the beta formation, calcium content, and STFI thickness are characterized for a normal 80 g/m² uncoated paper at a measurement scale of 1 mm².

Paper samples

In order to assess the variations of paper properties and the corresponding variations of spectral optical response with good accuracy, 27 paper samples with different values of grammage, density and filler content have been produced with property intervals chosen to represent the variations within a normal 80 g/m² uncoated paper. The paper samples were produced in a Formette Dynamique, which is known to give homogeneous paper with small spatial variations. The range of each paper property of the paper samples is shown in Table 1. The property ranges were selected from the intervals of variations assessed from a commercial 80 g/m² office paper at 1 mm² measurement scale.

The pulp was a mixture of two chemical pulps and consisted of 70% hardwood and 30% softwood, refined to 26°SR. The samples did not contain any fluorescent whitening agent or dye.

The different density levels were achieved by varying the pressure in the wet-pressing of the paper samples. In addition, to further increase the density span, one sheet of each sample was calendered. Soft nip calendering is known to produce a relatively even densification of the paper. Therefore, a soft-nip calender was used in this study, with a pressure of 32kN/m. Before including the calendered samples into the numerical analysis, they were compared to the uncalendered samples to see if they may be used in the same sample set. The resulting density and thickness intervals for the calendered paper sheets are shown in Table 2.

Measurements

Standard methods were used to assess the thickness, grammage and filler content of the paper sheets (ISO 534 1988; ISO 536 1995; ISO 1762 2001). The reflectance values of the samples were measured according to (ISO

Table 1. Ranges of paper properties of produced paper samples.

Paper property	min value	max value
Grammage	70.9 g/m ²	98.4 g/m ²
Filler content	20.8 %	29.7 %
Density	585 kg/m ³	783 kg/m ³
Thickness	102 µm	131 µm

Table 2. Intervals of paper properties of calendered paper samples.

Paper property	min value	max value
Density	716 kg/m ³	875 kg/m ³
Thickness	87.5 µm	116 µm

2469 2007) with a Lorentzen & Wettre Elrepho d/0 instrument. From the spectral reflectance the light scattering coefficient, s , and the light absorption coefficient, k , were calculated using Kubelka-Munk theory (ISO 9416 1998).

Numerical analysis

The samples used in this study vary mainly in paper structure (e.g. porosity and compression of fibers) and filler content. The variations of paper properties are therefore expected to mainly affect the light scattering coefficient, s , while the light absorption coefficient, k , is expected to show only small variations for the samples used. To verify this, a principal component analysis (PCA) was performed. The influence of the paper properties on the spectral values can be shown in the loading plots of the two first principal components. The PCA was also used to find spectral monotonicities in the spectral values, which can reduce the number of variables needed to represent the optical response.

For a detailed analysis, a partial least squares (PLS) method was used to derive an empirical model that relates the paper properties with the spectral optical response. To give the variables equal importance, the parameters were mean-centered and unit-variance scaled before the modeling. Possible models were evaluated in terms of the coefficient of determination, R^2 , and the root mean square error of prediction, $RMSEP$.

The coefficient of determination, R^2 , indicates to which extent the variations in paper properties and optical response are accounted for by the model. R^2 is defined by:

$$R^2 = 1 - \frac{\sum_i (y_i - y_{i,pred})^2}{\sum_i (y_i - y_{mean})^2}, \quad [1]$$

where y_i are the observed values, $y_{i,pred}$ are the predicted values, and y_{mean} is the mean of the observed data. The quantity R^2 ranges from 0 (no fit) to 1 (complete fit).

The predictability indicates how well the model predicts values for another set of data. The predictability can be evaluated graphically by plotting the values predicted by the model against measured values, or analytically by calculating a summarized error, such as the root mean square error of prediction, $RMSEP$, which is defined by:

$$RMSEP = \sqrt{\frac{\sum_{i=1}^n (y_{i,ref} - y_{i,pred})^2}{n}}, \quad [2]$$

where $y_{i,ref}$ are observations taken from a validation data set.

The samples used in the study were divided into two equally large parts: a training set, which was used for the calculation of the model, and a prediction set, which was used to evaluate the predictability of the model.

Possible non-linearities in the dependencies between paper properties and optical response were evaluated by comparing the R^2 -values and predictability of models when quadratic terms and cross-terms were included in the model. If the coefficient of determination, R^2 , and the predictability do not increase significantly with a non-

linear term included, the chosen non-linear term is concluded to be insignificant for the model. The significant contribution of each paper component to the optical response was evaluated in the same manner.

Results

Effect of calendering

In Fig 1, the mean value of the light scattering coefficient, s , is plotted against density for calendered and uncalendered samples. Larger density values give a lower light scattering coefficient when the samples are wet-pressed. However, calendering the dry samples with a pressure of 32 kN/m does not show any significant effect on the light scattering coefficient.

Principal Component Analysis

Fig 2 a) shows the PCA loading plot for the two first principal components when density, filler, grammage, and the spectral light absorption coefficient, k , are varied. The correlation between k and paper properties shown in Fig 2 a) is small. This can be seen from the fact that the three paper properties are near zero in the first principal component, which explains 91% of the variations.

In Fig 2 b), the PCA loading plot for the light scattering is shown. The first component explains 93 % of the variations. The filler content (Fi) has the strongest correlation to the light scattering coefficient, s , while density and grammage mainly affect the ratio between s -values at low wavelengths and s -values at high wavelengths. Density and grammage are strongly correlated, which indicates that the light scattering can be modeled with only filler content and one of the two properties without reducing accuracy.

The spectral light scattering coefficient, s , behaves monotonically: only the mean value and the slope of the scattering coefficient are affected by the variations in paper properties. This means that the spectral behavior of s can be represented by the mean value, s_{mean} , and a ratio between s at two different wavelengths representing the slope. In order to compensate for noise, the spectral filters for brightness and for Y are used to represent the two wavelengths 457 nm and 557 nm respectively, so that the quotient s_{457}/s_{557} is replaced by the less noise-sensitive quotient s_{R457}/s_{FMYC} .

Partial Least Squares Analysis

The PLS analysis shows that a linear model can be considered sufficiently accurate to model the dependence between paper properties and spectral light scattering for the samples used in this study. Including non-linear terms does not increase the coefficient of determination, R^2 , for the paper property intervals used in this work. The influence of grammage on the mean value of s is insignificant. Therefore, mean(s) can be modeled with density and filler content only, with two principal components. The ratio s_{R457}/s_{FMYC} is best modeled with all three paper properties and three components.

The absorption coefficient, k , could be modeled with a R^2 of 54% as best, which is considered to be too low to be separated from noise.

Fig 1. Mean value of the scattering coefficient, s , taken over wavelengths 400 nm to 700 nm for a subset of the uncalendered samples (•) and for the same set of samples calendered at 32 kN/m (×). The calendered samples have approximately the same values of mean(s) as the uncalendered samples.

A PLS model that is based on two components with linear relationships between density, filler content, and mean(s), has a coefficient of determination, R^2 , of 94%, and a prediction error, $RMSEP$, of 0.84.

A PLS model that is based on three components with linear relationships between density, grammage, filler content, and the ratio s_{R457}/s_{FMYC} has a coefficient of determination, R^2 , of 94%, and a prediction error, $RMSEP$, of 0.0017. The values s_{mean} and the ratio s_{R457}/s_{FMYC} can be calculated as:

$$s_{mean} = 1.34 \times f - 0.0325 \times \rho + 51.4 \quad [3]$$

$$\frac{s_{R457}}{s_{FMYC}} = 7.37 \cdot 10^{-4} \times f - 2.39 \cdot 10^{-5} \times w + 1.47 \cdot 10^{-4} \times \rho + 0.972 \quad [4]$$

where f is filler content in percent, w is grammage in g/m², and ρ is density in kg/m³. When the samples are calendered, the empirical functions changes slightly. The values s_{mean} and the ratio s_{R457}/s_{FMYC} are then calculated as:

$$s_{mean} = 1.28 \times f - 0.0462 \times \rho + 68.1 \quad [5]$$

$$\frac{s_{R457}}{s_{FMYC}} = 13.9 \cdot 10^{-4} \times f - 1.68 \cdot 10^{-5} \times w + 1.66 \cdot 10^{-4} \times \rho + 0.927 \quad [6]$$

The predictability of the PLS models is illustrated in Fig 3. The observed values in the prediction set are plotted against values predicted by the model. A good agreement between observed and predicted values shows that the model has a good predictability.

The effect the paper properties have on the s -value is illustrated in Fig 4. The measured sample has the density 725 kg/m³, the grammage 86 g/m², and 25% filler content. The figure shows the simulated effect on the spectral light scattering coefficient, s , when the filler content is increased or decreased with 4.5%, and the density is increased or decreased with 200 kg/m³ from the values of the measured sample. The light scattering coefficient, s , increases with increasing filler content and decreases with increasing density. The ratio s_{R457}/s_{FMYC} increases both when density is increased and when filler content is increased in the paper.

(a)

(b)

Fig 2. PCA loadings of the first and second principal components (p_2 vs. p_1) of the data set, including grammage (Gr), density (De), filler (Fi), together with spectral light absorption coefficient, k (a), with 91% of the variations explained in the first component, and spectral light scattering coefficient, s (b), with 93% of the variations explained in the first component. Values from every 10th nm are used in the analysis but for better readability of the graphs only values from every 80th nm are shown in the graphs.

Discussion

The linear dependence between the paper properties and the light scattering coefficient that has been shown in this study enables a reduction of the sample set if the same type of paper is to be evaluated with another measurement method.

For another paper grade, with a different pulp composition, a new set of samples probably needs to be produced and measured in order to accurately find the relations between paper properties and optical response. Since the light scattering coefficient, s , is dependent on the paper structure, the calendered and uncalendered sets show that the wet-pressing gives a more significant difference in the structure of the paper than the dry calendering.

(a)

(b)

Fig 3. Observed vs. predicted values of s_{mean} , (a), and of the ratio s_{R457}/s_{FMYC} (b), for samples in the prediction set. Both the results from the PLS model based on uncalendered samples (•) and calendered samples (x) are shown. The closer the data lies to the straight line shown in the figures, the better the agreement between observed and predicted values.

The light absorption coefficient, k , was found to vary very little. This may be due to the fact that the only differences between the paper samples were the amount of filler content and the structural properties, which mostly affect the light scattering in the paper.

The results in this study indicate the possibility to model the optical variations and perceived appearance of the paper from high resolution measurements of paper properties. To successfully develop such a method, further studies need to be made where the influence of measurement scale is quantified. Other possible extensions of this work are to include dye or fluorescent whitening agents, multiple layer structures, coated paper, a larger wavelength span, and other measurement geometries.

Fig 4. The simulated effect of the spectral s when density or filler content is increased. The center point is a sample with the density 725 kg/m^3 , grammage 86 g/m^2 , and 25% filler content. The dashed lines are responses calculated from the derived model for uncalendered paper when the filler content is increased or decreased with 4.5% and the density is increased or decreased with 200 kg/m^3 .

Conclusion

In this work, the simultaneous inter-dependence between density, grammage and filler content and the spectral optical response has been characterized with paper samples that represent the small-scale variations of paper properties that occur within a commercial 80 g/m^2 copy paper. Possible non-linearities in the relations between paper properties and optical response have been evaluated. The results show that the dependence between paper properties and spectral optical response can be represented by a linear model and that the optical response can be predicted from paper properties with good accuracy for the paper samples used in this study.

The models derived in this work are planned to be used together with high resolution measurements of paper properties and spectral optical response to characterize how the measurement scale influences the difference between the measured optical response and the optical response predicted from the model.

Acknowledgements

The project is financially supported by the European Structural funds, which are gratefully acknowledged. The authors also wish to acknowledge Henry Westin at Metsä Board Sweden AB, Husum and Jan-Erik Häggglund at MoRe Research AB for their valuable comments and experimental support.

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