A novel approach to quantify spatial coating-layer formation

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ABSTRACT: A straightforward correlation of coating thickness and base sheet surface height (topography) data does not clearly show the expected differences between a contour coat-like structure for curtain coating or a leveling coat-like structure for blade coating. We defined a measure to clearly distinguish between these different kinds of spatial coating-layer formation. Filtering in the frequency domain preserves only the large structures in the coating-layer thickness and the base sheet topography data. The coefficient of determination (R^2) and the slope of a linear regression model, between filtered coating thickness and base sheet surface height (topography), are the measures used to describe the leveling behavior of a coating layer. Three application systems—blade, film press, and curtain coating—were compared. Analysis of the original data showed only minor differences in the coating-layer formation between the application systems. For the filtered data where large structures are preserved, curtain coating gave an almost ideal contour coat, where coating thickness was hardly influenced by base sheet topography. The coating thickness in blade coating strongly depended on local base sheet surface height variations. A film press application of the coating layer resulted in a coating structure between curtain and blade coating.

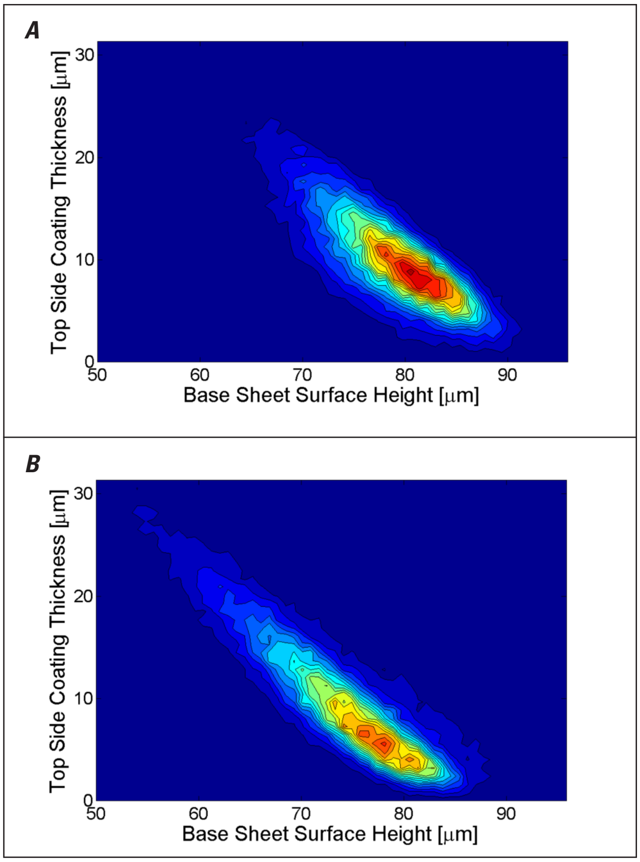
Application: Mills can better describe the differences caused by various coating parameters, such as coating application systems or variations in the pigment systems.

Targets in coating paper are two-fold and to some degree conflicting. On the one hand, a smooth surface has to be achieved and on the other hand, a uniform coating thickness has to be applied. Smooth surface and uniform coating thickness are both important for optical appearance and good printability [1-3].

Coating layer uniformity is influenced by base paper topography. A rough base paper surface will result in a wide range of coating thicknesses, whereas a smooth paper surface will give uniform coating mass distribution [4]. The application system used is also important in controlling coating-layer uniformity [5]. Remarkable coating thickness variations using blade coating would be expected, because irregularities in the base paper surface are leveled out during metering. This type of spatial coating-layer formation is the leveling coat, where a strong correlation between coating thickness and base sheet topography exists. Curtain coating would be expected to give a uniform coating thickness because there is no metering needed to remove excess coating color and this contour coat is barely influenced by base sheet topography.

Several methods are capable of quantifying the effect of base sheet topography on coating-layer uniformity (e.g., scanning electron microscopy [6, 7]; X-ray microtomography [8]; and serial sectioning combined with optical light microscopy [9]). All methods have basically the same measuring principle: local coating thicknesses and base sheet topography data are extracted from a digital representation of the paper sample to be analyzed. These methods differ only in their resolution and sample size to be investigated; the results obtained show similar trends [10].

Figure 1 gives an exemplary result that shows two typical local correlations between unfiltered coating thickness and



1. Local correlation of coating thickness data and base sheet surface height for curtain coater (A) and blade coater (B). The same type of base paper was coated with carbonate-containing coating color for both cases.

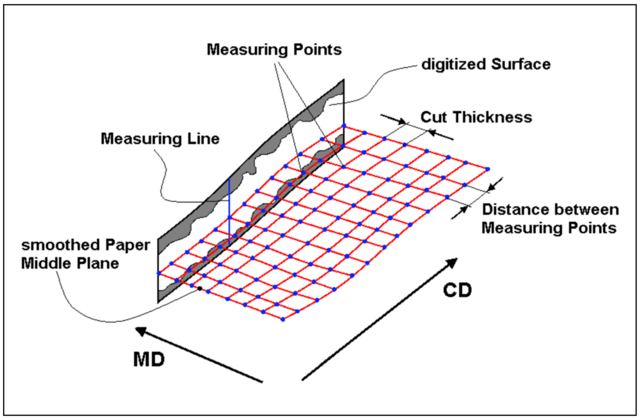
base sheet surface height data for a curtain coater and for a blade coater, coated on the same base paper. Data were obtained with a coating-layer analysis tool based on serial sectioning. Base sheet topography is expressed as height from a paper middle plane. Red areas indicate a high number of measurement values, blue areas a low number. Both application systems resulted in a similar slope of the regression line between coating thickness and base sheet surface height. Against the expectation for a contour coating process, the coating thickness applied with a curtain coater also showed a strong dependency on base sheet topography. There was only one difference between the two coating aggregates: for the curtain coater, the same coating thickness values were observed over a wider range of base sheet heights, giving a somewhat wider distribution. Better coverage of a curtain coater compared with a blade coater was evident in Fig. 1, but the influence of the application system on spatial coating-layer formation (contour or leveling coat) cannot be observed in a straightforward way from the plots. This pointed to the need for a measure to clearly evaluate spatial formation of coating layers.

Dahlström et al. [11, 12] used frequency analysis to study the effect of different length scales of the base sheet's surface height variations on local thicknesses of a coating layer applied with a laboratory coater that was equipped with a blade metering system. Coating thickness variations (i.e., coating-layer uniformity) were found to be influenced by two mechanisms.

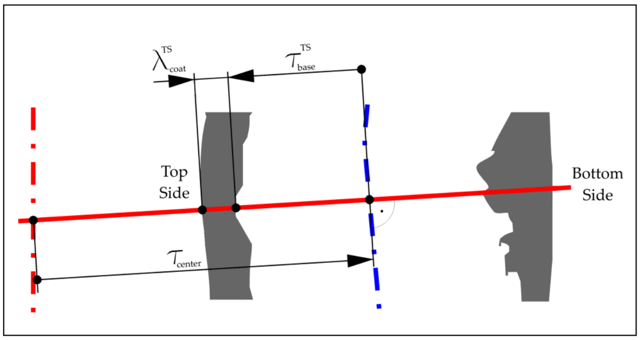
- At shorter wavelength or smaller structures, the coating layer behaves like a leveling coat structure where small voids at the paper surface are filled with coating color, creating a small-scale uniform surface.
- At larger wavelength or larger structures, the coating-layer thickness is not that strongly influenced by base sheet surface height. Random deposition of coating color is assumed to be a dominating effect at larger length scales of base sheet surface height, resulting in a coating layer with a contour coat-like structure.

The method we described uses a similar approach and also postulates that small structures in the base sheet surface (e.g., fibers) are filled with coating color independent of application system. Larger-scale structures in the base sheet surface (e.g., flocs) are the reason for typical coating-layer structures, as they are a leveling coat for blade coating and contour coat for the curtain coater. Filtering of coating thickness data and base sheet surface height data can be used to distinguish between void-filling effects at small-scale base sheet surface structures and contour coat or leveling coat-like behavior at larger scales.

Our analysis was based on coating thickness maps and base sheet surface height maps obtained with a serial sectioning technique, µSTRUCSCOP, developed at Graz University of Technology. These maps were filtered in a frequency domain, where a predefined cutoff wavelength was used to obtain small structures (highpass-filtering) and large structures (lowpass-filtering). The filtered data were then used for correlations of coating thickness data and base sheet surface height



2. Definition of a smoothed paper middle plane with a measuring grid for the extraction of thickness and topography data along measuring lines.



3. Definition of coating thickness and base sheet topography data along a measuring line. The measures are explained in Table I.

data. Coefficient of determination (R^2) and slope of a linear regression model were calculated and used as measures for spatial formation of a coating layer. The applicability of this approach was demonstrated on samples from a pilot coating trial, where identical coating colors were applied to the same base paper using blade, film press, and curtain coaters.

METHOD

Measurement of coating thickness and base sheet topography

The digitization concept and coating-layer segmentation are explained elsewhere [13,14]. A brief summary of the measuring principle follows.

The sequence of binary images obtained with coating-layer segmentation from paper cross sections was used to calculate a smoothed paper middle plane where a virtual measuring grid was drawn. This grid was defined by the slice (cut) thickness in the paper machine direction (MD) and a distance in the digitized paper cross section (paper cross direction [CD]) that is usually adjusted to the cut thickness, as **Fig. 2** illustrates. At the nodes of this measuring grid, a measuring line was placed perpendicular to the paper middle plane. Along this line, the local coating thickness, the local base sheet surface height and a location parameter were extracted, as **Fig. 3** shows.

		Blade Coating	Film Coating	Curtain Coating
λ^{TS}_{coat}	Top-side coating thickness	Leveling coat, thickness variations	←in between→	Contour coating, uniform thickness
τ^{TS}_{base}	Base sheet topography	Reduced roughness due to compression	←in between→	No roughness changes
τ_{center}	Paper curvature	–	–	–

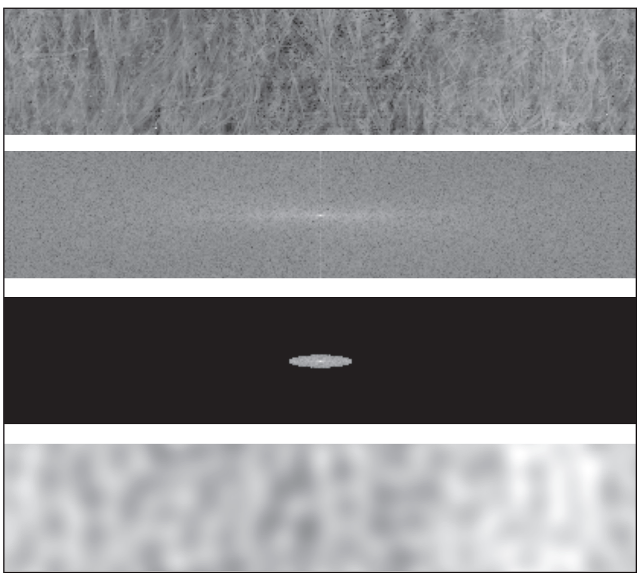
I. Definition of distance values extracted from coating segmentation results, with expected effects caused by different coating aggregates on these values.

Table I lists measures used in this paper and expected effects caused by different coating aggregates.

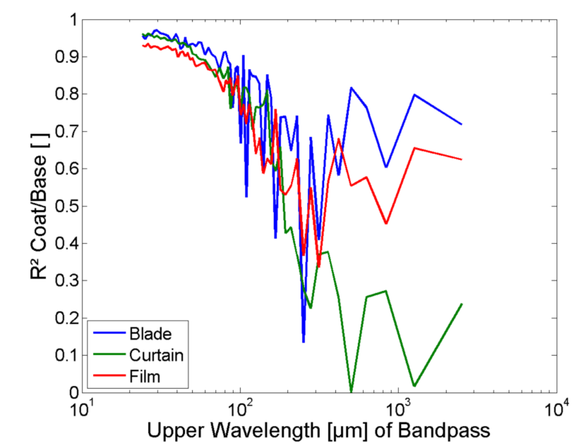
The data sets used for analysis covered an area of 5 mm² (5 mm CD x 1 mm MD). The slice thickness (i.e., the distance between the measuring lines) was 12 µm, giving 35.028 data points per digitized area (84 paper cross sections and 417 measuring lines per paper cross section). The resolution in the paper thickness direction was 0.4 µm per pixel. A minimum of three samples covering the area had to be analyzed to obtain statistically significant data.

Filtering in the frequency domain

Filtering in the frequency domain was performed in MATLAB by the use of standard operations [15]. **Figure 4** shows the main processing steps necessary: creating an original map of data; obtaining a corresponding frequency domain image after 2D-discrete Fourier transformation and the shift of the transformation's zero-frequency components to the center of the spectrum; applying a filter to the frequency domain image; and obtaining the retransformed map of data.

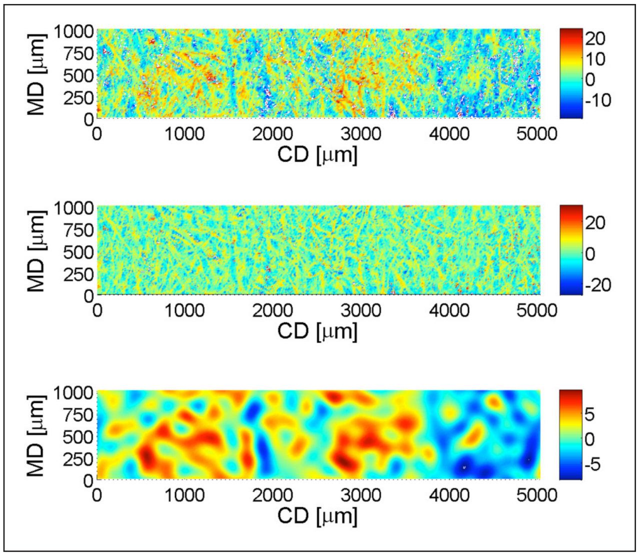


4. Basic operations of filtering in the frequency domain. This example shows a base sheet surface heights map. From top to bottom: original map of base sheet topography (surface height); frequency domain image after Fourier transformation and the shift of zero-frequency components to the center of the spectrum; lowpass-filtering (all frequencies above a cutoff frequency are set to zero); and reconstructed image.

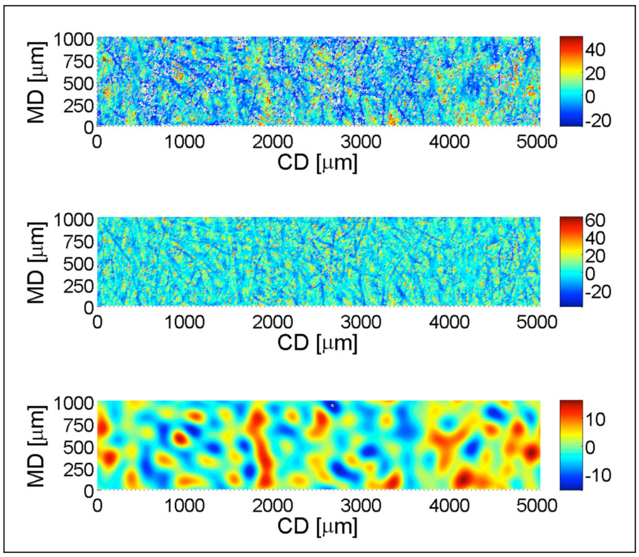


5. The coefficient of determination (R^2) of a linear regression model considering coating thickness and base sheet surface height as a function of the upper wavelength used in bandpass-filtering. At shorter wavelengths, a high R^2 can be observed for all coating aggregates. If larger structures, having a wavelength above 250–300 µm, are analyzed, then differences between the coating aggregates can be observed.

The cutoff wavelength used to differentiate between the void-filling effect of small base sheet structures and the coating-layer structure at larger scales was determined by calculating coefficient of determination (R^2) for a linear regression model of bandpass-filtered coating thickness and bandpass-filtered base sheet surface height. **Figure 5** shows R^2 as a function of the upper wavelength of the bandpass-filter kernels used; the lower wavelength was in all cases set to the upper wavelength for the filter kernel of the previously used smaller-filter kernel. At shorter wavelengths, the high R^2 values for all coating aggregates analyzed indicated a void-filling effect that occurred with all coating application systems. Above a wavelength range of 250–300 µm, calculated R^2 values between coating thickness and base sheet surface height began to differ for the coating aggregates. Blade coating still resulted in a high R^2 of about 0.5–0.7, while curtain coating gave R^2 values of 0–0.3. For all subsequent evaluations, the differentiation between small-scale and large-scale structures occurred at cutoff wavelength of 275 µm.



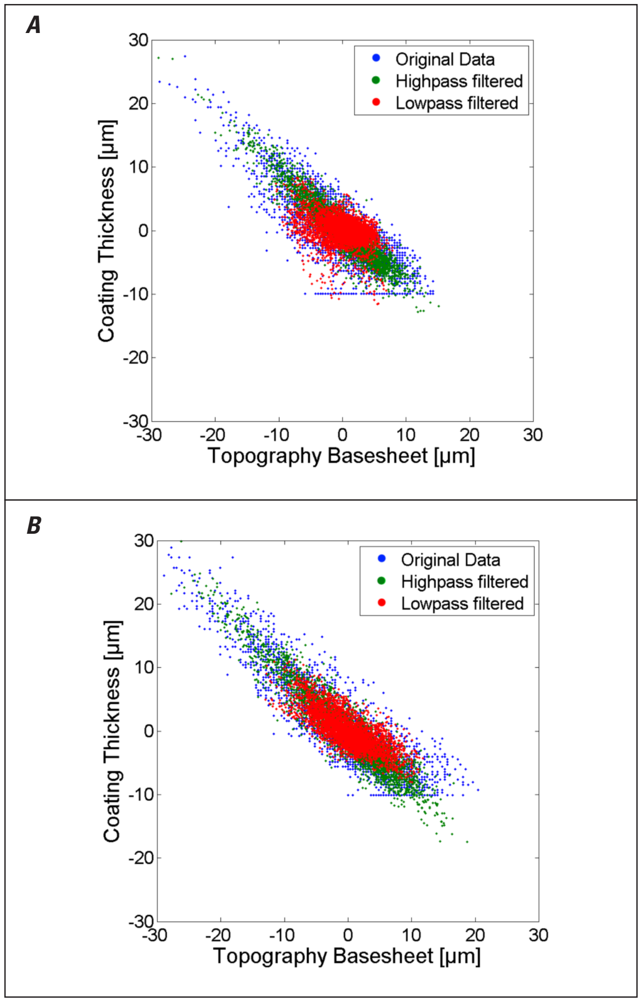
6. Base sheet height maps of a blade-coated paper. From top to bottom: original data map; remaining structures after highpass-filtering; and remaining structures after lowpass-filtering.



7. Coating thickness maps of a blade-coated paper. Top to bottom: original data map; remaining structures after highpass-filtering; and remaining structures after lowpass-filtering.

Figures 6 and **7** show typical maps of base sheet surface height data and coating thicknesses for a blade coated sample. The mean value was subtracted for visualization purposes in each case. Application of the filtering steps at a predefined cutoff-wavelength gave the following results:

- **Highpass-filtering to preserve only small structures below a defined cutoff-wavelength** – The shapes of individual fibers were the dominating structures in these images.
- **Lowpass-filtering to preserve only large structures** – Large structures, such as an area with higher coating thickness in the CD region between 4,000 and 5,000 µm, were preserved in the reconstructed map. In this case, a lower region in the base paper surface corresponded with a thicker coating layer, as was expected for blade coating.



8. Effect of highpass- and lowpass-filtering on correlations between coating thickness and base sheet surface height for curtain coater (A) and blade coater (B). Calculations were based on the same datasets presented in Fig. 1.

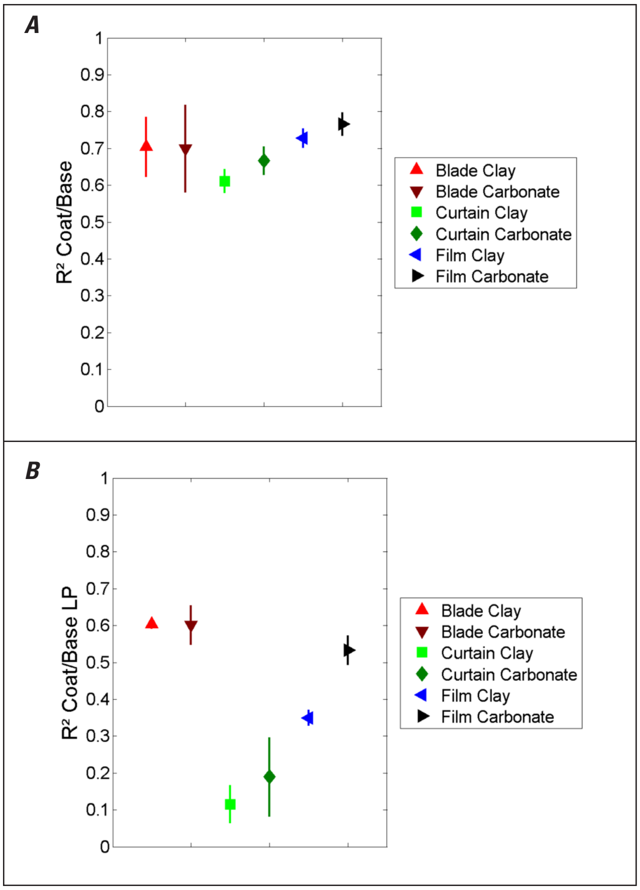
Figure 8 shows the effect of frequency filtering on the correlation of coating thickness versus base sheet surface height (topography) for the samples presented in Fig. 1 from a curtain coater and a blade coater. For visualization purposes, the mean values of coating thickness and base sheet surface height were subtracted from the original data. Blue dots in Fig. 8 correspond to the original input data. The same coating thicknesses were found over a wide range of base sheet surface heights for the curtain coater. The highpass-filtered data are indicated by green dots. Both curtain and blade coating showed a strong correlation between coating thickness and base sheet surface height. In agreement with recently published data by Dahlström et al. [11, 12], we interpreted this behavior as void filling or leveling of small base sheet struc-

Measure	Original Data	Highpass-Filtering	Lowpass-Filtering
R^2	0.6851	0.8564	0.1680
Slope	-0.6957	-0.8408	-0.2640

II. Measures of a (typical) curtain coating structure.

Measure	Original Data	Highpass-Filtering	Lowpass-Filtering
R^2	0.8050	0.8754	0.5802
Slope	-0.7909	-0.8646	-0.5558

III. Measures of a (typical) blade coating structure.



9. Influence of coating method and pigment type on the coefficient of determination (R^2) between coating thickness and base sheet surface height, for the original data (A) and for the lowpass-filtered data (B).

tures with coating color. The red dots indicate correlations of lowpass-filtered coating thickness and base sheet surface height data. Differences between curtain and blade coating were obvious. In curtain coating, base sheet surface height was a weaker influence on coating thickness, whereas the influence was more pronounced for blade coating. These findings are supported by calculated measures of R^2 and the slope of the regression line between coating thickness and base sheet surface height. **Tables II** and **III** present these measures for the curtain coater and the blade coater, respectively.

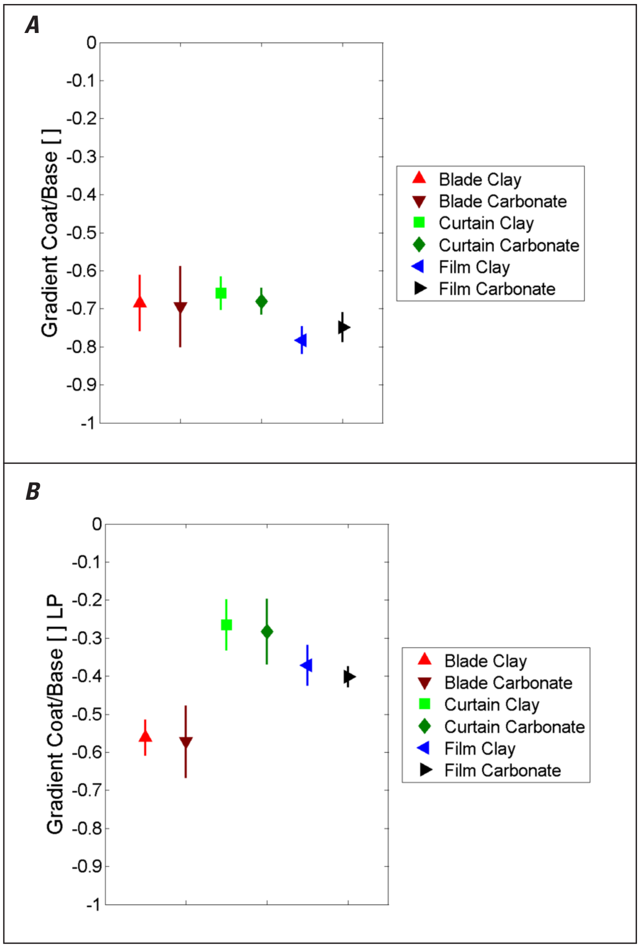
On the basis of these findings, we proposed to use R^2 values and the gradient of the correlations between lowpass-filtered coating thickness and base sheet surface height as measures to define spatial formation of a coating layer in a simple and straightforward way. Small structures, as obtained in highpass-filtering, do not show clear differences in spatial coating-layer formation between different application systems.

VALIDATION

To validate our approach, a woodfree base paper was coated at the Center of Excellence for Paper and Board Coating of Dow Europe GmbH (Horgen, Switzerland) with blade, film press, and slide curtain application systems. Two nearly identical coating colors—the first containing 100% carbonate and the second containing 100% clay—were used. The same coat weight was applied in each case. Detailed trial data, coating thickness distributions, particle orientation, mercury porosimetry analysis, and a detailed look at the coating layers with focus ion beam (FIB) cross section imaging were given by Salminen et al. [16]. Curtain coating resulted in a more uniform coating thickness distribution, indicating a contour coat-like formation. A higher porosity of the coating layer was obtained when clay was used. Blade coating gave a denser coating layer and high surface smoothness. The wider coating thickness distribution indicated a stronger leveling tendency of this application system. Spatial formation of a coating layer obtained in film coating was between that of curtain and blade coating.

A minimum of three sample areas covering 5 mm² (5 mm CD x 1 mm MD) were analyzed per paper sample with µSTRUCSCOP serial-sectioning method. Coating thickness data and base sheet surface height data were extracted at the nodes of a measuring grid in the CD–MD plane with a distance of 12 µm between nodes, giving about 35,000 measuring points per sample area. The resolution along the paper thickness was 0.4 µm per pixel. Coefficient of determination (R^2) and slope of the linear regression model, considering coating thickness and base sheet surface height data, were then calculated for each individual sample area for the original data and the lowpass-filtered data.

Figure 9 gives R^2 values for correlations between coating thickness and base sheet surface heights, for the original data and for the lowpass-filtered data with a 90% confidence interval. R^2 values for original data are located within a small range between 0.6 and 0.8. There is a significant difference between curtain and film-press coating, but blade coating cannot be distinguished from the others because of higher variability in the data expressed by the larger confidence intervals. Lowpass-filtering widened the range of R^2 values, between 0.1 for curtain coating and 0.6 for blade coating. Weak correlation in curtain coating is an indicator of a contour coat-like behavior,



10. Influence of coating method and pigment type on the slope of the regression line between coating thickness and base sheet surface height, for the original data (A) and for the lowpass-filtered data (B).

which was slightly more pronounced for clay coatings than for carbonate coatings. High R^2 values in blade coating are an indicator of a leveling coat-like behavior, as expected for blade coatings. Different pigments did not show an influence on the coating structure. Values for coating applied with the film press were between those of a curtain coater and a blade coater. As with curtain coating, clay-containing coating color gave a more contour coat-like structure than did carbonate-containing color.

Similar conclusions can be drawn for the slope of the regression line between coating thickness and base sheet surface height, which are shown in **Fig. 10** for the original data and for the lowpass-filtered data, again with a 90% confidence interval. The range of gradients obtained in the original data is even narrower than the R^2 value discussed before. After lowpass-filtering, correlations of the remaining structures of coating thicknesses and base sheet surface heights gave absolute gradients of about 0.6 for blade coatings, 0.2–0.3 for curtain coatings, and 0.4 for film-press coatings. The lower the gradient (the closer to zero), the weaker the correlation was between coating thickness and base sheet surface height. A distinction between coatings containing clay or carbonate is not possible in this case.

CONCLUSIONS

Concentrating the analysis of coating layers to larger structures (above ~250 µm) increases the informative value of the correlation between coating thickness and base sheet surface height. Small structures (e.g., voids between fibers) are filled with coating color independent from the coating application system. Larger structures (e.g., flocs) influence the relationship between coating thickness and base sheet surface height and define the final coating-layer structure.

Further work will concentrate on including additional information such as the variance of coating thickness distribution as a measure of coating-layer uniformity. Base sheet topography data will also be more closely analyzed with the aim of investigating structural changes induced by the application system. A more detailed analysis of differences in coating-layer structures obtained from different application systems, changes in coating-color compositions, or calendering will thus be possible. **TJ**

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The analysis of spatial coating-layer formation is an important field of research for several reasons at the Institute for Paper, Pulp and Fiber Technology at Graz University of Technology. In the current work, we dealt with the analysis of coating-layer properties such as coating-layer uniformity, coating-layer formation, coating holdout, and coating coverage. The analysis of these properties is usually limited to a small sample size, which limits the informative value of the results. We applied an automated serial sectioning technique that gave access to a large amount of data within a reasonable time and at a sufficient degree of detail.

Three-dimensional analysis of paper requires detailed knowledge of different disciplines such as microscopy, image analysis, statistics, and mathematics. For this work, we needed to have knowledge of Fourier transformation and filtering in the frequency domain.

The result most surprising to us was the small difference between curtain-coated and blade-coated (pilot) samples when untreated data were considered. The degree of clarity in the void-filling effect for the different application systems at small structures and the formation of coating-layer structures at larger length scales were unexpected.

This work can be helpful to mills because it allows for a better description of the differences caused by various coating parameters such as coating application systems or variations in the pigment systems. With novel measuring techniques, problems that have been hard to tackle in the past can be looked at in different ways and can be solved.

The method we described will be used to analyze other effects such as calendering on the spatial coating-layer formation, and changes in the small structures in the fiber-width scale will be analyzed in more detail.

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