QUANTIFICATION OF THE PENETRATION OF COATING PIGMENTS INTO THE BASE PAPER DETERMINED BY AUTOMATED SERIAL SECTIONING

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ABSTRACT

The penetration of coating pigments into the base paper is in clear contradiction to the goal of pigment coating which is to cover the base paper fibers. To determine its relevance and to obtain a better understanding of the influencing factors a proper characterization of the coating solids fraction penetrated the base paper is necessary. A high degree of penetration of coating material is equivalent to a poor coating holdout.

The presented approach uses three dimensional datasets represented as paper cross section images obtained with an automated serial sectioning technique. Image analysis is used to detect all regions in the paper cross section images that have similar color information as the coating layer. We distinguish between three types of coating: the coating on the paper surface, coating regions connected to the surface coating layer but hidden below some fibers (hidden coating) and discrete regions in the base paper with no connection to the coating surface layer (trapped coating). Two measures are calculated based on these types of regions: (1) an upper limit of the fraction of penetrated coating solids into the base paper by considering all regions detected as coating, including also fillers and (2) a lower limit of a fraction of penetrated coating solids by considering only those regions which are visibly connected to the coating on the paper surface. The higher the calculated fractions, the more coating color has penetrated into base sheet.

The applicability of this approach is demonstrated by analyzing the influence of base paper porosity and pigment variations (clay content) on coating penetration as a measure for coating holdout in a lab coating trial. A minimum of 10 – 20% of the total detected coating was found below the surface fibers (hidden coating) and was classified as penetrated coating in the base paper. A higher fraction of penetrated coating solids was found for the paper regions with the more open sheet structure. The clay containing coating color gave a somewhat better holdout than an only carbonate color.

KEYWORDS

COATING HOLDOUT; COATING PENETRATION; THREE DIMENSIONAL; BASE PAPER POROSITY; PIGMENT

APPLICATION STATEMENT

Penetration of coating pigments into the base paper is a matter of interest in paper coating since it is the target of coating to cover the base paper surface. Excessive coating pigment penetration impairs the surface properties of the coated papers. There are a number of approaches to reduce penetration of coating into the base sheet like e.g. reduction of base paper permeability, internal or surface sizing or changing the pigment system in the coating formulation. In order to assess the effect of these approaches a quantification of the amount of coating pigment penetrated into the base paper is desired.

INTRODUCTION

Coating holdout is generally defined as the base paper's resistance to coating solids penetration [1]. This property is important because coating pigments penetrated into the base paper do not contribute to the goal of surface treatment like covering surface fibers [2] or the contribution to the final gloss [3]. Also expensive brightening and hiding pigments penetrated into the base paper do not perform the desired function to the full extent [4,5].

Coating solids penetrate beneath some of the top fibers of the base paper, but normally not deeper than 15 to 20 µm, or the thickness of about two fibers [1]. The pore size was found to be a key issue in coating penetration. These have to be small enough not to be entered by solid pigments [1]. Coating solids penetrated totally into big pores of model substrates (larger than 3 µm) but not into pores having a width below 1.2 µm [6]. Alteration of the pore size by enhanced fiber swelling [7] or the application of a pre-coat to plug the pores and valleys of the base paper [8] prevents extensive coating penetration. Also an increased filler content to reduce pore size was found to influence coating penetration [2]. The addition of water retention agents and flow modifiers to the coating color or sizing of the base paper are used to control coating penetration into the base paper [4]. The coating application system influences coating penetration too, a high pressure in the nip at metered size press coating was found to facilitate coating penetration [9].

Also totally opposing views on this topic were discussed in the literature stating that the fraction of coating color penetrating into the base paper is not a problem at all [10,11].

A proper characterization of the coating solids fraction inside the base paper structure is necessary in order to decide whether coating holdout is a relevant problem or not. But there is no method available up to now which provides reliable measures:

- Coating holdout is usually inferred from properties of coated sheets, like oil absorption [1,3], brightness gain [4] or roughness and porosity [12].

- The reflectance of burnouts is used to evaluate coating holdout [1]. The idea is an enhanced contribution to brightness if coating remains on the paper surface. This approach however implies that coating holdout and coating layer uniformity are not independent from each other.

- Another way to evaluate coating holdout is the analysis of the exposed coating backside with roughness measurements or in more detail with electron microscopy. To expose the coating backside, the fibrous material has to be removed either by dissolving in a solution with cupri-ethylen-diamin (CED) [7,8,12] or low temperature ashing techniques [4].

- Confocal laser scanning microscopy (CLSM) is also used to estimate coating holdout [5]. Base paper and coating binders are stained with different fluorescent dyes. Intermixed regions in the obtained images indicate areas where coating (binders) penetrated into the base paper.

- Also cross section images of the paper samples are used to analyze coating holdout [1,2,6]. Lloyd et al. [13] presented an image analysis tool to quantify coating holdout from cross section images. They divided regions indicated as coating into three types: trapped coating (discrete regions in the base paper), hidden coating (visibly connected to a coating layer but separated by a layer of fibers) and the coating on the paper surface. Relationships between these three types are calculated and used as a measure for coating penetration and coated paper quality.

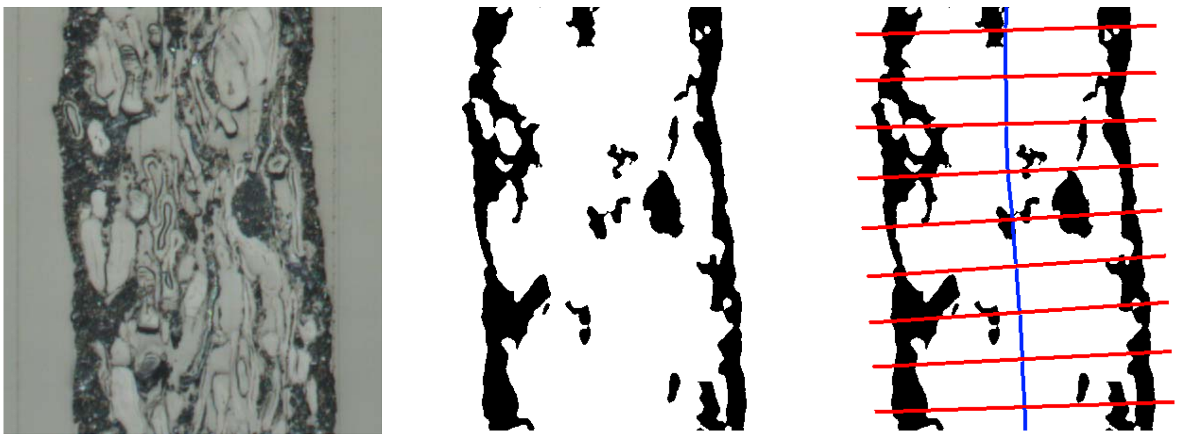
This paper introduces automated serial sectioning combined with image analysis as a tool to quantify coating penetration into the base paper in a representative way. A laboratory scale coating trial was carried out in order to evaluate the relevance of coating penetration and to analyze two of the possible influencing parameters: base paper porosity and pigment system in the coating color.

METHODS AND MATERIALS

Automated serial sectioning and detection of coating

The approach and analyses to be presented in this paper are based on paper cross section images obtained with an automated serial sectioning technique (μSTRUCSCOP) developed at Graz University of Technology. The digitization concept and coating layer segmentation are explained elsewhere [14,15]. A brief summary of the digitization parameters used in the analysis of coating holdout as well as an explanation of the segmentation results are given in the following:

A sequence of subsequent paper cross section images was obtained with automated serial sectioning. A cutout of a single paper cross section is shown in Figure 1 (a). The pixel size in these images is 0.4 μm; the cut thickness or the distance between the subsequent cross section images is set to 4 μm. From these cross section images, all regions having similar color information as the coating layer are detected. This results in a sequence of binary images where the coating layer is highlighted; a cutout is shown in Figure 1 (b). The following extraction of coating thickness data is performed along equidistant (~4 μm) measuring lines – red lines in Figure 1 (c) – which are placed perpendicular to a paper center line – blue line in Figure 1 (c).



(a) Cross section image. **(b) Detected coating layer.** **(c) Detected coating layer with measuring lines.**

Figure 1: Detection of coating layers in cross section images.

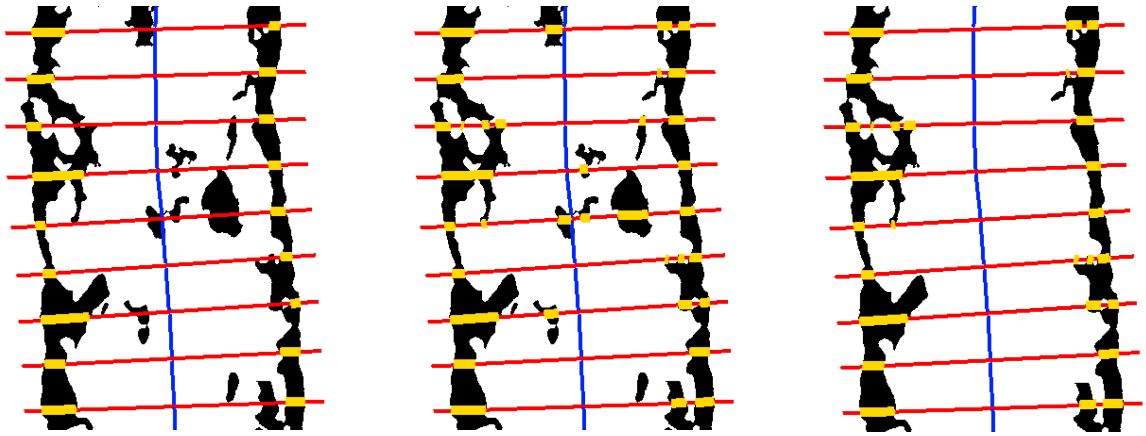
Definition and measurement of coating thicknesses

Similar to the approach presented by Lloyd et al. [13], three types of coatings were defined and their thickness measured from the binary images highlighting the regions detected as coating; see Figure 2. The yellow line segments indicate the individual types of coating:

- Thickness of regions detected as coating on the paper surface (C1).
  In a first step only the coating on the paper surface is considered. The coating thickness is determined by counting pixels. Counting starts at the outermost coating pixel on a measuring line (paper surface) and moves towards the paper center line. The distance to the first interruption is counted; see Figure 2(a). This is the standard procedure to measure coating layer thickness which e.g. was applied to quantify different other coating layer properties in the past [16].

- Thickness of all regions detected as coating (C2).
  In a second step all regions detected as coating are considered. The coating thickness results from a summation of the individual thicknesses (obtained by counting pixels) of all regions detected as coating along a measuring line – from the paper surface to the paper center line; see Figure 2(b). According to Lloyd et al. [13], this measure includes the coating on the paper surface, hidden coating and trapped coating. For papers containing fillers also filler agglomerates detected as coating regions can be included.

- Thickness of all coating regions connected to the paper surface (C3).
  The last step is used to measure the thickness of all regions detected as coating on the paper surface. This coating thickness is obtained in a way similar to the measurement of C2, but separated regions detected as coating in the base paper are removed before counting; see Figure 2(c). In accordance to Lloyd et al. [13], the coating layer on the paper surface and hidden coating are included.



(a) Data obtained at standard coating thickness extraction (C1). **(b) All regions detected as coating are considered (C2).** **(c) All regions connected to the coating on the paper surface are considered (C3).**

Figure 2: Definition of the coating thicknesses used to measure the fraction of penetrated coating. The yellow sections of the measuring lines denote the extracted coating thickness values.

Quantification of coating solids penetration

The three coating thicknesses – coating thickness on the paper surface (C1), thickness of all regions denoted as coating (C2) and the thickness of all regions linked to the paper surface (C3) – are used to define measures to quantify the coating penetration. At this stage of analysis, the fraction of coating penetrated into the base sheet is defined as a measure for the coating holdout: no penetration – good holdout.

In this first approach, the mean values of the measured coating thicknesses are used and two relations are calculated:

- All regions detected as coating below the paper surface (hidden and trapped coating) are defined as penetrated coating. This includes also incorrect detected regions in image segmentation like fillers in filler containing papers since a distinction between penetrated coating and filler agglomerates is often not possible. The coating in the paper is given as a fraction of the total detected coating – C2; see Figure 2(b):

$$Upper\ Limit = \frac{C2 - C1}{C2}$$

  The fraction obtained therefore indicates the upper level of penetrated coating solids into the base paper.

- From the paper surface separated but visibly connected regions (hindered coating) are defined as penetrated coating. The coating hidden in the base paper is given as a fraction of total detected coating connected to the paper surface – C3; see Figure 2(c):

$$Lower\ Limit = \frac{C3 - C1}{C3}$$

  The fraction obtained thus indicates the lowest level of penetrated coating solids into the base paper.

In general, the higher the calculated value, the more coating color has penetrated into the base sheet. The true fraction of penetrated coating color is somewhere between the two measures. A closer quantification of the true fraction is difficult because this requires knowledge of e.g. the amount of detected fillers considered as trapped coating (C2). In the following, these measures will be used to analyze the penetration behavior of coatings into the base paper.

Experimental

The influence of base paper porosity on the penetration behavior of coating solids was analyzed. Two different coating colors were applied with a laboratory web coater on one side of an only eucalypt pulp containing base paper which was produced on a pilot paper machine. The coating formulations are summarized in Table 1, Figure 3 shows the pigment particle size curves of the pigments used in the coating formulations. The change in the pigment system is to be considered a relatively minor change, since due to its small particle size and rather low aspect ratio Hydragloss 90 cannot be considered as the clay type yielding best possible coating holdout. The reason for this minor change in pigment system was to test the sensitivity of the quantification method.

		Coating I	Coating II
Hydrocarb 90 – ME (78%)	78%	100	60
Hydragloss 90 – EM (73%)	73%		40
PVA (BF-05, 6-98)	25%	0.4	
CMC (Finnfix 10)	12%	0.5	
Latex (Styronal D 628)	50%	11.0	
OBA (Leucophor VM fl)	100%	0.5	
Solids content		71.4%	68.7%
Viscosity (BV 100)		2050 mPa s	2150 mPa s
Coat weight		14.2 g/m²	14.3 g/m²

Table 1: Coating formulations as used to analyze coating penetration into base sheet structures.

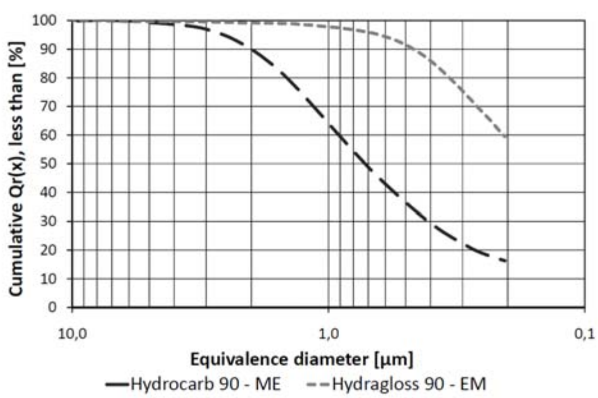
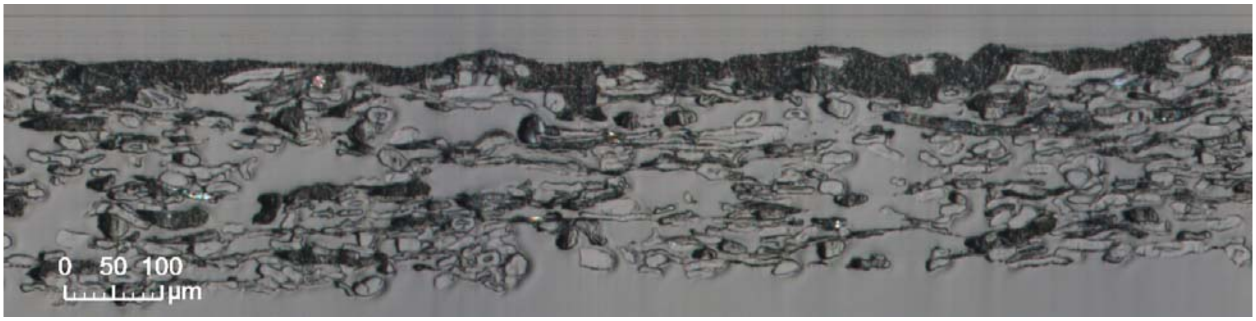


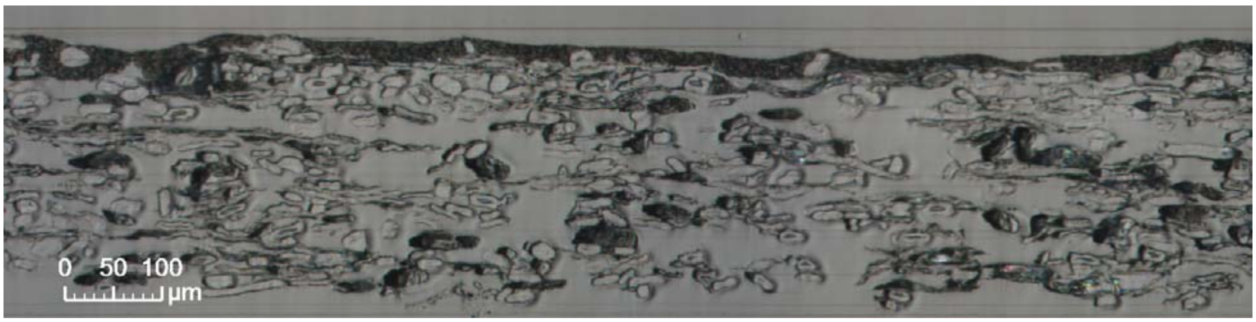
Figure 3: Pigment particle size curves of the pigments used in the coating formulations.

Strong variations of base paper porosity in terms of void volume between the individual fibers were found, a closer estimation revealed a distinct pattern visible in the look-through which most probably resulted from a suction roll in the production process.

Small sample areas (65 slices with a distance of 4 µm and a length of 2 mm giving a total area of 0.5 mm²) both in the low porosity and in the high porosity region of the base paper were digitized and prepared for coating layer analysis. This sample size was chosen because the regions with high and low base paper porosity are small. To get statistically meaningful results, 3 samples from different positions have been analyzed for each paper specimen. The results are given with a 90% confidence interval.



(a) Only carbonate containing coating color, high base paper porosity.



(b) 40% clay in coating color, high base paper porosity.

(c) Only carbonate containing coating color, low base paper porosity.

(d) 40% clay in coating color, low base paper porosity.

Figure 4: Effect of base paper porosity and coating formulation on coating holdout. An only eucalypt pulp containing base paper was used for the coating trials. Regions with low and high base paper porosity (voids between fibers) most probably resulted from the production process.

RESULTS AND DISCUSSION

The large differences in the porosity of the base paper are hard to measure directly but are clearly obvious from the cross sections, see the paper cross section images in Figure 4; (a) and (b) represent an open sheet with large pores between the fibers, (c) and (d) a dense sheet.

Coating thicknesseses

Figure 5 shows the measured coating thicknesses – 1) coating thickness at the paper surface (C1), 2) the thickness of all regions connected to the surface (C3) and 3) the thickness of all regions detected as coating (C2). The coating layer on the paper surface is significantly thinner on the regions with an open sheet structure than on regions with a dense structure. This difference is also true for the other thicknesses (C2 and C3) measured although they are not significant for the only carbonate containing color. Both coating colors give similar results for the particular measures and are therefore difficult to distinguish. The difference between the coating thicknesses C2 and C3 indicates a noticeable but not significant fraction of distinct regions detected as coating in the base paper. But a detailed look at the cross section images presented in Figure 4 reveals that there are hardly separated coating regions in the base paper. There are some fiber cross sections observable that have a similar appearance to coating and these are erroneously detected as coating during image analysis as well, see Figure 6 presenting all regions detected as coating in a cross section image. For this reason, C2 clearly overestimates the fraction of coating penetrated into the base paper. However, the increase from C1 to C3 indicates a remarkable coating penetration into the base paper in each case.

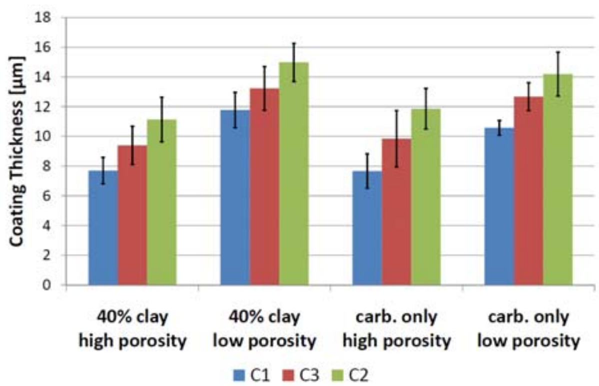


Figure 5: Influence of base paper porosity on measured coating thicknesses (C1, C2 and C3). High and low base paper porosity regions of a pure eucalypt containing base paper were coated at the same time. A similar coat weight was applied in all cases, see Table 1.

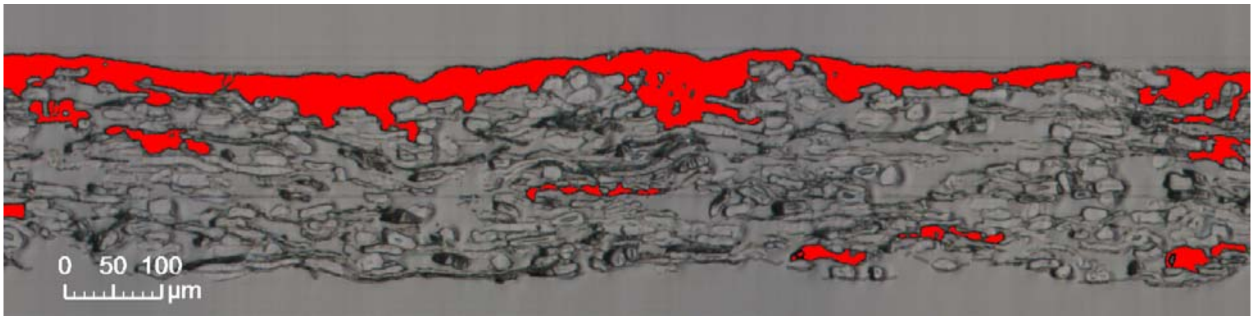


Figure 6: Cross section image (only carbonate containing color, on high porosity base paper region) with detected coating layer highlighted.

The presence of filler agglomerates could have a similar influence as the incorrect detected fiber cross sections on the measured coating thicknesses. This has to be analyzed in future experiments.

Fraction of penetrated coating

The measures used to explain the penetration of coating solids into the base paper are presented in Figure 7. In general, a minimum of 10 – 20% of the total detected coating has penetrated into the base paper. A more detailed look at the results reveals a higher fraction of penetrated coating solids for the paper regions with the more open structure for both coating colors. The differences between dense and open base sheets are more obvious for the clay containing coating color, both measures calculated indicate a significant lower fraction of penetrated coating solids in the base region with a low porosity. The only carbonate containing color shows significant differences between the open and the closed base paper structures at the upper limit of penetrated coating. The measure defined as the lower limit indicates also a better holdout at dense sheet regions but this is not significant anymore. There are also small differences between the coating colors observable. Despite the minor change in the pigment system the clay containing color gives a somewhat better holdout than the only carbonate color. As expected the large differences in base sheet porosity influence the penetration behavior of coating solids into the base paper, whereas the minor change in the pigment system is less important in this particular case. Since a high fraction of coating color cannot contribute to paper surface enhancement, coating holdout / coating penetration has to be treated as serious challenge in paper coating.

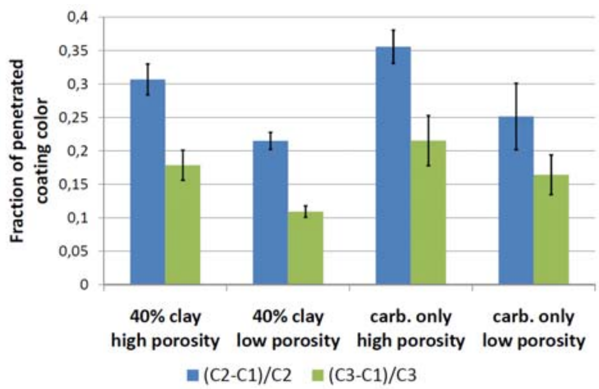


Figure 7: Effect of base paper porosity on the fraction of coating solids penetrated into the base paper. A pure eucalypt base paper was coated with equal coat weights in all cases, see Table 1.

CONCLUSIONS AND OUTLOOK

A method to quantify coating pigment penetration into the base paper is presented. Measures which span the range between the maximum and the minimum penetration of coating color solids into the base paper are defined.

The effect of base paper porosity and coating color formulations (changes in the pigment system) on the coating holdout expressed as coating penetration into the base paper is analyzed. The results show, that base paper porosity (remarkable changes in the base paper porosity) has a clear influence on the fraction of coating solids penetrating into the base paper. A clay containing coating color gives a somewhat better coating holdout than only carbonate containing colors. However all analyzed samples show a large fraction of the coating color penetrated into the base paper and for this reason coating holdout should be discussed as a relevant problem.

The results presented in this contribution are first trials and have to be validated with other methods and samples in future work. Also other approaches to analyze coating holdout based on serial sectioning have to be developed. For example, currently mean coating thicknesses are used for evaluation. The next step could be the use of local data (local coating thicknesses) which will result in a distribution of coating solids fractions penetrated into the base paper. Additional influencing factors like the effect of fillers on the results obtained have also to be analyzed in further experiments.

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