

Print quality of flexographic printed paperboard related to coating composition and structure

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ABSTRACT: Ink transfer and setting influence the achievable print quality and visual appearance. The pressure in the printing nip and the porosity of the substrate regulate the amount of ink that penetrates into a porous coating structure. The purpose of this study was to understand how print quality aspects could be related to ink penetration of water-based flexographic ink into coatings of differently engineered structures: calcium carbonate (GCC) of various particle size distribution (PSD), coatings with different amounts of latex binder, and coatings with various blends of GCC and kaolin.

Calcium carbonate with broad pigment PSD resulted in a lower print density compared to coatings of narrowly distributed particle sizes. Coatings of larger pore volume and greater dominating pore radius showed a higher amount of z-directional ink penetration. A high ratio of uncovered areas (UCA) could be detected for samples with high amount of latex. However, increased printing force eliminated these artefacts.

Increased printing force increased the print density to a higher degree than did a reduced ink viscosity for coatings with pure GCC. For coating layers containing both GCC and kaolin clay, decreased ink viscosity had a stronger impact on the print density than increased printing force. Print density was also affected by ink penetration, suggesting that the optical response is sensitive to the ink-substrate interaction layer. The result presented in this work also suggests that the print gloss decreases with increased amount of penetrated ink due to a higher rate of ink vehicle removal.

Application: By fine-tuning the coating composition and structure and by a proper selection of printing parameters, it was demonstrated that print quality defects can be addressed by fundamental studies of ink transfer and subsequent ink setting. The detailed understanding of mechanisms of ink-substrate interactions is a requirement for maintaining competitiveness in the packaging industry based on renewable materials.

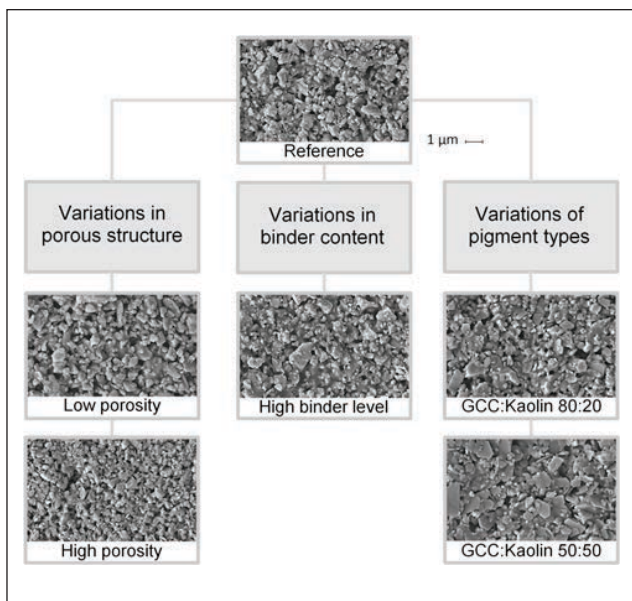
Competitiveness in the packaging industry increases the demands on end-user requirements and sustainability of the printing process. Coating is widely utilized in the paper industry to improve optical properties and printability. Even though smoothness, gloss, and printability improve after coating, the relatively rough base substrates cause local coating thickness variations, with subsequent gloss variations, at a sub-millimeter scale [1-5]. Calendering causes reduction of pore volume and average pore size [6]. Due to the thickness variations of the coating layer, variations in porosity can occur with low porosity regions, or closed areas, developing as a result of the consolidation and compression processes [7]. Such closed areas are often located in low coat weight regions, caused by the unevenness of the paper substrate [8].

The major constituent of a coating color is the pigment. Calcium carbonate, kaolin clay, talc, or a mixture of these are the most commonly used pigments. Commercial coating pigments are available in a wide range of particle sizes, particle shapes and particle size distributions. By mixing different pigments, coating layers with broad variations in porosity, struc-

ture and surface properties can be created.

High gloss of printed areas is often desired to give aesthetically attractive images. Print gloss is related to the rate of ink vehicle removal and the rate of ink levelling before it sets on the surface. Both the coating pore diameter and the pore density of the coating are factors that strongly influence the ink setting rate [9-12]. Smaller pores and higher pore density increase the ink setting capacity in case of a capillary driven process.

The amount of latex binder used in the coating recipe and the fraction of latex that eventually ends up on the surface affect the coating layer structure and porosity. This effect has been explored in several studies where both the appearance of the surface and distribution of the latex particles have been investigated [13-16]. Olsson et al. [17] showed how the dot gain and mottling were influenced by coating structure and ink viscosity. By means of changes in viscosity, it was demonstrated that the influence of coating structure variations could be overridden, allowing for local variations in ink setting. Barros et al. [18,19] studied the influence of topography on the occurrence of uncovered areas (UCA) and missing dots in



1. Overview of investigated coating structures, including scanning electron microscope (SEM) micrographs (top images).

flexographic printing on linerboard and showed that the depth and opening size of impressions were important factors for the UCA frequency.

Upon ink setting, a certain amount of ink penetration also takes place. Due to capillary forces and mechanical forces in the printing nip, ink pigments move downwards into the substrate together with the ink vehicle and form a mixed layer of coating material and ink pigment. A higher surface porosity often results in a faster ink setting, thus leading to a greater ink penetration in the z-direction and lower print gloss [20,21].

Recently, Kim et al. [22] studied ink transfer of functional nanoparticle inks by means of engineered nanoporous microstructures (“stamps”) to form ultrathin printing layers, a technology mainly developed for fabrication of printed electronic devices. As in conventional flexographic printing, the contact pressure between the nanoporous stamp and the substrate is crucial to form a uniform printing layer without loss of printed features or damage of the application unit.

Numerous publications concerning ink transfer and ink setting of oil-based or solvent-based offset and inkjet inks can be found in the literature [9,11,20], whereas rather few articles deal with this topic when it comes to water-based flexographic ink. This paper is thus an attempt to provide a broader picture about mechanisms controlling ink-substrate interactions in flexographic printing by comparing the findings in our previously published articles [23-25].

Print quality aspects related to ink penetration of water-based flexographic ink into pilot coated paperboard with variations in porosity and surface structure was the objective of this study. As a reference coating, a standard GCC-based coating color was prepared. Variation in porous structure was achieved by using calcium carbonate pigments of different particle size distribution (PSD), thus rendering coating layers

with either low or high porosity (**Fig. 1**), which were further treated by calendering. Impact of binder content was studied by varying the fed amounts of styrene-butadiene latex, medium level (reference coating) or high level; whereas, effects of pigment types were investigated from blends of calcium carbonate and kaolin clay.

These pilot-coated paperboard structures comprising a palette of varying surface porosity, surface roughness, and surface energy were then further printed in a laboratory flexographic printer, with ink viscosity and printing force as variables. The ink penetration was determined by visual evaluation of microscopic cross-sectional images and the print quality was measured as print density, fraction of uncovered areas, and print gloss. The print quality was strongly affected by the ink-substrate interactions, especially to the ink penetration in the z-direction. By fine-tuning the coating composition and structure and by a proper selection of printing parameters, it was demonstrated that problems with print quality defects can be overcome.

MATERIALS AND METHODS

Three-ply cartonboard (Duplex) with a basis weight of 179 g/m² with a core of unbleached sulfate pulp sandwiched between a bleached top layer and an unbleached bottom layer (supplied by BillerudKorsnäs AB; Frövi, Sweden) was used as the substrate for coating. The cartonboard was precoated with a 95/5 blend of calcium carbonate (GCC)/kaolin pigment at a coat weight of 11 g/m² using a stiff ceramic blade. The coating was performed on the pilot machine at BillerudKorsnäs AB in Frövi, Sweden, at a speed of 500 m/min using a bent ceramic blade held at a blade angle of 19°. Calendering of GCC-based coatings with varying PSD was performed online in a soft nip with line load of 100 kN/m using a rubber roll of hardness 65°Shore D and a steel roll held at a temperature of 100°C.

For the reference coating color, a standard grade GCC (Hydrocarb 90, Omya AB; Malmö, Sweden) was used. The latex used in all colors was a styrene-butadiene latex (Eka Polymer Latex Oy; Oulu, Finland) with T_g 24°C and particle diameter 120 nm. The thickener Sterocoll FD (BASF Paper Chemicals; Ludwigshafen, Germany) was added to all colors at 0.3 parts dry per hundred parts of pigment.

Variations in porous structure were achieved by using calcium carbonate pigments of either broad (Setacarb HG) or narrow (Covercarb 75) particle size distribution (both from Omya AB; Malmö, Sweden), thus rendering a coating layer with either low or high porosity (Fig. 1). Impact of binder content was studied by varying the fed amounts of styrene-butadiene latex binder: the medium level of 12 parts latex used in the reference coating was compared to a high level, corresponding to 18 parts per hundred parts of standard grade GCC. Effects of pigment types were investigated from blends of the standard grade GCC (Hydrocarb 90) and kaolin clay (Capim DG, Imerys Minerals Ltd.; Cornwall, UK) at 80:20 or 50:50 ratios respectively. Data for all coating formulations are given in **Table I**.

Coating	GCC Grade, pph		Kaolin Grade, pph		Latex, pph
Reference	Hydrocarb 90	100	-	-	12
Low porosity	Setacarb HG	100	-	-	12
High porosity	Covercarb 75	100	-	-	12
High binder level	Hydrocarb 90	100	-	-	18
GCC:Kaolin 80:20	Hydrocarb 90	80	Capim DG	20	12
GCC:Kaolin 50:50	Hydrocarb 90	50	Capim DG	50	12

I. Coating recipes.

Pore size distribution was characterized by an AutoPore III mercury porosimeter (Micromeritics; Norcross, GA, USA). The top surface structure and the percentage areas covered with latex and pigment, respectively, were evaluated from microscopic images (Jeol JSM-6700F cold field emission scanning electron microscope [SEM]; Tokyo, Japan).

The coated paperboard grades were printed with a water-based cyan ink (Sun Chemical Inks A/S; Skovlunde, Denmark), with a viscosity of 25 s (Zahn cup no. 2). Printing was done by a laboratory flexographic printer (IGT-F1 laboratory flexographic printer, IGT Testing Systems; Amsterdam, NL), using an anilox roll (volume 2.7 ml/m² and screen ruling 235 lines/cm) and a printing plate with hardness 57° Shore A and thickness 1.7 mm (Miller Graphics; Sunne, Sweden). Full-tone areas were printed at either 25 or 50 N at constant speed of 0.3 m/s. Impact of ink viscosity was studied by heating the ink from its normal temperature of use (23°) up to 50°C. The corresponding viscosities as measured by a controlled shear stress

rheometer (MCR 300, Physica Messtechnik GmbH; Stuttgart, Germany) were 52 mPas (23°C) and 31 mPas (50°C), respectively, at a shear rate of 100 s⁻¹.

Print density was assessed by a GretagMacbeth D19C densitometer (Regensdorf, Switzerland) at 10 points per sample. The fraction of uncovered areas (UCA) was evaluated by STFI Mottling Expert v1.2 (Innventia AB; Stockholm, Sweden) from scanned images of printed, full-tone areas (8 measurements). Gloss measurements of coated and printed surfaces were performed by means of a gloss meter (Zehntner glossmeter, 20°–75°; Shropshire, UK) at 75° (ISO 8254-1 “Paper and board — Measurement of specular gloss — Part 1: 75 degree gloss with a converging beam, TAPPI method”) at 10 points per sample. Ink penetration depth was evaluated by optical cross-section microscopy (Leica DMRX, Leica Mikroskopie & Systeme GmbH; Wetzlar, Germany) with assistance of software processing utilizing a method described elsewhere [23], which is illustrated in **Fig. 2**. The evaluation covered 12 measurement points on five cross-sectional images, i.e., 60 points in total for each printed sample.

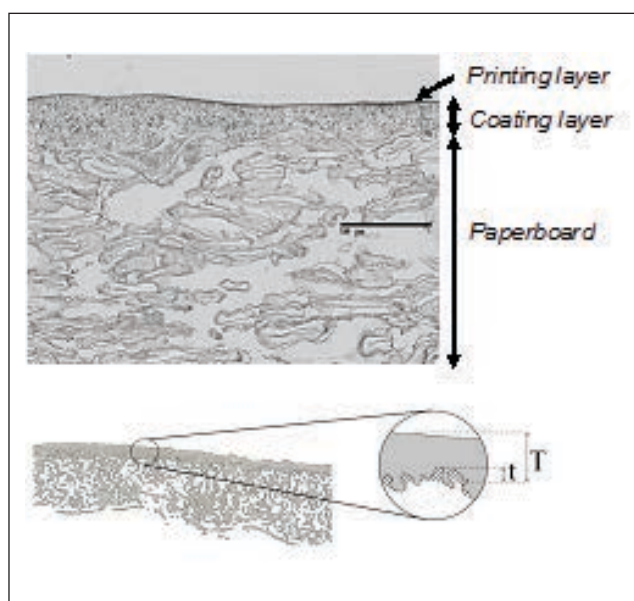
SEM (Jeol 6700F cold field emission SEM) was used to investigate the distribution of pigment and latex on the coated cartonboard surfaces. The percentage area covered with latex was determined by software analysis in a procedure described elsewhere [25].

RESULTS AND DISCUSSION

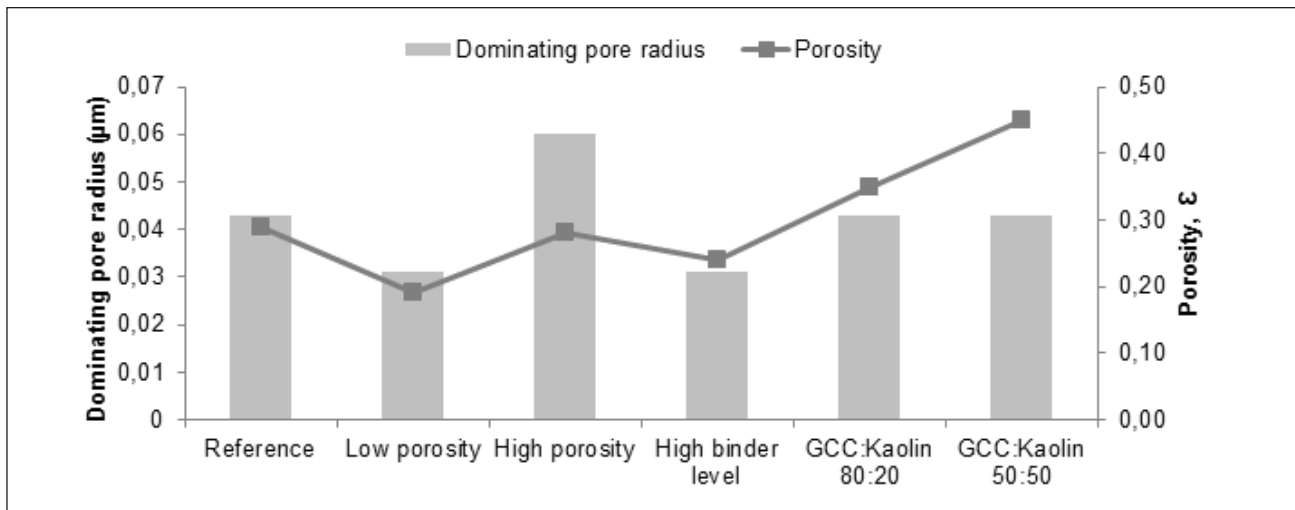
SEM images of all coated surfaces are shown in Fig. 1, while dominating pore radius and porosity determined by mercury porosimetry are summarized in **Fig. 3**. The impacts of porous structure, binder content, and pigment type are discussed under relevant headings in the following sections.

Variations in porous structure

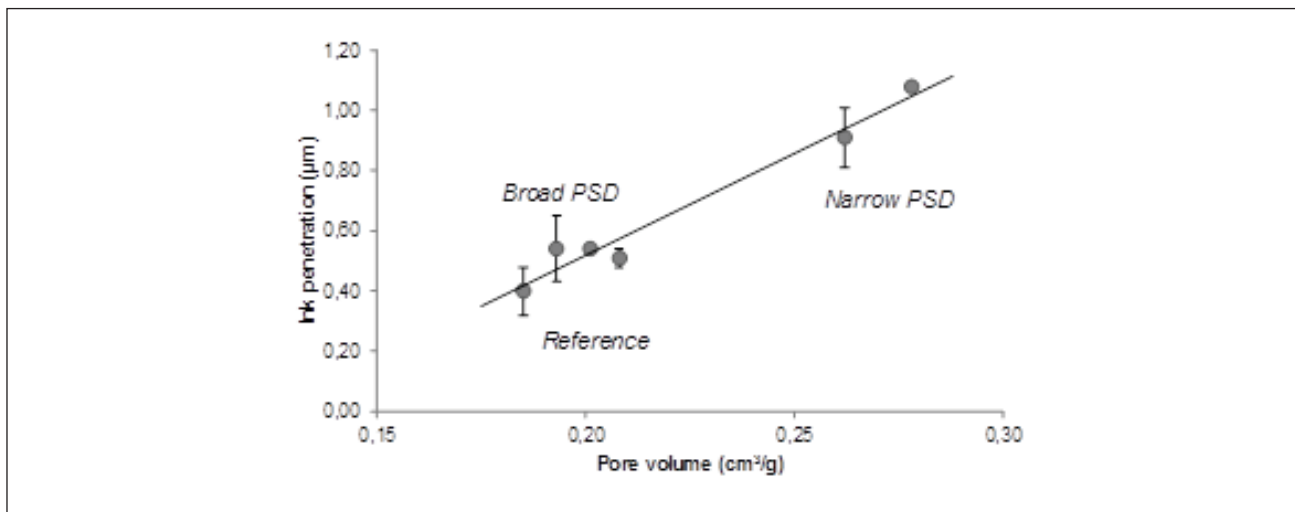
From the SEM images in Fig. 1, differences in surface structure depending on the choice of GCC grade are clearly seen. The dominating pore radius was, as expected, smallest for the coating containing the GCC grade with broad PSD and largest for the grade with narrow PSD, while the reference coating showed a pore radius between these two (Fig. 3). The GCC with broad PSD resulted in a coating layer porosity of 0.19,



2. Upper: cross-section image showing the printing layer, coating layer and paperboard structure. Lower: definitions of printing layer thickness (T) and ink penetration depth (t).



3. Dominating pore radius and porosity for different coating structures (GCC = ground calcium carbonate).



4. Ink penetration depth at a printing force of 50 N as a function of pore volume for GCC-based coatings with various particle size distribution (PSD). Average of 60 measurements with standard deviations. Data from Bohlin et al. [23] for both calendered and uncalendered surfaces.

whereas the GCC with narrow PSD and the reference coating both resulted in porosities near 0.28.

Figure 4 shows the ink penetration depth in micrometers plotted against pore volume in cm³/g evaluated for the three coatings printed at a printing force of 50 N, both before and after calendering. Calendering resulted in a significant reduction in pore volume, with a subsequent reduction in ink penetration for the reference coating and the coating based on the GCC grade with narrow PSD. The pore structure of the coating with broad PSD (which already showed small pore radius and porosity) turned out, however, to be less affected by calendering, which also was illustrated by an almost unchanged degree of ink penetration.

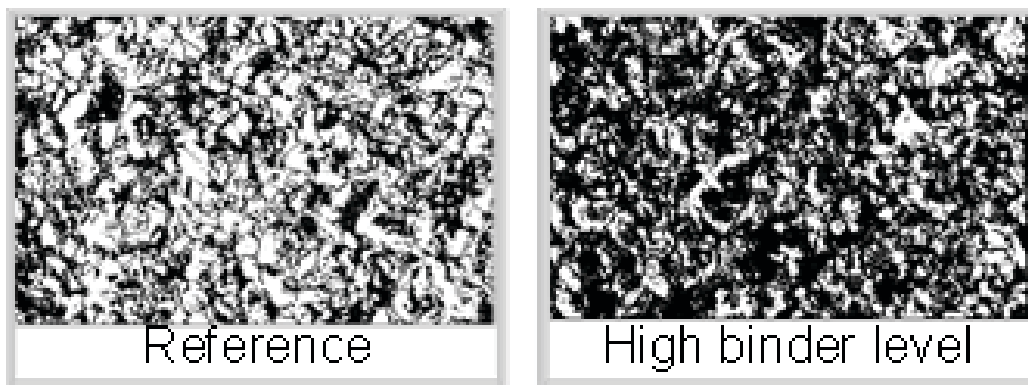
From Figs. 3 and 4, it is obvious that greater pore radius and larger pore volume of the coating layer formed by selection of a calcium carbonate grade with narrow particle size

distribution result in significantly higher z-directional ink penetration.

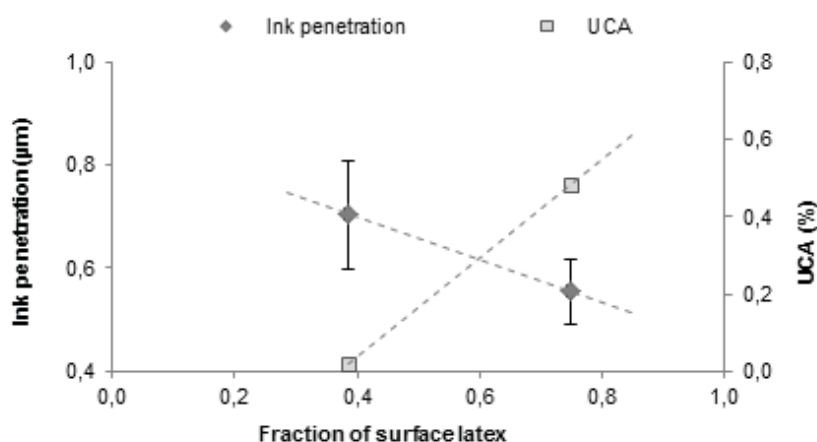
Variations in binder content

SEM micrographs of the top surfaces of the reference coating with 12 pph latex showed a coarse and porous structure consisting mainly of the GCC pigment (Fig. 1). Increasing the amount of latex to 18 pph resulted in enlarged regions comprised of a smoother texture, indicating a structure with GCC particles embedded in a latex film. This increased fraction of latex located near the top surface rendered a more closed surface structure. Figure 3 shows that the porosity decreased from 0.29 to 0.24 while going from 12 to 18 pph latex. At the same time, the dominating pore radius decreased from 0.043 to 0.031 μm.

The software processing method for estimation of percent-



5. SEM images of reference coating and coating with high binder level after software processing to determine the percentage area covered with latex (colored black). Compare with original images in Fig. 1. Data from Bohlin et al. [25].



6. Ink penetration depth and uncovered areas (UCA) at printing force 25 N as a function of surface fraction of latex for GCC-based coatings with varying latex content. Dotted lines are inserted as a guide for the eye only. Average of 3 measurements (surface latex), 60 measurements (ink penetration), and 8 measurements (UCA) with standard deviations. Data from Bohlin et al. [25].

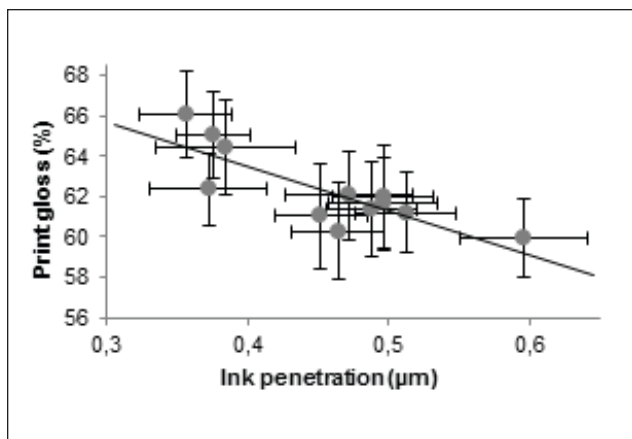
age area covered by latex and pigment, respectively, is illustrated in **Fig. 5** where the latex parts are colored black. The estimated fraction of the surface covered with latex increased from roughly 0.38 for the reference coating to 0.75 for the 18 pph sample.

This rather small increase of SB-latex in the coating recipe thus resulted in a tremendous increase in the fraction of latex located at the topmost surface. Higher fraction of surface latex in turn resulted in lower z-directional ink penetration (**Fig. 6**), which is easily explained by the more closed coating structure. The effect is strongest at the lowest printing force (25 N); increasing the printing force to 50 N aided in pressing more ink into the coating structure due to increased mechanical forces and a larger contact area between substrate and printing plate. The results are consistent with other studies

on relationship between nip force and ink transfer ratio [26]. Higher fraction of surface latex also resulted in an increased amount of uncovered areas (UCA), **Fig. 6**, due to local variations in wettability. However, increasing the printing force from 25 N to 50 N compensated for this effect, since no visible UCAs were detected at the higher printing force [25]. This points to the ambiguity in designing coatings for either higher printing speeds (i.e., quicker setting) or for minimizing print quality defects. Striving to retain ink on the surface for print density, less UCA, etc., by means of porous structure and surface energetics can be challenging when higher printing speeds are desired for multicolor printing [27,28].

Variations of pigment types

The change in surface structure by incorporation of higher



7. Print gloss as a function of ink penetration depth for GCC/kaolin clay-based coatings. Average from 10 measurements (print gloss) and from 60 measurements (ink penetration) with standard deviations. Redrawn from Bohlin et al. [24].

fractions of disc-shaped kaolin particles among the more sphere-shaped GCC particles is clearly shown in the SEM images (Fig. 1). Increased ratio of kaolin clay resulted in increased porosity, from 0.29 (reference) to 0.35 (GCC:Kaolin ratio 80:20) to 0.45 (GCC:Kaolin ratio 50:50), while the dominating pore radius remained constant at 0.043 μm (Fig. 3).

In **Fig. 7**, print gloss is plotted against ink penetration depth for the complete series of samples where pigment type was studied: reference coating and coatings with GCC:Kaolin ratio of 80:20 and 50:50—all printed at printing forces 25 N or 50 N with ink viscosity of either 52 mPas or 31 mPas. Increased ink penetration resulted in decreased print gloss (Fig. 7) due to a higher rate of ink vehicle removal and thereby a

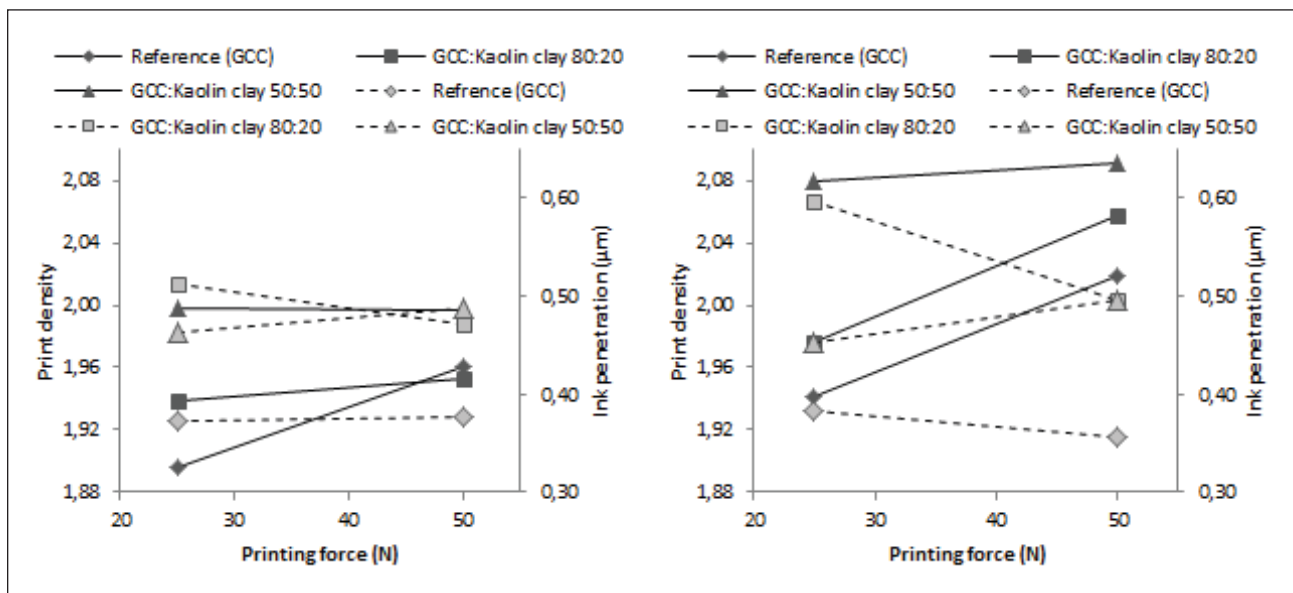
higher ink surface roughness. This lends credit to investigations made by Olsson et al. [29], where lower gloss and higher ink penetration resulted when the ink temperature was raised.

In **Fig. 8**, print density and ink penetration are plotted as a function of printing force (25 N and 50 N) for an ink viscosity of 52 mPas (23°C; left diagram) and 31 mPas (50°C; right diagram). Higher printing force did not increase the depth of penetrated ink to any greater extent; instead, higher printing force led to an improved distribution of the ink, with increased print density as a result. Higher printing force also resulted in a decreased amount of uncovered areas [25]. The effects were more pronounced at a lower ink viscosity (right diagram).

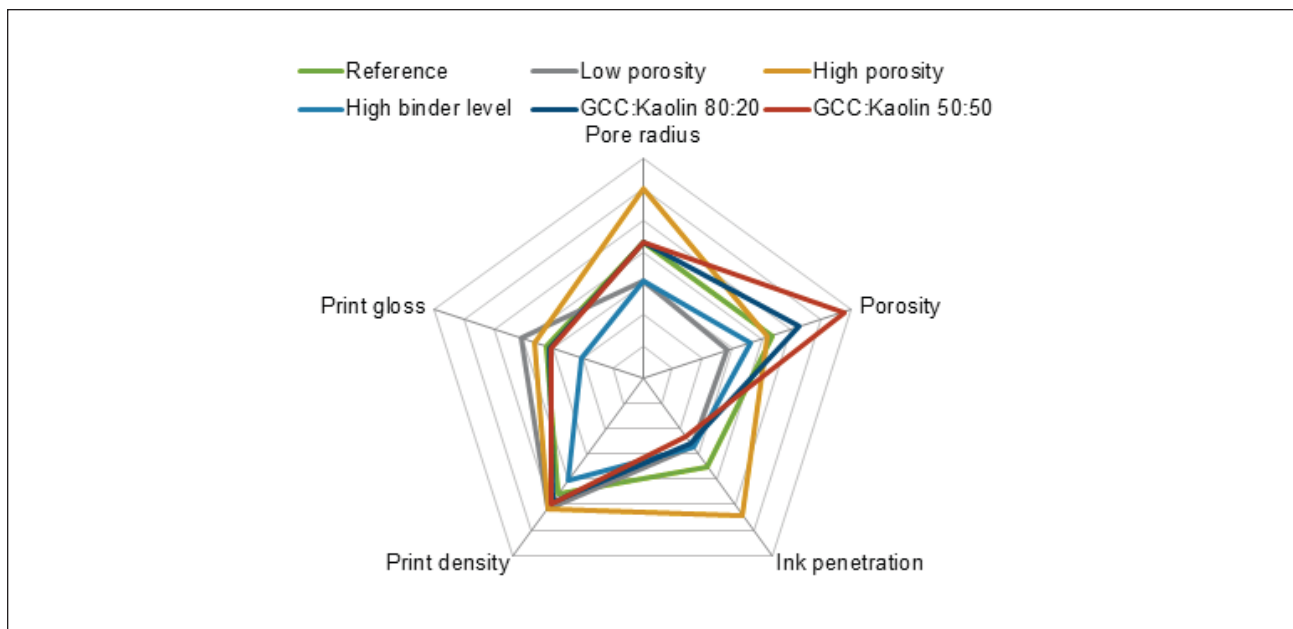
Overall impact of studied properties

Our attempt with this paper was to evaluate the overall impact on flexographic print quality by comparing the findings of three separate research studies. The pore radius and porosity were shown to differ considerably depending on the choice of pigment particle size, amount of binder, and type of pigment (Fig. 3). Furthermore, the ink penetration depth evaluated by our method from cross-section images of printed paperboard could easily be related to the observed variations in porous structure. A strong relationship between surface structure and ink-substrate interactions with subsequent impact on print quality was found.

The impact of various parameters in relation to each other is schematically depicted in the diagram in **Fig. 9**, where pore radius, porosity, ink penetration depth, print density, and print gloss are plotted (in arbitrary units) for the complete series of coating structures printed at a printing force of 50 N.



8. Print density and ink penetration as a function of printing force and ink viscosity for GCC/kaolin clay-based coatings. Data from Bohlin et al. [24]. Left diagram: ink viscosity ~52 mPas; right diagram: ink viscosity ~31 mPas. Average from 10 measurements (print density) and 60 measurements (ink penetration). Error bars are hidden to enhance readability of the diagrams. Lines are inserted as a guide for the eye. Solid lines: print density; dotted lines: ink penetration.



9. Schematic diagram showing the pore radius, porosity, ink penetration depth, print density and print gloss for the complete series of coating structures printed at a printing force of 50 N. Arbitrary units.

The porosity increases in the following order: low porosity < high binder level < high porosity < reference < GCC:kaolin ratio 80:20 < GCC:kaolin ratio 50:50. However, ink penetration depth was highest for the coating with the largest pore radius (i.e., the sample named high porosity), followed by the reference coating. This may be ascribed to a combined effect of capillary and mechanical forces, as discussed in our previous paper [23]. The samples showing smallest overall pore radius and lowest porosity (low porosity and high binder level) also were the ones showing lowest ink penetration depth.

Lower ink penetration with increased amount of kaolin can be related to an increased degree of tortuosity with the incorporation of flaky kaolin particles, even though the dominating pore radius was equal within this series.

The coating with high binder level showed the poorest print quality, both in terms of lowest print density and lowest print gloss among all investigated samples, which is understood by the negative impact from the high fraction of surface latex on ink transfer and setting.

Using a GCC grade with either broad or narrow PSD (i.e., low or high porosity), or a coating formulation with blends of GCC and kaolin, all resulted in slightly higher print density compared to the reference coating (Fig. 9). The highest overall value of print gloss was achieved by using the GCC grade with broad PSD, giving a coating layer of low porosity, which can be explained by the smooth surface of this coating created by the very small pigment particles [23]. Using a GCC grade with narrow PSD (high porosity) resulted in a higher print gloss than the reference coating, whereas the print gloss decreased slightly with increased fraction of kaolin clay.

Interactions between printing inks and substrates have

been the topic of some recent studies: Thorman et al. [30] developed a method for studying the absorption uniformity based on short-time absorption of a colored liquid, followed by blotting of excess liquid and characterization of the pattern of the stain. They analyzed a set of coatings with different compositions and could ascribe absorption nonuniformity to variations in coating layer thickness and/or wettability where different pore structures between thinner and thicker coating areas were claimed to affect the results.

The mercury porosimetry method has been used for decades for determination of porosity of coating layers. Recent approaches involve 3D-modeling of the pore structure by tomography: Aslannejad et al. [31] very recently presented a sophisticated method for modelling the three-dimensional pore space within a coating structure based on focused ion beam SEM. The authors reported a good agreement between mercury porosity measurements and the modelling based on SEM imaging and claimed the method to be useful for macroscale modelling of ink penetration during inkjet printing and, further, a valuable tool for optimization of the ink penetration depth and ink spreading on and within coated paper structures. Such an approach would also strongly add to the understanding of ink-substrate interactions when it comes to water-based flexographic inks used in our study.

Gomez et al. [32] developed a new method to study the distribution of ink components (oil and pigment) from cold-set inks in printed newspaper of thickness about 70 μm . Attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy analysis was performed on printed and unprinted sides of several consecutive layers separated by delamination of the paper. With this procedure, it was possible to determine the portion of each component at various

locations; oil was mainly found below half the paper thickness (i.e., penetration depth $>45\ \mu\text{m}$), whereas pigment was detected in the layers close to the printed surface (penetration depth $<15\ \mu\text{m}$). A drawback with this method lies in difficulties to differentiate components in the printing ink from components in the paper and coating layers due to overlapping peaks in the ATR-FTIR spectra.

Thus, further investigation of water-based flexographic ink-coated paper substrate interactions with the application of such newly developed analysis techniques will most likely provide deeper insights into these very intriguing aspects on print quality.

CONCLUSIONS

Overall, the results demonstrated that the print quality was strongly affected by the ink-substrate interactions. The ink penetration depth was related to the porosity of the coating; a coating layer with larger pore volume and larger pore size created by the use of a calcium carbonate grade with narrow particle size distribution resulted in higher ink penetration in the z-direction. The ink penetration depth was also related to the binder content in the coating color; higher fraction of

surface latex generated a more closed surface (lower porosity), thus hindering ink from penetrating into the coating structure.

The print quality was furthermore related to the ink penetration depth, which in turn was related to the pigment type in the coating color; increased ink penetration resulted in decreased print gloss. Higher printing force resulted in increased print density; for coatings with both GCC and kaolin clay (more porous structures), decreased ink viscosity had a greater impact on print density than increased printing force. This study has shown that by fine-tuning the coating composition and structure and by a proper selection of printing parameters, problems with print quality defects can be overcome. **TJ**

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ABOUT THE AUTHORS

Print quality of flexographic printed paperboard is an interesting area of research in the paper industry, and this research study summarizes and explains observations from several studies on the topic.

The most difficult aspect of this research was that paper is a nonuniform material that gives rise to variations in measured parameters. To address this, a broad number of replicates were tested. We found that many different parameters affect the final result in a complicated manner. The most interesting finding was the connection between ink penetration and print gloss.

Mills might benefit from this information in terms of their choice of materials and process conditions for optimization of product performance. Our next step is to explore new analytical techniques.

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