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DISTRIBUTION OF OFFSET INK CONSTITUENTS IN PAPER COATING AND IMPLICATIONS FOR PRINT QUALITY

Sanna Rousu[1], Patrick Gane[2] and Dan Eklund[3]

ABSTRACT

The implications arising from the distribution of offset ink components in a paper coating structure are presented. The work focuses on two mechanisms causing differential distribution of ink constituents, namely (a) adsorption chromatography, resulting from coating pigment chemistry and morphology variables, and (b) latex-oil diffusional interaction. Adsorption chromatographic fractionation and resultant distribution of ink constituents in a coating structure are shown clearly to affect print quality parameters like print gloss and density. Latex-oil diffusion and its dependent distribution, however, strongly affect ink setting dynamics but no observable effect was found on print gloss and density. This difference in effects is suggested to be due to the fact that adsorption delays the absorption of slightly polar oils, hence delaying the upconcentration of solved resins at the ink-coating interface, whereas diffusional absorption of the polar oils by latex, usually on a longer timescale occurs in volume terms away from the critical initial upconcentration region of ink setting and cohesive tack build.

Keywords: coating structure, absorption of liquids into bound porous networks, chromatography of inks, pigment surface chemistry, latex-ink interactions, ink-coating interactions.

INTRODUCTION

Support of a high quality print, short and flexible delivery with minimal environmental impact and maximum profit are continual challenges for both the paper and ink suppliers to the graphic industry. Statistics show that ink – coating interaction related problems like ink and coating build-up on printing blankets and mottle, are the major concerns associated with offset printing of coated paper (Pantel 1998), and that uneven absorption and distribution of the ink constituents is one of the primary causes for these problems. Therefore, a more comprehensive understanding of the factors controlling, and implications arising therefrom, of ink component distribution on print quality and defects, such as unevenness and mottle, or the runnability determining observables, such as back-trapping, show through, piling, dot gain, set-off or rub-off, is still needed. Both print quality and post-press functionality are strongly affected by the ink – paper interactions as they occur over a wide range of time-spans.

Absorption of offset ink oils into porous structures is always an important stage of ink setting and drying when printing on substrates like coated paper. With highly-heterogeneous materials like paper and inks, the absorption phenomenon is rather complicated as both chemical and physical phenomena occur simultaneously and variables known to be important for the end-result, e.g. the absorption rate, state of the absorbent and absorption path, are all interrelated variables. Capillary-driven transport is considered to be the major driving force for ink setting. Recent advances in this research area include the adoption of the inertial flow mechanism to explain the rate dependency of the short time-scale absorption in small capillaries of low aspect ratio like those of pigmented coating layer pores (Schoelkopf *et al.* 2000). The absorption pattern has been shown to be a complex function of the heterogeneous porous structure, leading to preferential pathways (Gane *et al.* 2000) and thus not a fully pore-filled absorption-front penetration. Another mechanism of absorption is the chemical diffusion of ink fluids into the latex part of the coating layer, which has also been a focus area over the years (VanGilder and Purfeerst 1994). As the absorbent, i.e. the ink, is a multi-component suspension, characterization of the absorption dynamics alone is not sufficient to describe the ink-paper interaction related causes for variations in printability and print quality. Optimally, ink pigments and resins remain on the paper surface, allowing controlled release of the low-viscous fluid phase for absorption into the coating structure. Furthermore, even the low-viscous absorbent includes a variety of chemically and physically differing constituents. The issue of fundamental interactions between ink and coating components and their influence on the final distribution of the multi-component ink on coated paper has attracted some in-depth research efforts during recent years (Wickman 1998, Ström *et al.* 1999, Preston 2002).

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[1] StoraEnso Fine Paper, P.O. BOX 196, FIN-90101 Oulu, Finland / sanna.rousu@storaenso.com
[2] Omya AG, CH-4665 Oftringen, Switzerland
[3] Åbo Akademi University, Porthansgatan 3, FIN-20500 Åbo, Finland.

This work is part of a larger project aiming at clarifying mechanisms behind the phase separation of offset ink constituents in contact with coated paper (Rousu 2002). In earlier publications, we have identified the specific interactions between different types of offset ink oils and coating pigment (Rousu *et al.* 2001), as well as the ink oil – coating latex interactions (Rousu *et al.* 2000, Rousu *et al.* 2002). The purpose in this paper is to summarize the practical relevance of these interaction mechanisms by determining the impacts on ink setting rate and print quality parameters such as print gloss and density, and to provide a rationale for why the various component separation mechanisms have different impacts on the print quality.

BACKGROUND

Parameters Affecting Ink Setting Rate

The offset ink oil-phase is the primary component absorbing into the paper coating structure, as evidenced in figure 1, where the ink oil-phase properties (surface tension/viscosity ratio) are seen to control the ink setting dynamics.

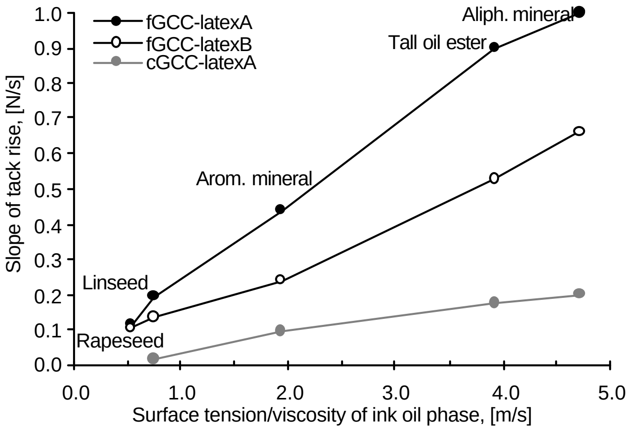


Fig. 1 The rate of ink oil phase absorption into coated paper is determined mainly by the surface tension/viscosity ratio of the ink oil-phase. Higher "slope of tack rise"-value signifies faster ink setting (measured with ISIT, see experimental), which is here measured for inks based on the different oils marked in the figure and for 3 coatings based on coarse GCC (cGCC), fine GCC (fGCC) and two latex types A and B (Rousu 2002).

Today's offset inks are based on a wide range of oils and oil blends of mineral and vegetable origin. The chemical and physical properties of these oils vary considerable, e.g. some measured viscosities can range from 2 to 60 mPas. In effect, ink oil viscosity was found to be the primary parameter affecting ink setting rate. Lower viscosity oils like mineral oils (containing aliphatic components preferably to aromatics, and low boiling point) and treated vegetable oils like tall oil ester (from pine) lead to fast ink setting. Untreated vegetable oils like linseed and rapeseed oils have high viscosity and therefore significantly slower penetration rate in the coating. In these controlled inks used to present the correlation in figure 1, the only variable was the oil type, and thus pigment and resin type and concentration were kept as closely constant as possible. It is therefore justified to conclude that slight variations in the inks´ oil-release properties, which are mainly controlled by oil-resin compatibility, do not have a significant contribution to the absorption rate dynamics.

When comparing the three different paper types in figure 1, we can see that coating pigment or latex type (at a concentration level of 10 pph of latex by weight of pigment) also strongly affect the ink setting dynamics for a given oil's surface tension/viscosity ratio. Coating pigment type affects ink setting mainly through porous structure changes; smaller pore size, higher porosity, increased surface area and lower tortuosity result in a faster ink setting rate. Latex properties, like lower T_g and gel-% as well as reduced solubility parameter through monomer composition, also contribute to faster ink setting.

Overall, all three variables 1) ink oil type, 2) coating pigment type and 3) latex type (at 10 pph) may have an equally strong influence on the ink setting dynamics. However, the setting dynamic itself is controlled by the underlying physics and chemistry of the interacting liquids and surfaces.

Adsorptive Chromatographic Separation of Ink Constituents

Even if the major part of the ink pigments and resins remain at the coating surface, as observed physically and shown by the correlations in figure 1, the absorbing phase itself includes a mixture of constituents with varied properties. Although these components are essentially miscible, their polarity may differ, ranging from practically non-polar mineral oil species to polar vegetable oils and resins. Coating pigments have been shown to cause adsorption chromatographic separation of these chemically differing constituents during their absorption into the coating layer. The hydrophilic dispersed coating pigment surface performs as a site for adsorption, and thereby retardation, of the polar components, i.e. vegetable oils and, on occasion when the pores are very large, ink resins, allowing the non-polar mineral oil species to absorb further into the structure.

From the ink perspective, the polarity difference is therefore the main attributable property to oil fractionation, whereas molecular size or viscosity difference is not a factor when considering species in a dissolved state (naturally, ink pigments and crystalline resin parts are excluded from absorption by size). From the coating side, the specific pigment surface area (m^2/g) is the most critical parameter describing the chromatographic separation activity of the solid phase, where increased separation is found to occur with larger specific surface area, i.e. generally with finer pigments. Increased pigment polar surface energy, mainly as a resultant of the amount and type of ionic dispersing agent, also assists polarity-driven liquid separation. On top of the influence from the pigment surface area and polarity, the separation tendency is moreover dependent on the time-frame available for the adsorption-desorption interactions, as shown in figure 2. The rate at which the absorption front progresses is shown to be the critical universal parameter for the separation of a given oil mixture (linseed + aliphatic mineral in this case) over a range of pigment structures.

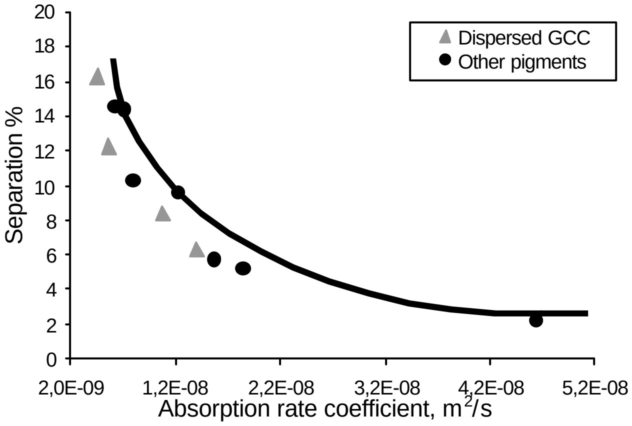


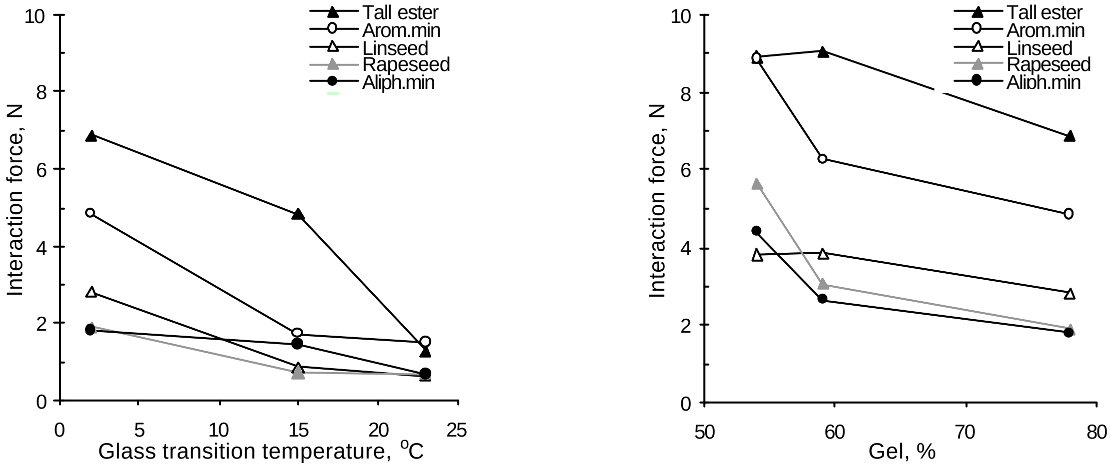
Fig. 2 The universal curve of separation of linseed and aliphatic mineral oils as a function of absorption rate over a range of porous pigmented coating structures (Rousu *et al.* 2001) derived from different pigment types and particle sizes.

In practical offset ink-coating contact, polarity-difference driving the separation of ink constituents is stronger at the initial stages of ink absorption when the fine pores with large surface area are first exposed to the absorbing liquid. Separation is less complete in large pore size and highly permeable coatings where the apparent absorption front progresses via a mechanism of drainage or percolation and there is a lack of surface area in the form of fine pores. With these types of coatings, on occasion, even the ink resins may penetrate excessively into the coatings and cause scuffing, chalking or other ink film strength related defects. Such coatings are formed especially when particle size distributions become two monosize or are too coarse.

Diffusive Differential Ink Oil – Coating Latex Interaction

Ink oil diffusion into latex is commonly acknowledged to be a significant mechanism affecting ink setting dynamics. At typical latex addition levels (~10 pph of latex per weight of pigment), the latex-oil diffusional interaction is anyhow a secondary mechanism mainly because it is a slower process than capillary-driven absorption. In effect, even a significant oil-latex interaction may not observably influence ink setting if the capillary-driven absorption is itself very fast (e.g. with very low viscous oil and fine pore size coatings). Generally, greater diffusion leads to faster ink setting, but as the diffusion also leads to physical swelling of the latex polymer some of the coating porosity can become blocked. This further complicates the prediction of the absorption dynamics and the path of the ink constituent in the heterogeneous coating layer. Furthermore the preference for certain oil chemistries, which otherwise tend to adsorb onto pigments, means that the action of interacting latex tends to assist the removal of otherwise retarded ink oil components from the ink into the coating, thus maintaining a more even spatial distribution of the oils as a function of coating penetration but not of course as a function of local coating constituent concentration.

Both the ink oil and the latex contribute to diffusional interaction. Oils with high solubility parameter, like vegetable oils and aromatic mineral oils, and low viscosity are most prone to diffusion. From the latex side a less rigid/crystalline network, resulting from low T_g and gel-%, and use of monomer composition of low solubility parameter, has a greater tendency to take up ink oils and to swell. Examples of the variation in interactions depending on the latex T_g and gel-%, and ink oil type are shown in figures 3 a) and b).



(a) **(b)**

Fig. 3 Degree of oil-latex interaction depends on both the latex and oil properties as shown here for a series of five oils and latex films of varying T_g and gel-% (Rousu *et al.* 2002). The latex films are formed on a synthetic substrate and the interaction force is measured using the separation force method for applied oil on the ISIT device (see under Experimental).

EXPERIMENTAL

Coating and Ink Materials

The experimental approach (from which also the early example data were shown above) is based on a systematic analysis of well-defined ink and paper coating systems. Model inks having a realistic known composition of commercial offset ink ingredients and the ink oil-phase is the single ink variable used. The studied ink oils were aliphatic mineral oil, aromatic mineral oil, linseed, tall oil ester, and rapeseed oil, as well as oil mixtures containing 25/75 %, 50/50 % and 75/25 % of linseed and aliphatic mineral oils respectively. The eight test inks included approximately 45 w/w-% of the respective ink oil or oil mix, 16 w/w-% of cyan pigment, 10 w/w-% of alkyd resin and 30 w/w-% hard resin (supplied by Sicpa Oy[4]).

Coatings were formulated and coated (~ 12 g/m^2) in laboratory scale on a smooth non-absorbent synthetic base sheet, Synteape[5], in order to have a macroscopically homogeneous coating structure. Pigment and latex types were studied at a constant latex addition level of 10 pph by weight of pigment in order to introduce controlled changes in the coating structure and chemistry. Pigments included GCC with varying particle size distribution, clays of different geographic origins, talc and PCC, which were investigated with a constant type of SB latex (see Appendix table A-1). Latexes included nine experimental types with controlled properties and several commercial grades (see Appendix table A-2), which were blended with a constant GCC type of pigment. The structures were analysed with

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[4] Sicpa Oy, Myllypuronkatu 30, FIN-33330 Tampere, Finland

[5] Arjo Wiggins Ltd. Arjo Wiggins Suomi, Itäranta 7 02110 Espoo, Finland

mercury porosimetry (AutoPore III[6]) and corrections applied using Pore-Comp [7] (Gane *et al.* 1995). The coatings were studied in their uncalendered state.

Ink Setting Rate

Ink setting was characterised as ink-on-paper tackification using the ISIT-device[8]. The ISIT method provides an analysis of the ink-paper interaction from the initial tack rise caused by the ink fluid phase absorption into the coating structure until the consolidation of the ink layer. The slope, or tack rise rate (N/s), is used as a measure for the mainly capillary-driven ink oil uptake by the coating. A standard procedure of ink application from a roller on the ISIT, pre-inked using an IGT ink distributor, was used for printing onto the coated paper samples. Three repetitions were performed for each ink – coating combination. A more detailed description of the device hardware and analysis procedure is found in the literature reference (Gane and Seyler 1994).

Print Quality

Print gloss and density were measured from a full-tone cyan surface printed for the ISIT measurement, i.e. using IGT distributor and ISIT print roller. The ink layer thickness was thus constant, approximately 1 g/m^2. The gloss of coated paper before and after printing was measured using the TAPPI 75° standard (T 480, om-92). Print density was measured using a standard optical measurement technique (D186, GRETAG).

RESULTS AND DISCUSSION

Surface Topography and Print Quality

In order to reveal the contribution of the differential distribution of ink constituents through adsorption chromatography or latex-oil diffusion (or both) on print quality, the strong surface topographic contribution needs to be first excluded. Paper gloss, i.e. the micro-smoothness, of the paper coating is known to be a critical factor for the offset printed gloss (Oittinen 1979). The print gloss-paper gloss relation is shown in figure 4 for all tested combinations of coating pore structure and chemistry as well as different inks. Higher sheet gloss naturally leads to higher print gloss. However, on top of this dependence comes a significant effect of other contributions; up to 20 units difference in gloss is observed for different coating–ink combinations with constant sheet gloss.

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[6] Micromeritics Ltd., USA

[7] Pore-Comp, Fluid Interactions Group, University of Plymouth, PL48AA, UK

[8] ISIT is a product of SeGan Ltd., Perrose, Lantyan, Lostwithiel, Cornwall PL22 0JJ, UK

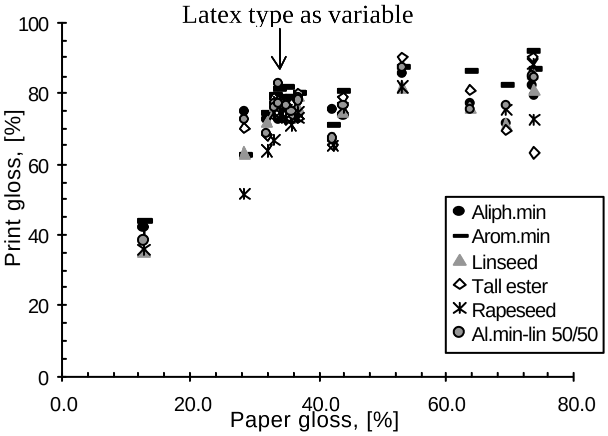


Fig. 4 Print gloss (inks based on the oils named in the legend) dependence on sheet gloss. Coatings were based on varying pigment and latex type and they were applied on micro-smooth base sheets.

In addition, print gloss does not increase with sheet gloss infinitely. There is a maximum at a sheet gloss of approximately 33-35 %, above which the ink cannot act to "smoothen" the coated paper surface to produce ever higher gloss. Above this critical sheet gloss the snap actually decreases with smoother paper surface, i.e. reduced pore size. The same has been found by Donigian *et al.* (1996), and has been explained by faster ink immobilisation with finer pores. Faster immobilisation allows a shorter time for ink filament levelling after the ink film split on application causing a rougher, less glossy print (Desjumaux *et al.* 1998, Preston 2002). In order to understand the variation in print gloss beyond this sheet gloss dependence, and hence independently of paper smoothness, the gloss data were normalised for the paper smoothness properties by dividing the delta gloss (print - sheet gloss) with the paper gloss.

Print density is primarily affected by ink layer thickness and smoothness, which is a function of both ink (e.g. pigment concentration) and coated paper surface properties, and their interaction. With a given amount of constant type of ink (g/m^2), higher print density is created with a smoother paper; or reversed, a greater amount of ink is required to reach desired print density with a silk/matt grade than with a glossy paper. As shown previously, the paper surface roughness also strongly influences the contour of the printed ink film or print gloss. Thereby, also print gloss and density are strongly related parameters. The similar analysis as was made for print gloss, in this case normalising to effective ink generated gloss, was thus followed for the print density values in order to extract and identify the influences of the specific ink-coating interactions independently of surface topographic and ink smoothening effects. Print density data were thus normalised by dividing by delta gloss.

Adsorption Chromatography - Influence on Print Quality

We first consider now the influence of the adsorption chromatographic separation of ink constituents on ink setting rate. This is done by comparing the ink setting rate for a series of inks formulated first with only one oil type and then a series of inks composed of a mixture of two oils that are known to fractionate (linseed and aliphatic mineral oils in 25/75, 50/50, 75/25 proportions). As seen in figure 5, we can conclude that the absorption behaviour is similar in both cases. Thereby adsorption chromatographic separation does not influence the absorption behaviour during ink setting, at least not in a macroscopically observable scale when measured as ink tackification dynamics.

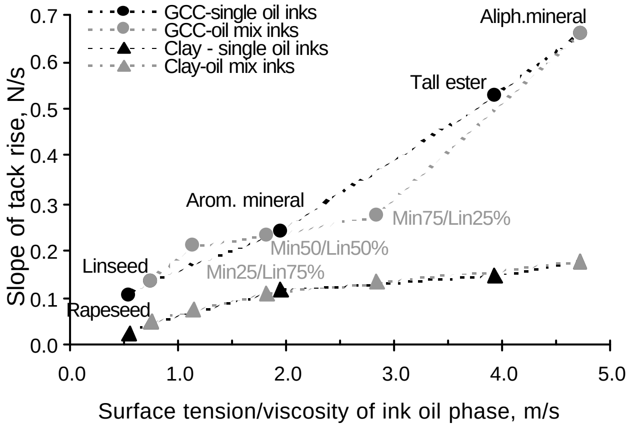
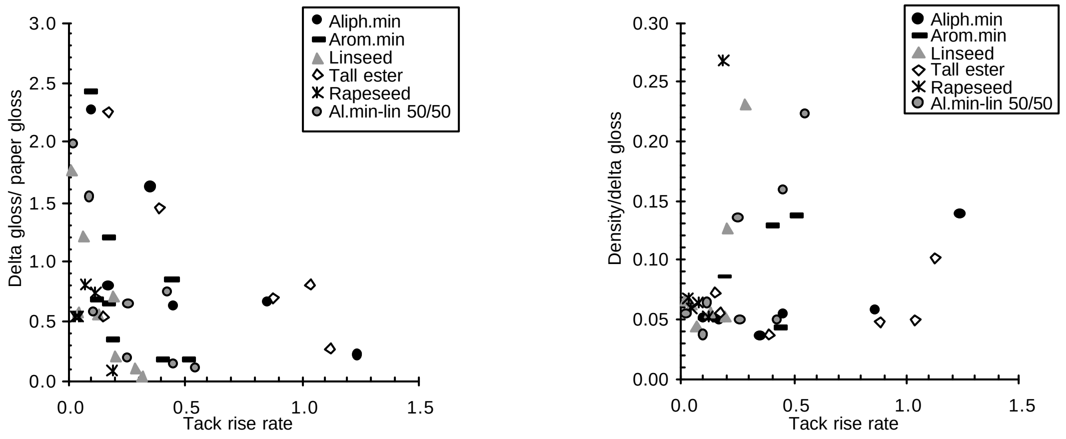


Fig. 5 Ink setting is determined by the surface tension and viscosity properties of the absorbing oil phase both in the cases of single oil and multi-oil inks. Thus, chromatographic separation of polar and non-polar ink oils during absorption does not observably affect the initial ink setting dynamics at 10 pph latex addition level.

The final normalised print gloss (a) and density (b) are plotted against the ink setting rate in figure 6 for different combinations of inks and coatings with variable pigments. Tack rise rate affects the final normalised print gloss and density differently for each ink; i.e. with a given ink setting rate as defined by the coating, the gloss and density depends on the ink type. As we are observing data that are normalised for the topographic difference between the paper and print and consider it here as a function of the absorption rate dependency of the coating, we can conclude that it is indeed the spatial location of the oil in the coating layer that causes the final print gloss and density differences.



(a)

(b)

Fig. 6 Influence of ink oil removal rate on (a) print gloss and (b) density as normalised for the paper and additive print gloss respectively. Coatings are formulated with a series of different pigments and 10 pph of a given latex.

When now comparing normalised gloss data for a series of inks based on different proportions of linseed and aliphatic mineral oils, as shown in figure 7, we can again see that the adsorptive chromatography is thus one mechanism contributing to gloss and density related variations. Inks separate into their own categories. In addition, the gloss values change consistently with the mineral to linseed oil proportion in the ink. By implication, it can be concluded that a more efficient separation of the oils results in a higher print gloss.

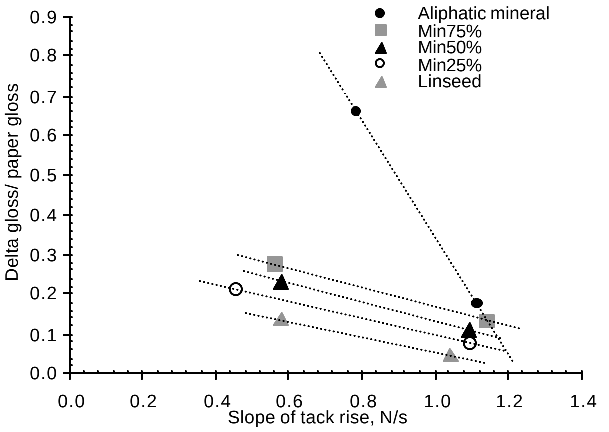


Fig. 7 Oil properties and their distribution in the coating layer affects the print gloss development, and may be the cause of gloss related mottle. Series of inks with varying aliphatic mineral oil and linseed content printed on two different coatings with varied pigment, one based on fine GCC and the other on English clay.

It can be concluded that the distribution of ink oils in the coating structure due to chromatographic separation could lead to print gloss and density variations in the print. Oils are drawn out differentially from an ink into a coating with variable pigment types, thereby creating differences in the print gloss and density values. In a heterogeneous paper, this may lead to spatial variations in the plane of paper, which could be seen as mottle or print gloss unevenness.

Oil Distribution in Latex and the Influence on Print Quality

The latex-oil interaction effect on ink setting dynamics is addressed in figure 8. From the ink side the viscous properties of the ink oil phase are still strongly dominating the ink setting dynamics, although slight "kinks" in the correlation are observable with strongly latex-interactive tall ester and aromatic mineral oil based inks. The differences coming from the coating latex types are anyhow much stronger.

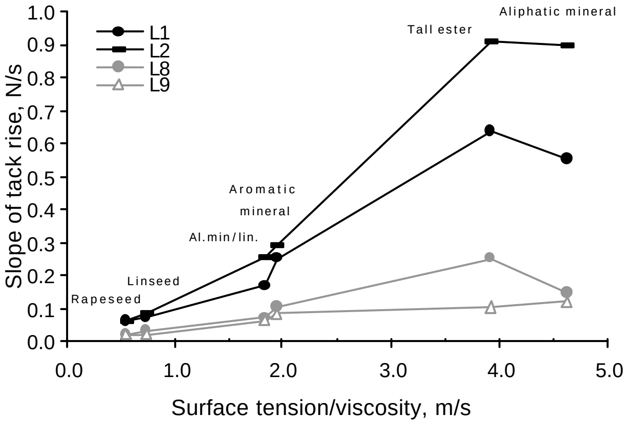
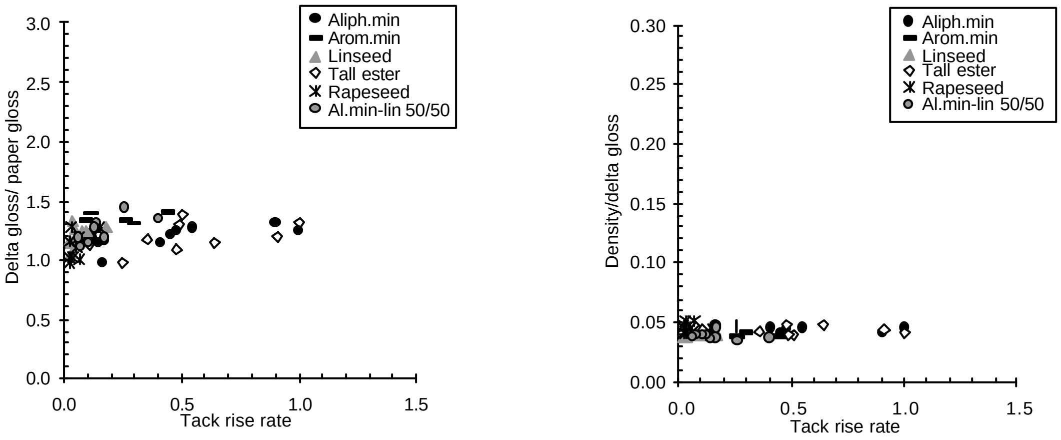


Fig. 9 Oil absorption from an ink measured as ink tackification on coated sheets with varying latex grade (fine GCC, 10 pph of latexes L1, L2, L8 and L9) is controlled by the surface tension and viscosity properties of the ink oil-phase. The influence of differential oil-latex interaction is observable with strongly latex-interacting oils, namely aromatic mineral and tall ester oil-based inks, as a discontinuity at surface tension/viscosity ratios of about 2 m/s and 4 m/s.

In figure 10 we now perform an analysis of the normalised print gloss (a) and density (b) data plotted against initial ink setting rate variations caused by latex-oil interaction. We note that all inks fall on top of each other. It can thus be concluded that differences in the distribution of ink oils in the latex do not influence the average print quality. It should be emphasized that this implication does not state that latex type as such is insignificant for print quality – this was seen not to be the case already in figure 3, where significant differences were observable depending on the latex type. The result here rather confirms that final print gloss and density are not affected by preferential uptake of a certain type of ink oil by the latex. Thus the distribution of any type of ink oil within the latex part of the structure is similar (or at least insignificant for the average print quality result) despite significant differences in the magnitude of interaction and ink setting dynamics. Large print gloss and density variations may nevertheless result depending on the latex type from changes in surface topography, coating structure and coating component distribution.



(a) **(b)**

Fig. 10 Influence of ink oil removal rate on (a) print gloss and (b) density as normalised for the simultaneous paper gloss and delta gloss development respectively. Coatings are formulated with fine GCC pigment and a series of variable latices at 10 pph. The inks contained the oils named in the legend.

CONCLUSIONS

The offset ink component distribution in coated paper, caused by differential interaction with coating pigment and latex, was identified to influence the ink setting rate and print quality. The main observations can be summarized as follows:

- Coating pigment type, dispersant concentration and surface area cause and control polarity-driven chromatographic fractionation of ink constituents. This influences print density and print gloss. The action of adsorption is to slow the absorption of polar oils.
- Coating latex – oil association may affect ink setting rate as strongly as capillarity driven absorption, but no observable influence on print gloss or density was found. The action of latex diffusional absorption is to increase the rate of polar oils.
- All interactions are time-dependent, and their relevance depends on the total time available for reactions. This is determined by the general absorption dynamics of the system.
- In a realistic heterogeneous paper structure many of the proposed mechanisms are present simultaneously, and may thus result in print unevenness, defects like mottle or print smoothness and gloss variations.

These observations allow us to conclude that the separation of polar and non-polar oils in the coating layer if associated with a strong retardation of polar oil, due to adsorption of the polar resin-solving component on the pigment, leads to deterioration of average print quality. Promoted absorption of the polar components which may occur via latex diffusional absorption reduces the impact of fast ink setting rate on print gloss and print density. This means that the concentration of the ink via a too rapid non-polar mineral oil loss, for example, leads to a retained solved resin within an otherwise immobilised ink layer. This can result mechanically in shrinkage and roughening effects during subsequent resin concentration within this matrix. Maintaining the right balance of polar and non-polar oils within the concentrating ink layer is thus concluded to be critical.

Commentary

It must remembered of course that these observations are not considered in the context of the impact of multiple nip impressions, as would occur on a multi-colour press. The action of rapid ink setting under the dynamics of film splitting can act to alter the conclusions in respect to the relative roles of adsorptive separation versus absorptive diffusion. However, the condition of the single colour before multiple nipping is defined by the observations made here.

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APPENDIX

Commercial paper coating pigments, commonly used by the industry, were used in the work. The properties of these pigments are presented in table A-1.

Table A-1 Details on coating pigments.

Product	Supplier	Particle size, % <2 µm	<1µm	BET area m^2/g
HC60	Omya AG	60	36	7.1
HC90	Omya AG	90	62	12.4
SC	Omya AG	97	80	16.1
CC	Omya AG	95	75	9.8
SW95	Imerys Ltd.	92	81	15.0
AG	Imerys Ltd.	94	83	21.0
AM90	Kaolin International BV	97	95	17.4
C10	FinnTalc	46	-	4.8
V40	Omya AG	85	63	8.8
A40	Specialty minerals Ltd.	median particle size 0.4		10.0

A series of model latexes, L1-L9, and commercial grade latexes were used. Details on the latexes are presented in table B-2.

Table A-2 Details on model and commercial grade latexes.

Abbreviation	Supplier	Monomers	Tg, °C	Gel, %	Particle size, nm
L1	BASF AG	SB	2	78	156
L2	BASF AG	SB	6	59	155
L3	BASF AG	SB	1	54	159
L4	BASF AG	SB(An)	12	70	148
L5	BASF AG	SB	15	78	155
L6	BASF AG	SB	23	77	152
L7	BASF AG	SA	7	65	153
L8	BASF AG	SA	24	59	156
L9	BASF AG	SA(An)	6	79	153
DL940	Dow chemical	SB	22	-	140
DL920	Dow chemical	SB	13	-	140
DL980	Dow chemical	SB	-7	-	130
S360D	BASF AG	SA(An)	5	-	-
SB370	Raisio Chemical Oy	SB	25	-	-
P5105	Eka Polymer Latex Oy	SB	-10	-	170
P7105	Eka Polymer Latex Oy	SB	17	-	170