

Dynamics of Ink Gloss: Influence of Ink Rheology on Leveling

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Abstract:

Gloss is an important aspect of a printed sample in terms of quality. Gloss dynamics have not been well documented yet. A good understanding of the influence of the ink rheological properties on gloss and its dynamics is not available.

The wet ink gloss evolution on Mylar film was recorded every 0.1 second, immediately after printing, and followed for 60 seconds. A specially designed gloss-meter directly mounted on a KRK was used to record the ink film gloss dynamics. Two inks with different setting rates were tested. Both inks were characterized on a rheometer in terms of steady and dynamic behaviors.

As the leveling proceeds with time, the ink film surface becomes smoother, increasing the ink film gloss. On plastic film, the high setting rate ink produces a lower print gloss development than the low setting rate ink. Rheological measurements reveal that the high setting rate ink has a higher viscosity and a faster elastic structure recovery for the range of shear rates involved during leveling. Thus, after printing on the plastic film, the higher viscosity of the high setting rate ink slows down the leveling and its faster elastic recovery limits the extent of leveling.

On thin films of paper coatings, at high pigment volume concentration, the two inks are set by capillary sorption of the ink mobile phase into the porous structure of the substrate and the ink rheology has little influence.

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On a coating containing a large content of latex that is non interactive with the ink mobile phase components, the ink leveling patterns are similar to the case of the plastic film. However, when printed on a coating containing a highly interactive latex with the ink components, the ink rheology is the most important factor to determine the gloss dynamics.

This results contribute to a better understanding of the ink gloss development as related to the ink rheological variables.

Introduction

The gloss is defined as the light reflection, at the same incident angle, from a surface.

Print gloss is a complex function of the coating (intrinsic gloss, roughness) but also of the ink formulation, the ink index of refraction, the ink amount, the printing speed and some parameters like the water application and the drying conditions.

Numerous examples in the literature show that the final printing gloss does not correlate with the coating gloss (Borchers, 1958; Fetsko and Zettlemoyer, 1962; Hecklau *et al.*, 1967; Kontschak, 1976; Oittinen, 1980; Donigian, 1996; Aspler, 1993).

Lavelle (1982) shows that the print gloss is a complex function of the ink composition and the substrate roughness. Lee (1994) gives the coating gloss and roughness relationship for coating colors. The roughness of an ink film is determined by either the roughness of the substrate or the leveling characteristics of the ink film. The ink film splitting in the printing Nip and the ability of the ink to level before drying have a tremendous effect on the print gloss.

Final gloss measurements are routine and gloss development has been studied for a number of years (Fetsko and Zettlemoyer, 1962; Oittinen, 1983). However, the mechanism behind gloss development is not well understood (Aspler, 1991). Fetsko and Zettlemoyer report the gloss of printed films as a function of time for coated and uncoated papers but due to equipment limitations, gloss could not be obtained during the first 10 or 20 seconds after printing. The influence of filaments on the gloss dynamics has been discussed and recently, the leveling of ink filaments on plastic Mylar surfaces was reported (Glatter *et al.*, 1996a).

Even if coating absorbency (and porosity measurements) are poor predictors of print gloss (Aspler and Lepoutre, 1991; Lepoutre and De Grâce, 1978), the final gloss depends on the balance between the rate of absorption of the fluid phase

into the coating which is not well understood yet and the rate at which the viscosity increases because of the ink setting (loss of the ink vehicle in the coating layer, oxidative polymerization of the binder). The importance of the ink-paper interactions are pointed out by Micale *et al.* (1983) and reviewed by Aspler (1993). H. Suzuki *et al.* (1996) show that the ink setting rate has a significant effect on print gloss in the first minute after printing. Oittinen (1976) examines the effects of ink properties on transfer and ultimate print gloss primarily through their influence on absorption and filtration. The print gloss is a function of the pore size of the coating and pore volume (Donigian, 1996; Cummings *et al.*, 1996; Desjumeaux and Bousfield, 1997; Arai and Nojima, 1997).

The type of latex is found to influence the rate of ink setting and the print gloss varies with the type of latex. It is known that the ink solvents (like petroleum distillate ink oils used as solvents for the resins) affect the latex polymer in the coating during printing by swelling or dissolving polymer films. A latex type with a great swelling and sorption results in a fast ink penetration that is to say a rapid ink tack-build rate (Kelly, Russell and Graham, 1971). This rate of tack-build is related to the degree of latex polymer solubility in the ink solvents and its influence on the ink-latex film interaction (Van Gilder and Perfeerst, 1994). A high ink tack-build rate (slope) is observed in the case of a highly interactive latex in contact with ink. The use of the less ink interactive latex in a pigmented paper coating contributes to an improved tail-pick resistance on a commercial 6-color sheet-fed press. The latex variables (latex binder, butadiene level, particle size and crosslink density) are very important and controllable factors in modifying the nature of paper and ink interaction in terms of ink tack-buildup (Triantafillopoulos, Lee and Ave'Lallemant, 1996).

The ink variables such as pigment type (composition, shape, size, distribution), pigment surface treatment, pigment volume concentration, and degree of dispersion influence the print gloss. Higher gloss readings are obtained from black inks containing high loadings of small size pigment particles (J. G. Shreckendgust, 1964). Vehicle properties (chemical composition, refractive index, compatibility of the components, wetting ability and both setting and drying times) contribute to print gloss. A poor degree of dispersion, over-pigmentation, film wrinkling or cratering during drying, inappropriate use of waxes (waxes may recrystallize into large particles at the ink surface and ruin film smoothness), use of some paste driers reduce print gloss by increasing surface roughness. The influence on the print gloss of the resin parameters and its solubility in the drying oil has been investigated (Apps, 1964; Fairfield, 1960; Williams, 1970).

Experimental Approach

Gloss Dynamics

The experimental device has been described by Glatter and Bousfield (1996b) in detail. Figure 1 is a schematic of the device. A laser light source (675nm, 1.0mW, M38, 920, Edmund Scientific) is mounted at a 75 degree angle from the vertical. The light reflects from a first surface mirror to the sample. The size of the light spot on the sample can be adjusted. However, we found that too small a spot would give different results every time because of the variation of gloss along the sample, even when printing on plastic films. We found a spot size of 5mm diameter to be large enough to give reproducible results. The light reflects off the ink film onto a 0.090² inch² light detector (OPT201, BurrBrown). A lens is used in front of the detector to capture more light from the film. The device collects gloss data every tenth of a second. The computer records the voltage signal (linearly dependent on the standard Tappi gloss) from the light detector. More details of the experiment and data collection are in Glatter (1996b).

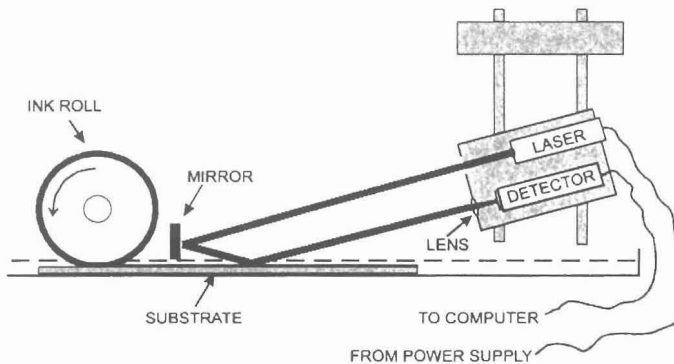


Figure 1. Schematic of experimental setup. The laser sends a beam of light that reflects off a mirror to the sample. The detector converts the light intensity to a voltage signal. The sample moves through the print nip and stops 10 cm after the nip.

The ink is applied to the inking rolls. The sample is mounted on the moving member. A print test is conducted. When the test is actuated, the operator also activates the computer to collect data. The sample stops, within 0.1 seconds after the printing nip and the gloss is recorded in time. The gloss value is sampled every 0.1 seconds. Four trials are averaged together to give a single gloss dynamic curve.

Because of questions with regard to transferring Tappi gloss values to the voltage values reported here, we will only report voltage values. Figure 2 shows that the Tappi gloss range of 50-70 correlates linearly with our 0-10 volts.

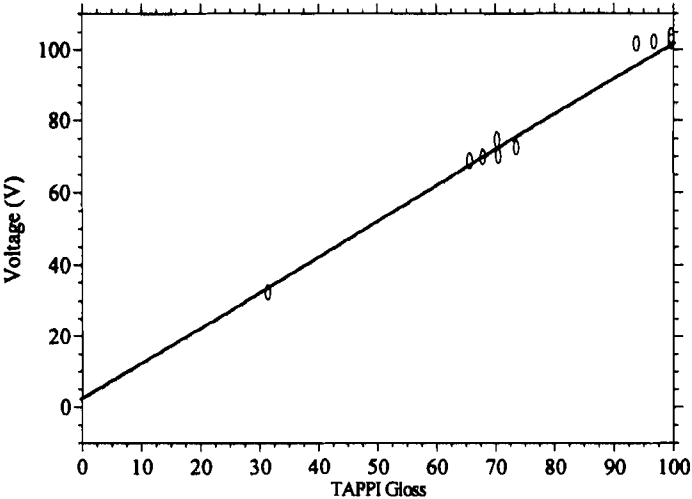


Figure 2. Linear correlation between the standard TAPPI gloss values and the acquired voltage values.

Inks

Two actual cyan sheet-fed offset inks were tested. In this work, they were referred as Ink A and Ink B. Ink A contains a small amount of diluent (mineral oil) and therefore has a high setting rate. Conversely, Ink B contains more diluent and has a lower setting rate.

Rheology Measurements

The main challenge of the rheological characterization of pigmented inks resides in the fact that these properties are time and strain history dependent (Amari *et al.*, 1995). This is mainly due to physical associations of pigments and their

ability to form a physical network. Actually, the time scales associated with these phenomena are also of prime importance. The best way to characterize ink rheology is therefore to try to simulate with the cone/plate rheometer the different strains the ink undergoes when running on a press and when leveling on the printed substrate.

During ink distribution on the rollers (both on a real press, or on the experimental KRK), the main deformations are: (i) extremely high shear in the nip and (ii) fast elongation during filamentation at the nip exit. These deformations are of such a magnitude that one can easily imagine every kind of physical association being disrupted by such strains. The interesting phenomena become therefore the magnitude and the time scales involved with the structure recovery of the ink after being transferred to the printing substrate. In order to further understand these phenomena and to try to correlate them with the dynamics of print gloss, flow, oscillation and creep measurements were effectuated on the studied inks. For each kind of measurement, we applied to the sample the highest shear rate possible before turbulent flow under the cone of the rheometer (20 s^{-1} with a $4\text{cm}/2\text{deg}$. cone), for 2 minutes. It is rather obvious that the stress and the related shear rate undergone by the ink in the nip are several orders of magnitude greater than this pre-shear. But we verified that the viscosity reached a pseudo-equilibrium before stopping the pre-shear. Therefore, if some trends are observed with such our standard pre-shear procedure, one can suppose that they will even be exaggerated in a real deformation on the press.

Every rheological measurement was performed at 25 deg.C and repeated 3 times for reproducibility. Generally, the 3 curves were superimposed.

Flow Measurements

The flow measurements will give the viscosity of the ink during structure recovery. A "down" stress ramp was applied to both inks after pre-shear, from 2500 to 1 Pa, with the stress decreasing exponentially during 3 minutes (Figure 3). In some aspects, this experiment simulates what the ink film is undergoing during leveling. Namely, leveling being a surface tension driven phenomena, the surface tension imposes a stress to the film, which is inversely proportional to the radius of curvature, and therefore which should decrease exponentially with time.

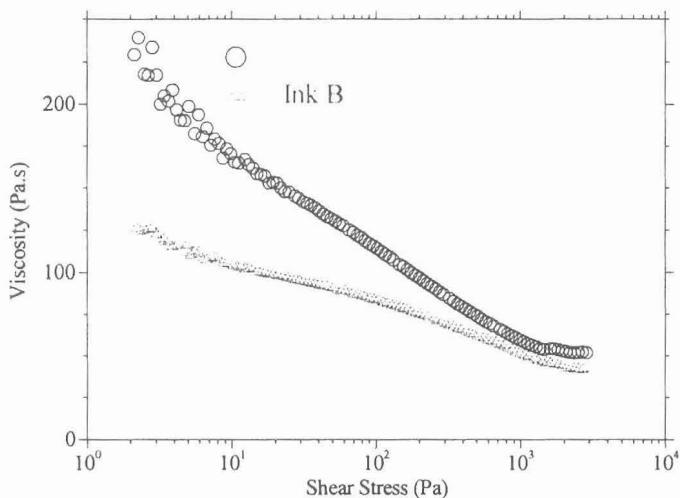


Figure 3. Flow experiment: down stress-ramp for Inks A and B.

If both inks have about the same viscosity at high stress, it is rather obvious that they do not show the same viscosity increase when decreasing the applied stress. The rate of viscosity increase is much higher for Ink A than for Ink B, indicating that some stronger associations take place in Ink A when the imposed stress decreases. This trend will be of prime importance for understanding the difference in leveling behavior of both inks.

Oscillation Measurements

The flow results suggest a different structure recovery for Inks A and B. In order to further investigate this phenomenon, some mechanical dynamical measurements were applied to the samples. After the standard pre-shear procedure (2 minutes at 20 s^{-1}), the viscoelastic parameters G' and $\tan\delta$ were recorded over 100 s, at 10 Hz and with the first measurement made after 2.5 seconds after stopping the pre-shear (Figures 4 and 5).

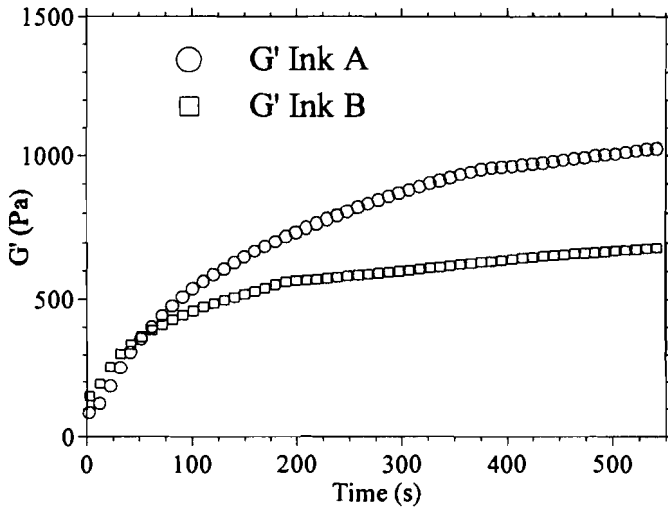


Figure 4. Structure recovery of Inks A and B after standard pre-shear. Elastic modulus vs. time.

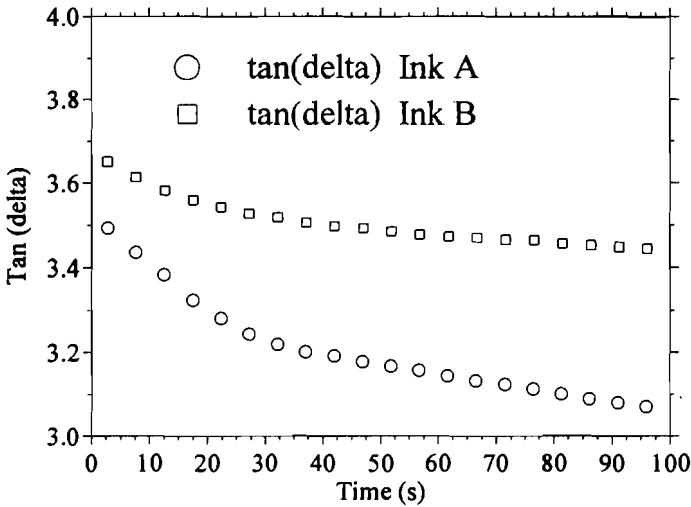


Figure 5. Structure recovery of Inks A and B after standard pre-shear. Loss tangent vs. time.

Such an experiment monitors the structure build-up of inks with time. Namely, looking at both G' and $\tan\delta$, one can see a drastic increase in G' along with an important decrease in $\tan\delta$ with time, which indicates that the ink is recovering some structure, certainly because of pigment reorganization and build-up of a physical network. Here again, it becomes obvious that Ink A has a tendency to restructure faster and more strongly than Ink B. Namely, looking at the initial slope on $\tan\delta$ curves, the rate of decrease in $\tan\delta$ of Ink A is twice that of Ink B (-0.011 s^{-1} vs. -0.0056 s^{-1}). Also of importance are the respective values of $\tan\delta$ for both inks. Actually, not only is Ink A restructuring faster than Ink B, but also its $\tan\delta$ value is lower than that of Ink B, indicating an ink with a higher degree of structure during the whole experiment. This will be of prime importance for the following dynamic gloss study.

Creep Measurements

In order to further investigate the rheological behavior of both inks right after high shear - i.e. when leveling starts after filamentation on the nip - a creep experiment is needed. Namely, such an experiment will give us some deeper insight into what happens right after shearing stops, i.e. in the first second of recovery. The following experiment was applied to both samples:

- pre-shear at 20 s^{-1} for 2 minutes
- applying zero-stress and recording % strain vs. time for the next 1000 s

A negative strain is undergone by the ink when the shear is suddenly stopped (Figure 6). This seems to indicate that the cone is coming back, due to some elastic return imposed by the ink sample. Such an effect can be interpreted by a relaxation of both the polymer matrix and the pigment network. Under shear, polymeric chains and pigments are aligned, certainly very close to the shear direction – at least to a certain extent, which depends obviously on the shear rate level. When the shear stops, both polymeric chains and pigments may have a tendency to move back to a more randomly position, the pigments starting to rebuild some sort of a physical network. The elastic return observed on Figure 6 is certainly a combination of both phenomena.

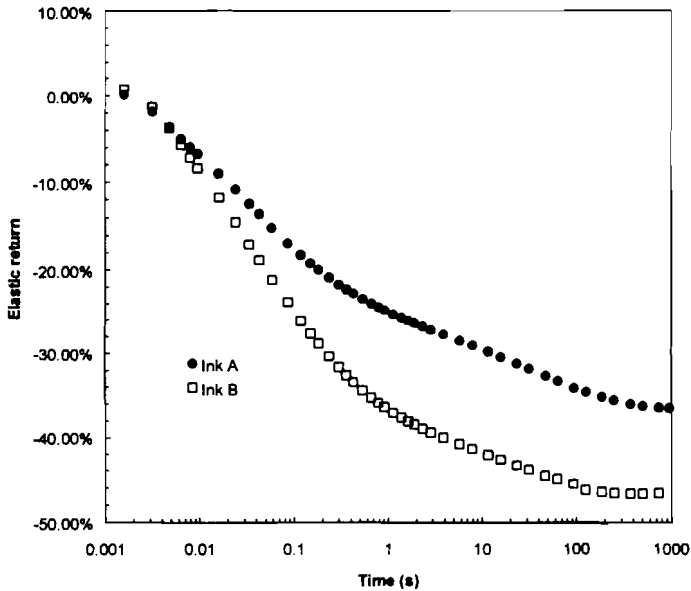


Figure 6. Elastic return of Inks A and B after shear stops.

The first observation concerns the time scale associated with this elastic return. One can see that it did not really stop until the end of the experiment, which indicates a quite long-time reorganization of the internal ink structure. When comparing both inks, it clearly appears that Ink B undergoes a larger return amplitude than Ink A (47 % vs. 33 %). This may be due again to the fastest recovery of Ink A along with its highest viscosity in the low shear stress region (Figure 3). Actually, if physical associations build up faster in Ink A than in Ink B, that will prematurely slow down the ink flow and therefore the long-time elastic return.

Another interesting creep experiment was performed with the aim of simulating to some extent the leveling process. Actually, after being submitted to high shear in the nip and to a large elongation during filamentation, the ink has to lay down and level on the printing substrate, the driving force for this phenomenon being the ink surface tension. The leveling stress imposed by the surface tension will decrease with time, the curvature becoming smaller with the leveling process. An estimated value of the initial stress applied by the surface tension to the ink film is calculated below and falls in the range 10 to 100 Pa. The following procedure was therefore applied to both inks:

- pre-shearing at 20 s^{-1} for 2 minutes
- applying a constant stress and recording % strain vs. time for the next 100s

Various stresses were imposed after the high pre-shear: 3, 30 and 300 Pa. Results are shown in Figures 7,8 and 9 with two different Y-scales, in order to see both the elastic return (negative strain) and the flow imposed by the applied stress (positive strain). The higher the imposed stress, the smaller is the return. Negative strain even disappeared for both inks with a stress of 300 Pa. Figures 7 and 8 clearly show that, whatever the imposed stress, the elastic return has a higher amplitude in Ink B than in Ink A. This confirms that some stronger physical associations may be established in Ink A very quickly after pre-shear stops and therefore may slow down the elastic return. Figure 9 describes what happens on the longer time scales, i.e. after 10 seconds. In fact, the trends observed here seem very relevant to the dynamic gloss development. On the long time scales, Ink B was proven to undergo much more deformation than Ink A: 3300 % vs. 2000 % at 100 s with a imposed stress of 30 Pa and 4500 % vs. 3000 % at 10 s with an imposed stress of 300 Pa. This trend confirms again the weaker structure of Ink B and suggests a higher ability of Ink B to be deformed under a given stress during the 100 seconds following the pre-shear stops.

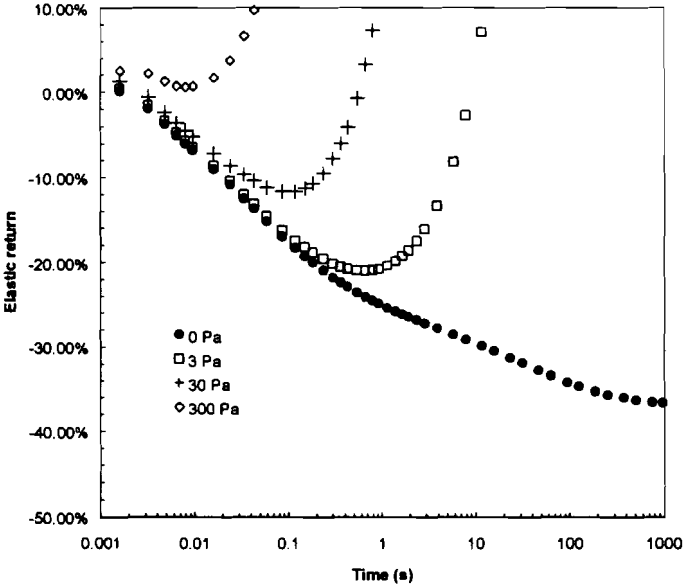


Figure 7. Short time elastic return of Ink A under constant stress: 0, 3, 30 and 300 Pa.

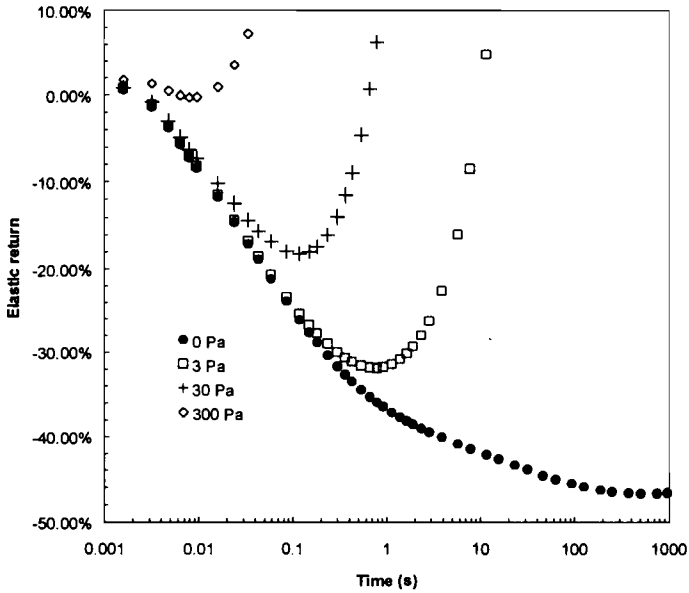


Figure 8. Short-time elastic return of Ink B under constant stress: 0, 3, 30 and 300 Pa.

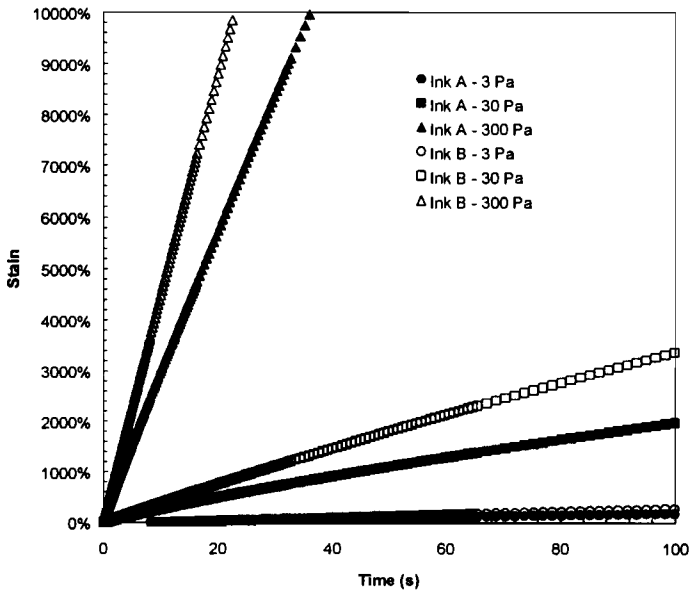


Figure 9. Long-time deformation of Inks A and B under constant stress: 3, 30 and 300 Pa.

Theoretical Development

The surface tension driven leveling of a fluid film on a planar substrate is described by Orchard (1962). In the limit of thin films, where the film thickness is smaller than the wavelength, an initially sinusoidal film profile follows the expression:

$$h = h_0 + \varepsilon_o \sin(2\pi x / \lambda) e^{-\alpha t}$$

$$\alpha = \frac{16\pi^4 \gamma h_o^3}{3\eta \lambda^4}$$

where h_o is the average film thickness, ε_o is the initial disturbance amplitude, λ is the wavelength of the disturbance, t is the time, γ is the surface tension and η is the viscosity. As the disturbance becomes large, surface tension forces become weaker along with more flow required to level the film. The stress that drives the leveling flow comes from surface tension and is the second derivative of the surface profile. The maximum stress is given by:

$$\sigma = \frac{4\pi\gamma\varepsilon_o}{\lambda^2} e^{-\alpha t}$$

The stress decays exponentially as discussed above and depends on surface tension, the initial disturbance amplitude and the disturbance wavelength.

For a viscosity of 50 Pa.s, a surface tension of 30 mN/m, a disturbance wavelength of 100 μm , and an initial amplitude of 1 μm , the initial stress is 38 Pa. The exponential factor is 1.3 s^{-1} . These values seem to correlate quite well with the experimental observations and conditions.

Experimental Model Latex Coatings

Different pigmented coatings were tested. They were prepared using high or low interactivity latex adding a plastic pigment (0.23 μm) inert to the ink mobile phase components. In this work, interactivity refers to the degree of softening or dimensional change of the latex film when in contact with the ink mobile phase components (low molecular weight fraction of polymer and solvent). The high interactivity latex absorbs the ink mobile phase very quickly and to a large

extent. In contrast, the low interactivity latex leads to greater ink hold-out during printing. The degree of cross-linking was established by determining the toluene-insoluble content (percent gel) of the latex film and it was eliminated as a factor affecting the latex film interactivity with the ink vehicle. In this work three methods were used to characterize the interactivity of the two different polymers: an indirect method using the solubility parameter, a direct method monitoring the degree of softening or plasticizing of the latex film in contact with the ink vehicle components and the rate of ink tack-build or slope of the latex film printed on the modified Vandercook press. The latex binder level was varied so that the pigment volume concentration (PVC) ranged from below to above the Critical Pigment Volume Concentration (CPVC). The pigmented latex coatings were prepared on uncalendered coated paper and the coatings were supercalendered. More information is given in Desjumaux and Bousfield (1998).

Experimental Results and Discussions

Inks A and B were printed on Mylar film for which no ink mobile phase removal is involved. Figure 10 shows the print gloss dynamics (voltage values as a function of the time) for the two inks printed on Mylar film for a printing speed of 4m/s and an ink film thickness of 4.0 μm . As time proceeds, the ink film levels on the substrate and the voltage values increase because the ink film surface becomes smoother. The rheological properties of the inks have a strong influence on the print gloss development over time. Right after the printing nip, some strong associations take place in Inks A and B as they level. Inks A and B recover some structure like a pigment reorganization and a physical network. However, Ink A recovers faster and more strongly than Ink B. It was noticed as well that Ink A has a higher degree of structure during the whole experiment. The stronger and faster recovery of Ink A as well as its higher degree of structure slow down the ink flow and limit the leveling to a lower extent. An important observation is that, while both inks have nearly the same high shear rate viscosity, the low shear rate viscosities differ by a factor of 2.0. Theoretically, the time to reach a gloss level of 6 volts for example is different by a factor of around 1.7. The theory states that the leveling time should scale linearly with viscosity. Therefore, the difference in leveling rates between the inks seems to be quite similar to the difference in low shear rate viscosity between the inks.

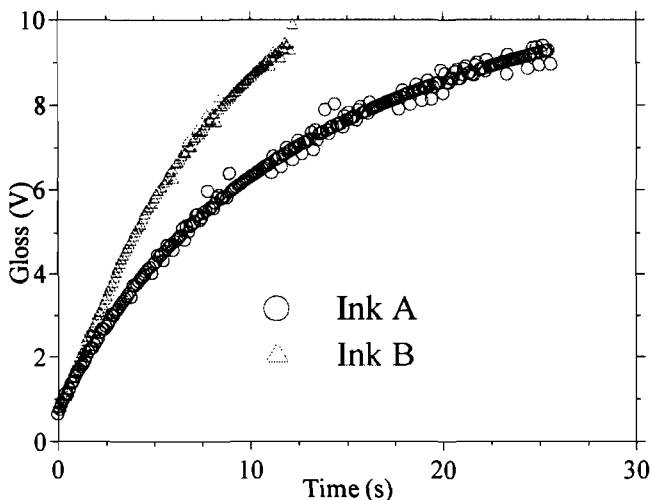


Figure 10. Print gloss development over time for Inks A and B on Mylar film. The voltage values (linearly dependent on the standard Tappi gloss values) are reported as a function of the time.

The print gloss development for both inks printed on the coating containing the low interactivity latex mixed with the plastic pigment at two different PVC levels is shown on Figure 11. Two different substrates were printed so that one would be a fast setting surface and the other, a slow setting surface. Previous measurements given in Desjumaux and Bousfield (1998) showed that the 45 PVC level surface is a slow setting surface: at this PVC level, the latex content is high at the surface. The low interactivity latex covers the surface and does not set the ink so that the ink has time to level. The 89 PVC level surface was showed to be a fast setting surface: at this level, the ink sees more pores than latex. The pores set the ink by capillary absorption and stop the ink leveling earlier (Desjumaux and Bousfield, 1998).

At both latex concentrations, for the low interactivity latex, both inks result in a comparable print gloss development.

At 45 PVC, we would expect to observe the same trend as the one on Mylar film because the coating layer should be rather inert to the ink. However, Mylar and pigmented coatings have different chemical and physical properties: the higher roughness of the coating and its permeability as well may stop the leveling earlier.

At 89 PVC, both Inks A and B do not level to a large amount. Ink A produces a slightly lower print gloss development because at any time as may be expected from its formulation, it contains less diluent. In this case, ink rheology is found to have little importance compared to the coating layer properties in terms of ink gloss.

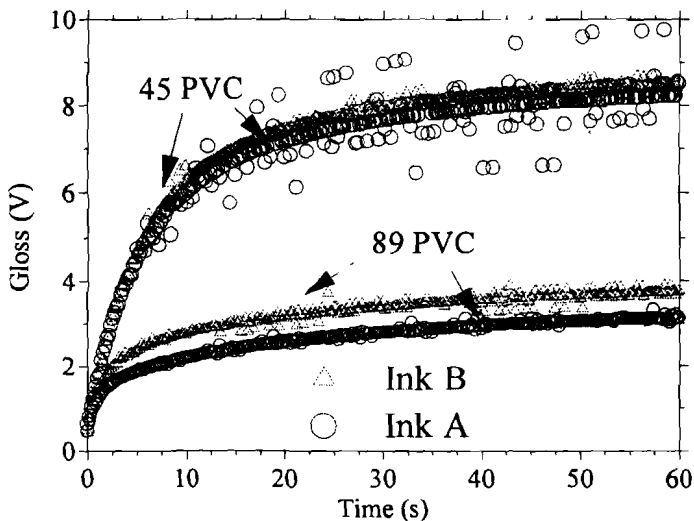


Figure 11. Print gloss development over time for Inks A and B printed on pigmented coatings containing the low interactivity latex mixed with the pigment at 45 PVC level (i.e. slow setting surface) and 89 PVC level (i.e. rapid setting surface).

Figure 12 shows the ink gloss development for Inks A and B printed on the coating containing the high interactivity latex.

The 89 PVC level surface is a rapid setting surface because the ink is set by capillary sorption of the ink mobile phase into the porous structure. The 77 PVC level surface was shown to be a slow setting surface because no significant capillary sorption or diffusion into the latex matrix is taking place (Desjumaux and Bousfield, 1998).

For both PVC levels, Ink A stops leveling earlier and produces lower gloss. However, the mechanisms responsible for this behavior are likely to be different for the two PVC levels.

At 77 PVC level, the surface is not rapidly setting. The ink and the difference in the ink rheology must be important.

At 89 PVC, Ink A containing less diluent which is quickly absorbed into the pore structure, stops leveling earlier and produces lower gloss.

However, even with the rheological and the surface characterizations, it is difficult to separate the influence of ink rheology from the effects of coating composition on these results.

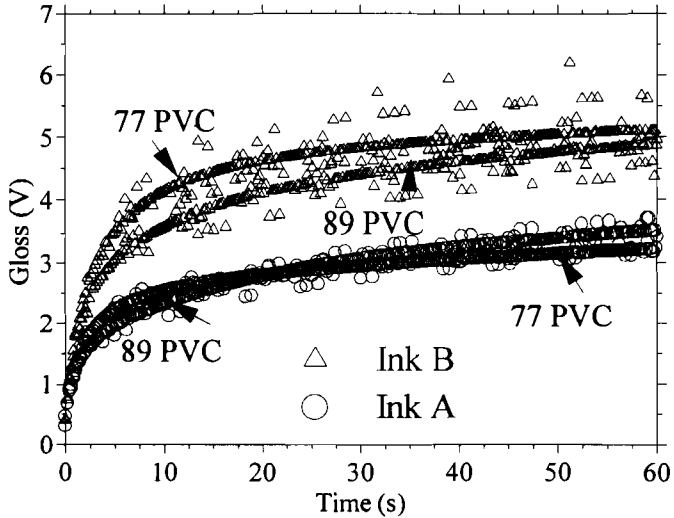


Figure 12. Print gloss development over time for Inks A and B printed on pigmented coatings containing the high interactivity latex mixed with the pigment at 77 PVC level (i.e. slow setting surface) and 89 PVC level (i.e. rapid setting surface).

Conclusions

Two model inks were characterized rheologically and on six different printing surfaces. Even though the inks had nearly the same high shear rate viscosity, their low shear rate viscosity and their rate of structure recovery after shear are quite different. The high setting rate ink had a higher viscosity at low shear rates and a faster and stronger elastic recovery.

Ink rheology did influence the gloss dynamics for the non-setting surfaces. The high setting rate ink gave the lowest ink gloss. On coatings containing the low interactivity latex, ink rheology did not influence the print gloss as much as the coating layer composition. On the coating containing the high interactivity latex, the high setting rate ink gave the lowest gloss and the ink rheology determines the ink film leveling on a slow setting surface.

Acknowledgments

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