Effect of Drying on Coated Paper Print Mottle

Françoise Forel* and W.J.Murray Douglas*

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Abstract: Many researchers have shown that among papermaking processes critical for coated paper print quality, drying is a key step. The objective of the present work is to investigate the potential for changing coated paper properties, especially its printability, by varying drying conditions, including through a new drying parameter. All current non-contact drying of coated paper occurs in an air environment. An experimental drying fluid, superheated steam, was used here at both stages of coated paper production, the base sheet and the coated sheet. The results for drying in superheated steam were compared to those with the conventional air drying fluid. Drying in superheated steam is of particular interest for materials such as paper and coatings for which the properties are affected by the glass transition of its polymer constituents. Paper coated and dried in the two drying fluids in a custom coating-drying facility developed at McGill University was printed on a Prüfbau press using cyan heatset ink in multiple-nip printing configuration. Mottle, evaluated quantitatively as a function of the scale of print nonuniformity, was significantly affected by the drying variables tested. Coated paper drying conditions affected the severity of print mottle, not the scale. Drying in superheated steam led to a more mottled sheet if used for drying the base sheet but gave less print mottle when used just for the coated sheet.

Theoretical Considerations: Print Mottle and Paper Surface Properties

Printing performance relates to the properties at the top of the coated paper. The variability or nonuniformity of these paper properties rather than their absolute values is important to mottle development. It is generally believed that back-trap mottle relates to ink setting (Plowman, 1994). The porous structure controls ink fluid phase transport in the coating and is therefore a key property affecting ink setting. The porous structure adjacent to the surface of the coating may be affected by surface chemical composition, specifically the surface binder content: higher binder content decreases surface porosity. Thus mottle has long been attributed to binder migration. This mechanism is quite possible for a soluble binder such as starch, but migration is rather more difficult in the case of

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* Chemical Engineering Department, McGill University, Montréal, Canada

a latex binder, a solid particle, considering the level of packing of pigment particles as consolidation of the coating progresses (Groves, 2001). In recent investigations using latex rather than starch as the coating binder, no indication of binder migration was found but there is evidence of nonuniform porous structure when mottle occurred (Kim-Habermehl et al., 1998b, Xiang and Bousfield, 2000b). On the other hand the extent of latex film forming rather than binder migration may affect print quality as noted by Yamasaki et al. (1993) and Xiang and Bousfield (2000b). Other components used in coating, such as surfactants or dispersants, may however be transported with water and thereby change surface chemical composition. Finally, coat weight local variation is often identified as an indirect cause of mottle (Matsubayashi and Saito, 1992, Engström, 1994, Eklund et al., 1995, Hashemi et al., 2000) because it affects both the porous structure and binder content.

Print Mottle and Drying Strategy

Among the many papermaking parameters possibly causing mottle - base paper, coating colour, coater type or calendering – the coating drying strategy is most often identified as highly important. Distance of the dryer from the coating application unit, drying rate and the temperature profile are variables directly affecting mottle. Several studies identified a coating solids content range where, to avoid mottle, evaporation rate and temperature should be kept below critical values (Norrdahl, 1991, Kim-Habermehl et al., 1998a among others). Based on such finding on-line drying profiles are used as print quality control tools. The existence of a critical solids content range results from the fact that the effect of drying conditions on coating structure varies with solids content. The coating consolidates in 3 steps delimited by two critical concentrations: first and second critical concentrations termed FCC and SCC (Watanabe and Lepoutre, 1982). When the coating is not yet immobilized (before FCC), drying rate affects coating porosity while after immobilization it does not have much effect. Temperature mostly affects the coating structure during drying before the SCC when latex film forming occurs. It was reported that temperature may still have some effect after the SCC through latex sintering (Xiang and Bousfield, 2000b). Because coat weight is locally nonuniform there is a local variation in moisture content during drying. A locally low coat weight area will reach immobilization faster than where the coat weight is locally higher. If drying rate or temperature is high around the immobilization point, the low coat weight area structure will be much less affected than where coat weight is higher. Consequently it is required to maintain low drying rate and low temperature around the immobilization point in order to avoid the development of local nonuniformity in coating structure.

Superheated Steam Drying

The process of drying paper in superheated steam has been pioneered in the laboratory of the authors, reported in several publications summarized by Douglas (1994). For drying in air, the wet material temperature at first rises slowly to approach the wet-bulb temperature, typically about 50-60°C. For drying in superheated steam at atmospheric pressure, thermodynamics requires that the material temperature jumps essentially instantaneously to 100°C. The use of superheated steam is therefore of particular interest for drying materials, such as paper, for which the properties are affected by the glass transition of its polymer constituents. The glass transition temperature of lignin, for example, is in the range above and below 100°C, depending on the moisture content.

During the production of LWC paper there are two stages at which the sheet could be dried in superheated steam: when drying the base sheet, and when drying the coated paper. Drying a lignin containing base sheet in superheated steam enhances the softening of the lignin, allowing for more development of fibre bonded area and hydrophobicity, Poirier et al. (1994), McCall and Douglas (1994), McCall et al. (1995). These changes in the base sheet may in turn affect the consolidation of the coating when the base sheet is coated. When drying the coating, deformation and film forming of a latex binder may be enhanced by the higher temperature experienced by the wet coating at the onset of drying in superheated steam, with this effect changing the bulk and surface structure of the coating.

Experimental

An investigation was made to determine the effect on coated paper print mottle resulting from: (1) choice of drying fluid, air or superheated steam, for drying the base sheet, (2) choice of drying fluid, air or superheated steam, for drying the coated sheet and (3) coated sheet drying fluid temperature. For these 3 parameters the 8 combinations of drying conditions used are presented in Table 1, along with specification of the neutral reference sheet for paper property comparisons.

The coated paper was produced under fixed conditions with respect to base sheet forming, to coating and to calendering typical of LWC grade specifications, thereby isolating the effect of the range of drying conditions tested. Paper was coated on one side with a suspension of clay and styrene-butadiene latex binder and dried in a custom designed coating-drying unit in our laboratory. This facility consists of a coater closely integrated with an impingement convection dryer which permits use of either the conventional air or the experimental drying fluid, superheated steam. Base sheets made on a Dynamic Sheet Former were dried in air or in superheated steam in the same facility. The coated sheets were calendered on a paper company laboratory soft-nip calender.

Table 1 – Drying conditions

Sheet Designation	Base Sheet Drying Fluid	Coated Sheet Drying Fluid	Drying Fluid Temperature
AA120	AIR	AIR	120°C
AA200	AIR	AIR	200°C
AS120	AIR	SHS	120°C
AS200	AIR	SHS	200°C
SA120	SHS	AIR	120°C
SA200	SHS	AIR	200°C
SS120	SHS	SHS	120°C
SS200	SHS	SHS	200°C
COM	Commercial paper – same base paper furnish – similar coating materials – produced in a mill		

Printing

Solid printing was performed on a Prüfbau Printability Tester at 23°C and 50% relative humidity with cyan heatset offset ink from Sun Chemical used for printing LWC grade. A custom multiple-nip printing configuration was used which consists of a printing unit followed by three printing units without ink thereby allowing for ink back-trapping, see Figure 1. This custom printing method parallels conditions on a commercial four-color offset press. The sequence of one pass on the two-nip Prüfbau followed by a second pass after replacing the inked units simulates four-nip commercial printing. The validity of this procedure has been demonstrated by Waech (1998) and Xiang and Bousfield (2000a). For each drying condition 5 replicate sheets were printed, while from each replicate sheet 2 independent images, 32.5mm x 32.5mm, were taken in reflected light for print mottle analysis. This procedure thereby provided 10 independent determinations of print mottle for each of the 8 drying conditions of Table 1.

The main characteristics of the experimental procedure are summarized in Table 2, with more details provided in Forel and Douglas (2002).

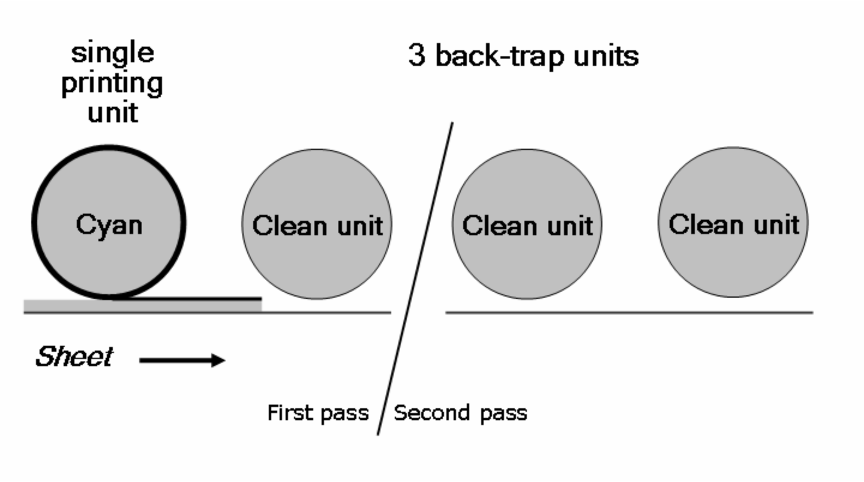

Figure 1 – Custom printing procedure

Table 2 – Characteristics of coated paper and print production

Base sheet production		
Dynamic Sheet Former, 40g/m^2	Impingement drying: AIR/SHS	Light hard-nip calendering
Coated sheet production		
Blade coating, 10g/m^2	Impingement drying : AIR/SHS	Soft-nip calendering
Clay / SB latex / CMC	Coating-to-drying delay time: 0.2s	100°C / 121kN/m
Printing		
Cyan Heatset Offset	Printing speed: 3m/s	Ink weight on plate: 3g/m^2
2-nip Prüfbau press: 2 passes	Load on first unit: 4.15MPa	Load on second unit: 3.13MPa

Results

General

Drying conditions significantly affected the printing pattern obtained with the multiple-nip printing configuration used. When in an earlier study, Forel and Douglas (2002), sheets had been printed in a single-nip printing configuration, very little print mottle was found. Our finding of significant print mottle only after multiple back-trap units is consistent with the experience of Xiang et Bousfiled (2000a). This now confirmed behaviour supports the hypothesis, that at least for the paper grade and ink type tested here, print mottle occurs during ink back-trap (i.e. re-transfer) subsequent to the printing and not during the initial ink transfer.

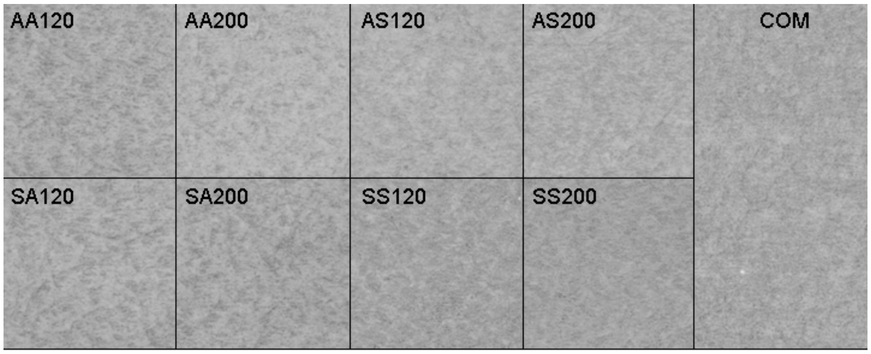

Figure 2 – Mottle of prints

Figure 2 provides example of the various mottling patterns observed with the range of drying conditions tested. Subjective evaluation of print mottle, inherently complex and therefore not standardized, is further complicated by dependence on the optical density of the print. We therefore obtained print mottle from a quantitative determination of mottle.

Instrumental Determination of Print Mottle

Quantitative determination of the nonuniformity of solid prints is likewise a complex problem for which no recognized standards yet exist. The objective technique which was used in this study to evaluate print mottle derives from the commercial “PaperPerFect” instrument developed at McGill University for the determination of paper formation nonuniformity. The PrintPerFect adaptation of this formation analyzer uses a similar algorithm for the measurement of solid print nonuniformity using reflected light, Bernié and Douglas (2001). The technique partitions the intensity of solid print nonuniformity into its components relative to the scale of the print nonuniformity for 9 values of scale of print mottle over the range of scale 0.4-18 mm. The specific values of scale of print mottle used here are 0.4, 0.6, 1.0, 1.5, 2.5, 4, 7, 11 and 18 mm. Thus with this method a determination leads not to a single-number index of print mottle, as in other print mottle evaluation instruments, but to a Print Mottle Line which provides the components of print nonuniformity as a function of the scale of print mottle. There are two basic reasons for partitioning print nonuniformity into its components over a range of scale of print mottle: first to account for the variable significance of scale of mottle with the nature and use of the print, and second to identify and control the causes of mottle.

These print mottle results are most easily interpreted when expressed relative to those for a standard sheet, leading thereby to values of intensity of print nonuniformity in the order of one. In this project, commercial coated paper made from the same pulp furnish and similar coating materials but produced in a paper mill, then printed in exactly the same way was used as the reference sheet for the components of print nonuniformity of our experimental coated paper. Thus the laboratory produced coated paper differs in two ways from the comparative commercial paper: the base sheet is made on a Dynamic Sheet Former and the coated sheet is produced on our laboratory coater-dryer facility. The higher the relative print nonuniformity component, the worse the mottle.

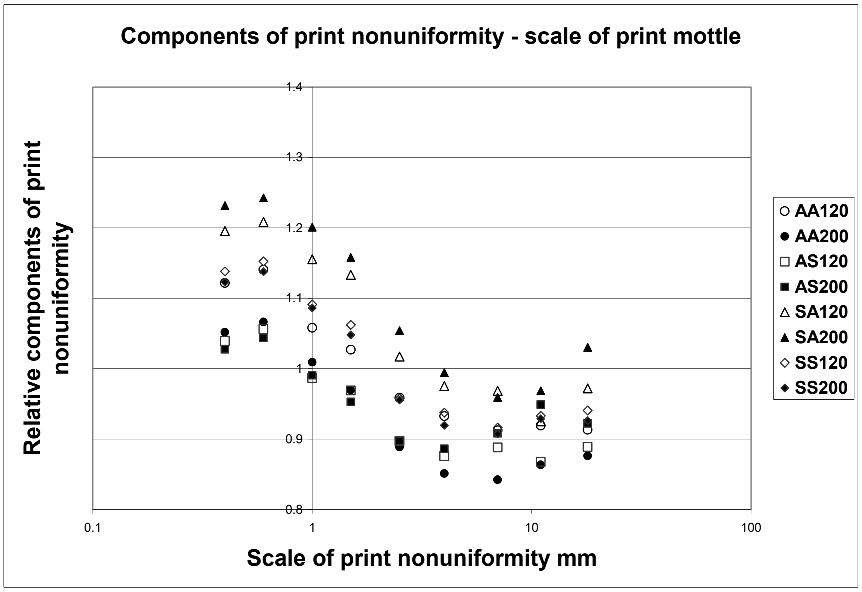

Figure 3 – Components of print mottle

In Figure 3 giving the print mottle results for the 8 drying conditions listed in Table 1, each component of print mottle represents the average of 10 independent measurements, i.e. 2 determinations on each of the 5 replicate sheets for each drying condition. With this method the print mottle is not given on an arbitrary scale. Thus on Figure 3 a value of 1.15 for a print mottle component means that, at this value of scale of print mottle, there is 15% more print nonuniformity than for the reference sheet. Similarly, a print mottle component of 0.9 means 10% less solid print nonuniformity than for the reference sheet. The experimental uncertainty in the determination of the components of print mottle is given in Table 3.

Table 3 – Uncertainty in determination of print mottle components

	Scale of print mottle, mm								
	0.4	0.6	1.0	1.5	2.5	4	7	11	18
σ	0.069	0.069	0.069	0.066	0.057	0.055	0.058	0.073	0.119

At each value of scale of mottle there are 8 distributions for that specific component of mottle, i.e. a distribution for each of the 8 drying conditions. As the dispersion of these distributions is not significantly different between the 8 conditions tested, at each value of scale of mottle it is valid to combine the 8 distributions in order to determine a single value of standard deviation, using standard statistical procedures. As the mean values of the print mottle components are in the order of one, i.e. from about 0.85 to 1.25, the coefficient of variation is therefore about the same as the value of standard deviation.

Scale of Print Mottle

Figure 3 reveals a complex relationship for this experimental paper between print mottle and scale of mottle. On one hand the mottle of the experimental paper relative to that for commercial paper varies with scale of mottle. The overall behaviour for the components of mottle is that near both the lower and upper limits of the range of scale of mottle there are plateau regions where the mottle of experimental paper relative to commercial paper is little affected by scale of mottle, while over the intermediate range of scale the intensity of mottle changes considerably with scale of mottle. Over the lower range of scale of print mottle the intensity of mottle of the experimental paper is greater than that for commercial paper, but over the upper range of scale the effect is just the opposite, i.e. mottle on the experimental paper is less than that on commercial paper. This switchover occurs in the range of 1-4mm scale of mottle, the exact value depending on the conditions used for drying the base sheet and the coated sheet.

On the other hand, between the various drying conditions the mottle of the experimental paper does not vary in scale but only in intensity. Examination of the results on Figure 3 indicates that, for the 8 drying conditions tested, for increasing scale of mottle from 0.4 to 7mm there is no crossing of the Print Mottle Lines, while for scale of mottle from 7 to 18mm there is only very limited crossing of Print Mottle Lines in a few cases (AA200, AS200 and SA200), such crossing being within the limits corresponding to the uncertainty in the determination from Table 3. The conclusion is that these drying conditions change the intensity of print mottle substantially but these changes are similar in magnitude over the 0.4-18mm range of scale of print mottle of this test method. Therefore for subsequent analysis the values of the print mottle components at the intermediate value of print mottle scale of 1.5mm is used as indicative of the effect of a drying conditions on intensity of mottle. With use of this 1.5mm component of print mottle as the basis, Table 4 lists the intensity of print mottle for the 8 drying conditions tested.

Table 4 – Components of print mottle at 1.5mm scale of mottle

Components of print mottle at 1.5mm								
AA120	AA200	AS120	AS200	SA120	SA200	SS120	SS200	COM
1.03	0.97	0.97	0.95	1.13	1.16	1.06	1.05	1

It is now possible to examine the hypothesis that the difference in base paper forming technique is the principal source of the consistent trend apparent in Figure 3 for the difference in print mottle between the laboratory and industrial coated paper. Thus Figure 4 shows the components of formation nonuniformity determined with transmitted light by the PaperPerFect method for the Dynamic Sheet Formed laboratory base paper using the same reference paper as for the print mottle determination of Figure 3, i.e. the equivalent commercial LWC base paper.

The Base Paper Formation Line is the average of 40 measurements: 4 for each of the 10 sheets tested. On Figure 4 both the Paper Formation Line for the base paper and the mean Print Mottle Line for the coated paper are shown on the same basis. Thus higher values indicate more nonuniformity, worse formation for the base paper and worse mottle for the coated paper. Figure 4 displays a significant similarity between the Paper Formation Line for the base paper and the mean Print Mottle Line for the coated paper. This finding supports the hypothesis that base sheet formation determines the scale of mottle while sheet drying conditions affect the amplitude of mottle.

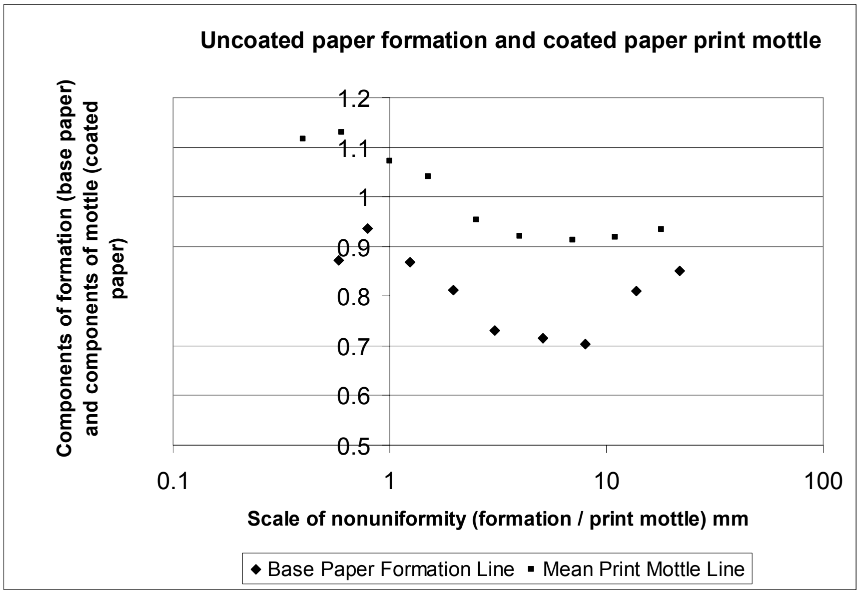

Figure 4 – Base Paper Formation Line and Mean Print Mottle Line

It is a basic characteristic of the mathematical procedure for partitioning formation or print mottle nonuniformity into components that the uncertainty of this determination increases towards the upper limit of scale. Thus it is quite possible that the base sheet formation becomes a plateau over the upper range of scale, just as seen for print mottle.

Intensity of Mottle: Effect from Base Sheet Drying

It is immediately apparent from Table 4 that the intensity of print mottle is greater for all 4 cases for which the base sheet was dried in superheated steam than for any of the 4 cases with the base sheet dried in air. The average value of the 1.5 mm print mottle component is 1.10 for the 4 cases with steam dried base sheets, and is 0.98 for the 4 conditions with an air dried base sheet. On average values of print mottle intensity from 40 determinations for each of these base sheet drying conditions, this difference of 12% is statistically significant.

As to why there is this clear difference in coated sheet print mottle when the base sheet is dried in superheated steam, the study of McCall et al. (1995) showed definitively that the mechanism by which the properties of mechanical furnish paper are affected is that by drying in superheated steam the wet lignin undergoes transition from the hard crystalline to soft amorphous state. In the latter form the micro-flow by the softened lignin results in better interfibre bonding and thereby produces a substantially stronger and more hydrophobic sheet. Another consequence is seen in the present study: when such a superheated steam dried base sheet is coated, the coated paper shows more print mottle. More research would be required to determine the mechanisms through which the steam dried, mechanical furnish base sheet leads to an average of 12% more print mottle intensity.

Intensity of Mottle: Effect from Coated Sheet Drying

With the 95% confidence limits at 1.5mm scale of mottle being about 0.05, the four cases with air dried base sheets listed in Table 4 show that there is no significant effect on print mottle coming from the effect of drying temperature 120 or 200°C. As for the effect of the choice of drying fluid for drying the coated sheet, air or superheated steam, there is a statistically significant trend for lower mottle with using SHS for drying the coated sheet. This conclusion is also supported by visual print evaluation. If a larger number of sheets were examined, the correspondingly smaller confidence limits would permit more precision in determination of these effects.

The effects from conditions of coated sheet drying on print mottle may be compared with effects with respect to ink coverage, optical density and print gloss, as summarized in Table 5.

Table 5 – Effect of drying conditions on printing properties

Sheet designation	**Ink coverage** %	**Optical density** -	**Print gloss** °
AA120	41.4	0.75	37.7
AA200	41.3	0.73	37.5
AS120	45.4	0.77	31.5
AS200	48.2	0.79	30.6
SA120	42.3	0.78	37.6
SA200	43.0	0.79	33.8
SS120	45.5	0.80	31.3
SS200	46.8	0.85	28.2
COM	46.9	0.80	27.1
Change to 200°C	+1.2%	+0.01	-2.0
Change to SHS	+4.5%	+0.04	-6.3

The average values of the changes due only to the use of drying at 200°C instead of 120°C, and due to the switch from drying in air to drying in superheated steam at the same temperature are also recorded in Table 5. As for the case of print mottle from the Table 4 results, the change in a drying fluid temperature has either negligible effect or a much smaller effect than a change in drying fluid. For the switch of drying fluid from air to the experimental fluid, superheated steam, significant changes are seen in Table 5. For drying in steam, the average ink coverage increases from 42.0 to 46.5%, the optical density from 0.76 to 0.80, and print gloss decreases from 36.6 to 30.4°, i.e. ink coverage and optical density improved but print gloss decreased. Further observation of printed sheets with light microscopy also revealed noticeable differences in ink distribution between sheets for which the coating was dried in superheated steam versus those in air, as illustrated in Figure 5.

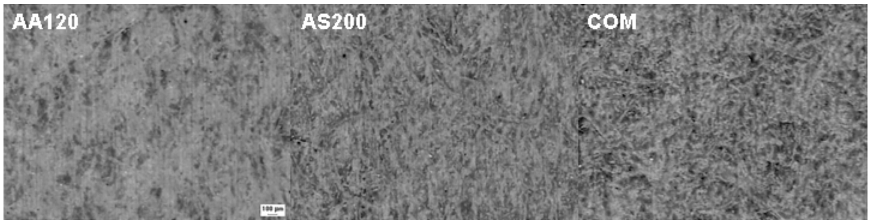

Figure 5 – Ink distribution at microscale – individual image width ~ 3mm

There are several arguments supporting the idea that coating surface porosity is responsible for the differences arising from conditions used to dry the coating. First of all, mercury porosimetry results indicate higher porosity and a wider pore size distribution when the coating was dried in SHS. Secondly SEM images of uncalendered coated sheets clearly showed a more open and disrupted surface for sheets dried in superheated steam. These findings are consistent with the lower print gloss and higher ink coverage reported in Table 5. Finally, a different surface porosity was proposed earlier, Forel and Douglas (2002) to explain, for a single-nip printing configuration, the finding on superheated steam dried coated sheets of a pattern of white dots through inspection of the prints by optical microscopy and SEM.

Using SHS for Coated Paper Grade Drying

We have investigated the effect on print mottle of using superheated steam for the drying of the LWC coated paper grade. For production of base sheet, drying in superheated steam should not be used as mottle is worsened. A switch from drying in air to drying in superheated steam would also change paper production costs from such effects as changing the drying rate, elimination of the loss of energy associated with the discharge of humid air to the atmosphere etc., but such aspects are beyond the subject of the present paper. By contrast, using SHS only for drying the coating drying improves mottle. Although the sample size is small in the present exploratory study, with this reservation the improvement in print mottle from drying coating in superheated steam is somewhat better at the 95% confidence limit. Also, visual observation of print mottle supports this finding.

Summary and Conclusions

1. Coated paper print mottle was determined for base paper formed on a Dynamic Sheet Former then coated and dried in the laboratory. All measurements were compared to the same grade of paper formed and coated commercially. Testing involved 8 combinations of drying conditions involving both the base paper and the coated paper. Print mottle was determined quantitatively by a new, very informative method in which solid print nonuniformity is partitioned into its components over the range of scale of mottle 0.4-18mm.
2. Large effects on print mottle were found to derive from both the switch between machine-formed base sheet formation and that from a Dynamic Sheet Former, and from changing base sheet or coated sheet drying conditions.
3. Variation of components of mottle with scale of mottle was found to relate to the variation of formation components with scale of formation.
4. By contrast, drying conditions affected the intensity of mottle rather than its distribution relative to scale of mottle. Superheated steam drying of the base sheet made print mottle worse while superheated steam drying of the coated sheet decreased print mottle.

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