PRINT MOTTLE OF WOOD-CONTAINING PAPER:
THE EFFECT OF FINES AND FORMATION

by

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A thesis submitted in conformity with the requirements
for the degree of Doctor of Philosophy
Graduate Department of Chemical Engineering and Applied Chemistry
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0-612-41218-0
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Doctor of Philosophy

1998

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Abstract

The spatial unevenness of print density, print mottle, is due to the small-scale variation in the ink transfer and in the ink penetration depth. Print mottle is affected by both paper properties and printing conditions, and is a principle factor affecting the printability of paper. The objectives of this study were to develop a sensitive method to describe print mottle, to conduct a fundamental study of the effects of paper properties on print mottle of wood-containing paper, and to develop a model to predict the print mottle tendency of paper. The principle pulp used in the study was thermomechanical pulp and the printing method used was an IGT proof printer to simulate offset printing.

A texture analysis method, based on the spatial gray level dependence method (SGLD), was developed and used to evaluate the structural characteristics of both the mass density map and the print density map of handsheets, dynamic former sheets and machine made paper. Texture features of homogeneity, contrast and correlation, combined with the
coefficient of variation (CV) gave a complete description of the uniformity. Structural variation and print density variation due to changes in orientation, wire marks and uneven mixing were identified by the texture analysis method.

Four types of sheets: handsheets, dynamic former sheets, pilot machine sheets and commercial newsprint were used to determine the effects of paper structure on print density. The fines content level of the handsheets and dynamic former sheets were controlled to 0%, 20%, 30% and 40%. Formation was varied by changing the settling times, i.e. the time before drainage. While the fibre orientations in the dynamic former sheets were changed by varying the jet/wire ratios. Print mottle was affected by the fines content level, as well as the fines distribution, and by the formation level. The handsheet results indicated the importance of not only the absolute amount of fines, but also the uniform distribution of fines on print mottle. The study on oriented samples showed that sheets with 30% fines gave the best printing results, and the structure of these sheets was close to that of commercial newsprint. The principle print mottle factor in wood-containing paper was shown to be the uniformity of ink penetration, a conclusion supported by a pointwise study of the dependence of the local print density, mass density and ink amount for newsprint sample.

A model was developed based on a two-layered Kubelka-Munk model to relate print density variations to other variations. The model is able to predict the experimental observations such as a 30% fines content in sheets made with a dynamic former is an optimal fines level for offset printing.
Acknowledgments

To my supervisor, Professor David Kuhn for the excellent guidance and support.

To my committee members: Professor David Goring, Professor Mark Kortschot and Professor C.T.J. Dodson for their invaluable suggestions and recommendations.

To the defense committee members: Dr. Joe Aspler, Professor G. Allen and Dr. Z. Tan for their many wonderful questions and suggestions.

To the friendly people at Abitibi-Consolidated Technology Center in Mississauga, Xerox Research Center in Mississauga, Paprican in Montreal and the state key laboratory of Pulp and Paper Engineering in South China University of Technology for letting me use their facilities and assisting me with my experimental work.

To my friends in the Department of Chemical Engineering and Applied Chemistry: Hai-Hui Lin, Karen Liu, Tie Mao and Ramin Farnood, for their helps and encouragement.

To my parents and my relatives for their generous support and encouragement.
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Glossary of Symbols

A  coverage factor (section 2.5)
    ink penetration area (chapter 6)

a1, a2, a3 and a4  constant

B  number of “black” zones (section 2.1.2.1)
    immobilization factor (section 2.5)

CV  coefficient of variation

CON  contrast

COR  correlation

ENG  energy

D  optical contrast between the printed and unprinted surface

d  diameter of pore ((section 2.3.2.2)

G  roughness value (section 2.3.1.3)

h  ink penetration depth (section 2.7)

h_w  whole ink penetration depth

h_s  penetration depth of solvent after leaving the printing nip

HOM  homogeneity

K  proportional constant (section 2.3.1.3)

K_i  light absorption coefficient of ink

K_p  light absorption coefficient of paper

k  rate of recovery parameter (section 2.5)

PMN  print mottle number

p  applied pressure (section 2.3.2.2)
    nip pressure (section 2.7)

p (i,j)  normalized probability of gray level, or print density, or mass density pair i, and j

Q  air flow rate (section 2.3.1.3)
$R$ reflectance
$R_\omega$ reflectance of an opaque pad of the unprinted paper
$R_p$ reflectance of the printed paper placed over a pad of unprinted paper
$R_0$ reflectance of unprinted paper under black background
$R_q$ reflectance of the reverse side of printed paper under opaque pad background
$R_{cm}$ reflectance of mixed layer
$R_{exp}$ reflectance of unprinted paper
$r$ correlation coefficient (chapter 5)
pore size (chapter 6, chapter 2)
$S_m$ scattering coefficient of mixed layer
$S_i$ light scattering coefficient if ink
$S_p$ light scattering coefficient of paper
SND sum of the absolute optical density difference among all neighboring zones in the density map
$t$ nip dwelling time (section 2.7)
$U_i$ print evenness
$V$ ink volume (section 2.7)
$W$ number of “white” zones (section 2.1.2.1)
$W_m$ grammage of the ink penetrated layer
$x$ ink weight on the plate (section 2.5)
$y$ ink weight on the sample (section 2.5)
$Z$ Caliper of paper
$\beta$ whole grammage (section 2.6)
$\beta^-$ zonal grammage (section 2.2.3.1)
$\beta^+$ average grammage (section 2.2.3.1)
$\rho_p$ apparent density of paper (section 2.3.2.1)
$\rho_r$ solid phase density of paper (section 2.3.2.1)
\[ \varepsilon \quad \text{void factor of paper} \]
\[ \gamma \quad \text{surface tension of the wetting liquid (section 2.3.2.2)} \]
\[ \tau \quad \text{tortuosity} \]
\[ \eta \quad \text{fluid viscosity (section 2.7)} \]
1. INTRODUCTION

Print density is the optical contrast between a printed and unprinted area. The spatial unevenness of print density is called print mottle and it is considered to be the most important factor affecting printability [1,2]. A mottled image is spotted or blotched in appearance due to the small-scale variation in the ink transfer and/or in the penetration depth of the ink. Although print mottle is a well known problem, there is no standardized, or commonly accepted method to measure print mottle.

Print mottle is affected by both paper properties and printing conditions. The process of ink transfer and setting can be described as follows [3]: during printing, a hydraulic impression acts on the ink and forces it into the voids in the surface of paper; some of the ink then penetrates into the porous structure of the paper. At this stage, the ink penetrates as a homogenous mixture of the ink pigments and vehicle oil. The remaining free ink film is then split at the exit of the printing nip [3,39]. Once the printing pressure is released, the movement of the ink pigments will slow down and the vehicle oil will be drawn into the paper structure by capillary penetration and spreading. There are three factors controlling the interaction between ink and paper in monochrome printing [3,39]: 1) contact between the ink film on the printing plate/blanket and the paper, which mainly depends on the topography, printing pressure and speed; 2) immobilization of ink in the paper, which mainly depends on the porous structure of paper, printing pressure and ink rheology; and 3) splitting of the free ink film, which is affected mainly by the printing speed and ink rheology. The above factors indicate the importance of paper properties, such as surface topography, porous structure, and formation on the uniformity of printing.
Several studies on the effects of formation and other paper properties on print mottle for specific paper grades and printing methods have been conducted [4,5,6,7,8,9,10,11]. However, the fundamental principles underlying these effects are not clearly understood.

It is known that fines play an important role in paper made from mechanical pulps. The fines are essential for both the strength and optical properties of paper. To date, studies on the effects of mechanical pulp fines on printing properties of the offset printing process have mainly focused on the problems such as picking or linting, and print-through. Although print mottle is another important print quality factor, there are few fundamental studies of the effect of fines content on the print mottle of wood-containing paper. It is generally believed that the formation and surface smoothness of paper will be improved with the increasing fines content. It is expected, therefore, that a higher fines content will decrease the problem of print mottle.

The present study was carried out in order to better understand the underlying fundamental effects of paper properties on print mottle. The objectives of this study were: 1) to develop a sensitive method to measure print mottle; 2) to study the effects of paper properties, such as formation and fines content, on print mottle; 3) to develop a model to predict the above effects on print density and its uniformity.

Non-oriented and oriented sheets were made from 100% thermomechanical pulp (TMP) and offset printed using an oil-based carbon black ink. This work focuses not only on the
overall print mottle tendency but also on the pointwise dependence between local print density, ink transfer and local mass density. An analytical model of the effect of formation and fines content on the print mottle was developed and qualitatively compared with the experimental data.

Chapter 2 presents relevant literature on printing processes, interrelationship of ink and paper, and effects of paper properties on print density and print mottle. It also addresses issues regarding some important paper structural properties such as formation, porosity and fines content. Chapter 3 presents the initial parameter identification. Experimental methods are described and a new print mottle measurement method is developed in Chapter 4. Chapter 5 discusses the overall print mottle tendency of both non-oriented sheets and oriented sheets with different formation and fines contents, and the pointwise dependence between local print density, ink transfer and local mass density. An analytical model of the effects of fines and formation on print mottle is developed, and its application is discussed in Chapter 6. Conclusions and recommendations are given in Chapter 7.
2. LITERATURE REVIEW

The literature review is arranged into three sections: Section 1 presents information on printing processes, and focuses on the effects of paper properties on print density and print mottle; Section 2 addresses issues regarding some important paper structural properties such as formation, porosity and fines content; and Section 3 discusses the ink and paper interrelationship, and presents models developed in this area.

2.1 Print Mottle of Paper

Paper products are printed by either the traditional impact printing processes of letterpress, flexographic, offset and gravure printing or by electronic non-impact printing processes. While the letterpress printing process prevailed in the 60’s and 70’s, currently, offset printing accounts for over 70% of the circulation printing [12]. This section of the review will focus on the studies of print mottle on both letterpress and offset printing processes. Some background knowledge on printing processes will be presented first, followed by a discussion on the effects of paper properties on print density and its uniformity for certain grades of paper and printing processes.

2.1.1 Printing Processes

In the letterpress method, a hard raised surface is inked and the image is printed on the paper under high pressure, as shown in Figure 2-1 [13]. This is the earliest form of printing, and has been replaced by other methods due to its poor print quality.
Figure 2-1. Schematic diagram of the letterpress printing process configuration [13]

Offset printing is an indirect printing method, see Figure 2-2 [13]: the inked image is transferred from the plate to a rubber blanket and then to the paper. The surface of the plate and blanket is flat. Chemicals are applied to the printing plate to make the printing image areas receptive to oil-based ink and repellent to water, while the non-image areas are water-receptive and ink-repellent. A number of advantages are attributed to the offset method, including lower cost plate, shorter make ready time and better print quality due to a better contact as compared with the letterpress method.
In the flexography printing process, the image carrier is a raised flexible rubber or photopolymer plate. The principle of this method is similar to the letterpress process as shown in Figure 2-3 [13]. Flexographic inks are rapidly drying, highly fluid-like water-based inks, and these inks are dried by evaporation or absorption with or without the aid of heated air. There are fewer paper problems, such as picking, in flexography than in other printing processes. However, smooth and level surfaces are essential for good halftone detail and low printing impression with minimum mottle and dot distortion [14].
Gravure is printed from a sunken image, which is lower than the non-printed surface of the printing plate. The copper printing plate or cylinder is engraved with microscopic cells. A low viscosity ink fills these cells and is then transferred to the paper as shown in Figure 2-4 [13]. This printing method requires paper of exceptional smoothness to enable a uniform surface contact between printing plate and paper, so that the ink residing in the recessed cells can be transferred properly. The economy of gravure printing process is associated with the scale. For a relatively small scale operation, the cost is much higher compared with other printing processes.
2.1.2 Print Density and Print Mottle

Print density, $D$, is the optical contrast between the printed and unprinted surface and is defined as,

$$ D = \log \left( \frac{R_\infty}{R_p} \right) $$  \hspace{1cm} (2-1)

where $R_p$ is the reflectance of the printed paper placed on top of a pad of unprinted paper, and $R_\infty$ is the reflectance of an opaque pad of the unprinted paper [14]. Print mottle is defined as the variations in print density over a printed area, where there is no optical density variation in the original image.

2.1.2.1 Measurement of Print Mottle

Although print mottle is a well-known problem, there is no standardized or commonly accepted method to describe print mottle. Some researchers simply ranked the mottle
status of the samples by subjective ranking [1,11]. Several studies have been conducted using image analysis and data analysis techniques. The image of the printed paper is captured by a charge-couple device (CCD) camera or a scanner and digitized. The variations in print density are characterised by different statistics measures [2,4,5,9,11]. Vanya [2] introduced a semi-imperid parameter: print mottle number (PMN) for newsprint evaluation. PMN is determined from Equation 2-2 [2],

\[
PMN = \{(B_{0.03} + W_{0.03})/100\} \times 100 + \{(SND \times 100)/13.68\} - 100 \\
+ \{(B_{0.04} + W_{0.04})/(B_{0.02} + W_{0.02})\} \times 100
\]

(2-2)

where PMN is the print mottle number; B is the number of "black" zones; W is the number of "white" zones; subscripts 0.02, 0.03 and 0.04 are the thresholds used to define the "black" and "white" zones. For example, if the print density of the zone is 0.02 lower than the average print density, it is defined as a white zone according to the 0.02 threshold. SND is the sum of the neighboring differences, or the sum of the absolute optical density differences among all neighboring zones in the density map. The coefficient of variation and the standard deviation of print density have been used for print mottle measurement by Kajanto [4], as well as Miwata, et al. [11]. However, coefficient of variation of print density is an overall evaluation of the printing unevenness, and does not take into account the texture structure of the print, i.e., the local variations between neighboring zones. The location of the zones is a crucial factor characterizing the printing uniformity. Figure 2-5 [15] shows a series of images with the same coefficient of variation. It is obvious that the appearances of these images are different from one another, but they all have the same coefficient of variation (13.4%).
Thus, the coefficient of variation alone is not sufficient to describe the uniformity of printing accurately.

![Images with a coefficient of variation of 13.4%, (a) an uneven image, (b) a random image, (c) a pattern image [15]](image)

Figure 2-5. Images with a coefficient of variation of 13.4%, (a) an uneven image, (b) a random image, (c) a pattern image [15]

Jordan [30] developed a index, specific perimeter, to take into account the size distribution of the nonuniformities. Specific perimeter is defined as the length of the median-density contour line per unit area of sample.

### 2.1.2.2 Print Mottle and Paper Formation

Print mottle is the variation of the print density resulting from the uneven distribution of ink at a small scale. It is clear that the absorption properties and surface topography are essential factors affecting ink transfer. The role of formation on print density uniformity may be due to its effects on the variations of porosity, surface properties and ink absorption of paper [4].

The smoothness and porosity of a paper are significantly dependent upon the uniformity of its formation. An paper with poor formation may have “hills” and “valleys” on the surface of paper, especially before calendering. Kajanto [4] demonstrates a correlation
between the formation of wood-free offset paper and the print quality. A similar relationship has been found in Vanya’s work on newsprint [2]. Calendering will level out and eliminate the surface hills which improve the smoothness of paper, and as a result, reduce print mottle [4]. However, studies indicate that this improvement is limited, especially for hard-nip calendering sheets [4,7]. These results imply that not only surface smoothness, but also other properties, such as porosity and local mass density are important.

In terms of the effects of pore structure on print mottle, Miwata [11] investigated the influence of the micro-porous structure of the coating layer on the print mottle of high grade coated paper. The results show no relationship between these two parameters. On the other hand, Xiang and co-workers [69] conclude that the presence of the closed areas is the most important factor for backtrap mottle. A study on the ink distribution was conducted on the scale of a single fibre, using a scanning electron microscopy (SEM) technique [16]. This work studied the effect of local topography of a fibre on the ink transfer, as well as the effect of ink types on the penetration of ink into the interfibre structure. The results provide a qualitative understanding on the mechanism for ink transfer, and indicate that the penetration of ink into the interfibre voids is limited by the high viscosity of the offset ink. Thus, this result may indicate that some of the previous studies [17,18] overestimated the ink penetration under capillary forces. Further study is needed in order to draw a conclusion on the effect of pore size and distribution on print mottle.
2.2 Formation of Paper

Two scales of mass density variations exist in paper: small-scale and large-scale variations. Small-scale variations are due to natural fibre flocculation and flocculation induced by high frequency pulsation. Large-scale variations are caused by the variability in the manufacturing process [19]. Formation refers to small-scale grammage variations and is defined by the International Standards Organization as “the manner in which the fibres are distributed, disposed, and intermixed to constitute the paper” [19]. Formation is a fundamental property and it affects nearly every other paper property and its uniformity. As discussed in the previous section, formation is also an important factor affecting print mottle.

2.2.1 Methods of Determining Grammage

Formation was first evaluated by the “look-through” method. In this method, the uniformity of the sheet is subjectively evaluated by visually judging the pattern of paper under transmitted light. The first formation tester was introduced more than sixty years ago by Davis [20] to correlate the distribution of light transmission of paper (optical density) to the mass distribution of paper. The application of light absorption methods is limited since the light transmission of paper is affected not only by the mass but also by other factors such as coating, fillers, and bonding [21,22,23]. These shortcomings can be overcome by the beta radiography method [24,25,26]. In this method, the absorption of beta rays depends mainly on the mass due to the very low scattering power of beta rays. A sheet of paper is placed in contact with a uniform radiation intensity source (usually carbon-14 labeled poly-methylmethacrylate). The transmitted radiation through the paper
is recorded on an x-ray film. This film is then developed and the radiograph map of the paper sample is analyzed by a microdensitometer or an image analysis system.

Other imaging techniques such as soft x-ray microradiography and electrography are also used for paper formation measurement [25]. Tomimasu, et al. [25], conducted studies to compare the above four imaging techniques and concluded that both beta radiography and electrography are suitable for formation measurement in terms of good resolution and contrast.

2.2.2 Sample Size and Aperture Size

Since the size of a fibre flocs can vary from 1 millimeter up to a few centimeters, it is essential to select a sample size that is large enough to include all floc sizes and small enough to exclude large-scale variations. A measuring size from 50-100 mm is considered to be suitable for formation investigation [21,27].

The measuring area is divided into small inspection zones, and the mass density data of each small zone are recorded. The size of inspection zone is called the aperture or zone size. The variance of mass density over the measuring area decreases with increasing aperture size [26,28]. An extremely small aperture size will focus on the variation between single fibres; and a large aperture size will mask the variation within the zone and decrease the resolution of the measurement. In general, aperture sizes lay in the range of between 0.1 mm and 1 mm [21].
2.2.3 Characterization of Paper Formation

Once the mass density map of the paper is obtained, the next step is to provide a description of the uniformity of this map. Several techniques have been introduced, including statistical analysis, frequency analysis, and texture analysis.

2.2.3.1 Coefficient of Variation (CV)

The intensity of the grammage variations can be described by standard deviation and coefficient of variation. The standard deviation is not a dimensionless function and is not suitable for comparing samples with different grammages. The coefficient of variation is the ratio of the standard deviation over the mean grammage [26],

\[ CV(\tilde{\beta}) = \frac{\sigma(\tilde{\beta})}{\tilde{\beta}} \]  \hspace{1cm} (2-3)

where \( \sigma(\tilde{\beta}) \) is the standard deviation, \( \tilde{\beta} \) is zonal grammage and \( \bar{\beta} \) is the average grammage. This parameter provides information on the extent of the overall variation of mass density, but this parameter by itself fails to describe the spatial arrangement of mass density. Dodson [26] uses the CV over a whole range of inspection zone sizes as a "fingerprint" of the mass density distribution of paper.

2.2.3.2 Frequency Analysis

Frequency analysis focuses on the contributions of different floc sizes to the variations of mass density [23,29]. A sheet of paper is scanned at a constant speed and the signal is recorded. When the sensor passes a small floc, high frequency variations are recorded;
while for a large floc, low frequency variations are obtained. This signal is then analyzed using Fourier analysis and the power spectrum, i.e., the frequency spectral density $P(v)$ of the variance of the signal over different frequencies are used for formation measurement. This method is sensitive to periodic variations such as wire marks of paper. But for paper samples with stochastic variations, it is difficult to interpret the frequency curve, which made it impossible to use this method as an on-line control unit.

2.2.3.3 Texture Analysis

Texture analysis focuses on providing information on the textural structure of paper [30,31]. Jordan et al. [30] introduced the specific perimeter, which is the sum of the total length of the contour of the median mass density area, to emphasise the average size of the flocs.

Cresson et al. [15,31] present a texture analysis of paper mass distribution using the spatial gray level dependence method (SGLD) [32,33,34] to analyse a digitized video beta radiographic (VBR) image of paper. In this method, the digital image is scanned along the four principle directions: horizontal, vertical, left diagonal and right diagonal. The probability of occurrence of various predefined gray level transition patterns is computed and stored in intermediate matrices. Based on the information provided by these co-occurrence matrices, five texture features: energy, contrast, correlation, entropy and homogeneity are determined to describe the mass distribution in paper. The significance of these five features is presented in Table 2-1. The advantage of this method is that it accounts for the spatial arrangement of flocs.
<table>
<thead>
<tr>
<th>Features</th>
<th>Properties measured</th>
<th>Significance for paper formation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Low</td>
</tr>
<tr>
<td>Energy</td>
<td>Uniformity</td>
<td>Poor</td>
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<tr>
<td>Contrast</td>
<td>Transitional irregularities</td>
<td>Good</td>
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<tr>
<td>Correlation</td>
<td>Linearity</td>
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<tr>
<td>Entropy</td>
<td>Disorder</td>
<td>Good</td>
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<td>Homogeneity</td>
<td>Uniformity, emphasise on the</td>
<td>Poor</td>
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<tr>
<td></td>
<td>contribution of diagonal entries</td>
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</tbody>
</table>

Table 2-1. Significance of the Second-Order Statistics Features [15]

2.3 Paper Structural Properties

The surface of paper is the primary bearer of the print. An ideal paper surface for printing should be a surface that is able to accept, retain and present the ink to the observer in an optimum manner. This means that paper should be smooth enough to provide good contact, and the pore size should have a suitable mean size and uniformity to absorb ink evenly and provide good visibility of ink. The porous structure of paper depends on the components of paper and their spatial arrangement. These components include fibres, fines, fillers and coatings, and the spatial arrangement is influenced by the formation of paper and the degree of pressing and calendering.
2.3.1 Characterization of Surface Roughness of Paper

2.3.1.1 Surface Topography Profile

Surface topography measurements [35,36,37] scan the topography of a paper surface using a stylus or other devices such as lasers, and then present the information as 3-D profiles and contour maps of the paper. Surface roughness can thus be calculated from these data. The measurement results of these methods are quite accurate; however, the drawbacks of these methods are that the measuring areas are small ranging from several hundred micrometers to several centimetres, and the measuring procedure may also be time-consuming.

2.3.1.2 Fractional Contact Area

Smoothness of the surface of paper is determined by the fractional contacting area between the sample and a reference plate, under certain defined static or dynamic pressure conditions [38]. For example, the Chapman smoothness tester [38] measures the optical contact between a paper sample and a glass surface. The smoothness of a paper is determined by the percentage of the contacting areas, represented as the white areas in Figure 2-6 [38].
Figure 2-6. A view of the optical contact between the paper and the glass in the Chapman smoothness tester [38]

2.3.1.3 Air flow measurements

In the pulp and paper industry, the surface roughness of paper is commonly measured by air flow methods, which measure the air flow between the paper surface and a reference plate under specific conditions. The Sheffield, Bendtsen and Parker-Print-Surf (PPS) instruments are in this category. The Park-Print-Surf, shown in Figure 2-7 is currently considered to be the superior instrument [14], since the configuration of this instrument is closest to the printing nip. In this method, the paper is clamped against a rubber blanket plate, air is forced under constant pressure between the central annulus and the paper
sample. The air flow rate, $Q$ (ml/min), is measured and converted to a roughness value, $G$, in $\mu m$ using Equation 2-4 [40],

$$G = kQ^3$$  \hspace{1cm} (2-4)

where $k$ is a constant. The rubber blanket simulates the printing blanket in offset printing, and the measurement width of 51 $\mu m$ is approximately the size of a halftone dot.

Figure 2-7. Schematic diagram of the Parker-Print-Surf instrument [40]

2.3.2 Characterization of the Porosity of Paper

2.3.2.1 Void Fraction of Paper

The porosity of paper can be characterized by the void fraction, $\varepsilon$, defined as the ratio of the volume of the void in the paper over the total volume of paper. Void factor can be
determined from the density of the solid phase of paper $\rho_f$ and the apparent density of paper $\rho_p$.

$$\varepsilon = 1 - \frac{\rho_p}{\rho_f}$$  \hspace{1cm} (2-5)

2.3.2.2 Pore Size Distribution

A more completed description of the porous structural of paper is given by the pore size distribution of paper. The porous structure of paper is complicated due to the irregular shapes of pores and the interconnection between pores. The measurement of the pore size distribution is based on the assumption that paper may be treated as a material with a series of cylindrical pores [41,42].

Mercury porosimetry evacuates the air from paper, and mercury is forced into the pores of the paper sample under pressure [41]. Mercury fills the large pores first, and with stepwise increases in pressure, mercury fills the smaller pores. Applying Kelvin's equation, the pore radius corresponding to each pressure can be calculated from the contact angle, the surface tension of mercury and the applied pressure. The disadvantage of this method is that it requires very high pressure to access the small pores in the paper. Furthermore, this method gives the distribution by volume, which is proportional to the second or third power of the pore radius [41].

The disadvantage of the mercury method can be overcome by the dioxane method, which was introduced by Corte [41,42]. Instead of monitoring the stepwise penetration of liquid, this method measures the stepwise permeation of a gas through the pores. The
Coulter® Porometer [42] is based on this principle. The specimen is first immersed in a low surface tension wetting liquid so that the liquid fills in all the pores of the sample by capillary suction. Then the sample is placed into a holder with an air supply. This instrument monitors the expulsion of the wetting liquid from the pores by dry air with increasing pressure. The pore flow and number distribution over the range of 100 micrometers to 0.05 micrometers is determined from the flow rate and the pressure by,

\[ d = \frac{40\gamma}{p} \]  \hspace{1cm} (2-6)

where \( d \) is the diameter of pore in \( \mu \text{m} \), \( \gamma \) is the surface tension of the wetting liquid in mN/m, and \( p \) is the applied pressure in millibar. Therefore, the pore size distribution is given by the number frequency of pores, which is superior to the mercury method.

Corte [41] showed that the pore size distribution of paper can be approximated by a logarithmic normal distribution using the dioxane method, which was later confirmed by Dodson [43] using theoretical modeling.

2.3.2.3 Air Permeability

In practice, the porosity of paper is usually approximated by the air permeability of paper. Air permeability is measured by detecting the air flow through the paper structure under defined pressure. A group of instruments such as Bendtsen, Sheffield, Gurley and Parker-Print-Surf are based on this principle. Dalphond and Koller [44] showed that the data of Sheffield, Gurley and Parker-Print-Surf are correlated to each other very well, whereas, the data measured by Bendtsen correlates less well.
2.4 Fines

Fines strongly affect paper structure and properties. A commonly accepted definition of fines is the fraction of fibres passing through a 200-mesh screen (76 μm slots) in a Bauer-McNett classifier. Some investigators have used other cut-off sizes such as 150-mesh or 0.2 mm. Mechanical pulps for printing grades contain 25% to 50% fines depending on the paper grade and the defined cut-off size of fines. Gavelin [45] showed that TMP fines are mainly long and thin fibrils in agreement with Mohlin's observation that TMP fines contains mainly ribbon-like particles [48]. Fines of mechanical pulps play an important strength and optical role in paper. Mohlin [46,47,48] recorded a drop in all strength properties when the fines fraction is removed from TMP pulp. She also showed an increase in both the scattering coefficient and the absorption coefficient in paper with increasing fines content. In terms of surface topography, handsheets [46] made with white water recirculation, which maintains higher fines content, have a more closed and less rough surface compared to handsheets made by the standard procedure. Silveria et al., [49] investigated the structural functions of fines by halogenating the fines with bromine and then observed the position of the fines faction in the embedded handsheet using a backscattering imaging technique. It was found that fines have the following four functions: 1) to fill in the interfibre voids and bridge the gaps in the long fibre structure; 2) to deposit on the surface of fibres if the shape of the fines is thin and long; 3) to act as small fibres; and 4) to form a bond between fibres.

Research on the effects on fines content on the printability of paper is limited. It is generally believed that surface smoothness will improve with increasing fines content,
while the porosity of paper will decrease. Both these changes will help ink holdout, and as a result, provide a better print with less ink. Isono and Hasuke [50] studied the effect of fines retention on print density of woodfree paper, and found that low print density spots are the areas where fines are removed from the paper surface during dewatering. However, most of the fines studies were under poorly controlled conditions: typically, paper samples were collected from different mills, which introduced many other variables other than fines content. This may result in contradictory results, e.g. De Grace [51] showed that print-through increases with increasing fines content based on a comparison of 19 North American newsprint samples, while Giertz [45] reached an opposite conclusion based on a study based on European newsprint. A better understanding of the effects of fines on ink transfer and penetration is necessary.

2.5 Modeling of the Ink and Paper Interrelation

The printing process is complicated and printing quality is affected by many paper properties, ink properties, surface phenomenon, etc. The printing process is controlled by several competing factors: contact between ink and paper, immobilization of ink and splitting of the ink film. Walker and Fetsko [3,55] developed an ink transfer model based on.

\[
y = A \left[ b B + f(x - bB) \right]
\]  
(2-7)

where \( A = e^{-kx} \), and \( B = 1 - e^{1-b} \); \( x \) is the ink weight on the plate, \( y \) is the ink weight on the sample, \( k \) is the rate of coverage parameter, \( b \) is the immobilization or absorption parameter, \( f \) is the free ink film split parameter, \( A \) is the coverage factor and \( B \) is the
immobilization factor. At low ink levels, the coverage parameter $k$ dominates the process, since the paper surface grooves or pores are deeper than the thickness of the ink film. The transfer of ink decreased with increasing roughness due to the reduced contact. At commercial levels of amount of ink on the printing plate, the coverage parameter $k$ and the absorbency parameter $b$ are of equal importance, and the splitting parameter $f$, which depends on the ink properties, begins to influence the fractional transfer. At higher ink levels, the pores on the surface will be completely filled, as the thicker ink film is forced into the pores by the hydraulic pressure created in the printing nip. The amount of ink immobilized increases with an increasing ink film thickness up to a maximum value, which depends on the surface porosity of the paper under printing compression, and in this case, the splitting parameter $f$ begins to dominate [55].

Several modified models of the Walker and Fetsko equation have been suggested by other researchers [52,53,54,55,70]. Mangin and co-workers [55] examined the Walker-Fetsko equation, as well as the modified forms of this equation, using the ink transfer data from 29 printing conditions with letterpress newsprint. They concluded that their data fits the original form of Walker and Fetsko equation best. They also recommended adding a fourth parameter to the equation to describe the coverage factor, which improved the fit of the data significantly. Zang [70] introduced a new splitting factor, which changes with the amount of ink on the printing disk. It was reported that the new three-factor equation using this definition of splitting factor fits the experimental data very well.
2.6 Modelling of the Print Based on Kubelka-Munk Theory

Kubelka-Munk theory [56] modified by Van den Akker [57] is widely used in the pulp and paper industry to calculate the optical properties of paper, including the print density of paper. Several models of the effects of paper properties on printing have been developed.

Bristow [58] derived an equation to calculate ink penetration depth based on Kubelka-Munk theory. The ratio of the grammage of the ink penetrated layer ($W_m$) over the whole grammage, $\beta$, was estimated using the Bristow Equation, which is:

$$\frac{W_m}{\beta} = \ln \left\{ \frac{(1-R_0 R_\infty)/(1-R_p R_\infty)(1-R_q R_\infty)}{[(1-R_0 R_\infty)(1-R_p R_\infty)(1-R_q R_\infty)]} \right\} / \ln \left( \frac{(1-R_0 R_\infty)/(1-R_0 R_\infty)}{1} \right) \tag{2-8}$$

where $R_\infty$ is the reflectance of unprinted paper under opaque pad background;

$R_0$ is the reflectance of unprinted paper under black background;

$R_p$ is the reflectance of printed paper under opaque pad background;

$R_q$ is the reflectance of the reverse side of printed paper under opaque pad background.

Therefore the average ink penetration depth can be obtained by simply measuring the four reflecting parameters of paper listed above.
Pauler [59] developed a model to explain the effects of ink and paper interaction on the relationship between print density and ink quantity. This model assumes that ink penetrates into the paper to a given depth, and within this layer, the ink is distributed uniformly and no ink is set on the surface of the paper. A diagram representing this assumption is reproduced in Figure 2-8 [59]. According to Kubelka-Munk theory, the author assumed that the light scattering coefficient and light absorption coefficient of the mixed layer are the weight averages of those of the ink and paper in this layer. The form of this model is given in.

\[
R_p = \frac{R_{am} \left( \frac{1}{R_{am}} - R_{ap} \right) \exp[S_m W_m \left( \frac{1}{R_{am}} - R_{ap} \right)] + \frac{R_{ap} - R_{am}}{R_{am}}}{\left( \frac{1}{R_{am}} - R_{ap} \right) \exp[S_m W_m \left( \frac{1}{R_{am}} - R_{ap} \right)] + (R_{ap} - R_{am})} \tag{2-9}
\]

The model shows that the reflectance of the printed paper \( R_p \) is a function of the reflectance of the unprinted paper, \( R_{ap} \), the reflectance of the mixed layer, \( R_{am} \), the scattering coefficient of the mixed layer \( S_m \) and the amount of paper occupied by the mixed layer \( W_m \). The shortcoming of this model is that it assumes that the depth of ink penetration layer is uniform over the paper, which differs from the results obtained by the observation under the microscope. An uneven ink penetration layer is typical in real printed paper.
Wahren et al. [60] developed a theoretical model relating the print density and mottle of solid print to the optical properties of paper, the ink, the amount of ink and the unevenness of the ink distribution. The primary assumption of this model is that ink distribution is governed by a stochastic process relating to paper properties. This model is based on the local validity of the Kubelka-Munk set of formulae and assumes the printed paper consists of two layers: a uneven layer of ink and a paper layer. The print evenness $U_i$ is defined as the coefficient of variation of the local grammage of the ink layer. However, this model can not show how the parameter of paper structure affects the mass distribution of the ink layer.

Kajanto [61] modified the Wahren method and developed a theoretical model to predict the effects of various factors on the uniformity of print density based on the two-layered Kubelka-Munk theory. A schematic diagram is shown in Figure 2-9 [61]. The principle of the model is similar to that of Wahren's [60] model, but takes into account ink penetration. This model assumes that the printed paper consists of a homogeneous ink-paper-mixed layer and a paper layer, and there is no ink setting on top of the surface of
the paper. A simplified form of the model is given in equation 2-10. The relative change in print reflectance, $dR/R$, is described as a function of the relative changes of the ink transfer to the paper ($dy/y$), the grammage of the ink penetrated paper layer ($dW_m/W_m$), the light scattering coefficient of paper ($dS_p/S_p$) and the light absorption coefficient ($dK_p/K_p$). Thus

$$\frac{dR}{R} = a_1 \frac{dy}{y} + a_2 \frac{dW_m}{W_m} + a_3 \frac{dS_p}{S_p} + a_4 \frac{dK_p}{K_p},$$

(2-10)

where $a_1$, $a_2$, $a_3$ and $a_4$ are constants and can be calculated if the average ink amount, the grammage of the ink penetrated layer, the scattering coefficient and absorption coefficient of ink and paper are known (see Appendix II).

Figure 2-9. Two-layered model of the printed paper [61]
This model shows that the amount of ink, the ink penetration depth and the optical properties of paper are important to print density. However, the model, can not be used to predict the printing performance of paper, since there is no relationship between paper properties and the parameters used in the model, especially, the “black box” variable \( W_m \), the grammage of the penetrated layer. It is impossible to measure this parameter directly. Therefore it is important to develop a method to express \( W_m \) in terms of measurable paper properties and printing conditions. This relationship is also important for the fundamental understanding of the ink-paper interaction.

### 2.7 Ink Penetration into Paper

Ink penetration is a key factor that controls ink distribution. The penetration of ink under pressure in the press nip may be modeled by the Kozeny-Carmen equation [62],

\[
h = \frac{V}{A} = \frac{\sigma}{2\tau} \sqrt{\frac{p}{\eta}}
\]

where \( h \) is the ink penetration depth, \( V \) is the volume entering an area \( A \) during the nip dwell time \( t \), \( R \) is the average pore radius, \( p \) is the nip pressure, \( n \) is the fluid viscosity, \( \tau \) is the tortuosity of the pore structure and \( \varepsilon \) is the sheet void fraction. Unfortunately, this relationship has the following shortcoming [62]: the assumption of uniform parallel cylindrical tubes does not describe paper structure well; it assumes that the fluid viscosity remains constant during immobilisation.

The present study was carried out in order to understand better the underlying fundamental effects of paper structural properties on print mottle. 100% TMP pulp was
used as the furnish for a detailed experiment to study the effects of formation and fines content on print mottle and to develop a model to predict the effects of these structural properties on print density and its uniformity.
3. PARAMETER IDENTIFICATION

A series of preliminary experiments were conducted to optimize experimental conditions and to identify key parameters for further investigation. A Wood-Pulps Network of Centers of Excellence (NCE) standard pulp (TCMP) was used to investigate the effects of formation and fines content on print mottle. Handsheets with and without fines were made under different settling times to vary the formation of paper, and then calendered using a hard-nip calender. Settling time is the length of time before drainage, and the longer the settling time, the more the chances that fibres entangle with each other. Standard paper properties: caliper, porosity, surface roughness, brightness, opacity, scattering and absorption coefficient, gloss and compressibility were measured. The top side of the sheets were then printed on a IGT AIC2-5 Printability Tester to an average print density of 1.0. The print mottle was evaluated by an image analyzer developed by Abitibi-Consolidated Inc. and the formation of the same printed area was measured using video beta radiography. The two parameters, print mottle and formation, were also measured using the Paprican Micro-Scanner. Finally, the printed and unprinted paper samples were investigated under a scanning electron microscope (SEM) to obtain the surface and z-direction topography, as well the ink penetration depth. All measurements and printing were conducted in a Tappi standard temperature and humidity room. Measurement methods and techniques identified as useful for further study are described more fully in Chapter 4.
3.1 Experimental Methods

3.1.1 Pulp Preparation

The TCMP pulp sample used was first hot disintegrated in a Domtar hot disintegrator according to CPPA standard C.8P to remove latency. The fibre length distribution was determined using a Bauer-McNett classifier, according to Tappi Standard T233 cm-82. The Bauer-McNett classifier was also used to separate the long fibre factions and fines, which was defined as the fraction of fibre passing through the 200 mesh screen. The long fibre factions was obtained by combining all the fibres retained at each screen and the fines was collected by restoring and settling the white water passing through the final (200 mesh) screen.

3.1.2 Handsheet Preparation

Paper sample sets were made at two levels of formation and two levels of fines content, 0% (fines removed) and 25% fines, i.e. four sets in total. All handsheet were made to an oven-dry grammage of 60 g/m² according to the CPPA Standard C.4 modified as follows:

1) Handsheets were made using an AMC square handsheet maker with a sheet size of 30 cm x 30 cm. For handsheets with 25% fines content, white water recirculation was used to maintain the fines content.

2) Handsheets were hard-nip calendered in a laboratory calender under 300 PSI pressure at 3 m/s.
3) Two settling times, the time before draining water, were chosen to vary the formation level of the handsheets at each fines content level.

3.1.3 Paper Characterization

Caliper of paper was measured by a linear variable differential transformer according to Tappi Standard T411. The smoothness and porosity of the samples were measured using a Parker Print-Surf Model PPS78 according to CPPA Standards C.159 and C.156 respectively. Compressibility of paper was determined by the ratio of the Parker-Print-Surf roughness under 10 kgf/cm² to that under 5 kgf/cm².

Brightness and opacity of the unprinted paper samples were measured using a TecniBrite™. Brightness refers to reflectance measured through a blue filter (475um) and opacity refers to the ratio of the reflectance of the paper sample backing by a black background to that backing by a pad of paper sample. The scattering power and the absorption power of the sheets were also measured according to CPPA standard E.2. Gloss was measured using a HunterLab glossmeter according to CPPA method E.3P.

Formation, as described by the coefficient of variation, of the printed paper samples was measured by the beta radiography technique developed by the University of Toronto [26]. For comparison, formation was also measured using the light transmission based Paprican Micro-Scanner. In this method, the size of the nonuniformity is represented by the specific perimeter, which is defined as the length of the median-density contour line
per unit area of sample. The higher the specific perimeter, the finer grained or more uniform the grammage.

The surfaces of the printed samples were photographed using the SEM technique. The samples were pre-treated by gold coat for 2 minutes to increase conductivity. To observe the z-direction profiles, the sample was first embedded into a resin, then the sample block was smoothed using a strong alkali solution [63]. After this treatment, the sample block was microtommed and the thin section was observed under the SEM.

3.1.4 Printing of Paper

The samples were printed in an AIC2-5 IGT printer using an oil-based newsprint ink. Three printing speeds: 0.25m/s, 0.5m/s and 1m/s were used to perform the printing. Print mottle was measured using the print mottle tester in Abitibi-Consolidated Inc., 24 hours after printing. The print mottle number, as defined in Equation 2-2 and Equation 3-1, was used to evaluate the unevenness of the print. The higher the print mottle number, the more uneven the solid print. The print mottle was also characterized by the specific perimeter which was measured with the Paprican Micro-Scanner.

\[
PMN = \left\{ \frac{\left(B_{0.03} + W_{0.03}\right)}{100} \times 100 \right\} + \left\{ \frac{\left(SND \times 100\right)}{13.681} - 100 \right\} + \left\{ \frac{\left(B_{0.04} + W_{0.04}\right)}{\left(B_{0.02} + W_{0.02}\right)} \times 100 \right\}
\]  
(3-1)
In the PMN method, the inspection zones of the printed image were classified into black, gray and white cells according to the print density difference between this zone and the average print density. B is the number of "black" zones; W is the number of "white" zones; subscripts 0.02, 0.03 and 0.04 are the threshold used to define the "black" and "white" zones, e.g., if 0.02 is set to be the threshold, a zone with a density higher than the average print density by 0.02 or more will be considered as a black zone; on the other hand, if the density value is lower than the average by 0.02 or more, this zone is a white zone. SND is the sum of the neighboring differences, or the sum of the absolute optical density differences among all neighboring zones in the density map.

3.2 Results and Discussion

As mentioned at the beginning of this chapter, the objectives of the preliminary study were to optimize the experimental conditions and to identify the key parameters for further study. In this section, the optimization of experimental methods and conditions is discussed, followed by a discussion of the results of the parameter identification study.

3.2.1 Papermaking Method

The samples were made using a square handsheet maker. Compared with the standard British handsheet maker, the square handsheet maker has the advantages of a large enough sheet size, 30 cm x 30 cm, for IGT printing and the wet pressing and drying conditions are more similar to machine-made paper. However, some shortcomings of this square handsheet maker were discovered in the experimental process: 1) Strong edge
effects in the sheets were found. 2) Due to the set up of the flexible forming wire, air bubbles often leaked into the system resulting in a low proportion of good quality sheets. 3) The white water recirculation pump was under-powered resulting in exceptionally long drainage times. Due to these shortcomings, subsequent to this preliminary study, handsheets were made using the standard British handsheet maker and the dynamic former.

3.2.2 Paper Property Measurement

The measurement results of the standard properties of the sheets are listed in Table 3-1. The results indicate that large-scale measurements can identify the surface structural differences between the samples with different fines content, but these types of measurements can’t detect differences caused by formation, i.e. settling time. A more sensitive measurement, such as a measure of the pore size distribution is required.

<table>
<thead>
<tr>
<th>Sample % fines, settling times(s)</th>
<th>Caliper (µm)</th>
<th>PPS (µm)</th>
<th>Porosity (mL/min)</th>
<th>Compressibility</th>
<th>Opacity</th>
<th>SC*</th>
<th>AC**</th>
<th>Gloss</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% (5s)</td>
<td>172</td>
<td>5.43</td>
<td>2761</td>
<td>1.26</td>
<td>91.13</td>
<td>40.5</td>
<td>3.18</td>
<td>4.05</td>
</tr>
<tr>
<td>0%(120s)</td>
<td>166</td>
<td>5.39</td>
<td>2755</td>
<td>1.25</td>
<td>91.30</td>
<td>40.8</td>
<td>3.25</td>
<td>3.75</td>
</tr>
<tr>
<td>25% (5s)</td>
<td>118</td>
<td>4.43</td>
<td>177</td>
<td>1.36</td>
<td>95.20</td>
<td>53.9</td>
<td>3.85</td>
<td>4.50</td>
</tr>
<tr>
<td>25%(120s)</td>
<td>120</td>
<td>4.86</td>
<td>166</td>
<td>1.33</td>
<td>94.86</td>
<td>54.1</td>
<td>3.82</td>
<td>4.0</td>
</tr>
</tbody>
</table>

* scattering coefficient (SC)
** absorption coefficient (AC)

Table 3-1. Paper properties of square handsheets
The surface SEM images of the 25% samples, a good formation sample, with a settling time of 5s and a bad formation sample with a settling time of 120s, are shown in Figure 3-1 (a) and (b). It is clear that the surface of the poor formation sample is rougher and less uniform compared with that of the good formation sample. Figure 3-1 (c) and (d) show the z-direction profiles of the corresponding samples. It appears that the fibers and fines distribute much more evenly in the good formation sample than that in the poor formation sample (see Figures 3-1(c) and 3-1(d)). SEM is a useful method to view the surface topography of the paper, however, it is hard to get results that are statistically significant from this kind of measurement due to the small size of the zone observed which was several hundred microns in length. It was also found that strong alkali solution was not suitable for pre-treatment of the printed samples for Z-direction SEM study, as claimed in the literature [63], since the solution washed out some of the ink particles.
Figure 3-1. SEM images of the 25% fines content samples:
(a) surface of a sample with 5s settling time,
(b) surface of a sample with 120s settling time,
(c) Z-direction profile of a sample at 5s settling time,
(d) Z-direction profile of a sample at 120s settling time.
3.2.3 Printing Conditions

Three printing speeds were tested and it was found that 0.5 m/s was the most suitable printing speed for handsheets. At the higher speed of 1 m/s, some fibres were pulled out from the surface of the paper, especially, the samples without fines, due to poor surface strength. This kind of print unevenness is out of the scope of the present study. At a lower speed of 0.25 m/s, the printing process is too slow compared with a commercial printing process and this may cause differences in ink transfer, ink immobilization and ink splitting.

3.3 Effect of Fines Content and Formation on Print Mottle

As discussed above, formation of paper was determined by both coefficient of variation of mass density and specific perimeter. Print mottle was evaluated by the print mottle number (PMN), as well as the specific perimeter. As listed in Table 3-2, as the settling time increases, the level of flocculation increases for both 0% and 25% samples. The formation marginally improves at the 5s settling time as the fines content is increased from fines free to 25%. Surprisingly, at the 120s settling time, the fines free samples are more uniform than the 25% fines content samples. This may due to the longer drainage time of the 25% samples in the square handsheet maker: the under-powered pump in the white water recirculation extends the drainage time up to 75 seconds longer than that of the fines free samples. As a result, the fibres have more time to entangle with each other.
This result is supported by a comparison of the SEM images of 0% and 25% samples at 120s settling time, see Figure 3-2 (a) and (b). At the 120s settling time, the overall structure of the 0% fines sample is very open, with a relatively uniform distribution of long fibres compared to the 25% fines content sample, where the light weight areas appear to be more open than the flocculated areas. This phenomenon indicates that the addition of fines in paper forming is more complicated than simply reducing the average fibre length of the pulp samples and filling in pores.

As listed in Table 3-2, specific perimeter of mass density is primarily dependent on fines content and to a lesser extent on the settling rates in this study. For 0% fines content samples, the specific perimeter is lower at longer settling times, indicating a coarse structure. For the 25% fines content samples, the difference in specific perimeter with settling time is minor. For different fines content levels, it appears that specific perimeter is related to the average fibre length, which is in agreement with a previous study [30]: higher specific perimeters are obtained for samples with 25% fines.
Figure 3-2. SEM images of the z-direction profiles: (a) a 0% fines content sample at 120s settling time, (b) a 25% fines content sample at 120s settling time.

As listed in Table 3-2, it appears that specific perimeter of the print density is dependent strongly on fines content. Although differences in samples at different formation levels are evident, specific perimeter results did not distinguish differences between samples, which is not in good agreement with visual observation. Since specific perimeter is sensitive to fines content and less so to formation level, this parameter will not be used in this study to investigate the effects of both fines and formation on the print mottle.

The differences of the uniformity of print density are reflected in the PMN number: print mottle improves with formation improvement at each fines content level. The samples made from the whole pulp with 25% fines print better compared with 0% fines content samples at each formation level. This may be due to a better contact between the fines-containing paper and the printing disk. PMN appears to be a good general indicator of the print unevenness except that the standard deviation of the measurement results are relatively high, from 5 to 50, indicating that the repeatability of this method is relatively
Furthermore, this is a semi-empirical parameter and a more fundamental method is desirable to describe the features of print mottle.

<table>
<thead>
<tr>
<th>Sample % fines content, settling time</th>
<th>CV mass density (%)</th>
<th>Specific perimeter mass density (mm⁻¹)</th>
<th>Specific perimeter print density (mm⁻¹)</th>
<th>PMN</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% fines (5s)</td>
<td>7.48</td>
<td>2.33</td>
<td>4.15</td>
<td>59.60</td>
</tr>
<tr>
<td>0% fines (120s)</td>
<td>7.85</td>
<td>2.07</td>
<td>4.14</td>
<td>77.26</td>
</tr>
<tr>
<td>25% fines (5s)</td>
<td>7.22</td>
<td>2.68</td>
<td>4.49</td>
<td>49.06</td>
</tr>
<tr>
<td>25% fines (120s)</td>
<td>8.5</td>
<td>2.72</td>
<td>4.46</td>
<td>74.34</td>
</tr>
</tbody>
</table>

Table 3-2. The evenness of mass density and print density of square handsheets

In summary, it was found that both formation and fines content affect the printing uniformity of paper. More detailed parameters are required to better understand the principle effects of paper structure on print mottle, particularly, a more sensitive method to describe print mottle is needed. New methods are proposed in the following chapter. Traditional large-scale measurements of the paper structural properties do not adequately describe the underlying causes of print mottle variations.
4. EXPERIMENTAL METHODS

The details of the experimental methods will be described in the following sections in this chapter. The experimental methods were based on the key parameters identified in the parameter identification stage, as described in chapter 3. In addition, a sensitive method was developed to describe print mottle and to evaluate the effects of fines and formation on the print mottle of wood-containing paper. The pulps used was 100% TMP couch trim from the Grand Falls Mill, NFLD., Abitibi-Consolidated Inc. Four types of sheets: handsheets, dynamic former sheets, pilot machine sheets and commercial newsprint were made or collected. These samples covered a wide range of formation, fines content levels and fibre orientations. The sheets were hard-nip calendered before printing. Standard paper properties, i.e. caliper, surface smoothness, porosity, gloss, etc. were measured. The samples were printed using an IGT proof printer on the top side of the sheets to an average print density of 1.0. Both the print mottle of the print and the formation of the paper were evaluated using the second order statistics texture analysis features. Other common printing properties were also recorded. All the measurements and printing were conducted in a Tappi standard temperature and humidity room.

4.1 Sample Preparation

4.1.1 Separation of Fines and Long Fibre Fraction

Sample sheets were made from 100% TMP couch trim from the Grand Falls Mill, NFLD., Abitibi-Consolidated Inc. Latency of the furnish was removed using a Domtar disintegrator according to CPPA standard C. 8p. The fines fraction was separated from
the furnish using a FLYGT Float Wash Fractionator with a 200-mesh screen [64]. The long fibre fraction was retained. The white water passing through the 200-mesh was restored and the fines was collected after 36 hours of settling\textsuperscript{1}. The specific surface area of the fines fraction was measured by turbidity measurement\textsuperscript{[60]} using a Paprican turbidity tester\textsuperscript{2}.

4.1.2 Sheet Preparation

4.1.2.1 British Handsheet

The furnish used to make standard handsheets was the long fibre and fines fractions of the TMP couch trim separated at Paprican. The fines fraction was artificially blended with the long fibre fraction at 0%, 20%, 30% and 40% by weight. Standard British handsheets (50 g/m\textsuperscript{2}) were made according to CPPA Standard C.4. using a standard British handsheet maker with white water recirculation to maintain the fines level in the handsheets. The first eight to ten handsheets of each sample set were discarded until a constant sheet weight was obtained. At this stage, the fines content in the handsheets was the same as that in the furnish. Unpressed handsheets were redispersed and the fibre length of each fines content level was measure using a fibre quality analyzer (FQA) [60]. In order to vary the formation level at each fines content level, three settling times, i.e. the time before drainage, were selected as 5 s, 60 s and 120 s. The longer the settling time, the more the chances that the fibres could entangle with each other.

\textsuperscript{1} The separation was conducted at Paprican Laboratories in Pointe Claire, Que.
\textsuperscript{2} The turbidity measurement was conducted at Paprican Laboratories in Pointe Claire, Que.
The handsheets were then trimmed to a size of 14.0 cm x 6.0 cm and hard-nip calendered using a Beloit Wheeler Laboratory Calender at 65 °C under a linear pressure of 52.5 KN/m (300 pounds per inch).

4.1.2.2 Dynamic Former Sheet

TMP sheets with a grammage of 50 g/m$^2$ at 0%, 25%, 35% and 45% of fines levels were made using an Noram Auto-Dynamic Sheet Former. A picture of this equipment is shown in Figure 4-1. The machine cabinet [61] contains a metal drum carrying the forming fabric, a stock projection system, a stock flow pump, a storage tank with a mixer, and a monitoring and controlling system. This equipment simulates the headbox delivering stock onto a moving fabric. The nozzle, which simulating the headbox, travels at a constant speed across the width of the forming fabric for a preselected number of sweeps. The forming fabric is laid along the interior of the metal drum, and this drum spins around its axis while the paper is formed. The pulp at a consistency of 0.5% was spread on the rotating wire through the nozzle at a flow rate of $1.2 \times 10^{-3}$ m$^3$/min. Three rotation rates of the wire: 950, 1100 and 1250 rpm were chosen to vary the jet/wire ratio at each fines content level. The angle of the jet was set to 20° and the distance between the jet and the wire was set to 10 mm. The wires used were standard polypropylene 60-mesh forming fabric. In order to obtain samples with different degree of wire mark and drainage mark, the samples were made using either a single fabric or double forming fabrics. In the case of a single wire, a clear mark was observed in the samples. The drainage procedure for all the samples were controlled at the same drainage time and rotational speed of the drum using the machine default program No.6. Then the sheet
was transferred from the fabric to the metal board and wet pressed in the presser under a pressure of 137 KN/m² (20 PSI). The sheet together with the metal board was placed on top of the dryer to dry. The original size of the sheets is 22 cm x 90 cm, which was then trimmed to 7 cm x 30 cm and hard-nip calendered at 65°C under a linear pressure of 52.5 KN/m (300 pounds per inch).

Figure 4-1. A picture of the auto-dynamic sheet former: 1. nozzle, 2. forming fabric, 3. forming and drainage drum

4.1.2.3 Pilot Scale Paper Machine Made Sheet

Paper was made using a multipurpose pilot Fourdrinier paper machine at the South China University of Technology. This machine has a Fourdrinier forming section with a top former as shown in Figure 4-2. The pulp used was a mixture of 30% CTMP and 70% recycled newsprint. The fibre length distribution of this pulp was measured using a fibre quality analyzer. The consistency of the pulp in the air-cushioned headbox was 0.3%. The jet/wire ratio was set to 0.92 at a machine speed of 145 m/min. The paper was made at a newsprint grammage of 45 g/m². The width of the paper roll was 40 cm, and the

---

3 South China University of Technology dynamic former
sheets were trimmed to a size of 30 cm (machine direction) x 7 cm (cross machine direction) and hard-nip calendered at 65°C under a pressure of 137 KN/m².

Figure 4-2. (a) Schematic diagram of the pilot scale paper machine: 1. Air-cushioned headbox, 2. Forming wire, 3. Top former, 4. First press section, 5. Second press section, 6. Sample collecting point

(b) A picture of the pilot scale paper machine
4.2  Paper Property Measurements

4.2.1  Surface Roughness and Porosity of Paper

The surface roughness and porosity of the sheets were measured using a Parker-Print-Surf instrument (PPS78 Digital Model) according to CPPA Standard E6 and E7, respectively.

Detailed surface images of sheets were made using the SEM technique. A sample was cut to a size of 1 cm diameter and mounted on the support. The sample was then coated with a thin layer of gold with a coating time of 2 minutes. The surface image was captured using a SEM system at an intensity of 10 KV.

Complete pore size distributions of the samples were measured using a Coulter @Porometer [46]. A specimen was first immersed in a low surface tension wetting liquid, provided by the Coulter company, so that the liquid filled in all the pores of the sample by capillary action. Then the sample was placed into a holder with one side pressurized with air. The expulsion of the wetting liquid from the pores with increasing dry air pressure was monitored. The flow rate and the pressure were measured and converted to a measure of the number distribution of pores as discussed in Section 2.3.2.2.

4.2.2  Formation of Paper

The video beta radiographic (VBR) technique was used to measure the formation of paper [19]. This method combines beta radiography, image processing and data analysis. In this method, a printed handsheet sample and a calibration wedge were exposed to a C^{14}
beta source for 30 minutes and the attenuated rays were recorded on an X-ray film. The developed film was then digitized using an image analysis system and the gray levels of the image were converted into a grammage matrix. The mean, coefficient of variation of grammage and the floc grammage were then calculated using Dodson’s method [26].

4.2.3 Other Paper Properties

The grammage of the samples were determined according to CPPA Standard A.4. The caliper was measured following the CPPA Standard C.5. The zero-span strength in both the machine and the cross-machine direction of the oriented sheets was tested using a Pulmac™ instrument. The reflectance of the sheet, before and after printing, was determined by a Micro Technibrite™ opacimeter. Other optical properties of the paper, including the scattering coefficient, the absorption coefficient and the opacity were also measured using this instrument according to CPPA standards.
4.3 Printing of Samples

The samples were printed using an AIC-5 IGT printer to a target print density of 1.0. A rubber printing disk was selected to simulate the soft printing blanket of offset printing. The ink (Rieger™ news black 27175) used was a typical offset news black ink with 18% carbon black pigments. The printing of paper was performed on the top side of the samples. The printing speed was set to 0.5 m/min and the printing pressure to 625 N/m. The size of the printed area was 25 cm x 5 cm. The print density was measured using a densitometer 12 hours after printing.

4.3.1 Ink Amount Transferred

The amount of ink transferred to the paper sample was determined by measuring the weight of the sample before and after printing. To confirm results, the amount of ink on the printing disk before and after printing was also recorded. The analytic balance used to weigh paper samples is accurate to 0.0001 g.

4.3.2 Average Ink Penetration Depth

The average ink penetration was determined following the Bristow model described in Chapter 2. The reflectance of the printed and unprinted paper under different background was tested using the Micro Technibrite™ opacimeter.
4.4 Print Mottle Measurements

Print mottle was measured through a combination of image analysis and data analysis. A schematic diagram of the experimental setup is shown in Figure 4-3. The printed image was placed under a uniform reflecting light source. The image of the printed paper was scanned by a charged coupled device (CCD) camera and digitized. In order to get rid of the noise of the background, 16 images of each print were captured and the final image used was the average image of these 16 images. The 2 cm x 2 cm measuring area was divided into small inspection zones scaling from 0.1 mm to 2 mm. The gray level of the image in each small zone was recorded. A series of standard print density plates provided by Optest Equipment Inc. (P/N 395310) were used to calibrate the correlation between the gray level of the image and the print density of the image.

Figure 4-3. Experimental setup of print mottle measurement
Three statistical methods were used to analyse the data: the print mottle number (PMN), the coefficient of variation of print density (CV) and the spatial print density dependence method.

The spatial print density method developed in this study is based on the spatial gray level dependence method (SGLD) as applied to texture analysis [15,31,33,34]. Two steps are associated with this method as shown in the flowchart in Figure 4-4: in step 1 was to compute the intermediate co-occurrence matrices, and in step 2 was to extract the textural features. The co-occurrence matrices of the print density along the four principal directions (horizontal, vertical, right diagonal and left diagonal) are generated based on the print density map. Each entry of the co-occurrence matrix is the probability of a certain pair relationships occurring between an inspection zone and its neighboring zone along the scanning line. The print density range was set from 0.7 to 1.3 and the threshold for print density difference was set to 0.02. This threshold setting resulted in 900 possible relationship patterns. A simplified example of the computation of the intermediate matrices is presented in Appendix I.
SPDD was used to determine the textural features: energy, homogeneity, correlation and contrast of the printed image. The definitions of these features are shown as,

\[
Energy = \sum_i \sum_j [P(i,j)]^2 \tag{4.2}
\]

\[
Contrast = \sum_i \sum_j (i - j)^2 [P(i,j)] \tag{4.3}
\]

\[
Correlation = \sum_i \sum_j (i - \mu_i)(j - \mu_j)[P(i,j)/\sigma_i \sigma_j] \tag{4.4}
\]
\[ Homogeneity = \sum_i \sum_j \frac{1}{1 + (i - j)^2} \times [P(i, j)] \] (4.5)

where \( P(i,j) \) is the normalized probability of gray level pair \( i \) and \( j \), and

\[
\sum_i = \sum_{i=1}^{i=N_G} \quad \sum_j = \sum_{j=1}^{j=N_G}
\]

With \( N_G = \) number of print density or mass density, and

\[
\mu_i = \sum_i [\sum_j P(i, j)],
\]

\[
\mu_j = \sum_j [\sum_i P(i, j)].
\]

### 4.5 Local Correlation Measurement

The local mass density, ink transfer and print density of the samples were measured and compared. There were three experimental steps: 1) print the paper sample and the corresponding IGT sheet, 2) capture the images of the printed paper sample, and the corresponding IGT print, and 3) use the VBR technique to determine the mass density map. The key element to this experiment is to carefully register the corresponding areas of these images and the translation of the gray levels to print density, ink transfer and local mass density.

#### 4.5.1 Ink Transfer Measurement

A paper sample was printed on an IGT proof press with a labelled printing roller (see Figure 4-5 (a)). Since the ink amount transferred to the paper varied locally, the ink layer
left on the printing roller after the first print was uneven. A sheet of cast-coated IGT paper, a paper with high surface smoothness, superior formation and extremely high density, was printed using the remaining ink on the printing roller (see Figure 4-5 (b)). The second print left a thin and uniform layer of ink on the printing roller. The image recorded on the second sheet could then be considered as an inverse image of the ink pattern transferred to the first sheet.

The IGT sheets were chosen as the second sheets because of the high surface smoothness and density of these sheets. This type of structure will leave most of the ink on the surface of paper, and as a result, will give a better correlation between ink transfer and the print density (that is, in this case, the penetration of ink is negligible). The calibration curve of the ink transfer to the IGT sheet and the print density was developed based on a set of 10 IGT sheets printed with different amounts of ink. The weight of the printing roller before and after printing was measured to determine the total amount of ink transferred to the IGT paper. A calibration curve was obtained based on these data. In addition, a uniformity study was conducted on the 10 IGT sheets to evaluate whether the printing roller or the blanket introduced variations. The results indicated that there is no variation caused by these printing set-ups.
Figure 4-5. (a) A Typical printed paper surface, and (b) its corresponding second print on the IGT sheet

4.5.2 Print Surface and Corresponding IGT Image Capture

A standard 2.5 cm x 1.25 cm Avery® label was attached to the edge of the triangular label and the base printing line. This label helped register the printed and the VBR image of the same paper sample, which will be discussed more fully later. It would have been ideal to first develop the VBR image of the printed sample and then do the image analysis on these three images at the same time. However, for safety reasons, the image analysis of the printed sample must be performed prior to beta-ray exposure of the sample.

A CCD camera was used to capture the images of the printed surface of the paper sample, and the corresponding cast-coated IGT sheet under reflected light conditions. The images of a set of 10 Optest Standard Print Density cards were also captured under the same lighting and camera conditions to generate a calibration between gray levels and actual
print densities ranging from 0.05 to 1.23. The image size was 98 mm x 60 mm and the pixel size was 0.090 mm$^2$. A triangular pattern together with the printing edge was used to register the first and second prints. The upper left corner of the adhered label was defined as the lower right corner of the 20 mm x 20 mm area of interest. The gray level value of each pixel was recorded. The gray level data of the first print were then converted into print densities, while the gray level data of the second print were translated into ink transfer to the IGT sheet. The local amount of ink transferred to the first print can then be calculated, knowing the total amount of ink on the printing roller before printing. Finally, the cross correlation coefficient of these two sets of data, the local print density and the local amount of ink transfer, was determined.

4.5.3 VBR Image Analysis

The video beta radiographic (VBR) technique as described in Section 3.2.2 was used to measure the local mass density of the samples. Samples were printed and labelled prior to analysis. The label was clearly defined on the VBR film and used to identify the area of interest. The gray levels of the image were converted into a grammage matrix.
5. EXPERIMENTAL RESULTS AND DISCUSSION

The effect of paper properties on the print mottle of wood-containing paper was investigated experimentally for both random paper and oriented paper. Second order statistics were used to quantify and to correlate paper formation and print mottle as a function of fines level. As a basis for discussion, printing uniformity will be discussed as a function of paper properties.

5.1 Fibre and Fines Properties

5.1.1 Properties of the Raw Material

The species composition of the 100% TMP couch trim was 90% black spruce and 10% Douglas fir, and there were no furnish additives. The fibre fractions of the pulp measured by the Bauer-McNett classifier is given in Table 5-1.

<table>
<thead>
<tr>
<th>Fibre fractions</th>
<th>Bauer-McNett weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R14</td>
<td>9.5</td>
</tr>
<tr>
<td>P14/R28</td>
<td>30.0</td>
</tr>
<tr>
<td>P28/R48</td>
<td>20.8</td>
</tr>
<tr>
<td>P48/R100</td>
<td>10.2</td>
</tr>
<tr>
<td>P100/R200</td>
<td>4.7</td>
</tr>
<tr>
<td>P200</td>
<td>25.2</td>
</tr>
</tbody>
</table>

Table 5-1. Fibre fractions of the TMP couch trim
The fines was defined as the pass 200 mesh screen fraction, i.e. 76 μm width or less, and accounts for 25% of the furnish. The fibre length distribution of the couch trim sample measured by a fibre quality analyzer (FQA) is shown in Figure 5-1. The fibre length has a lognormal distribution ranging from 0.05 mm to 3.75 mm with approximately 50% (arithmetic) fines (<150 μm). Note that the cutting size of fines measured by FQA is twice of that of the definition, this is due to the lower measurement limit of the FQA is 76 μm. So the FQA fines content data is used only as a reference.

![Fibre length distribution of TMP couch trim](image)

**Figure 5-1.** Fibre length distribution of TMP couch trim: (a) full distribution and (b) partial distribution showing the fibre length over the range from 0 to 0.5 mm. Mean length (arithmetic): 0.68 mm, Mean curl: 0.035, Mean kink index: 0.46, Fines content (<150um): 50% (arithmetic).
5.1.2 Properties of Fines

The TMP pulp was separated into a fines fraction and a long fibre fraction using a Paprican FLYGT Float Wash Fractionator as described in Chapter 4. A 200 mesh screen was used to separate the fines fraction from the furnish. The optical specific surface of the TMP couch trim fines, measured by turbidity was 10.1 m²/g, indicating these fines have relatively high specific surface areas.

The fines fraction was artificially blended with the long fibre fraction at different weight percentages to make sheet samples using both the British Standard handsheet maker and the Noram Dynamic Sheet Former, as described in Chapter 4. The fines fraction was paste-like due to the high storage concentration. Mechanical agitation could not completely re-disperse the fines in the fines/long fibre blended suspension. As a consequence, the blended pulps contained more and larger coherent fines flocs than the original couch trim. Although this re-mixing procedure differs from the normal papermaking process, it is similar to the industrial circumstances involving mixing with fillers or the other types of pulps. This re-mixing operation also provides us the opportunities to investigate the importance of fines distribution on print mottle. A detailed discussion on this issue will be given later in this chapter.
5.1.3 Fibre Properties of the Handsheets

The fibre properties of each unpressed handsheet sample set are shown in Table 5-2. The results show that the mean fibre length becomes shorter with higher fines content, as expected. The fines fraction added was mainly the portion of fibres smaller than 150 µm. measured by the FQA. The fibre length distribution of the reference newsprint furnish is similar to that of the pulp sample with 30% fines. The mean curl and kink indexes of each sample are very similar, indicating that the fibre properties of each sample do not change with increasing fines content.

<table>
<thead>
<tr>
<th>Fines added (%)</th>
<th>Fines &lt; 150 µm (%)</th>
<th>Mean fibre length (mm)</th>
<th>Mean curl</th>
<th>Mean Kink</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>7.5</td>
<td>0.81</td>
<td>0.032</td>
<td>0.45</td>
</tr>
<tr>
<td>20</td>
<td>19.0</td>
<td>0.73</td>
<td>0.034</td>
<td>0.40</td>
</tr>
<tr>
<td>30</td>
<td>27.0</td>
<td>0.63</td>
<td>0.032</td>
<td>0.42</td>
</tr>
<tr>
<td>40</td>
<td>41.0</td>
<td>0.51</td>
<td>0.032</td>
<td>0.43</td>
</tr>
<tr>
<td>Newsprint</td>
<td>35.0</td>
<td>0.61</td>
<td>0.035</td>
<td>0.46</td>
</tr>
</tbody>
</table>

Table 5-2. Fibre properties of the handsheet samples

5.1.4 Fibre Properties of the Oriented Sheets

Blended furnishes were also used to make dynamic former sheets. Compared with the handsheet samples, a slightly higher fines content was added to the furnish, since there is no white water recirculation system in the dynamic former to maintain the fines content.
Table 5-3 lists the fibre property analysis of the dynamic former sheets. These results are similar to those of the handsheet samples.

While making the oriented sheets, it was found that the drainage holes in the drum left marks on the sheet using a single forming fabric. To eliminate the drainage marks, a second forming fabric was added on top of the first forming fabric; a double fabric setup also helped to maintain the fines content on the resulting sheets, especially at the 40% fines content level. Most of the printing tests were performed on the sheets made on the double forming fabrics. Some single fabric sheets with drainage marks were also used to test the sensitivity of the image analysis method, i.e. the texture features.

<table>
<thead>
<tr>
<th>Fines added (%)</th>
<th>Fines &lt; 150 μm (%)</th>
<th>Mean fibre length (mm)</th>
<th>Mean curl</th>
<th>Mean Kink</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Double*</td>
<td>Single**</td>
<td>Double*</td>
<td>Single**</td>
</tr>
<tr>
<td>0</td>
<td>9.5</td>
<td>8.3</td>
<td>0.74</td>
<td>0.84</td>
</tr>
<tr>
<td>25</td>
<td>22.1</td>
<td>20.5</td>
<td>0.63</td>
<td>0.68</td>
</tr>
<tr>
<td>35</td>
<td>28.3</td>
<td>26.2</td>
<td>0.56</td>
<td>0.59</td>
</tr>
<tr>
<td>45</td>
<td>39.8</td>
<td>31.7</td>
<td>0.46</td>
<td>0.55</td>
</tr>
</tbody>
</table>

* Double: two forming fabrics  
** Single: one forming fabric

Table 5-3. Fibre properties of the samples made with the dynamic former
5.2 Standard Paper Properties

5.2.1 Surface Properties of Handsheets

5.2.1.1 Density and the Parker-Print-Surf Measurement

At the same average grammage of 45 g/m², paper samples without fines are bulkier than fines-containing samples as listed in Table 5-4; the density of 0% fines sample is much lower than the other samples. The 0% fines samples have an open structure, 2538 mL/min porosity; while the 40% fines samples have a closed structure, 52 mL/min porosity. The porosity of the reference newsprint is approximately eight times greater than that of the 40% fines samples. This structural difference is also illustrated by the series of SEM images in Figure 5-2. As the fines content increases, the fines fill into the pores between the stiff TMP fibres and produce a denser structure. There is much less variation in porosity with changes in formation, i.e. settling times, compared to changes in fines content. At each fines content level, a flocculated sample is expected to have a higher porosity and porosity variation than a uniform sample. The Parker-Print-Surf measurement device could not distinguish this change, see Figures 5-3 and 5-4. The error bars covers the range of ± two standard deviations. The results of the t-test on the surface roughness and porosity data show that there are significant difference between samples of different fines content levels, but there is no significant difference between samples of different formation (settling time) levels at each fines content level.
Table 5-4. Properties of handsheets made at 5s settling time

At each fines level, the surface smoothness of the good formation samples, 5 second settling time, are close to that of the poor formation samples, 120 second settling time, especially, for the samples with high fines content. Under the same calendering conditions, the surface roughness of the handsheets decreases as the fines content increases from 0 to 40%. The surface roughness of the newsprint sample is very close to that of the 30% fines content handsheets; however, the porosity of the newsprint is higher than that of the respective handsheet. This difference may reflect the different forming methods.
Figure 5-2. SEM images of handsheet surfaces: (a) 0% fines content, (b) 20% fines content, (c) 30% fines content, and (d) 40% fines content
Figure 5-3. The effect of fines content at different settling times on the surface roughness of handsheets as measured by the Parker-Print-Surf instrument (the error bars covers the ranges of +/- two standard deviations).

Figure 5-4. The Effect of fines content at different settling times on the porosity of paper measured by the Parker-Print-Surf instrument (the error bars covers the ranges of +/- two standard deviations).
5.2.1.2 Pore Size Distribution

The actual pore size distributions of the samples were measured using a Coulter®
porometer. The distribution curves illustrate, as shown in Figure 5-5, that the pore size
distribution is lognormal, in agreement with Corte and Dodson [43]. The mean and
coefficient of variation (CV) of the pore size of each fines content sample are listed in
Table 5-5. The results are in agreement with the Parker-Print-Surf (PPS) results that the
mean pore size decreases with increasing fines content and mean pore size is not as
strongly sensitive to formation. At each fines content level, the CV of the pore size
decreases as formation improves. The samples with fines have higher CV than samples
without fines due to the existence of the small fines flocs, as discussed in the previous
section, as well a much lower mean pore size.

<table>
<thead>
<tr>
<th>Fines Content (%)</th>
<th>Good formation (5 s)</th>
<th>Poor formation (120 s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean (μm)</td>
<td>CV</td>
</tr>
<tr>
<td>0</td>
<td>8.45</td>
<td>41.8</td>
</tr>
<tr>
<td>20</td>
<td>1.79</td>
<td>42.1</td>
</tr>
<tr>
<td>30</td>
<td>0.71</td>
<td>45.2</td>
</tr>
<tr>
<td>40</td>
<td>0.55</td>
<td>47.2</td>
</tr>
<tr>
<td>Newsprint</td>
<td>1.21</td>
<td>38.3</td>
</tr>
</tbody>
</table>

Table 5-5. The effect of fines content and formation on the pore size distribution of
handsheets and newsprint sample.
Figure 5-5. A typical pore size distribution curve of a 30% fines content handsheet sample

5.2.2 Surface Properties of Oriented Sheets

5.2.2.1 Density and the Parker-Print-Surf Measurement

At the same average grammage of 45 g/m², the oriented paper samples with 0% fines are bulkier than the other oriented paper samples as listed in Table 5-6. The caliper of 0% fines sample is much higher than the other samples, which is in agreement with the handsheet samples.
At each fines level, the surface smoothness of both sides of the oriented samples is listed in Table 5-6. The wire side of the sample shows a much lower smoothness than that of the top side, especially for paper samples with fines. More fines accumulate at the top side of the paper, which improves the smoothness of that side of the paper. Furthermore, the top side is in contact with a metal plate, and the wire side is in contact with a felt during wet pressing. The metal plate will further re-distribute fines on the top surface of the paper. In support of this observation is the minor effect that the fines content has on the roughness of the wire side. On the top side, the paper sample without fines is much rougher than that of all other samples with fines. Increasing the fines content from 20% to 40% does not change the surface topography dramatically, in contrast to the result for
the handsheets. Density and roughness of the pilot machine sheets are similar to those of the reference newsprint, see Tables 5-4 and 5-6.

5.2.2.2 Pore Size Distribution

The pore size distribution of the oriented samples was measured using a Coulter® porometer. The mean and CV of the pore size of each fines content sample are listed in Table 5-7. At each fines content level, the mean and the CV of the pore size of the sheets made by the dynamic former is lower than those of the good formation handsheet samples. The results indicate that the structure of the oriented sheets is denser and more uniform compare to the handsheet at each fines content. The mean pore size of the pilot machine sheet is similar to that of the reference newsprint, but the CV is higher than that of the newsprint due to the poor formation of the sheet.

The mean and CV of the pore size of the sheets made using two forming fabrics are lower than those of the sheets made using one forming fabric. This indicates that the sheets formed using two fabrics have more uniform structure due to the better retention and distribution of fines. This finding indicates the importance of the pattern, the size and shape of the slots or holes in the forming fabrics on the quality of the resulting paper.
<table>
<thead>
<tr>
<th>Fines Content (%)</th>
<th>Pore size distribution</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean (μm)</td>
<td>CV</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Double*</td>
<td>Single**</td>
<td>Double*</td>
</tr>
<tr>
<td>0</td>
<td>9.31</td>
<td>9.52</td>
<td>27.4</td>
</tr>
<tr>
<td>20</td>
<td>1.43</td>
<td>1.56</td>
<td>29</td>
</tr>
<tr>
<td>30</td>
<td>0.63</td>
<td>0.77</td>
<td>38.2</td>
</tr>
<tr>
<td>40</td>
<td>0.4</td>
<td>0.43</td>
<td>39.7</td>
</tr>
<tr>
<td>PL</td>
<td>1.25</td>
<td></td>
<td>42.3</td>
</tr>
</tbody>
</table>

* Double: sheets made using two forming fabrics
** Single: sheets made using one forming fabric

Table 5-7. The effect of fines content and formation on the pore size distribution of the oriented sheets and South China University pilot machine sheets (PL).

### 5.2.3 Optical Properties of the Sheets

Standard optical properties, such as scattering coefficient and absorption coefficient of paper are used as indictors of potential print quality. Scattering coefficient and absorption coefficient both increase with increasing fines content due to the large specific surface area of fines, see Table 5-8. However, at each fines content level, the effect of formation on the scattering and absorption power is not clear. The measuring area (1 cm in diameter) smoothes out the smaller scale variations.
In general, the previous two sections discussed the standard paper properties describing the surface and density, as well as optical properties of paper. The effects of fines content on these structural properties are clear, however, the effects of settling time (formation) can not be distinguished using the large-scale standard instruments, such as Parker-Print-Surf. The pore size distribution measurements do reflect the higher variation of the poor formation samples. In the next section, the effects of forming conditions and furnish on the structure, especially the mass distribution of paper will be discussed. Then, a discussion will follow on how this diversity of properties affects the printing performance of paper.

### Table 5-8. Optical properties of sheets

<table>
<thead>
<tr>
<th>Sample</th>
<th>Scattering power</th>
<th>Absorption power</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Handsheet</td>
<td>Oriented</td>
</tr>
<tr>
<td>0% fines</td>
<td>1.21</td>
<td>1.39</td>
</tr>
<tr>
<td>20% fines</td>
<td>1.78</td>
<td>1.97</td>
</tr>
<tr>
<td>30% fines</td>
<td>1.89</td>
<td>1.99</td>
</tr>
<tr>
<td>40% fines</td>
<td>2.18</td>
<td>2.2</td>
</tr>
</tbody>
</table>

#### 5.3 Second Order Statistics Properties of Sample Sheets

As mentioned in Chapter 4 on experimental methods, two parameters: the fines content and the formation level were controlled. Four fines content levels were selected and these levels were retained in the resulting sheets using white water recirculation. Three settling
times were chosen to vary the formation levels at each fines content level in handsheets, and three jet/wire ratios were selected to vary the degree of fibre orientation in oriented sheets.

Ranking the handsheets on the light table was difficult since for some samples, the flocculation scale was high but the flocculation intensity was low, and the paper sample looked "cloudy". For other samples, on the other hand, the flocculation scale was low but the intensity was high, and the samples appeared to be "grainy". To overcome the difficulty in comparing formation at different measurement scales, a more reliable method, the video beta radiography (VBR) technique, was used to achieve a more accurate assessment.

Coefficient of variation (CV) of the mass density and print density, as well as the texture features: energy, contrast, correlation and homogeneity were used to determine the formation of the samples from the density map, and the print nonuniformity of the print density from the print density map. The four texture features have been defined in Chapter 4, and a brief review is given as follows:

\[
\text{Energy} = \sum_i \sum_j [P(i,j)]^2
\]

\[
\text{Contrast} = \sum_i \sum_j (i-j)^2 [P(i,j)]
\]

\[
\text{Correlation} = \sum_i \sum_j (i-\mu_i)(j-\mu_j)[P(i,j) / (\sigma_i\sigma_j)]
\]

\[
\text{Homogeneity} = \sum_i \sum_j 1/[1 + (i-j)^2] \times [P(i,j)]
\]

where P(i,j) is the normalized probability of mass density (or print density) pair, i and j.
\[
\sum_{i=1}^{N_G} = \sum_{i=1}^{N_G} \\
\sum_{j} = \sum_{j=1}^{N_G}
\]

with \(N_G\) = number of mass density (or print density), and

\[
\mu_i = \sum_{i}[\sum_{j} P(i, j)] \\
\mu_j = \sum_{j}[\sum_{i} P(i, j)]
\]

\(P(i, j)\) is the frequency of the occurrence of a specific pair \((i, j)\). Energy is a measure of the uniformity of a structure with a maximum value of 1 indicating a totally uniform structure and a minimum value of \((1/N_G)^2\) indicating a maximally nonuniform structure. When people subjectively rank formation and print uniformity, they more heavily weight the large local difference of mass and print density. As shown in Equation 4-2, energy weights each occurrence pair evenly without taking into account the amount of difference, which is different from the human visual ranking.

Homogeneity provides a more relevant measure of the uniformity of the structure, since as defined in Equation 4-5, the contribution of a large difference between zones (large \((i-j)\)) is more lightly weighted in terms of the contribution to the uniformity of the structure. And as a result, in this study, only homogeneity is used as a feature parameter for further discussion on the uniformity of mass density and print density.

Contrast (CON) and correlation (COR) were used as indicators of the intensity and scale of the structural arrangement of the mass density map for each sample, respectively. As
defined in Equations 4-3, contrast is the moment of inertia of the co-occurrence matrices, and it focuses on the differences between the neighboring zones. For a perfectly uniform structure, the value of contrast is 0, and this value will increase with increasing neighboring differences, (i-j). This parameter is a measure of the intensity of differences. Correlation is a feature focusing on the scale of the structure and varies between 0 and ±1. The COR value is 0 for a totally uniform structure, and increases with an increase in the scale of the disorder in the structure.

5.3.1 Properties of Handsheets

5.3.1.1 CV of Mass Density

The changes of CV of mass density at the 1 mm scale with settling time is shown in Figure 5-6; as the settling time increases, the formation deteriorates at each fines content level and the level of flocculation increases. This is expected since longer settling time gives the fibres more chances to entangle with each other. At the highest fines level, i.e. 40%, these changes with settling times are minor, as a result of its long drainage time at all formation levels. The 40% fines content pulp drains 10 times more slowly than that of the other pulp samples, see Table 5-9. At longer drainage times, there is more opportunity for fibres to entangle; however, the rate of entanglement is expected to decrease with time. Therefore a settling time of 120 seconds has a significantly reduced effect on fibre entanglement for the 40% fines content sample since the drainage time is already long, i.e. 70 seconds.
Figure 5-6. The effect of settling time at different fines content levels on the CV of mass density of the handsheet samples (the error bars covers +/- two standard deviations).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Drainage time (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% fines</td>
<td>5.0</td>
</tr>
<tr>
<td>20% fines</td>
<td>7.0</td>
</tr>
<tr>
<td>30% fines</td>
<td>12.0</td>
</tr>
<tr>
<td>40% fines</td>
<td>70.0</td>
</tr>
</tbody>
</table>

Table 5-9. The drainage times of pulps

5.3.1.2 Texture Features of Mass Density

In this study, every 0.5g/m² difference was set to be one level for texture analysis. The inspection zone size is 1 mm. This scale is selected according to the fact that human eyes can pick up a 0.02 print density difference from a normal reading distance in this scale.
Figures 5-7, 5-8 and 5-9 show the effects of settling time on homogeneity, contrast and correlation value, which focus on the uniformity, intensity and scale of formation, respectively.

The trend of homogeneity shows that the uniformity of the structure decreases with settling time at each fines content level. And this is confirmed by a t-test on the homogeneity data which shows that the homogeneity of each fines content sample at different settling time is different, except that this effect is less significant for samples with 30% fines. Although the CV of the 40% fines samples are quite similar for different settling times, the homogeneity of the sheets changes dramatically with longer settling times. This changes in homogeneity indicates that some local arrangement changes within the sheet with longer settling time and these types of local arrangement change can not be pick up by using a single CV value.

The contrast and correlation features provide description of the intensity and scale of the flocculation. The results indicate that for 0% fines content samples: at 5s settling time, the flocs are loose and large; at 60s settling time, the flocs increase both in size and density; and at 120s, the floc density increases dramatically but the scale of the flocs remains at the 5s settling time level. This forming pattern also applies to the 20% fines content samples. At a higher fines content level of 30%, at a 5s settling time, the paper structure is dense and flocculated with a much higher contrast and a similar correlation value in comparison to the 0% fines content sample. The high concentration of paste-like fines form coherent flocs that are not completely separated by mechanical agitation. It
appears that the flocs suspended in the pulp increase the density of fibre/fines flocs in the paper. At longer settling times, the flocs of fibre and fines join together to make bigger flocs without significantly changing the floc density, as indicating by the small changes in contrast and homogeneity values. At the 40% fines content level, the correlation value increases weakly with settling times and is lower than the corresponding lower fines content level, suggesting that the fines interfere with the ability of long fibres to form flocs with dense centers.

In general, at short settling times, the handsheets with higher fines content have higher floc density and poor formation. At longer settling times, both floc density and scale increase for low fines content samples, while, for high fines content samples, it is mainly a merging of small flocs with a moderate increase in correlation. For paper samples with fines, the existence of the fines flocs is an important characteristic of the handsheets. This results in larger but less dense flocs at higher fines content levels than those that at a lower fines level. At the 40% fines level, the fines appear to interfere with the long fibre fraction and the formation of dense flocs. The texture features present more information on the uniformity, the scale and the intensity of the flocs than that of the CV formation measure alone.
Figure 5-7. Settling time vs. homogeneity of handsheets with different fines content levels and at 1 mm scale.

Figure 5-8. Contrast vs. settling time for handsheet samples at different fines content levels and at 1 mm scale (the error bars are of +/- two standard deviations)
5.3.2 Fibre Orientation of Oriented Sheets

Three jet/wire ratios were used to produce sheets with different degrees of fibre orientation at four fines content levels using a Noram dynamic sheet former [61]. The jet flow rate is 1.32 m³/min, which produces an effective jet velocity of 790 m/min. Three wire rotation speeds: 950 rpm, 1100 rpm and 1250 rpm were chosen to vary the ratio. The
effective wire speeds on top of a 3 mm water wall were 847, 981 and 1115 m/min, resulting in jet/wire ratios of 0.93, 0.8 and 0.71, respectively.

The ratio of the zero-span strength in the machine direction over that of the cross machine direction was used as an indicator of the average fibre orientation in the sheets. A highly oriented sheet has a high zero-span ratio due to a large proportion of fibres aligning close to the machine direction, which provide a high strength in the machine direction. Figure 5-10 shows the effects of jet/wire ratio on fibre orientation. The results show that the zero-span ratio increases with decreasing jet/wire ratio (all ratios are less than one in this study) at each fines content level, indicating, as expected, that the greater the difference between the jet and wire velocity, the greater the orientation of the fibres along the machine direction. A t-test of the data also conforms that at each fines content level, the zero-span ratios at each jet/wire ratio are significantly different. Minimum zero-span ratios are achieved at the 30% fines level at each jet/wire ratio, however the underlying fluid dynamics, which led to this is not evident.
The contrast and correlation of the oriented samples measured in the machine and cross machine direction are shown in Figures 5-11 and 5-12, respectively. In agreement with Luner [15], the contrast in the cross machine direction is higher than along the machine direction. The t-test shows that the contrast and correlation in the machine direction are different from those of the cross machine direction. This difference is not significant for 40% fines content samples, as expected, due to the existing of a large portion of furnish, the fines, do not have the preferred orientation.

In this study, the ratio of the contrast value in the cross machine direction to that in the machine direction, CON(CD)/CON(MD) has been introduced and used as an indicator to evaluate the degree of the orientation of the sample. The results listed in Table 5-10 show that a higher contrast ratio is correlated with a lower jet/wire ratio, i.e. a higher difference
between jet velocity and wire velocity. This contrast ratio will be used to identify print density orientation as well. The correlation in machine direction is higher than in the cross machine direction, see Figure 5-12, which indicates structures aligning more linearly in the machine direction.

![Graph showing comparison of contrast in machine direction (MD) and cross machine direction (CD).](image)

Figure 5-11. Comparison of the contrast in machine direction (MD) and in cross machine direction (CD), (the error bars are of +- two standard deviations).
Figure 5-12. Comparison of the correlation in machine direction (MD) and in cross machine direction (CD), (the error bars are of ± two standard deviations).

<table>
<thead>
<tr>
<th>Fines Content (%)</th>
<th>Contrast Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mass Density</td>
</tr>
<tr>
<td></td>
<td>0.93</td>
</tr>
<tr>
<td>0</td>
<td>0.7</td>
</tr>
<tr>
<td>1.17</td>
<td>1.23</td>
</tr>
<tr>
<td>20</td>
<td>0.93</td>
</tr>
<tr>
<td>1.02</td>
<td>1.25</td>
</tr>
<tr>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>0.93</td>
</tr>
<tr>
<td>1.08</td>
<td>1.22</td>
</tr>
<tr>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>0.93</td>
</tr>
<tr>
<td>1.01</td>
<td>1.07</td>
</tr>
<tr>
<td>0.7</td>
<td></td>
</tr>
</tbody>
</table>

Table 5-10. The contrast ratios of the oriented sheets

5.3.3 Oriented sheets made using a single forming fabric
Paper formed using a single forming fabric at 0.93 jet/wire ratio shows clear drainage marks on the mass density map, and a typical VBR image of a 20% fines content sample is shown in Figure 5-13. It appears that there are stripes of light weight areas along both diagonal directions. Table 5-11 lists the differences between sheets made using a single forming fabric and a double forming fabric. The results show that although the appearance of these two types of sheets are different, the CV of samples 1 and 2 are similar at inspection zone sizes from 0.1 mm to 1 mm. The structural differences of these two sheets are indicated by the difference in the contrast values at high resolution ranges (from 0.1 mm to 0.4 mm). With resolution lower than 0.5 mm, these differences start to diminish due to the average effect of the large inspection zone size. The high contrast value of the single forming fabric sample representing a large grammage transition between the light weight stripes and the heavy weight stripes. The correlation values of these two types of paper are close to each other, which indicates that there is an underlying diamond pattern in the double fabric formed sheet but the less severe drainage in this case leads to less abrupt changes in density. Double fabric made papers were used for printing evaluation. As shown in Table 5-11, it is not a coincidence that the peak contrast value for both the single fabric and double fabric forming sheets shows around 0.4 to 0.5 mm zone size which is the size of the screen mesh, 0.423 mm or 60 mesh per inch.
Table 5-11. Comparison of the 20% fines content sheets made by a single fabric and a double fabric at 0.93 jet/wire ratio.

<table>
<thead>
<tr>
<th></th>
<th>Single fabric, zone size (mm)</th>
<th>Double fabrics, zone size (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.1  0.2  0.4  0.5  1.00</td>
<td>0.1  0.2  0.4  0.5  1.00</td>
</tr>
<tr>
<td>CV</td>
<td>5.8  5.63  5.20  5.04  4.20</td>
<td>5.93  5.77  5.33  5.10  4.20</td>
</tr>
<tr>
<td>CON</td>
<td>37.9 86.3 140.5 135.0 88.8</td>
<td>32.5 79.8 129.2 135.0 89.5</td>
</tr>
<tr>
<td>COR</td>
<td>0.91 0.78 0.58 0.55 0.54</td>
<td>0.92 0.78 0.58 0.52 0.51</td>
</tr>
</tbody>
</table>

In summary, there are four different types of sheets used in the print mottle study: handsheets, dynamic former sheets, pilot machine made sheets and commercial newsprint. The differences between the handsheets and the dynamic former sheets are not only due to the orientation effects, but also the existence of small fines flocs in the
handsheets. This effect has relevance to certain industrial situations in which small fines flocs may be formed when mixing different pulps or when a large amount of fillers are added to the furnish. In the dynamic former made sheets, differences are found between the sheets made with a double fabric or a single fabric. One extreme dynamic former sheet case is the 40% fines samples, where the overall structure of this paper is so closed that the mean pore size of these samples are smaller than a typical ink particle. The structure of pilot machine paper and commercial newsprint is similar, except that the formation of the pilot machine made paper is poorer. The results also show the ability of the texture analysis on identifying the difference between samples which can not be fully described by measuring the CV alone.

5.4 Print Mottle

In this section, coefficient of variation of print density and the structural print density parameters: homogeneity, contrast and correlation are used to describe the print unevenness of paper. The oriented sheet results are discussed first as a basis, then the results of the handsheets are discussed.

5.4.1 Print Mottle of Oriented Sheets

5.4.1.1 CV of Print Density

Figure 5-14 shows a general improvement in print density with an improvement in the CV of mass density. The correlation coefficient, r, was used to determine the interdependency of these two densities. The correlation coefficient, r, is defined as
\[
\sum_{y=1}^{m} \sum_{x=1}^{n} (P(x,y) - \overline{P(x,y)})(G(x,y) - \overline{G(x,y)})
\]
\[
\frac{r}{\overline{\delta_p(x,y)} \overline{\delta_o(x,y)}}
\]

where \( P(x,y) \) and \( G(x,y) \) are two data sets, and \( \delta_p(x,y) \) and \( \delta_o(x,y) \) are the variance of data set \( P(x,y) \) and \( G(x,y) \), respectively. The value of \( r \) is 0.498, indicating that these two parameter are related but there are other variables affecting print density as well.

The reason of this correlation is that hard-nip calendering of the samples before printing reduces the unevenness in the paper thickness and improves the smoothness of paper surface. However, the variation in grammage causes variation in density and the variation in pore size due to poor formation is retained following calendering. In paper with poor formation, different areas vary in roughness and density. While printing on a low density area, the contact between the print blanket and the paper is poor, ink penetrates more easily into the paper, and less ink is set on top of the surface. As a result, a light print density area is observed. In a high floc density area, on the other hand, there is good contact between the blanket and the paper, and a shallow depth of penetration of the ink; this leads to a dark print area. Variation in the ink distribution results in an uneven print density and a deterioration in print mottle.

Figure 5-15 is similar to Figure 5-14, except that it shows not only the CV of double fabric formed sheets but also those of single forming fabric sheets. At each fines content
level, the CV of print density of single fabric formed samples are higher than that of corresponding double fabric formed samples, although the CV of mass density are similar. This difference is likely due to the differences in pore size distribution as listed in Table 5-7. The CV of the pore size of the single fabric formed sheets is higher than that of the double fabric formed sheets due to the lost of fines through the drainage holes.

5.4.1.2 Textural Features of Print Density

In this study, every 0.02 difference in print density was set to be a print density level in calculating the co-occurrence matrix for texture analysis. 0.02 print density is selected since it is the smallest print density difference human’s eyes can identify in the normal 18” reading distance [2].

Homogeneity

Generally, as shown in Figure 5-16, the homogeneity of print density improves with the homogeneity of mass density at each fines content level. As expected 30% fines content samples led to the most uniform printing, which reflects by the higher homogeneity values at each formation homogeneity level. This is due to the fact that the samples have good surface smoothness and relatively uniform pore size distribution, see Section 5.2, which satisfies the need for both good contact between paper and printing disk, and uniform ink penetration. The effect of mass distribution on the 40% fines content samples is minor, which indicates by a close to zero slope of the data of 40% fines samples, see Figure 5-16. The print uniformity remains constant and ranked between the quality of 20% and 30% fines content samples. The samples with 40% fines have a very
closed structure with a mean pore radius of 0.4 μm, which is smaller than the average ink particle size. This may decrease ink transfer and most of the ink particles will set on the surface of paper. Since the ink is not absorbed properly, the ink particles may be relocated by the printing nip pressure and become independent of the mass uniformity. Similar situation is observed in the pilot machine made paper, the slope of the data, see Figure 5-16, is lower than those of lower fines content and more open samples. This phenomenon is also due to the very closed structure of the PL sheets as what happened in the 40% fines content sheets.

Figure 5-14. CV of print density vs. CV of mass density at 1 mm of oriented sheets made by dynamic former and PL is the pilot machine made sheets.
Figure 5-15. CV of print density vs. CV of mass density at 1 mm of oriented sheets made by dynamic former and PL is the pilot machine made sheets, S0% and S20% are the samples made using a single fabric, the fines content levels are 0% and 20%, respectively.
Contrast

Contrast focuses on the contribution of the neighboring differences, which is important for subjective ranking. Figure 5-17 shows that at each fines content level, a higher contrast in mass distribution leads to a higher contrast in print density, although at higher fines content level, the effect of local differences in grammage does not strongly effect the local print density arrangement. As shown in Figure 5-17, the slopes of the data of 30% and 40% fines content are lower than those of lower fines content samples. This lower sensitivity to the contrast of mass density, combining with the best homogeneity of print density, indicates that 30% fines content is an ideal furnish to make printing paper.
The slope of the PL samples are between the low fines content and high fines content samples.

Figure 5-17. Contrast of mass density vs. Contrast of print density of oriented sheets made by dynamic former and pilot machine made sheets (PL) at 1 mm scale.

Correlation

In contrast to other structural parameters, the effect of local correlation of mass density on the correlation value of print density is independent of the fines content, as shown in Figure 5-18. A larger flocculated area in the original paper will result in a larger area of uneven print, even for samples made from different furnishes and by different paper making methods, see Figure 5-18.
Figure 5-18. Correlation of mass density vs. Correlation of print density of oriented sheets made by dynamic former and Pilot machine made sheets (PL) at 1 mm scale.

Orientation

Orientation is another factor associated with machine made sheets. Table 5-12 shows that both mass and ink particles tend to distribute along the machine direction. The orientation of print density is less than the corresponding orientation of mass density, indicating that the printing process smooth out some of the mass orientation effects.
Table 5-12. Orientation of mass density distribution and print density distribution

<table>
<thead>
<tr>
<th>Fines Content (%)</th>
<th>Contrast Ratio</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mass Density</td>
<td>Print Density</td>
</tr>
<tr>
<td>0</td>
<td>j/w=0.93</td>
<td>1.26</td>
</tr>
<tr>
<td></td>
<td>j/w=0.7</td>
<td>1.44</td>
</tr>
<tr>
<td>20</td>
<td>j/w=0.93</td>
<td>1.22</td>
</tr>
<tr>
<td></td>
<td>j/w=0.7</td>
<td>1.47</td>
</tr>
<tr>
<td>30</td>
<td>j/w=0.93</td>
<td>1.22</td>
</tr>
<tr>
<td></td>
<td>j/w=0.7</td>
<td>1.45</td>
</tr>
<tr>
<td>40</td>
<td>j/w=0.93</td>
<td>1.18</td>
</tr>
<tr>
<td></td>
<td>j/w=0.7</td>
<td>1.24</td>
</tr>
</tbody>
</table>

5.4.2 Print mottle of handsheets

CV and Homogeneity of Print Density

Figure 5-19 shows the effect of CV of mass density on the CV of print density for handsheets. The correlation coefficient, r, is 0.15, which indicates that formation is not the only factor affecting the ink and paper interrelationship. The results of the texture analysis indicate that the print mottle of paper is affected by both the formation and the fines content. Print uniformity improves as the formation of paper is enhanced at each fines content level, although as shown in Figure 5-20, handsheets with fines have lower homogeneity compared with handsheets without fines. This is in contrast with the oriented sheet measurements. This difference may be attributed to the difference in the forming method.
As discussed in the section of the properties of fines in this chapter, there are fines flocs existing in the re-mixed furnishes. In the dynamic former, the high jet flow through the small nozzle breaks fines flocs and helps to disperse the fines in the wet web structure. However, in the case of handsheet forming, without a high shear force to break and disperse the fines, these fines flocs are retain in the resulting paper. The average pore size is very small in areas with fines flocs, whereas some large pores remained in the paper structure in areas where the fines flocs can not completely fill the interfibre pores. As a result, the CV of the pore size is higher than those samples made by the dynamic former or the newsprint sample, see Tables 5-5 and 5-8. In the fines rich areas, the pores are so small that most of the ink is held out on the surface and these areas appear as dark areas. On the other hand, the loose long fibre and fines areas absorb ink easily and appear as light areas. The high contrast value of the mass distribution for good formation (5s settling time) samples with 30% and 40% fines, as shown in Figure 5-21, confirms this phenomena.

**Contrast and Correlation**

The slopes of the contrast data the high fines content samples, 30% and 40%, as shown in Figure 5-21, are higher than those of lower fines content samples. This result shows that a stronger effect of formation on print uniformity at higher fines content level. This higher sensitivity is also due to the contribution of the fines flocs existing in the paper as discussed in the last paragraph, which causes a large ink penetration variation even with small changes in formation.
This result indicates that not only sufficient fines are needed for good print quality, but also the fines in the furnish must also be well dispersed for good print quality. With 0% fines samples, the overall structure of the paper is so open that it facilitates uniform ink penetration, i.e. at both light weight and highly flocculated areas and compensates for the effect of poor formation. Figure 5-22 shows the unevenness scale, or the correlation of the print and the mass density are related, but the relationship is not as strong as the case for the oriented sheet samples.

Figure 5-19. CV of print density vs. CV of mass density of handsheets at 1 mm
Figure 5-20 Homogeneity of print density vs. CV of mass density of handsheets at 1mm scale

Figure 5-21. Contrast of print density vs. CV of mass density of handsheets at 1mm scale
5.4.3 **Ink Amount and Ink Penetration Depth**

The ink amount transfer and the ink penetration are the factors controlling print density.

Table 5-13 shows that samples without fines need more ink to reach the target print density. The ink amount for samples with fines continues to decrease with an increase in fines content. A thicker ink film in the printing disk provides a better coverage of the paper surface to compensate for the rough surface. The ink penetration depth is much deeper in the loose structure of the fines free samples and more ink is required to provide the visibility of the ink.

![Figure 5-22. Correlation of print density vs. CV of mass density at 1mm scale](image-url)
<table>
<thead>
<tr>
<th>Sample</th>
<th>Ink amount on disk (g/m²)</th>
<th>Ink amount transfer (g/m²)</th>
<th>Average ink penetration Depth (um)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% fines</td>
<td>3.9</td>
<td>2.1</td>
<td>34</td>
</tr>
<tr>
<td>20% fines</td>
<td>3.3</td>
<td>1.6</td>
<td>26</td>
</tr>
<tr>
<td>30% fines</td>
<td>3.1</td>
<td>1.4</td>
<td>24</td>
</tr>
<tr>
<td>40% fines</td>
<td>3.1</td>
<td>1.35</td>
<td>23</td>
</tr>
</tbody>
</table>

Table 5-13. Ink amount required by each handsheet sample to reach the target print density of 1.0
5.5 Pointwise Correlation

The previous discussion is based on the overall evaluation of the effects of structural properties on print mottle. In addition, a better understanding of the local interrelationship between paper structure and the ink transfer and penetration is important to the study of print density. The experimental methodology of this pointwise study was fully described in Chapter 4 and is summarized in the following paragraph.

A newsprint sample was printed using a printing disk carrying a known uniform layer of ink, then a IGT sheet was printed using the remaining ink. Due to the double coating layer on the IGT sheet, the ink transferred to the IGT sheet is held out on the paper surface. The uneven print density shown on the IGT sheet was used to determine the local amount of ink transfer to it, and as a result, the local amount of ink transferred to the corresponding newsprint sample was determined based on a known original amount of ink on the printing. The correlation between print density and ink amount on the IGT sheets were obtained by printing 10 IGT sheets using different amounts of ink on the printing disk. The local correlation between print density and mass density was obtained by comparing the print density map and the VBR image of the paper.

Figures 5-23, 5-24 and 5-25 are the calibration curves of print density vs. gray level, IGT print density vs. gray level, and mass density vs. gray level, respectively. The measuring area for each calibration data point was 1 cm x 1 cm.
Figure 5-23. Calibration curve of print density vs. gray level

Figure 5-24. Calibration curve of ink amount transferred to the IGT sheet and the print density of IGT sheet
Figure 5-25. A typical calibration curve of grammage vs. gray level over the range of 15 to 100 g/m²

5.5.1 Pointwise Dependency

The correlation coefficient, r, as defined by Equation 5-1, was used to determine the interdependency of any two of the local grammage, local print density and local ink amount. A 0.45mm square inspection zone was chosen for the pointwise comparisons, and the measuring area was 2 cm x 2 cm. A typical plot of the print density vs. local grammage of a newsprint sample is shown in Figure 5-26. Three more replicate experiments were conducted and the results, as listed in Table 5-14, are in agreement with the above sample.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>r (print density vs. local grammage)</th>
<th>r (print density vs. ink transfer)</th>
<th>r (local grammage vs. ink transfer)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>0.52</td>
<td>-0.08</td>
<td>0.089</td>
</tr>
<tr>
<td>S2</td>
<td>0.47</td>
<td>-0.12</td>
<td>0.11</td>
</tr>
<tr>
<td>S3</td>
<td>0.45</td>
<td>-0.09</td>
<td>0.075</td>
</tr>
</tbody>
</table>

Table 5-14. The coefficients of correlation of local parameters at 0.45 mm scale
For the newsprint samples shown in Figure 5-26, the correlation coefficient between local print density and mass density is 0.62, considering the large sample size, 1936 data points, the dependency of these two densities are strong. The trend of data shows that generally, print density locally increases as local grammage increase, i.e. an area of high print density is usually an area of high grammage.

It is known that any factors affecting ink transfer, ink retain and ink visibility will affect print density. For hard nip calendared sheets, such as newsprint, the correlation between local print density and local mass density may be explained as follows. In a hard nip calendared newsprint sample, the caliper of the sheet is relatively uniform over the whole sheet. Consequently, the difference in local mass density determines the difference in local density and compressibility. In the higher mass zones, the density is high and the pores in these zones are small. It is difficult for ink particles to penetrate into the paper structure, and as a result, more ink particles are held close to the surface of the paper increasing the visibility of the ink. Therefore, local high mass density areas appear as high print density areas and conversely, local low mass density areas appear as low print density areas.
Figure 5-26. Local print density vs. local mass density at 0.45 mm scale

A weak correlation \((r=0.1)\) between the amount of ink transferred to the paper and the print density indicates that ink transfer is not a dominate factor affecting print density for newsprint, see Figure 5-27. This is in agreement with a previous study [14] which showed that a print density of 1.0, a set point in this study, is close to the maximum print density that newsprint can reach \((= 1.2)\). In this case, print density is less sensitive to the amount of ink transferred, as long as the ink amount is sufficient to cover the surface of paper. Furthermore, as discussed in the last section, the visibility of ink particles depends on the location of particles in the paper structure. It is possible that in light weight areas, the structure of paper is so open that the majority of the ink particles penetrate deeply into the structure independently of ink transfer. This possibility is supported by the fact that the ink amount required to reach a print density of 1.0 is almost doubled for a 0% fines content sample compared to a high fines content sample. The overall openness of the 0% fines content sheets dramatically decreases the visibility of the ink particles, which is
similar to what happen in a light weight area in a flocculated sheet.

The small negative correlation value (-0.06) between the local grammage and the amount of ink transfer indicates that ink transfer is not affected by the local mass density, see Figure 5-28. The amount of ink transfer depends on the surface smoothness and the immobilization of ink. A locally smooth area, usually a high mass density area, provides a good contact between the paper and the printing disk and helps ink transfer. Conversely, since the ink penetration in this area is low, i.e. the amount of ink immobilized into the system is low, and as a result, at the exit of the printing nip, the free ink film tends to adhere to the printing disk more and less ink is transferred to this area. The above two factors are competing factors which controls the local ink amount transfer. For a relatively smooth sample, such as newsprint, the effect of local mass density on the ink transfer is minor. The negative correlation coefficient implies that in some high mass density area, a shallow ink penetration depth will contribute to a higher print density, even if the ink transfer is low. This also indicates the importance of ink penetration in wood-containing paper. In general, it has been demonstrated that a pointwise dependency study is a useful tool in determining the key factors that affect print quality.
Figure 5-27. Print density vs. ink amount transferred at 0.45 mm scale

Figure 5-28. Local grammage vs. ink transfer at 0.45 mm scale
6. PRINT MOTTLE MODEL

6.1 Introduction

Kajanto [61] developed a theoretical model to predict the effects of various factors on the uniformity of print density based on two-layered Kubelka-Munk theory as shown in Figure 6-1. The model assumes that the printed paper consists of a homogeneous ink-paper-mixed layer and a paper layer, and there is no ink setting on the surface of paper. This model is most suitable for printed samples with little ink setting on the top of the surface of paper, such as newsprint samples. In order to determine the relative importance of different parameters on printing uniformity, Kajanto derived the following Equation 6-1. The relative change in print reflectance, \((dR/R)\), as described in the beginning of Chapter 2, which is related to the uniformity of print density, is described as a function of the relative changes of the ink transfer to paper \((dy/y)\), the grammage of the ink penetrated paper layer \((dW_m/W_m)\), the light scattering coefficient of paper \((dS_p/S_p)\) and the light absorption coefficient \((dK_p/K_p)\). From the definition of optical print density \(D = \log(1/R)\), it can be further derived that the relative change in print density \((dD/D)\) is related to \(dR/R\) by the expression \(dD/D = (1/\ln(R)) \times dR/R\). So \(dR/R\) reflects the uniformity of printing as described above. The governing equation is,

\[
\frac{dR}{R} = a_1 \frac{dy}{y} + a_2 \frac{dW_m}{W_m} + a_3 \frac{dS_p}{S_p} + a_4 \frac{dK_p}{K_p}
\]  

(6-1)
where $a_1$, $a_2$, $a_3$ and $a_4$ are constants and can be calculated if the averages of $y$, $W_m$, $S_p$, $S_i$, $K_p$ and $K_i$ are known, see Appendix II. $S_i$ is the light scattering coefficient of ink and $K_i$ is the light absorption coefficients of ink.

All the terms of Equation 6-1 are directly measurable except the “black box” variable $W_m$, the grammage of the penetrated layer. It is desirable to develop a method to express $W_m$ in terms of measurable paper properties and printing conditions. This relationship is also important for the fundamental understanding of ink-paper interaction. In the following section, an equation was derived for determining $W_m$ as a function of printing conditions and paper properties. The general approach is to predict the penetration depth based on the modified Kozeny-Carmen equation. In subsequent sections, the new form of the print mottle model is used to interpret the data sets presented in the previous chapter.

Figure 6-1. A two-layered model of the printed paper [61]
6.2 Model Derivation

Newsprint printing inks consist of ink pigments and vehicle oil. The ink penetration process can be divided into two stages as shown in Figure 6-2. In the first stage, ink penetrates into the paper surface as a homogenous mixture of ink pigments and vehicle oil under the printing pressure. Once the printing pressure releases, a chromatographic separation between ink particles and vehicle oil takes place due to differences in affinity, solubility and size. In this stage, the movement of ink particles ceases and the vehicle oil continues to penetrate deeper into the paper structure by capillary suction and spreading [62]. Since the black ink particles play a dominant role on the print density and uniformity, the further penetration of vehicle oil is neglected and only the first stage penetration is of interest. Thus the ink penetration depth \( h \) discussed in subsequent sections refers to \( h_w \) only, as defined in Equation 6-2.

![Figure 6-2 Ink penetration: \( h_w \) is the penetration depth of the ink particles and solvent, \( h_s \) is the further penetration depth of the solvent.](image)

The mean penetration depth of ink into an area, \( A \), under an external pressure is governed by
where $h$ is the mean penetration depth, $r$ is the effective pore radius, $\varepsilon$ is the average void fraction, $p$ is the printing pressure, $t$ is the dwelling time, $\eta$ is the viscosity of ink and $\tau$ is the tortuosity ratio of paper. There are three factors associated with the paper structure: the porous radius, the void fraction and the tortuosity ratio. Tortuosity ratio is a material related constant and is ranging from 2 to 12 for different types of paper [62].

The void fraction is correlated to the mass density by

$$\frac{\beta}{Z} = \rho_f \ast (1 - \varepsilon) \tag{6-3}$$

where $Z$ is the total thickness of paper, $\beta$ is the grammage and $\rho_f$ is the density of fibre.

By rearranging equation 6-3 and substituting it into equation 6-2, one obtains

$$h = (1 - \frac{\beta}{Z\rho_f}) \frac{r}{2\tau} \sqrt{\frac{lp}{\eta}} \tag{6-4}$$

and

$$W_m = \frac{\beta}{Z} (1 - \frac{\beta}{Z\rho_f}) \frac{r}{2\tau} \sqrt{\frac{lp}{\eta}}. \tag{6-5}$$
For a hard-nip calendered sheet, the caliper of the paper sample, $Z$, is constant. If the sheet is then printed under a constant printing pressure and dwelling time, the relative change of $W_m$ becomes

$$\frac{dW_m}{W_m} = \frac{(Z\rho_f - 2\beta) \, d\beta}{(Z\rho_f - \beta) \, \beta} + \frac{dr}{r}$$

Upon substitution of the expression for $dW_m/W_m$, equation 6-1 becomes

$$\frac{dR}{R} = a_1 \frac{dy}{y} + a_2 \frac{(Z\rho_f - 2\beta) \, d\beta}{(Z\rho_f - \beta) \, \beta} + a_3 \frac{dr}{r} + a_4 \frac{dS_p}{S_p} + a_4 \frac{dK_p}{K_p}$$

### 6.3 Qualitative Discussion of the Model

The parameters used in the print mottle model of the TMP newsprint are listed below in Table 6-1 and are typical for sheets made from mechanical pulps. Based on the values listed in the table, the coefficient $a_1, a_2, a_3, and a_4$ are found to be -0.43, 2.2, -0.41 and -0.00068, respectively. It is clear that $a_2$ is significantly greater than other coefficients. This indicates that the relative change of $W_m$ is the dominant factor affecting the change of print density. This result is in agreement with the experimental results of pointwise correlation which indicating that local print density is related to local mass density, which controls ink penetration. The subsequent discussion will therefore focus on the behavior
of $dW_m/W_m$ under various conditions. The ideal paper structure for minimizing print mottle should be a structure with a minimal $dW_m/W_m$ with paper structural property changes.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Assigned Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average ink transfer</td>
<td>$\gamma$</td>
<td>1.6 g/m²</td>
</tr>
<tr>
<td>Average grammage of the ink penetrated layer</td>
<td>$W_m$</td>
<td>10.5 g/m²</td>
</tr>
<tr>
<td>Average scattering coefficient of paper</td>
<td>$S_p$</td>
<td>0.038 m²/g</td>
</tr>
<tr>
<td>Average absorption coefficient of paper</td>
<td>$K_p$</td>
<td>0.0068 m²/g</td>
</tr>
<tr>
<td>Average scattering coefficient of ink</td>
<td>$S_i$</td>
<td>0.0175 m²/g</td>
</tr>
<tr>
<td>Average absorption coefficient of ink</td>
<td>$K_i$</td>
<td>3.45 m²/g</td>
</tr>
</tbody>
</table>

Table 6-1. Newsprint printing parameters

### 6.3.1 The Effect of Fines Content on $dW_m/W_m$

Equation 6-6 shows that there are two factors that affect the relative change of the grammage of the ink penetrated layer, $W_m$: the change in grammage of paper ($d\beta/\beta$) and the change in pore size of paper ($dr/r$). As mentioned in the previous section, an ideal paper sample for uniform printing is the one in which the change in the grammage of the
penetrated layer, \( dW_m/W_m \), is small, even if there are relative large changes in paper structure, such as a large changes in \( d\beta/\beta \) or \( dr/r \).

It is known that \( d\beta/\beta \) and \( dr/r \) are related when caliper, \( Z \), of the sheet is constant, such as in the case of a hard-nip calendered sheet. Since as the local grammage \( \beta \) increases, the local mean pore radius \( r \) must decreases, i.e. grammage and pore radius are negatively correlated. \( dW_m/W_m \) changes only moderately with changes in \( d\beta/\beta \) and \( dr/r \).

### 6.3.1.1 The Effect of Fines Content on Print Mottle

The following discussion of the effect of fines content on print mottle is based on the effect of the void fraction on print mottle since typically fines content primarily determines the void fraction. The ratio \( \frac{(Z\rho_f - 2\beta)}{(Z\rho_f - \beta)} \) is defined as \( C_1 \) and is determined by the void fraction according to Equation 6-3. The impact of changes in void fraction on the changes in the grammage of the ink penetrated layer is now considered.

If the void fraction is set to 0.5, \( C_1 \) becomes zero and \( \frac{dW_m}{W_m} = \frac{dr}{r} \). In this case, the relative change of the grammage of the penetrated layer is entirely controlled by the variation in pore radius. For void fractions greater than 0.5, i.e. an open sheet, \( 1>C_1>0 \), the variation in grammage gradually becomes dominant in determining the variation of the grammage of the penetrated layer. For paper with a relatively closed structure, such as a high fines content sheet, the void fraction is less than 0.5, and in this case, \( C_1<0 \). Thus the relative
change of grammage becomes a negative contributing factor, that is, changes in grammage and pore size contribute to changes in $W_m$, instead of competing against each other. In general, for hard nip calendered sheets, there is an optimum fines content which will result in a minimum relative change in the grammage of the penetrated layer with certain changes in grammage and pore size.

6.3.1.2 Comparison Of The Trends of the Experimental Data and the Model Prediction

Two sets of sheets with different fines content levels are used for comparison: the handsheets and the dynamic former sheets. For sheets made using the dynamic former, as shown in Figures 5-14 and 5-15, the print density uniformity improves with increasing fines content from 0% to 30%, and then decreases at 40% fines content. The samples with 30% fines produce the best uniformity of printing, based on the homogeneity and CV criteria. The model supports this experimental trend and is discussed below.

The void fraction $\varepsilon$ for each sample is determined based on the measured grammage and caliper of the paper and is listed in Table 6-2. The coefficient $C_1$ of each sample is calculated and, as shown in Table 6-2, $C_1$ decreases from 0.51 to -0.25 as the fines content increases from 0% to 40%. As discussed in the above section, there is an optimum fines content where $dW_m/W_m$ is minimized for certain changes in grammage and pore size. In this case, the ideal fines content must be lower than 40%, since $C_1$ has already turned negative at 40% fines content. When $C_1$ becomes negative, both changes
in grammage and pore size contribute to the changes in $W_m$, and as a result, a relatively small unevenness in the paper structure will cause a relatively large change in print density. Consequently, 40% fines content samples do not result in the best printing quality. With the fines content increases from 0% to 30%, the relative contribution of $d\beta/\beta$ to print mottle decreases, since $C_1$ decreases from 0.51 to 0.08. At 30% fines content, the effect of formation on print mottle is minor. This may explain why the contrast of the print density is independent of the contrast of the mass density of the sheet as observed in Figure 5-17. Since the CV of the pore size distribution of the 30% fines sample is low, i.e. $dr/r$ is small, the model predicts low $dW_m/W_m$ in agreement with experimental results.

Figure 5-15 shows that at each fines content level, the sheets made using a single fabric have much higher unevenness than those made by a double fabric, although the formation level of these two types of sheets are similar. This may be explained by the model as follows: since the $dr/r$ of the single fabric sheets are much higher than those of the double fabric sheets at each fines content level, see Table 5-7, $dW_m/W_m$ for a single fabric sheet should be much higher than that of a double fabric sheet.

The print mottle model is based on an ink penetration model, and the oriented sheets with 40% fines content tests the limits of this model. As listed in Table 5-7, the mean pore size of 40% fines samples is 0.4 $\mu$m which is smaller than the mean ink particle size of 0.5 $\mu$m. It is expected that, overall, ink particles will not be able to penetrate into the
paper structure. Without ink penetration, the influence of the formation i.e. homogeneity, of the unprinted sheets in the $W_m$ is less than for other paper samples. This may explain the independence of the homogeneity of the print density on the homogeneity of the mass density of 40% fines content sheets.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fines Content(%)</th>
<th>Void fraction</th>
<th>$C_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dynamic former sheet (D-0)</td>
<td>0</td>
<td>0.65</td>
<td>0.51</td>
</tr>
<tr>
<td>Dynamic former sheet (D-20)</td>
<td>20</td>
<td>0.59</td>
<td>0.30</td>
</tr>
<tr>
<td>Dynamic former sheet (D-30)</td>
<td>30</td>
<td>0.51</td>
<td>0.08</td>
</tr>
<tr>
<td>Dynamic former sheet (D-40)</td>
<td>40</td>
<td>0.42</td>
<td>-0.25</td>
</tr>
<tr>
<td>Newsprint</td>
<td>25</td>
<td>0.66</td>
<td>0.47</td>
</tr>
<tr>
<td>Pilot machine made paper</td>
<td>28</td>
<td>0.62</td>
<td>0.45</td>
</tr>
</tbody>
</table>

Table 6-2. The effects of fines content on the void fractions and $C_1$ of oriented sheets

For handsheets, the print uniformity of the samples with high fines contents of 30% and 40% is worse than that of 0% and 20% samples, as shown in Figure 5-19. This trend is opposite to that of the dynamic formed sheets which has been discussed above. This may be due to the structural difference between handsheets and dynamic former sheets as illustrated in the schematic diagrams shown in Figures 6-3 and 6-4. As discussed in Chapter 4, the fines flocs in the handsheets will result in a large pore size CV. The void fraction $\varepsilon$, and corresponding $C_1$ coefficient as determined by Equation 6-2 are listed in Table 6-3 and the coefficient $C_1$ of each sample are then calculated using Equation 6-2. $C_1$
decreases from 0.53 to -0.22 as the fines content increases from 0% to 40%. Since $C_1$ for 30% fines is so small, the relative change of $W_m$ mainly depends on the relative change of pore size as shown in Equation 6-6. Compared with the samples with low fines content and without fines, the CV of pore size for 30% and 40% fines samples are higher, which results in a worse print.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fines Content(%)</th>
<th>Void fraction</th>
<th>$C_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Handsheet (H-0)</td>
<td>0</td>
<td>0.68</td>
<td>0.53</td>
</tr>
<tr>
<td>Handsheet (H-20)</td>
<td>20</td>
<td>0.60</td>
<td>0.33</td>
</tr>
<tr>
<td>Handsheet (H-30)</td>
<td>30</td>
<td>0.53</td>
<td>0.11</td>
</tr>
<tr>
<td>Handsheet (H-40)</td>
<td>40</td>
<td>0.45</td>
<td>-0.22</td>
</tr>
</tbody>
</table>

Table 6-3. The effects of fines content on the void fractions and $C_1$ of oriented sheets
Figure 6-3. Schematic diagram of a dynamic former paper

Figure 6-4. Schematic diagram of a handsheet
6.3.2 The Effect of Formation on Print Mottle

It is known that the relative change of pore radius $\frac{dr}{r}$ is proportional to the relative change of grammage $\frac{d\beta}{\beta}$ for a paper with constant caliper, such as the case for a hard-nip calendered sheet, i.e. $\frac{dr}{r} \propto \frac{d\beta}{\beta}$.

In this case, equation 6-6 becomes

$$\frac{dW_n}{W_n} = M \frac{d\beta}{\beta} \quad (6-8)$$

where $M$ is a regression constant related to paper structure. This relationship indicates that a higher variation in the grammage of paper will result in a higher variation in the grammage of the ink penetrated layer, which will cause a higher variation in the reflectance of the printed image.

The experimental results presented in the experimental section indicate that printing uniformity improves as the formation of paper is enhanced at each fines content level. This trend is in agreement with the trend predicted by the model.
7. CONCLUSIONS AND RECOMMENDATIONS

A fundamental study of the effects of paper structural properties on print mottle was conducted. The primary pulp used in the study was TMP pulp and a IGT proof printer was used to simulate offset printing. The main conclusions are summarized in the following sections.

7.1 Measurement of print mottle and paper uniformity

A second order statistical method, based on the spatial gray level dependence method (SGLD), was developed and used to evaluate the structural characteristics of both the mass density map and the print density map of handsheets, dynamic former sheets and machine made paper. Texture features of homogeneity, contrast and correlation, combined with the coefficient of variation (CV) give a complete description of print mottle and formation. This new method is more sensitive to both intensity and scale changes than using coefficient of variation alone. Structural variance and print density variance due to changes in orientation, drainage marks and uneven mixing can be identified by this second order textile analysis method. In general, this new method offers several advantages over traditional methods.

7.2 Prediction of print mottle

A model was developed based on a two-layered Kubelka-Munk model to relate print density variations to structural variations. This model is in a usable form and no fitting
parameters are required. In this study, the model is able to predict the experimental observations, e.g. 30% fines content oriented sheets are ideal for printing, the differences of the printing quality between the sheets made by double fabrics and a single fabric, and the poor printing quality of handsheets with poor fines distribution. This model will be a useful tool to qualitatively relate measurable paper properties to print uniformity. This model can also identify the relative importance of the printing conditions, paper uniformity and ink penetration to print mottle for different samples.

### 7.3 Effects of fines content, formation and orientation on print mottle

Print mottle is affected by both the formation level, the fines content of the sheets and fines distribution. During the fines separation process, fines flocs form which persist when fines and long fibre are blended and during the British handsheet making process. The existence of small fines flocs in the handsheet results in a higher CV of pore size in the paper. In the fines-rich areas, the pores are so small that most of the ink is held out on the surface and appear as dark color areas. The loose long fibre areas absorb ink easily and appear as light color areas. High contrast values were recorded for handsheets with high fines content, which supports this observation.

This finding emphasizes that not only the amount of fines but also the uniform distribution of fines is important for a uniform print density. This indicates the importance of proper mixing and storage of different types of pulps in industrial processes.
At each fines content level, print mottle increases as formation deteriorates. The effects of formation are the most dramatic for the 40% fines content sample, a small change in formation causes a relatively large change in print density. This may due to the effect of the long drainage time, which forms flocs with dense centers and this center become denser and denser with longer settling time. In this case, pore size CV is high, even with a small change in grammage. This unevenness in structure is less seriously for a dynamic former sheet, since the high shear force acting on the pulps at the exit of the nozzle helps to break the fines flocs and re-distribute the fines.

The structure of oriented sheets changes with fines content with similar trends to that of handsheets. Oriented sheets with 30% fines give the best printing results, although the surface roughness of all oriented sheets are similar. The key factor for uniform printing in wood-containing oriented or machine made paper is the uniform penetration of ink. For an extreme case which dose not have ink penetration, the oriented samples with 40% fines, the effect of formation on print density is minor since the structure is very closed and the average pore size is smaller than the average ink particle size. In this structure, most of the ink particles were held out on the surface of paper, and decreased the dependence of the printing unevenness on the structure of paper.

A pointwise study of the dependence of the local print density, mass density and ink amount for a typical wood-containing paper, newsprint, shows that only the mass density and print density are locally correlated. The high mass density areas usually appear as
high print density areas, since more ink particles are held out close to the surface of paper due to the inability of ink to penetrate in these high mass density areas for hard-nip calendered newsprint samples. This result indicates the local ink penetration has a stronger influence on local print density than on local ink amount transfer.

The traditional large-scale measurements of paper surface and density properties can identify large changes in the structure caused by different fines content, but can not distinguish structural differences caused by differences in formation. Homogeneity, contrast, correlation and coefficient of variation together provide a comprehensive description of structural differences important to print quality.
The following are recommended for the further study of the effect of paper structural properties on print mottle of wood-containing paper:

- Investigate the relationship between the texture features: energy, homogeneity, contrast and correlation, and the size of the inspection zones. It may be possible to identify the "fingerprint" of each structure through this study.

- Extend the texture study based on different pre-defined mass density and print density transition patterns, possibly to identify suitable measurement interval that agrees with objective ranking.

- Expend the experimental study to include soft-nip calendered sheets and to establish a model for soft-nip calendered sheets in which the density of the paper is constant over the whole sample area.

- Investigate the pointwise correlation between print density, mass density and ink amount at different average print densities.
8. REFERENCES


42. Operation manual of Coulter® Porometer.


47. Mohlin, U-B., “Properties of TMP fractions and their importance for the quality of printing papers, Part 1: Large variations in properties within fractions are observed”, Svensk Papperstidn, 83(16), P461(1980).


66. Operation manual of the Optest Fibre Quality Analyzer


Appendix 1: Sample calculation of the intermediate matrices of print density

The first step of the spatial print density dependence method is to scan the print density map in four principal directions and to calculate the co-occurrence matrix. Here a simply example will to given to demonstrate the process.

For demonstration purpose, a small section of a sample with only three different print densities are used. There are 4 x 4 inspection zones in the sample as shown bellow:

```
<table>
<thead>
<tr>
<th>1.02</th>
<th>1.00</th>
<th>0.98</th>
<th>1.02</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>1.00</td>
<td>1.02</td>
<td>1.00</td>
</tr>
<tr>
<td>1.00</td>
<td>0.98</td>
<td>0.98</td>
<td>1.02</td>
</tr>
<tr>
<td>1.02</td>
<td>1.02</td>
<td>1.00</td>
<td>1.02</td>
</tr>
</tbody>
</table>
```

**Figure A-1: print density map**

Print density difference 0.02 was set to be a print density level, in order to obtain a co-occurrence matrix in the horizontal direction, the image was scan along the horizontal direction. In this sample, as shown in Figure A-2, the entry of the horizontal matrix corresponding to row 1 and column 1 is the number of pairs of print density (0.98, 0.98) along the scanning direction. There is only one pair found and the entry is 1. Through this method, the entry of the matrix was obtained, for example, the transition for 1.02 as first point to 1.00 as second point happened twice, and the entry to the co-occurrence matrix is 2 for column 3 and row 2 is 2, as shown in Figure A-2.
Print density of the second point

<table>
<thead>
<tr>
<th></th>
<th>0.98</th>
<th>1.00</th>
<th>1.02</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>1</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>1.00</td>
<td>2</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1.02</td>
<td>0</td>
<td>3</td>
<td>1</td>
</tr>
</tbody>
</table>

**Figure A-2: The horizontal co-occurrence matrix (scanning direction: left to right)**

Then the co-occurrence matrices were symmetricized as shown in Figure A-3 and normalized to get the probabilities as shown in Figure A-4. All the horizontal related information are now in the matrix and texture features can be calculated according to the definition shown in Chapter 4.

Print density of the second point

<table>
<thead>
<tr>
<th></th>
<th>0.98</th>
<th>1.00</th>
<th>1.02</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>2</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>1.00</td>
<td>3</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>1.02</td>
<td>2</td>
<td>4</td>
<td>2</td>
</tr>
</tbody>
</table>

**Figure A-3. The symmetric horizontal matrix**
### Print density of the second point

<table>
<thead>
<tr>
<th></th>
<th>0.98</th>
<th>1.00</th>
<th>1.02</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>0.083</td>
<td>0.125</td>
<td>0.083</td>
</tr>
<tr>
<td>1.00</td>
<td>0.125</td>
<td>0.083</td>
<td>0.167</td>
</tr>
<tr>
<td>1.02</td>
<td>0.083</td>
<td>0.167</td>
<td>0.083</td>
</tr>
</tbody>
</table>

**Figure A-4. The normalized probability matrix**
Appendix 2: Definition of the constant a1, a2, a3 and a4 [61]

The constant a1, a2, a3 and a4 in Kajanto's model are defined below. These constants can be calculated if the averages of y, W, S, S, Kp and Ki are known.

\[
a1 = A' \frac{W_m^2}{C_3} (C_1 + C_4 b \frac{S_p}{W_m}) + \frac{D}{(y + W_m)^2} (D_1 + D_3 b \frac{S_p}{W_m}) - E
\]

\[
a2 = A' \frac{W_m^2}{C_3} (C_2 + C_4 b \frac{y}{W_m^2}) - \frac{D}{(y + W_m)^2} (D_2 + D_3 b \frac{y}{W_m^2}) - B
\]

\[
a3 = -A' \frac{W_m^2}{C_3} (C_4 + \frac{DD_3}{(y + W_m)^2}) (1 - b \frac{y}{W_m})
\]

\[
a4 = A' \frac{W_m^2}{C_3}
\]

where

\[
C_1 = k_p S_i + k_i S'_p W_m \approx k_i S'_p W_m
\]

\[
C_2 = k_p S_i + k_i S'_p y \approx -k_i S'_p W_m y_m
\]

\[
C_3 = (y S_i + W_m S'_p) W_m \approx S'_p W_m^2
\]

\[
C_4 = (k_i y + k_p W_m) W_m \approx k_i y W_m
\]

\[
D_1 = (S'_p - S_i) W_m \approx S'_p W_m
\]

\[
D_2 = (S'_p - S_i) y \approx S'_p y
\]

\[
D_3 = (y + W_m) W_m
\]
\[
c = \frac{k_i}{S_i}
\]
\[
A = [1 - \exp\left(\frac{-S_m(W_m + y)}{R_{m\omega}}\right) + (R_p - R_{m\omega}) \frac{S_m(W_m + y)}{R_{m\omega}^2} \exp\left(\frac{-S_m(W_m + y)}{R_{m\omega}}\right)]
\]
\[
A' = A(1 - \frac{1 + C'}{(2C'' + C'^2)^2})
\]
\[
B = (R_p - R_{m\omega}) \frac{S_m}{R_{m\omega}} \exp\left(-S_m(\frac{W_m + y}{R_{m\omega}})\right)
\]
\[
D = (R_p - R_{m\omega}) \frac{W_m + y}{R_{m\omega}} \exp\left(-S_m(\frac{W_m + y}{R_{m\omega}})\right)
\]
\[
E = (R_p - R_{m\omega}) \frac{S_m}{R_{m\omega}} \exp\left(-S_m(\frac{W_m + y}{R_{m\omega}})\right)
\]