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Studying Ink Penetration with Microscopic and Spectroscopic Techniques

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Abstract. *Distribution of ink jet ink in paper substrates and the consequences of ink penetration for printing color reproduction have been studied by combining microscopic image processing with spectroscopic analysis. The study focused on the effects of the composition of uncoated paper, for five laboratory papers plus two commercial products, all consisting of similar pulps but with different combinations of additives. In particular, it was observed that hydrophobizing internal size agents significantly reduced ink penetration, while their effect on paper optical properties was negligible. This observation thus made it possible to study experimentally the pure effects of ink penetration. Pairwise comparisons of prints on such laboratory papers with identical optical properties revealed remarkable impacts of ink penetration on optical density, causing color saturation reduction and color shift. These experimental observations confirmed the theoretical predictions. © 2006 Society for Imaging Science and Technology.*
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INTRODUCTION

Ink absorption by the substrate is a necessity for ink setting in almost all types of printing, and naturally depends on the physical and chemical properties of inks and substrates and their bilateral interactions. Penetration of ink components can be significant over a range of time scales, from the first stages of ink transfer and drying by absorption and evaporation through to long-term stability. A high grade paper often consists of one or more coating layers on a base paper, thus allowing greater control over ink penetration and increased scope for optimization of the combination of print quality indexes affected by it. For uncoated grades, however, inks are directly received by the base paper in which colorants (dyes and pigments) have increased freedom to move together with the liquid carrier until the combination of liquid evaporation and absorption into pores immobilizes the colorants. This process can contribute to serious ink penetration problems that significantly affect, for example- ,tone and saturation of a printed color and color gamut.

Studies of ink penetration related issues have long been important topics for offset printing and cover a wide range of aspects, such as ink setting, trapping and rub-off, print gloss and print density, print unevenness and mottle, print through, etc.[1,2] Studies of analogous issues for ink jet printing are relatively sparse in the literature, although have increased in recent years.[3–8] A variety of experimental techniques measuring penetration of ink components into print media have been developed, with their sensitivity, accuracy, and thus suitability depending on the types of ink and substrates under consideration. In general, their measurement principles can be classified as either spectroscopic or microscopic. Spectroscopic techniques include surface or cross-sectional analysis using Ultraviolet (UV)-visible,[9] Fourier-transform infrared (FTIR)-attenuated total-reflection, electron spectroscopy for chemical analysis, x-ray photoelectron spectroscopy, or time of flight-secondary ion mass spectroscopy, as well as depth-profiling using FTIR photoacoustic spectroscopy or confocal Raman. Microscopic techniques include surface or cross-sectional microtome analysis using optical or fluorescence microscopy, scanning electron microscopy[10] or transmission electron microscopy, as well as depth profiling using laser scanning confocal microscopy.[11] Microscopic and spectroscopic techniques are complementary, and a combination of these can provide the most complete information regarding ink distribution in substrates and its impact on printing color reproduction.

Theoretical studies that provide principles for experimental practices and interpretations to experimental obser-

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vations have also attracted scientists worldwide.[12–18] Among others, Bristow and Pauler[13,14] proposed a method based on Kubelka-Munk (K-M) theory[19,20] to estimate depth of ink penetration from spectral reflectance measurements, provided the ink distributes homogeneously with an uniformly equal depth of penetration. Yang proposed an alternative model that requires fewer spectral measurements.[21] For inhomogeneous ink penetration, the K-M theory has to be extended, as recently suggested by two of the current authors.[17,18] Moreover, a theoretical revision to the Kubelka-Munk theory, incorporating light scattering and absorption, has been made.[22] Such a revision brings out the mutual dependence of the phenomenological absorption and scattering coefficients, K and S, on the intrinsic ones, and enables the revised model to be applicable to a system with a wide range of absorption influences, as is the case for ink penetration.

In our recent article[23] we described a novel method, based on microscopic imaging and image processing plus statistical analysis, which facilitated detailed characterization of ink penetration. In the current paper this study is extended by combining results from the microscopic technique with spectroscopic ones, with a focus on the correlation between ink penetration and color reproduction. The outcomes of this study enrich our knowledge of the effects of ink penetration, serving as guidance for improving print quality.

METHODOLOGY

Substrates and Prints

The study addressed uncoated fine paper. Seven types of paper were used as print substrates, including two commercial paper products and five laboratory-made papers (hand sheets), all seven of grammage $80 g/m^2$. The hand sheets were specially produced at MoRe Research in Sweden using the Formette Dynamique. They consisted of a mixture of 60% birch (24 °SR) and 40% pine (19 °SR) kraft pulp (abbreviated W), with or without calcium carbonate filler (20% precipitated calcium carbonate, abbreviated F), and with or without internal size [approximately 0.3% alkenyl succinic acid anhydride, abbreviated S], and also together with typical dry strength, drainage, and retention additives. By mistake, three of the five hand sheets also contained fluorescent whitening agents [(FWA), abbreviated X]. The compositions of these hand sheets can thus be abbreviated as, for example, $W_1F_1S_0X_1$ and $W_1F_0S_1X_0$, representing hand sheets consisting on the one hand of pulp (W_1), filler (F_1), and FWA (X_1), and on the other pulp (W_1) and internal size (S_1). The pulp, type and amount of filler, type of internal size and Cobb value thus obtained, and additives were all chosen to be similar to those of the commercial samples. Thus, hand sheet $W_1F_1S_1X_1$ is most similar to the commercial papers. However, none of the hand sheets were surface sized, in distinction to the commercial samples.

The printing was performed with a Desktop HP 970 printer that controls the ink level in six steps. The printed ink amount (number of ink droplets fired) was controlled in such a way, through home-developed software based on printer command language, so as to be proportional to the ink levels. In the earlier investigation it was verified that the dry ink amount indeed increases linearly, from approximately 0.8 to 5.2 g/m^2 from level 1 to 6.[23] Full tone color patches were printed on each paper with primary (C,M,Y) and secondary (R,G,B) colors, i.e., only involving the dye-based inks and not the pigment-based black (K).

Spectroscopic Measurements

Spectral reflectance values (R) of the prints were measured, employing a spectrophotometer (L&W Elrepho) that covers a spectral range from 400 to 700 nm in steps of 10 nm. A stack of hand sheets having the same material composition as the printed one was used as backing in each of the measurements. To avoid the influence of fluorescence in the measured spectra, an UV filter cutting off illumination wavelengths below 420 nm was employed, consequently restricting the range to 420–700 nm. Since reflection optical density, defined as $D=-\log_{10}(R)$ is commonly used in the graphic arts industry, the reflectance results are presented in this form.

Microscopic Imaging

A piece of printed patch was first embedded in methacrylate resin LR White (AGAR Sci.), after which the embedded block was sliced by a microtome (Reichert-Jung 2050) equipped with a diamond knife (DIATOM Histo) to a thickness of 2 μm. The slices were collected dry (since presence of water would risk redistributing the ink) and mounted on microscope slides under cover glasses using immersion oil. The sections were examined in a microscope (ZEISS Axioplan) with transmitted bright field illumination using a 20× objective lens. As the sections typically did not lie sufficiently flat on the slide over their imaged length, a focus series was collected at each of two separate locations on the sample. A digital camera NIKON DXM1200 was used to record the images, with resolution of 0.425 $\mu m/pixel$. Each focus series was subsequently merged into one image using the "Average-Focus" function in the software package "Easy Image Analysis 2000" from Techno Optik AB. All images were stored in files of Jpeg format, of size $1280 \times 1024 \times 3$, corresponding to the width (pixel, in the direction along the section) and height (pixel, in the direction of paper thickness) of the image, and three color channels, respectively. The height was later reduced to 400–500 (pixels) after removing a large portion of background (the embedding polymer) from the image.

Image Processing

Image processing (thresholding) formed the basis for the ink-penetration analysis, classifying interfaces between embedding polymer and paper sheet and between the portion of paper with ink penetration and the rest. The image thresholding was carried out column wise (see Fig. 1). As image intensities vary dramatically from column to column, subsequent statistical treatments of the data obtained was necessary.

Let $y_1(x)$ and $y_2(x)(y_2 > y_1)$ be the rows (heights) of the upper and lower surfaces of the paper sheet (see Fig. 1),

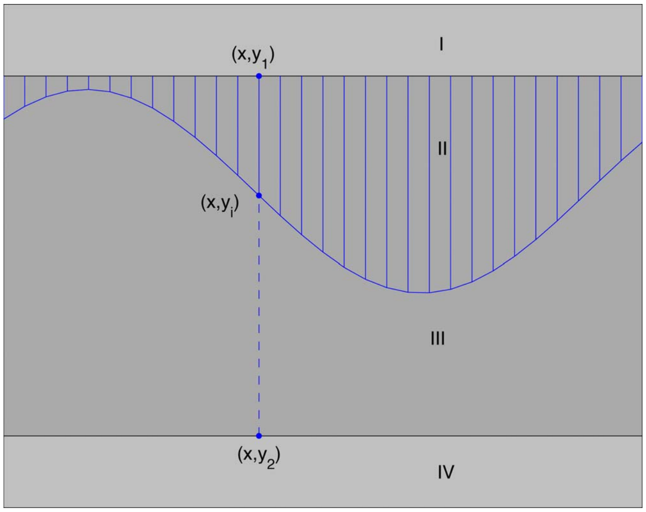

Figure 1. A schematic diagram of ink penetration consisting of four subareas. Areas I and IV are the layers of embedding polymer material, while II and III are the inked and noninked portion of the paper sheet, respectively.

respectively, with x denoting an image column. We assume that the interface between the portion with penetrated ink and the rest of the paper lies at $y_i(x)$. The depth of ink penetration (as a percentage of the local paper thickness) along the column is then given by

$$d_i(x) = \frac{y_i(x) - y_1(x)}{y_2(x) - y_1(x)} 100\ \% \ . \tag{1}$$

If the area of ink penetration has uniform ink concentration but variable depths as suggested in Fig. 1, more detailed characterization of penetration as, for instance, ink density in the substrate, can be obtained by means of statistical treatment. Further details of the image processing are to be found in our recent report.[23]

RESULTS AND DISCUSSION

In this section, ink penetration and its effects on printed color are examined, using both microscopic and spectroscopic techniques that reflect different characteristics of the penetration. First, the dependence of ink penetration on substrate composition (specifically presence of filler and internal sizing agent) is addressed. Second, the impact of this penetration on printed color appearance, in terms of optical density, is analyzed.

Dependence of Ink Penetration on Substrate Composition

Figure 2 depicts images of cross section of two types of hand sheets. The material compositions of these sheets differ slightly, in that $W_1F_0S_0X_0$ contains no performance additives, while in $W_1F_0S_1X_0$ the pulp fibers are hydrophobized with internal sizing agent to reach a Cobb60 value of 19 g/m^2. Figure 2 exhibits clearly differences in both depths and forms of ink penetration. In $W_1F_0S_0X_0$, the ink distributes fairly uniformly around the fibers, whereas in $W_1F_0S_1X_0$

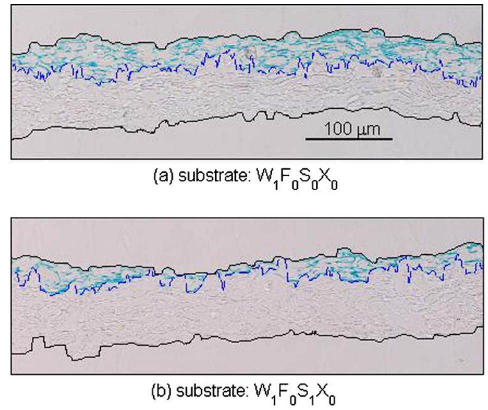

Figure 2. Image classifications for top and bottom surfaces of the paper sheets (solid lines) as well as interfaces of ink penetration (dashed lines). The samples are printed with (cyan) ink level 3 on hand sheets (a) $W_1F_0S_0X_0$ and (b) $W_1F_0S_1X_0$, respectively.

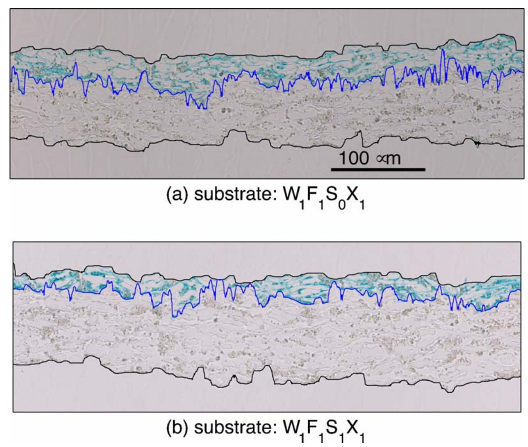

Figure 3. Dependence of ink penetration on paper composition. Two types of hand sheets, $W_1F_1S_0X_1$ (in particular, with filler), and $W_1F_1S_1X_1$ (containing both filler and internal size), are printed with ink cyan and ink level 3.

the distribution displays significantly greater variations. Further, image analysis reveals that the average depth of penetration in $W_1F_0S_0X_0$ is much larger (31%) than that (18%) in $W_1F_0S_1X_0$, indicating the expected strong impact of internal sizing in reducing penetration of the water-based ink. While the print on the former sample is relatively uniform, its internally sized counterpart ($W_1F_0S_1X_0$) displayed a short-scale print mottle texture, consistent with the uniformity and nonuniformity, respectively, of the penetration in the cross-sectional images.

Fillers (typically the minerals calcium carbonate or kaolin clay) that have strong light scattering strengths are often used to replace a fraction of the pulp, both for reasons of economy and to improve the paper's reflectivity, brightness and opacity. It is, therefore, practically relevant to examine the effect on ink penetration of internal sizing agents in the presence of filler. Figure 3 displays the cross sections of two (printed) hand sheets containing filler and fluorescent whit-

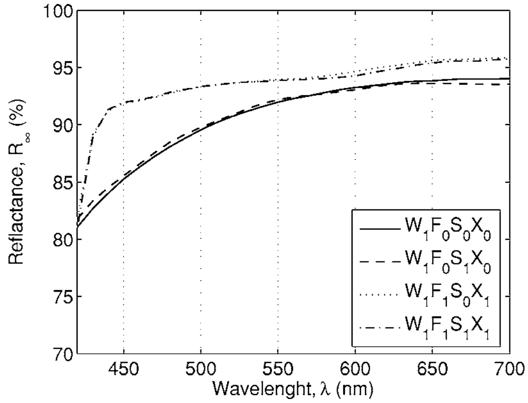

Figure 4. The spectral reflectance values of hand sheets (infinitely thick) of the four different material compositions namely $W_1F_0S_0X_0$, $W_1F_0S_1X_0$, $W_1F_1S_0X_1$, and $W_1F_1S_1X_1$.

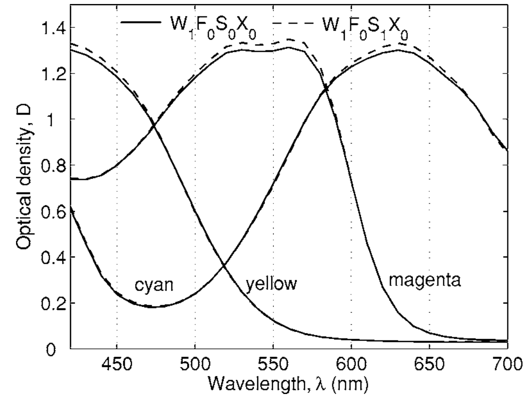

Figure 5. The optical density spectra of prints (primary colors) on two unfilled hand sheets, namely $W_1F_0S_0X_0$ and $W_1F_0S_1X_0$. The prints on the former have larger depth of ink penetration.

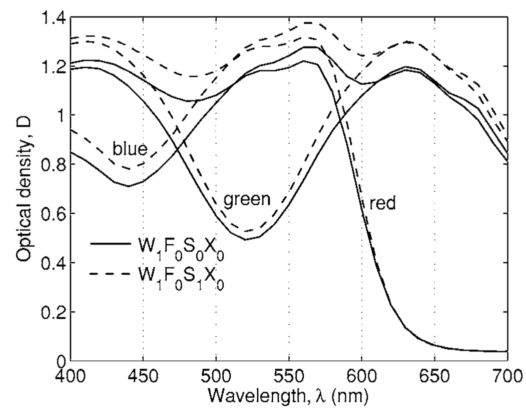

Figure 6. The optical density spectra of prints (secondary colors) on the two unfilled hand sheets $W_1F_0S_0X_0$ and $W_1F_0S_1X_0$. The prints on the former have larger depth on ink penetration.

ening agent, namely $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$, with the latter also containing the internal size. Once again, the significant impact of internal sizing agent on reducing ink penetration, is apparent. Moreover, it is worth noting that Figs. 2(a) and 3(a) share a great similarity, implying that presence of fillers (here calcium carbonate) has a limited effect on ink penetration.

Impact of Ink Penetration on Print Optical Density

Studying experimentally the pure effect of ink penetration on optical density (or spectral reflectance) of a print is a difficult task in general. The difficulties lie in finding substrates of identical optical properties but with different penetration behavior. This section is devoted to exploring these possibilities.

Figure 4 plots the spectral reflectance of stacks of hand sheets with different material compositions, mimicking infinitely thick media. These four curves fall into two pairs, depending on whether or not they contain the filler. This is in line with the above-mentioned expectations that presence of filler significantly improves the reflectivity and opacity of the hand sheets. On the contrary, addition of internal sizing agent makes essentially no difference at all to the optical performance of the hand sheets, while significantly reducing ink penetration. These experimental findings facilitate isolation of the effect of ink penetration on color reproduction, i.e., without any accompanying changes in paper optical properties to complicate interpretation.

Figure 5 depicts the optical densities of the prints of primary colors, using $W_1F_0S_0X_0$ and $W_1F_0S_1X_0$ as the hand-sheet substrates, i.e., the pair compared in Fig. 2. Since both prints contain the same amounts of dye, their differences in ink penetration depth imply, therefore, involvement of different amounts of the paper materials (i.e., fiber). This results in remarkable differences in the light propagation in the mixed ink paper layers since the fibers have a relatively strong scattering ability. Consequently, this affects the optical density and color of the prints.

As seen from Fig. 5, the prints have essentially identical optical densities in the transparent bands, while they differ somewhat in the absorption bands. Although the density deviations are relatively small in magnitude, they reveal systematic trends, i.e., the prints (on $W_1F_0S_1X_0$) with less ink penetration generate colors of higher optical densities than those on the substrate ($W_1F_0S_0X_0$) with greater penetration. This deviation becomes much larger for prints with secondary colors (see Fig. 6), due to the higher ink amounts (approximately twice that of the primary) and correspondingly larger differences in penetration depths. These observations have a straightforward explanation. As the ink itself scatters little light, reflection occurs to all intents only when the light reaches the paper material (fiber). For a print without ink penetration the reflected light is that portion of the incident light that passes twice through the whole thickness of the ink layer, causing significant loss of the light and therefore higher optical density in wavelengths of low transparency. In the case of significant ink penetration, an ink paper mixed layer exists. Light can then be reflected before it passes through the entire ink containing layer, resulting in a higher reflectivity or a lower optical density. Ink penetration depth makes little difference when light passes though a transparent band, but a large difference for a band of low transpar-

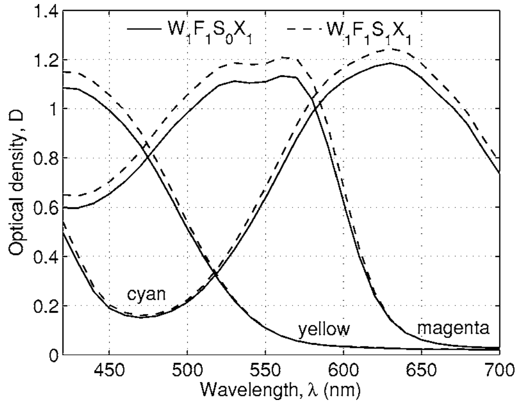

Figure 7. The optical density spectra of prints (primary colors) on the two filled hand sheets $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$. The prints on the former have larger depth on ink penetration.

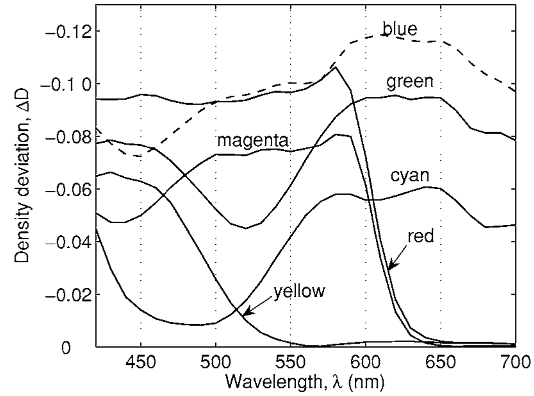

Figure 9. Density deviation caused by differing depths of ink penetration into the two filled hand sheets $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$. For clarity, a dashed line is used for blue.

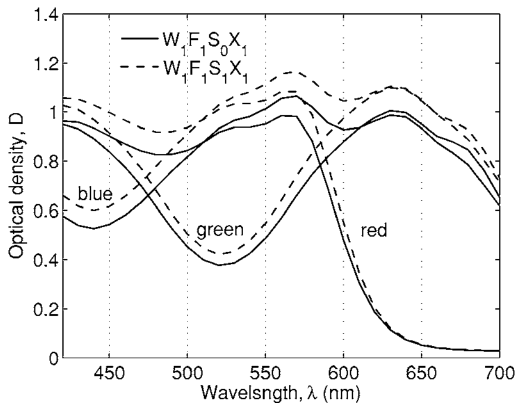

Figure 8. The optical density spectra of prints (secondary colors) on the two filled hand sheets $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$. The prints on the former have larger depth on ink penetration.

ency. These experimental observations are in line with the theoretical predictions made by two of the current authors.[17,18]

Figure 7 displays the corresponding optical density results for the primary colors on the hand sheets $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$, i.e., the pair containing strongly scattering filler compared in Fig. 3. Due to this stronger scattering compared to the unfilled pair $W_1F_0S_0X_0$ and $W_1F_0S_1X_0$, the differences in optical density, again resulting from different depths of ink penetration, now become more evident. In addition to the absorption bands, for magenta and yellow, there are remarkable differences even in the transition bands between the absorption and transparent bands. Since the transparency of cyan is reduced even in its transparent band, the difference exists across the entire spectral range. Again, these observations confirm the theoretical predictions.

Similar behavior is observed from the prints of secondary colors (Fig. 8). Again as a consequence of the larger amount of ink printed, contributing to greater depth of penetration, the differences become even more significant relative to the primary colors. Compared to the prints on substrates without filler (Figs. 5 and 6), the presence of filler clearly leads to an overall drop of the optical density (Figs. 7 and 8) for all colors.

To more clearly quantify the effects of ink penetration on color reproduction, the density deviation (ΔD) caused by differing depths of ink penetration are depicted in Fig. 9. For the two substrates $W_1F_1S_0X_1$ and $W_1F_1S_1X_1$, for example, the density deviation is defined as the difference between the optical densities of the prints on these substrates, i.e.,

$$\Delta D = D(W_1F_1S_0X_1) - D(W_1F_1S_1X_1). \qquad (2)$$

As the depth of ink penetration in the unsized sheet is larger than that in its sized counterpart, the print has lower optical density and so the density deviation is negative. The deviation exhibits a clear wavelength dependency, namely a greater reduction in wavelengths of low transparency. The color red, for example, has significant density deviation in the blue and green bands due to strong light absorption there, while the deviation in the transparent band (red) is negligible. From a color reproduction point of view, the density deviation (reduction) caused by ink penetration results in lower color saturation. On the other hand, the significant variation of the impact from a transparent band to an absorption band results in a significant color shift, as has been pointed out by the authors.[8]

SUMMARY REMARKS

Ink distribution in paper substrates and the effects of ink penetration have been studied by combining microscopic image processing with spectroscopic analysis. This combination reveals correlations between form/depth of ink penetration and printed color appearance.

The characteristic of internal sizing, significantly reducing ink penetration while little affecting paper optics, makes it possible to isolate the effects of ink penetration. Pairwise comparisons for prints on substrates of identical material compositions except for addition of internal sizing agent show remarkable impact of ink penetration on optical densities. The optical density of a printed color, in wavelengths of low transparency, can be strongly reduced by ink penetration. This in turn causes color saturation reduction and color shift. The impact of ink penetration increases proportionally to the increasing power of scattering of the substrate and to the decreasing transparency of the ink. These experimental observations are in line with the theoretical predictions made previously by some of the authors.

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