

# The Influence of Internal Paper Sizing on Water Base Ink Printability

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Abstract: Rotogravure printing inks containing HAP's (Hazardous Air Pollutants) and VOC's (Volatile Organic Compounds) have long been targeted by the EPA for regulation and control. An alternative is water base inks which can be used as direct substitutes for solvent base inks. Implementation of water base ink technology, however, requires a new balance between ink properties, paper properties, and press conditions. The work presented here addresses the interactions between water base gravure inks and paper substrates having a variable level of hydrophobicity. Uncoated papers were made under controlled conditions on a pilot papermachine at different levels of internal alkaline sizing. They were supercalendered and printed in a pilot-plant rotogravure printing press with water base inks. Except for one case where groundwood pulp was added, all papers were woodfree. Results indicated that there were only small variations in the physical properties of woodfree papers; smoothness and compressibility remained unchanged. Although delta gloss continually increased with sizing, print density remained constant up to a certain sizing level, thereafter decreased with increasing sizing levels. Above the critical level, print smoothness (the area of voids in solid prints) also increased substantially. Micrographs of the solid areas illustrated qualitatively that not only a screening defect exists at high sizing levels, but also the air voids are distorted, indicating uneven substrate coverage. These defects were attributed to poor in-plane wetting and spreading of the water base ink over highly hydrophobic surfaces. The groundwood-containing paper demonstrated significant fiber swelling, very high voids area, and low print density. Some additional comments are being made regarding press runnability and printability.

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## INTRODUCTION

### Background

Gravure is the second largest method for printing, particularly popular for high-volume publications. Currently there are 26 publication gravure printing facilities operating in the US, with a publication product value in excess of \$3.5 billions. Examples of publications printed include magazine, catalogues, Sunday magazines and newspaper inserts. Notable gravure printed products are the National Geographic Magazine, TV Guide, Readers Digest, USA Weekend, and Parade magazines.

Publication gravure printing products are printed with solvent based inks, where the primary solvent is toluene. Toluene is not only a volatile organic compound (VOC), but it is also classified as a hazardous air pollutant (HAP) under Title V of the EPA Clean Air Act. Today efficiency rates for VOC/HAP reduction at publication printing facilities are 92%, proposed by the EPA March 14, 1995 (60 FR 13664). However, the 1991 EPA Toxic Release Inventory reports that the total toxic release for the printing industry was 57 million pounds. These releases were in the form of fugitive air emissions and transfers (i.e., other environmental contacts). Data were unavailable for 1995. However, totals can be estimated by factoring a 3% annual growth between 1991 to 1995 for the gravure publication industry (GAA 1995). Given these increases, the total toxic releases for 1995 can be estimated to more than 64 million pounds. Solvent recovery is the current primary means for control of fugitive emissions from publication gravure printing operations. Gravure presses are encapsulated and ventilated, with a solvent recovery of about 92%, while new inks reduce the residual solvents by over 50 percent (Anon., 1995). The solvent recaptured from the recovery process is either sold back to the ink supplier or reused within the printing facility as a reducing solvent for the production inks. In spite the high recovery rates, toluene represents greater than 85% of the total toxins reported to the EPA by the printing industry. A typical average size plant consumes 27 million pounds of solvent (toluene) annually. At the efficiency rate of 92% the total release to the environment is about 2 million annually after controls. Therefore, there is still an incentive to economically replace toluene with solvent-less inks without compromising gravure print quality.

The benefits for introducing new, friendly to the environment inks are multiple. By reducing the levels of HAPs and VOCs, gravure plants would have more regional flexibility as they can expand into new areas presently not permitted. Additionally, safety is enhanced as risk of fires and explosions would be drastically reduced or eliminated. Costs for installation and press maintenance would be reduced by at least 10% and insurance fees would be lower. Environmentally friendly technologies immediately help meet maximum achievable control technology (MACT) requirements without adding additional solvent recovery equipment.

There are several alternatives to obtaining low VOC, non-HAP inks. At present,

water base ink technology is the only practical method available for toluene replacement (Mueller, 1992; Cunningham, 1995). Water is an ideal substitute for the ink solvent, being neither a VOC nor classified as a HAP. Water base inks are a direct substitution to solvent, with little or no production modifications necessary to implement their use.

The industry has struggled for many years attempting to implement water publication gravure. The driving force for implementing water base publication gravure is the need to expanding press facilities while keeping emissions within acceptable limits. It is typical to switch a single color, black or yellow, from solvent to water base in order to meet VOC and HAP emission requirements. In printing a single color down onto an uncoated web, water based black has been used. In multicolor printing on surface treated webs, the first color down, i.e., yellow, is switched to water base because it represents a significant amount (about 40%) of the total ink transferred to paper.

However, good press runnability and printability are not easily obtainable when printing with water ink under publication gravure conditions. Runnability is being associated with the capability to process wide webs of papers at speeds as high as 15 m/s and yet reproduce an image with acceptable print quality. Printability is defined as the ability of the paper, exclusive of all other factors, to accept and retain an image of ink applied by the gravure print cylinder. While missing dots on a rotogravure printed substrate limit printability, runnability with a water base ink is inhibited by the dimensional stability and roughening of the basesheet, problems which cause wrinkles and registration. Print quality is defined as the control of the paper-ink interactions that optimize the final quality of the printing product (Mueller, 1992), but its interpretation many times depends on subjective measurements. Therefore, papermakers, printers, advertisers, and readers disagree on how to rate print quality with the exception of one evaluation. This exception is variation of ink gloss, expressed as ink mottle, due to the uneven setting of the ink film onto the paper surface. Variations in ink gloss and film thickness can be greatly increased with the use of water base inks in lightweight publication rotogravure printing. This is mainly due to fiber swelling which causes a reduction of ink gloss and an increase in distortion of the tonal dots in the printed image (Ginman, 1983; Micale et al., 1989).

Water base inks present unique issues with respect to press runnability and print quality which do not appear with solvent inks. Resins of water based inks do not have the same physical and chemical characteristics as resins used in solvent based inks (Mueller, 1992). While in the gravure cells, water base inks tend to dry out fast, so that a specially designed cells geometry is required for optimum ink transfer. On paper, drying requirements between the two ink systems are different since water needs 3 to 5 times more heat for evaporation than solvent (Rapport, 1992). At room temperature, a solvent based ink has an evaporation rate of 180 seconds, while water based inks have evaporation rates as high as 1102 seconds (Ginman, 1983). Water base inks would not only require an increased drying capacity, but they also promote distortion of the image, especially for the case of printing low basis weight or absorbent papers.

Distortion, sometimes appearing as dot irregularity, arises from capillary penetration and absorption of water into the sheet where excessive penetration, in combination with the lateral movement (or spreading) of gravure inks, leads to blur images (Bassemir, 1990). This is particularly evident when printing with solvent base inks on top of a substrate which has been previously exposed to a water base color. Trapping of colors following a water base ink station therefore exhibits print surface irregularities. As a result, print images with water based gravure inks lack the depth or clarity of solvent based inks, in the extreme case causing misregister and poor print gloss. However, there are some reports that suggest better transfer of water based inks on alkaline papers and more uniform smoothness with fewer missing dots (Mueller, 1992). Additionally, the polar water vehicle naturally has a strong affinity for cellulose fibers and higher surface tension than solvent, so that control of its spreading and penetration into the paper are difficult. Surface smoothness and fast initial ink setting by absorption of water into paper also affect drying speed, rub resistance, print finish and gloss (Hutchinson, 1980). It is therefore the interaction between ink and basesheet which plays the most critical role in determining print quality.

In addition, physical properties of the paper substrate are significant in gravure printing. Smoothness, for instance, is one of the most critical substrate parameters because it helps ink transfer from the gravure cells onto paper (Beri, 1985). It is required at both the microscopic and macroscopic level for good printability. A common problem with the use of water base inks in rotogravure printing is that water enters the fibrous network of the paper, causing fibers to swell and surface roughness to increase. Roughened surface areas, in turn, lead to mis-transfer onto the substrate (skip), or result in poor halftone dot formation. Furthermore, undesirable fiber deformation changes the compressibility of the substrate and inadvertently influences the dimensional stability of the paper web. Compressibility is a vital paper requirement for gravure printing as it aids in substrate-cylinder contact and promotes ink transfer. Additionally, color trap is degraded as the paper surface deforms unevenly, causing poor color balance and reproduction.

The following section reviews important aspects of paper and ink interaction as it influences gravure printability and print quality. Because this interaction is critical to water base gravure printing it deserves a more in-depth discussion. The focus is on the fundamental properties of both ink and paper and the mechanisms involved in gravure ink transfer and setting.

### **Paper-Ink Interaction in Water Base Gravure**

Transfer and setting of the ink on a permeable and absorbent substrate involve complex interactions which are difficult to predict and control. Ink transfer from gravure cells to paper occurs mainly by hydraulic impression (Lyne and Aspler, 1982). The paper in rotogravure printing press is subject to a significant impression

load, as high as 250 pli (pounds per linear inch). The waterbased ink is hydraulically impressed into the paper, penetrating both the surface and inner cellulose fibers. Subsequent wetting, spreading, adhesion and absorption play an important role on ink setting and print quality of the final product. Although ink transfer can be effectively optimized by electrostatic assist at the press (Beri, 1985), mechanisms of ink setting are more difficult to control as the specific interaction between ink and paper is not known *a priori*. Furthermore, it should be noted that, under gravimetric conditions, the liquid ink spreads equally fast in the in-plane direction as it penetrates along the depth (z-direction) of the paper. In-plane spreading of the ink has implications in wicking and halftone dots definition.

The dynamic contact angle of the ink onto the paper influences wetting and spreading and, consequently, manifests gravure printability and print quality. Differences in wetting have been demonstrated with acid and alkaline water base inks on acid and alkaline papers (Triantafillopoulos et al., 1992). Alkaline inks on alkaline papers give good wetting which improves print quality. Currently, inks used in practice are on the alkaline range of pH. The trend in papermaking, on the other hand, is replacing traditional acid with alkaline papers in publication grades. Because of differences in wetting and drying characteristics, water base inks have different press and paper requirements for ink transfer than solvent base inks.

During water base gravure, water enters the fibrous paper matrix, causing an increased surface roughness by swelling the fibers (Triantafillopoulos et al., 1992; Sprycha and Hruzewicz, 1995). Roughening arises from penetration and molecular diffusion of water in to the paper, both processes being significantly rapid contribute to debonding of the paper, stress relaxation and fiber swelling (Skowronski et al., 1986). A thorough review on the topic was presented by Aspler and Beland (1994). The amount of swelling is proportional to the mechanical fiber content of the paper, the wall thickness of the fiber species, and the amount of water and heat applied to the paper. Longer time periods during transfer and drying promote swelling. Papers made from chemical pulps swell less than paper made from mechanical or chemimechanical pulps due to less absorbency. More ink transfers onto papers with high absorbency and therefore more ink is available to penetrate into the substrate. Treatment of groundwood and TMP pulp fibers to reduce their absorbency has been shown to decrease fiber swelling (Nurminen and Sundholm, 1995).

Internal paper sizing to improve hydrophobicity is a common and effective method used to reduce absorbency and control the spreading of ink onto paper. This treatment inhibits wetting by increasing the ink contact angle or reducing water absorptivity and has been proven beneficial to print quality (Hutchinson, 1980). Internal sizes function by absorbing onto cellulosic fibers and making them more hydrophobic by decreasing the capillary penetration and in-plane spreading rate. However, a balance needs to be maintained because excessive water repellency can cause ink transfer problems (Ginman, 1983). Additionally, it is important to control the polar repulsions at the

interface between ink and paper since strong repulsive forces can lead to poor wetting (Etzler et al., 1995).

The present paper discusses the influence of internal paper sizing and its affect on paper properties for water base gravure printing. Sizing was achieved with alkyl ketene dimer (AKD) which reacts with cellulose at the dryer section of the papermachine to form an ester covalent bond. Optimum sizing is obtained at a pH range of 6.5 to 8.5, hence the process is being called alkaline sizing, and requires several hours after the paper has been manufactured. An attempt is being made to correlate different sizing levels with gravure print quality from printing on a pilot-plant press.

## EXPERIMENTAL APPROACH

Woodfree paper was produced under controlled conditions in a pilot-plant papermachine. The pulp furnish was a blend of 16% softwood and 84% hardwood kraft. This furnish was then slack-sized to hard-sized by increasing the concentration of AKD internal sizing. Six levels of sizing were used. In the seventh condition 27% stone groundwood was added to the kraft furnish. Table 1 shows the selected sizing loads for each papermaking condition. In all conditions the pH was controlled at 7.5 and sizing levels were documented using a Hercules Size Tester (HST), see Table 1. Higher loadings of the sizing agent result to slower HST dye absorption (i.e., longer time periods) and represent more hydrophobicity. A basis weight of 55 g/m<sup>2</sup> was targeted at 5% final sheet moisture. Three nips were used to calendered the papers on-machine, providing PPS (Parker PrintSurf @ 10 kg<sub>f</sub>/cm<sup>2</sup>) smoothness of 3.0-4.0. The papers were then supercalendered with 5 nips at 1700 pli at a steam temperature of 175-180°F. Table 2 shows the final smoothness values for each paper.

The next phase of the project involved printing the papers on the Moser™ sheet gravure proof press and the pilot-plant Cerutti™ rotogravure web printing press. The proof press was used to establish ink parameters such as viscosity and color demand before scaling up to the printing press. Printing conditions were as follows:

1. 1200 feet per minute production rate
2. Oven dryers were set to 160 degrees F, @ 9000 cfm nozzle velocity
3. Impression 125 pli @ 3/8 nip flat with a 85 durometer (Shore A) roller
4. ESA (electrostatic assist) was applied to 4kV @ 1.4 mA.

**Table 1.** Sizing levels for each papermaking condition.

Conditions	AKD Loading (Lbs/ton)	HST (s)
1	0	0
2	6	75
3	10	117
4	14	170
5	18	207
6	24	268
7	40	459

**Table 2.** Smoothness of sheets for each papermaking condition (pooled standard deviation = 0.08).

Condition	Smoothness (Parker PrintSurf @ 10 kg <sub>f</sub> /cm <sup>2</sup> )
1	3.11
2	2.73
3	3.08
4	2.75
5	2.80
6	2.70
7	2.74

The ink selected for this project was a commercial publication water base gravure ink. The ink had a pH of 9.0, typical for a alkaline water base ink. The printing viscosity was 20s on a Shell #2 efflux cup. Both magenta and cyan inks had 50% solids by

weight with no more than 2.5% total VOC.

A two color test form with cyan and magenta were printed on the papers. The image on the test form had solid 100% coverage blocks, tones of 25%, 50%, 75% and traps a both solids and tones. The engraving cell configuration for the cyan printer was compressed with a 140° diamond angle and a depth of 32  $\mu$ . The engraving for the magenta was an elongated cell with a depth of 30  $\mu$ .

Several performance criteria were selected to quantify the effect of internal paper sizing on printability. Ink transfer from the gravure cells to the paper surface was carefully monitored. Wetting of the ink film on the paper surface as well as the penetration of the ink into the fiber structure had a direct affect on dot formation and solid lay. Following are tests which were selected to quantify the printability.

### **Gloss**

Paper and ink gloss were measured by a light reflectance meter at 60°. Delta gloss was also calculated to determining the ink penetration into the paper. The delta gloss is the difference in gloss between paper gloss and ink gloss.

Typically high gloss values are associated with high ink hold out of the paper. However, too high of ink hold can result in printing mottle, as exhibited by poor solid ink formation. Gloss values which are too low are ascetically unappealing and can cause scuffing and poor rub resistance. Poor dot definition can also result from too much ink penetration into the fiber surface. Also, halftone dots printed with water base inks are very likely to distort as they migrate in and around the absorbent cellulose fibers.

### **Smoothness and Compressibility**

Good gravure printability is dependent to a large extent on the smoothness and compressibility of the substrate. Gravure is a direct contact method of printing where the ink from very small engraved cells must transfer to the paper surface. The engraved cells on a gravure print cylinder can range from 15 to 250  $\mu$  in width, depending on screen ruling and desired color. Transfer is further complicated due to the concave ink meniscus which forms as a result of the doctor blade wipe prior to impression. If small voids or surface defects are present on the substrate surface the ink filled cells may transfer only in part or not at all resulting is "skipped" dots or dot distortion. Electrostatic assist (ESA) is an effective tool for improving transfer by bulging the ink meniscus and improving wetting of the ink to the substrate surface. In that sense, ESA



is effective in reducing the amount of missing dots but it is not as successful in improving tonal dot detail.

Substrate smoothness can be measured several ways. The two most common methods are based on air leak and stylus profilometry. Paper smoothness is typically measured using the air leak method, such as the Parker PrintSurf (PPS) or Sheffield. The Parker PrintSurf was the chosen smoothness measurement method for this project. Readings were taken with the PPS at 10  $\text{Kg}_f/\text{cm}^2$  and 20  $\text{Kg}_f/\text{cm}^2$ . Higher readings represent a rougher substrate.

Compressibility also influences gravure printability. During gravure printing, the substrate is being subjected to impression between the hard chrome print cylinder and the soft rubber impression roll. It is desirable for the substrate to “compress” in the nip in order to improve its contact area with the ink filled engraved cells. Compressibility can be measured by calculating the difference between the PPS 10  $\text{Kg}_f/\text{cm}^2$  and 20  $\text{Kg}_f/\text{cm}^2$  smoothness values.

### **Ink Optical Density**

An X-Rite 418 densitometer was used to measure the density of the printed solid area. High density is desirable for vivid color reproduction and good gloss development. For years, printers have used densitometers to measure color and other print attributes, such as dot area and trap. The holdout properties of the paper have a significant influence on the density of the ink. In this project optical density was used, along with gloss, to quantify the absorptivity and hold out properties of the papers.

### **Print Smoothness**

Print smoothness can be judged by evaluating the formation of the individual dot structure and solid ink lay. In a solid image print smoothness can be associated with the term mottle. In a halftone dot area poor print smoothness can also be described as “rough” or “grainy” printing. In gravure, the dot must spread and maintain its original formation as closely dictated by the engraved cell geometry as possible. Paper surface properties such as smoothness, wettability, holdout and surface tension have a major influence on print smoothness.

Until recently print smoothness was a qualitative evaluation and subject to opinion by the observer. Fortunately today there are objective image analysis techniques available. Image analyzers, like the one used in this project, are capable of quantitative measurements of an image smaller than one micron in size. An individual half tone dot can be measured for density profile, circularity, perimeter area, and a host of other physical attributes. Additionally, the capability to simultaneously measure

characteristics of hundreds of halftone dots offers a major statistical benefit.

## RESULTS AND DISCUSSION

There are some general statements worth noting with regards to press runnability. Common problems with water base gravure printing are web wrinkles and distortion of paper integrity, as observed from the backside. The latter corresponds to the defects sometimes also called wicking and puckering. Press tension control and dimensional stability of the web are important factors in avoiding these defects. Ink transfer is usually no problem with the help of electrostatic assist and the optimum design of gravure cells for easy transfer at high press speeds. Drying with water base inks is much more critical than drying of solvent base inks. Poor drying can cause transfer of ink to the idler rollers and image distortion, as well as generates problems in the folders. Some of these process variables, however, are difficult to simulate well in a pilot-plant printing press where runs are relatively short. This is particularly true when considering the rise of ink temperature, and subsequent reduction in ink viscosity, occurring during long commercial press runs. As a result, some operational issues encountered in commercial facilities when they implement water base gravure are unpredictable from scale up trials.

Physical properties of the papers tested demonstrated little variation with increasing hydrophobicity, i.e., increased AKD sizing level. The most significant changes occurred when groundwood was used during papermaking (sizing level of 459s). The paper smoothness for all samples having variable sizing levels was between 2.7 and 3.1  $\mu$  (Fig. 1). There was a slight, maximum 10%, decrease in smoothness with an increasing sizing level for papers made out of chemical kraft pulp. The coefficient of variation for this measurement was 2 percent. More rough sheets were produced at the sizing level of 117s with all-chemical pulp fibers and at the level of 459s with the pulp containing 27% groundwood. There were small differences in compressibility values of the papers with variable sizing levels (Fig. 2). The pooled standard deviation of the compressibility factor was 0.08, so that the differences are considered negligible.

Although physical paper properties illustrated small variations, there were significant differences between papers in print properties. Smoothness measurements after printing are an indirect indication of paper roughening. This is demonstrated with PPS results for magenta prints in Figure 1. In most cases there was an increase in roughness with printing, the effect being more pronounced with the use of groundwood pulp where there was a 12% increase in roughness after printing.

Delta gloss increased continually with raised internal sizing levels of woodfree papers (Fig. 3), while print density remained constant up to a certain sizing level, thereafter decreased (Fig. 4). At sizing levels below 170s, improved print gloss can be realized, while the print density remains constant. For example, increasing sizing by 127% (i.e., from 75s to 170s) improved print gloss in the trap of cyan over magenta by approximately 43% at a constant ink density. As shown in Fig. 1-2, both of these papers had similar paper smoothness and compressibility. Because of their lower absorptivity of the water base inks, the more hydrophobic paper requires a longer period of time for the ink to dry and print gloss increases.

The drop of print density at sizing levels above 170s is significant. When hydrophobicity continually increases above a critical sizing level, wetting is being inhibited and uniform spreading of the ink on the paper is not possible. This is illustrated best by quantifying the size and frequency of occurrence of "voids" in printed solid areas, depicted in the print smoothness graph in Fig. 5, and by micrographs (Fig. 6 and 7). High values of print smoothness suggest that there is more area depleted of ink coverage, i.e., more voids, at sizing levels above 117s. The difference between the woodfree and wood-containing papers is substantial, the latter twice the voids area of woodfree papers. Print density (Fig. 4) drops as more paper is exposed through the ink film at high sizing levels and the print appears to have mottle. The trend is not visible in the print gloss graph because, although density corresponds to the relative light and dark areas of a print, gloss reflects the combined influence of both the paper and ink film.

Quantitative results presented in Fig. 4 and 5 are in agreement with qualitative observations in micrographs of solids magenta prints. The small voids appearing white in Fig. 6 are un-inked areas which the liquid ink was unable to bridge in order to form a solid film area. This defect appears in the form of a "screen" pattern, and it is commonly referred to as screening. It is generally caused by worn out engraving cylinders, high ink viscosity, or poor wetting. The last mechanism is probably responsible for the patterns illustrated in Fig. 6. In addition to screening, higher magnification (Fig. 7) reveals that the void areas have an irregular perimeter. This is typically a problem with rough substrates and/or poor in-plane spreading of the ink. The second mechanism is probably the one responsible for distortion of void images in Fig. 7. When the paper becomes extremely hydrophobic, water base inks are spread unevenly in the plane of the paper resulting to poor formation of a solid image

Some additional comments deserve attention based on experience with water base gravure in the pilot-plant scale. A whole-system approach is needed in order to optimize water base gravure printing. Paper and ink properties and press conditions have to be coordinated synergistically for good printability and print quality. Once the optimum balance has been reached, water base prints have quality comparable to solvent base gravure. From the standpoint of paper properties, some hydrophobicity is required to slow down excessive swelling of fibers due to water penetrating the base paper. However, there is an certain level of hydrophobicity above which print quality deteriorates quickly. This is presumably due to substantial reduction in wetting and spreading which cause mottle, image distortion, and loss in print density. Although excessive ink penetration in the z-direction (depth) of the paper is undesirable, uniform in-plane spreading of the ink is preferred for dot fidelity and good solid image formation.

Recently, there have been significant improvements in ink formulations and press conditions. New inks have been developed which have less than 3% VOC, dry well at 2000 fpm, and provide excellent gloss and image reproduction. Additionally, changes in the color printing sequence, i.e., YMCK changing to KCMY, have been proven to improve print quality through control of ink lay down. Gravure cylinders for water base are designed with small cells, high line screens and shallow diamond cutting angles (i.e., 120° to 140°). Printing presses have also been modified with more intense drying, higher hot air flow, and more effective temperatures controls.

## CONCLUSIONS

Internal sizing of uncoated papers improves water base printability and print quality. Although fiber rising is always present with water base gravure, the effect can be minimized by making the paper more hydrophobic. Sizing of woodfree papers does not significantly influence physical properties of paper, assuming that papermaking and supercalendering conditions remain constant. However, print quality changes with an increasing sizing level. Delta gloss continually increases, while print density remains constant up to a certain sizing level, then reducing substantially. The area of printing voids (print smoothness) also increases above the critical sizing level. Main quality problems are screening and irregular coverage of the paper surface arising from poor in-plane wetting and spreading of the ink.

The groundwood-containing paper demonstrated major differences from the woodfree papers. Although it was sized at the highest level, it had more tendency for fiber swelling and the highest print smoothness. Its print density was also low. It is however difficult in this case to attribute poor print quality to a single cause because both paper toughening and poor wetting play detrimental roles.

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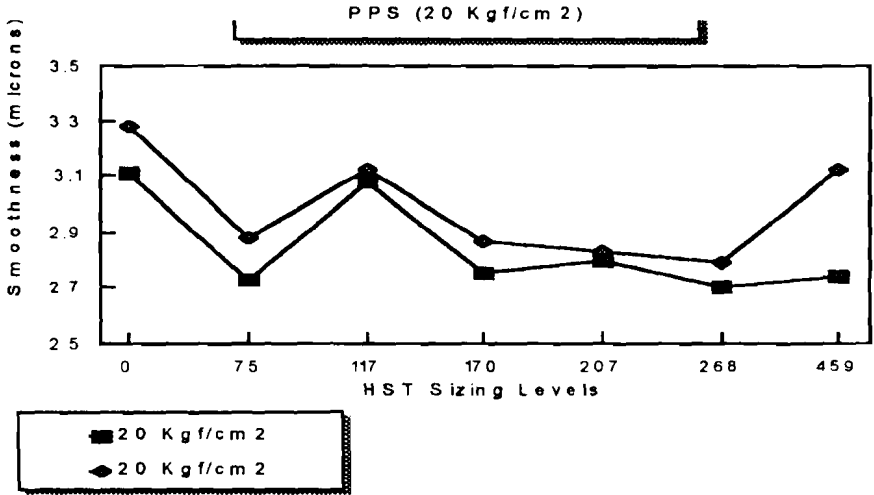


Figure 1

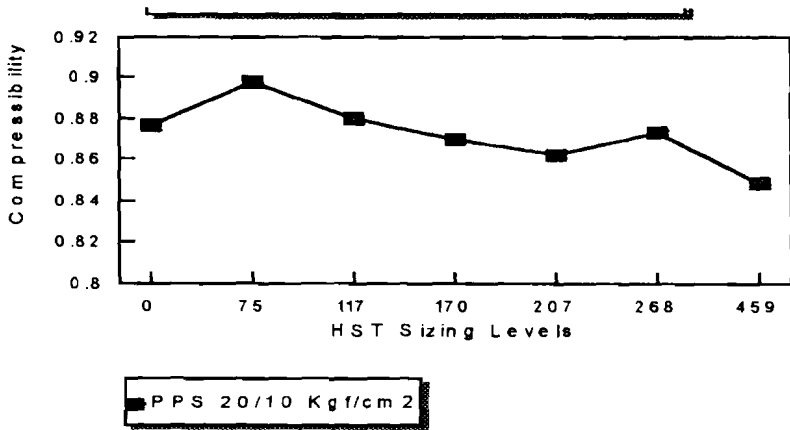


Figure 2



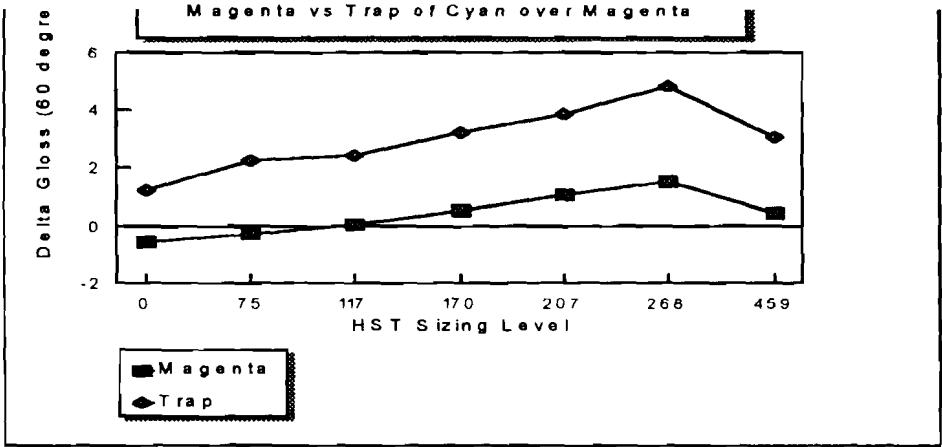


Figure 3

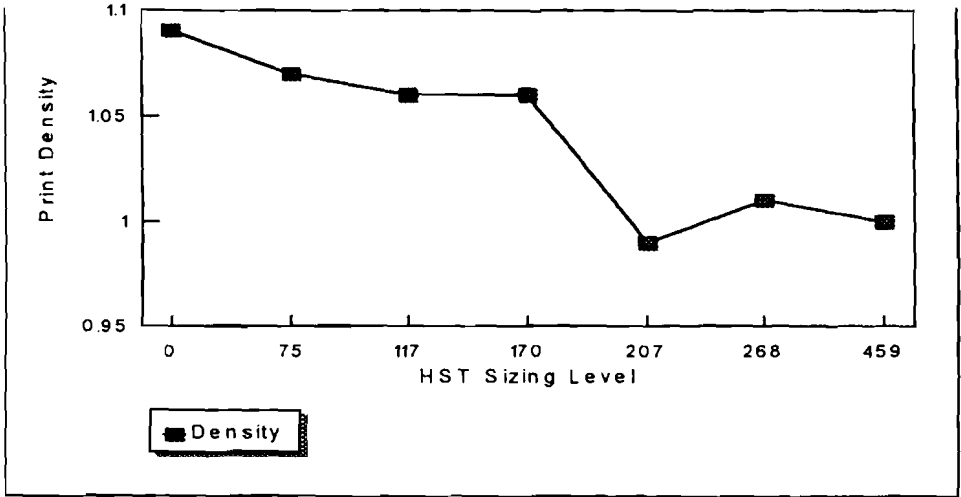


Figure 4

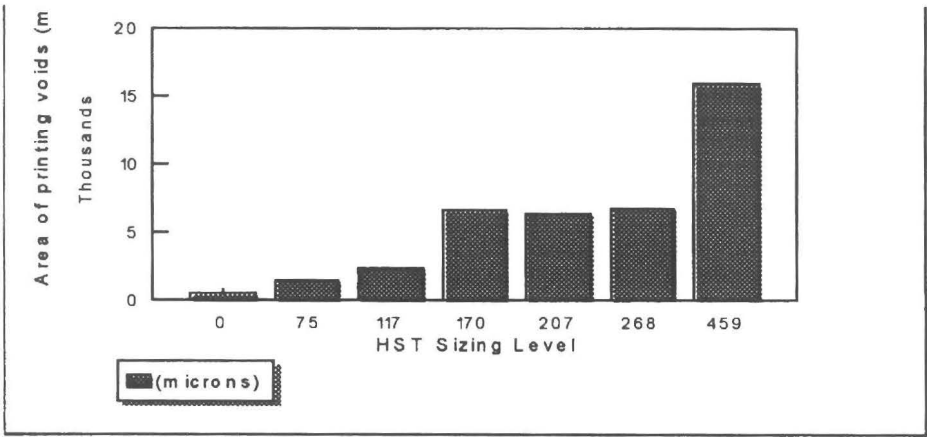


Figure 5

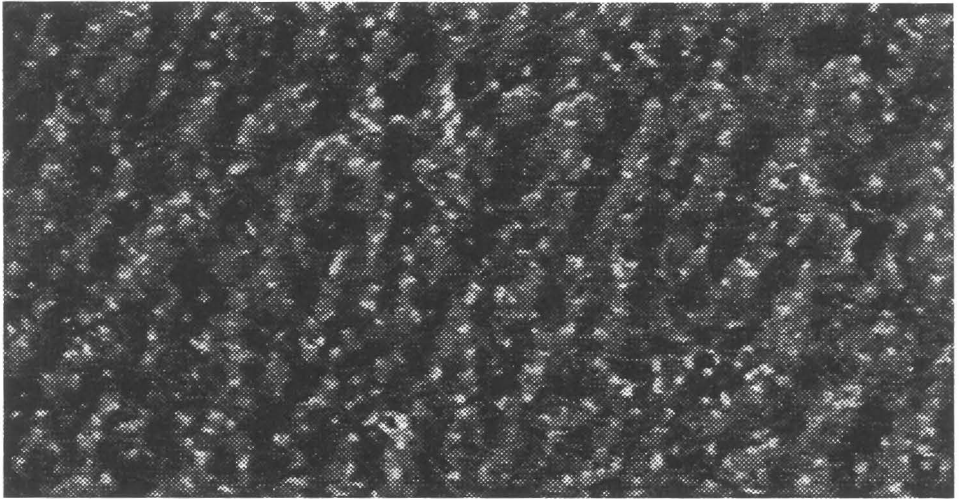


Figure 6