Influence of Pigment Particles on Gloss and Printability for Inkjet Paper Coatings

Hyun-Kook Lee, Margaret K. Joyce, and Paul D. Fleming
Department of Paper Engineering, Chemical Engineering and Imaging
Center for Coating Development
Western Michigan University
Kalamazoo, MI 49008

Abstract

Previously, we have shown that coatings formulated with fumed silica pigments are capable of producing glossy inkjet media, despite the presence of these coating cracks, caused by shrinkage of the coating layer. However, their high cost and the inability to run them at high solids (>30% solids) limits their commercial application. Raising the coating solids with the addition of conventional pigments reduces the amount of shrinkage that occurs during drying, and thereby reduces the coating cracks, improving the final gloss of the coating layer.

In this research, the solids of fumed silica coating systems were increased by adding co-pigments of different particle sizes. Fumed silica was used as the main pigment and precipitated calcium carbonate (PCC) and ultra fine ground calcium carbonate (UFGCC) were used as co-pigments. The ratio of fumed silica to co-pigments were 100:0, 90:10, 80:20, 70:30, 60:40, 50:50, 0:100. These blends of pigments were previously shown to produce high gloss and good printability. The coatings were applied using Meyer rods to obtain a coat weight of 10g/ m^2 on one side. The optical brightness, printability, ink density, and ink gloss were compared. Based on the findings, it is concluded that the co-pigment systems had optical properties and printing qualities as good as the mono pigment coatings.

Introduction

Most high-end publication printing and photofinishing employ a glossy paper because the readers associate gloss with high surface smoothness and print quality. Gloss is associated with high surface smoothness[1] and commonly regarded as a smoothness index. However, it does not always correlate with physical smoothness, since it is possible to have a glossy surface that is quite rough. For example, some photo glossy inkjet paper has high gloss although it has rough surface. However, the average observer instinctively tends to downgrade the rougher paper, even though the gloss is the same.

Previous studies by Do Ik Lee[2] have shown that the gloss of a paper coating is mainly affected by the following factors:

- The effect of the particle size and shape.
- The effect of particle size distribution.
- The effect of colloidal stability.
- The effect of binder level.
- The effect of drying conditions.
- Effect of binder composition.

Product development of inkjet printing papers has accelerated greatly to meet the rapidly growing market for inkjet[3-6] printing. Advancements in inkjet printing technology have also placed new demands on the glossy substrate that produces photo quality images[3,5,6]. To meet these requirements, papermakers are turning to pigmented coating systems. In the glossy inkjet coating field, fumed silica is used. The particle size of these glossy pigments is typically in the range of 100-300 nm. This particle size range offers possibilities for glossy inkjet applications.

Research by Lee[7,8] and Ramakrishan[9] showed that fumed metallic oxide pigments are capable of producing semi-gloss and high-gloss inkjet papers with acceptable print quality, after calendering. Both researchers found the gloss of fumed alumina pigments to be higher than fumed silica. An important finding of Ramakrishan's studies was that the gloss of the inkjet papers increased with an increase in silica particle size. Smaller particle silicas were shown to produce coatings that contained severe coating cracks. Ramakrishan concluded that the presence of these coating cracks increased the microroughness of the papers, consequently reducing the gloss of the silica coatings. Coating cracks were not observed in the scanning electron microscopy images of the alumina-coated papers. These results correlate with previous findings by Do Ik Lee[2], which showed that shrinkage during drying is the cause of surface roughening and loss of coating gloss. Do Ik Lee attributed the roughening of the coated surface to the shrinkage of the binder and removal of coating waters upon drying.

Basic Theory

On plain surfaces like glass, when a beam of light strikes the surface part of the light is reflected from the surface and the remainder is transmitted and scattered[1]. The reflection and refraction that can occur are illustrated in Figure 1. The relationship between the incident and

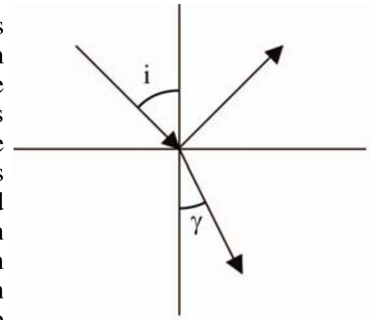

Figure 1. The angle of refraction γ is governed by Snell's law.

refracted angle is given by Snell's law[1] (Equation 1):

$$\sin i / \sin\gamma = n, \qquad (1)$$

where n is a constant known as the refractive index. Since γ can be changed depending on the material of the plane, the refractive index can be also changed.

Due to the angles i and γ, the intensity of light reflected from surface affecting the surface gloss is fixed by Fresnel's law[1].

$$I_{rft}/I = [\sin^2(i-\gamma)/\sin^2(i+\gamma) + \tan^2(i-\gamma)/\tan^2(i+\gamma)]/2, \qquad (2)$$

where I is the intensity of the incident beam, I_{rft} is the intensity of the reflected beam. For normal incidence, the equation may be written in the form:

$$I_{rft}/I = [(n-1)^2/(n+1)^2]. \qquad (3)$$

If n is approximately 1.5, about 4 % of the incident light is reflected and if n is about 2.4, about 17 % of the incident light is reflected.

For wavy surfaces, Chinmayanadam[10] tried to account theoretically for the reflection from rough surfaces by hypothesizing that the reflecting elements of an optically rough surface are distributed according to the probability law, and he derived the expression:

$$I = e^{-(8\pi^2\cos^2 R)/\alpha\lambda^2}, \qquad (4)$$

where R is the angle of viewing and α is a constant having dimensions of length^{-2}. For moderate angles of incidence (up to 54°), it shows good agreement with practice. For higher angles, Chinmayanadam falls back on an empirical formula, which has been found to give good agreement with observed values:

$$I = e^{-(\alpha\tan^2 R + b\cos^2 R/\lambda^2)} + (1 - e^{-\alpha\tan^2 R})e^{-(c\cos R + d\cos^2 R)/\lambda}. \qquad (5)$$

Gloss is the degree to which the surface stimulates a perfect mirror in its capacity to reflect incident light. From a study of the different existing gloss instruments and properties that they measure, six types of gloss have been defined[1,11,12]; specular gloss, sheen, contrast gloss, absence-of – bloom, distinctness –of- reflected image gloss, and absence-of-surface texture gloss. In the paper industry, specular gloss (reflection) is sought[11]. Specular reflection refers to the portion of incident light that is reflected from the surface of an object with an angle of reflection equal to the angle of incidence (Figure 2).

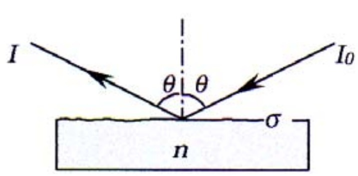

Figure 2. Light path of specular reflectance.

From Figure 2, the light path on the surface at certain incident and reflect angle (θ) is seen[13]. Specular gloss is related to the ratio of the incident light intensity (I_0) to reflected light intensity (I), and more exactly defined as the ratio of the luminous flux reflected by the test surface into a specified angle and luminous flux from the standard reflecting surface. Therefore, specular gloss can be formulated as:

$$G_s(\theta) = \rho_v(\theta)/\rho_0(\theta) \times 100, \qquad (6)$$

where $\rho_v(\theta)$ is the Fresnel Equation of the test surface, $\rho_0(\theta)$ is the Fresnel Equation for the standard, and θ is the incident light angle. From the Fresnel theory, the equation of the reflectance of opaque or black glass with a perfectly smoothnesssurfaceisshownby[13,14]:

$$\rho(\theta,\lambda) = \frac{1}{2}\left[\left(\frac{\cos\theta - \sqrt{n(\lambda)^2 - \sin^2\theta}}{\cos\theta + \sqrt{n(\lambda)^2 - \sin^2\theta}}\right)^2 + \left(\frac{n(\lambda)^2\cos\theta - \sqrt{n(\lambda)^2 - \sin^2\theta}}{n(\lambda)^2\cos\theta + \sqrt{n(\lambda)^2 - \sin^2\theta}}\right)^2\right], \qquad (7)$$

where $n(\lambda)$ is the refractive index at wavelength λ.

The equation for rough surfaces like paper is as follows:

$$\%Gloss = I/I_0 = f(n, \theta)\exp[-(4\pi\sigma\cos\theta/\lambda)^2] \qquad (8)$$

where I_0 and I are the specularly reflected and incident light intensities,

f(n, θ) is the Fresnel coefficient of specula reflection as a function of refractive index n and angle of incident light I, σ as the surface roughness (standard deviation of the surface profile), and λ is the wavelength of incident light.

From Equation 8, the paper gloss is determined by the incident angle of light, incident light wavelength, refractive index, and surface roughness of the paper.

According to TAPPI standards[15], gloss is defined as the 75° spectral reflectance of light at λ=0.55 µm. Based on this definition, coating gloss is optimized by increasing the refractivity of the coating layer and minimizing the roughness of the coated surface layer.

$$\text{TAPPI } 75°\text{gloss} = 384.6 f(n,75°)\exp\{-(4\pi\sigma\cos 75°/0.55)^2\} \qquad (9)$$

From the Equation 9, TAPPI 75° gloss is dependent on the refractive index and surface roughness. Theoretically, sheet gloss should be responsive to the effective index of refraction of a surface. It is the index of refraction that depends on both void volume and the material present in the coating. Increasing the void volume should decrease the index of refraction and thus the gloss.

The ideal surface should catch and reflect the light, when the light source is behind the observer and at a high angle of incidence, but should absorb the light when viewed at a low angle with the light source in front. The proportion of light at the glare angle need not be much greater than light reflection at the glare angle to cause appreciable glare. Gloss can be reduced by coating the paper with highly opaque pigments, provided that the pigment-coated surface is not too highly finished.

Materials

Silica

The ink used for inkjet printing is usually a water-soluble-dye based system. Comparisons between ink jet printing with water-soluble dye and water dispersible pigments are given elsewhere[16,17]. Most inkjet inks consist of 90% water[18]. Therefore, the paper must absorb water quickly to avoid wicking and maintain sharp edge acuity. The highly-absorbent, porous nature of silica pigments make them an excellant pigment for inkjet printing papers. These pigments quickly absorb the vehicle and set the toner, which maintain edge acuity, proper color balance and saturation. The excellent water absorptivity of high-structure silicas minimizes banding caused by the improper absorptivity of the paper for the ink. High water absorptivity also decreases the drying time for print.

Polyvinyl alcohol

In comparison to latex and natural binders, polyvinyl alcohol (PVOH) is the strongest binder[19,20]. Polyvinyl alcohol is an excellent film former and is resistant to wetting by oils, greases, and organic liquids[19,20]. Since it is a water-soluble synthetic binder, the properties of polyvinyl alcohol can be controlled precisely over a wide range of values. Although water soluble, it can be made water-resistant. It forms clear, colorless, non-blocking, abrasion resistant, yet flexible, films at room temperature. PVOH shows outstanding adhesion to cellulose, and provides high pigment binding strength. It is compatible with other hydrocolloids, natural and synthetic binders, pigments and other commonly used additives in the paper industry. PVOH provides unique enhancement of fluorescent whitening agents[21,22], which are commonly used in inkjet papers. It is used in clear and pigmented paper and paperboard coatings. Paper grades using PVOH include release liners, carbonless, flexible packaging, and high brightness papers.

The quality of the record obtained in an inkjet recording process depends highly on the properties of the ink and the recording paper. The recording sheet must absorb the ink rapidly and at the same time precipitate the ink dye on the sheet surface. From two properties, the ink property reduces the tendency for set-off (i.e. transfer of the ink from the paper to sheet handling rollers and the like) whereas the recording sheet property insures that images having high optical density are obtained. Unfortunately, there is a problem with these two properties compromising each other. Papers having high absorbency allow the ink to penetrate deeply into the paper, decreasing the printing density of the image at the surface. They also show feathering, poor edge acuity, and show-through. Papers with low absorbency, such as highly sized papers, provide good optical density by retaining the ink at the paper surface, but have set-off problems because the ink vehicle is not absorbed rapidly[23-26].

For high-grade coated paper, to which major attention regarding print gloss is directed, print gloss develops frequently irrespective of sheet gloss. Since the 1960's, it has been stated that print gloss is more normally associated with holdout of ink vehicle on the stock surface, rather than directly with the smoothness of the stock or ink film, because it is the vehicle holdout that contributes to the smoothness of the final print and to print gloss. Leveling of an ink film and menisci development, among ink pigment particles, play an important role in achieving print gloss. In the printing industry, leveling of an ink film is the primary mechanism[24].

In order to study the relationship between gloss and pigments, surface modifications to increase the refractive index of the silica pigments should also be studied, because refractive index can affect the gloss of the coated paper. Additional studies are also needed to determine if composite pigments can be formulated that would enable the coating solids of the formulations to be increased. To minimize shrinkage, coating solids greater than 55% solids should be targeted to reduce the difference between application solids and the coating's immobilization solids point. Since the immobilization solids point is defined as the point at which the free drainage of coating water to the basesheet ceases, raising the application solids will reduce the amount of free water lost to the basesheet upon its application and metering.

Experimental Design

This project is divided into two phases: (1) relative sediment volume (RSV) of coatings studies, (2) drawdown coating studies, characterization of the optical properties, printing properties after calendering. One fumed silica and PCC and UFGCC were used. The physical properties of pigments are shown in Table 1.

Table 1. The Physical Properties of Pigments as Supplie.

Sample	Solids Content	Specific Gravity	pH	Refractive Index	Particle size (nm)
FS	30%	1.20	10.0-10.3	1.46	225
PCC	70%	2.72	9.0-10.0	1.58-1.63	544
UFGCC	75%	1.92	9.0-10.0	1.58	600

The ratio of fumed silica + PCC and fumed silica + UFGCC were 100:0, 90:10, 80:20, 70:30, 60:40, 50:50, and 0:100.

The binder used in the coating formulation was a partially hydrolyzed and low viscosity, polyvinyl alcohol (Airvol 203, Air Products Inc.). This polyvinyl alcohol was chosen to increase the coatings solids by minimizing the interaction between pigments and PVOH. Polyvinyl alcohols with a high degree of hydrolysis are known to interact more strongly with silica pigments, thus limiting the coating solids that can be feasibly prepared. Stable solutions of polyvinyl alcohol were prepared at 30% solids by adding the required amount of dry PVOH powder to cold water under agitation and heating the mixture to 185º F. The solution was held at this temperature for 35-40 minutes to assure complete dissolution and hydration of the PVOH. A defoamer was then added (Foam master VF, Henkel, Inc.). The solids content of the solution was measured using a CEM Labwave 9000 microwave moisture analyzer. The solution was cooled to 40º F before adding the slurried pigments at a slow rate of agitation. The coatings were mixed for 20-30 minutes and the pH and viscosity measured. Coatings were prepared at one pigment-to-binder ratio (7:1). In the previous research, the ratios of pigment to binder were 4:1 and 5:1 but in this research, the binder ratio was reduced because PCC and UFGCC with low binder demand were added.

The viscosities of the coatings were measured using a Brookfield RVT digital viscometer (#5 spindle @ 100 rpm). Twenty-five ml of each coating was centrifuged at 25,000 RPM for one and one half hours to determine its Relative Sediment Volume (RSV) at 30% solids.

Drawdowns

The coatings were applied to the uncoated base paper using several different Meyer rods. The Meyer rods were selected to enable 10g/m^2 of coating to be applied. The samples were allowed to air dry, until they were dry to the touch. The samples were then cut into 5-inch diameter circles using a press punch. The coat weights of the samples were then determined, taking the difference between the weights of the uncoated and coated samples after being dried in a CEM Labwave 9000 microwave moisture analyzer. The samples were then conditioned in a humidity-controlled room, before performing optical and physical property measurements.

The brightness of the papers was measured using a Technidyne Brightness meter, TAPPI procedure T 452 om-92. Gloss was measured using a Hunter 75º gloss meter according to standard TAPPI procedure T 480 OM-92. The surface roughness and permeability of the sheets were measured using a Parker Print Surf (PPS) tester (TAPPI T555 pm-94).

The samples were printed on a Canon S450 inkjet printer using a proprietary test print pattern created with Adobe software. The print gloss and print density of the samples were then measured. Print gloss was measured using a Gardener 60º Micro-Gloss meter. Print density was measured using a BYK Gardner densitometer.

The samples were calendered on one side through 3 nips at 123 kN/m at and 60°C.

Result and Discussion

Figure 3 shows that RSV (relative sediment volume) of each coating. From the graph, the RSV of fumed silica and UFGCC coatings decreased as the addition of a UFGCC increased.

The following equation was applied to quantify the RSV:

① =weight of water/density of water
+ weight of pigment/ density of pigment
weight of water/① = % volume of water
100 - % volume of water = % volume of pigment
Volume of pigment = Total volume of coating x
% volume of pigment/100
Then, RSV = separated water from centrifuge
machine/ volume of pigment

Figure 3 shows how RSV was changed by the addition of PCC and UFGCC.

In the case of FS and PCC coatings, RSV volume did not change much until 30% PCC was added. It is believed that the aragonite (needle shape) particles of PCC did not reduce the internal void volume of the fumed silica of grape-cluster shape. Therefore, the coating layer absorptivity of 70% fumed silica and 30% PCC coating maintains the same level of 100% fumed silica coating. After 40% addition of PCC, RSV decreased because the particles of PCC filled the internal void volume of fumed silica. In the case of FS and UFGCC, RSV decreased from the 10% addition of UFGCC. From the data, the rhombohedral shape of UFGCC more readily reduced the internal void volume of the fumed silica.

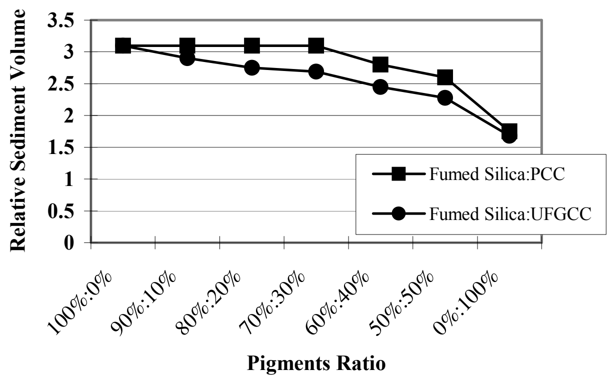

Figure 3. Relative Sediment Volume comparison of each coatings(Coat weight:10 gsm, Calendered samples, Pigment:Binder =7:1).

Figure 4 shows the relationship of brightness to the addition of UFGCC and PCC.

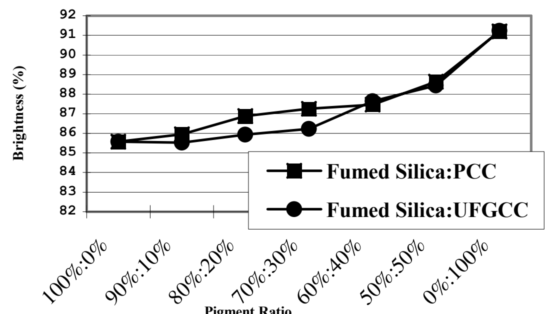

Figure 4. Brightness Comparison of PCC and UFGCC coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1)

As the addition of PCC and UFGCC increased, the brightness value increased.

The brightness value of fumed silica and PCC coating was higher than that of fumed silica and UFGCC. Brightness was influenced by the amount of light scattered and absorbed. Since scattering is influenced by the surface area of the pigments and the number of air-to-pigment interfaces, the fumed silica and PCC coating may have more air voids in the coating structure enabling more light to be scattered.

Figures 5 and 6 show the relationship between the glosses and roughness of each coating. From these figures, it is seen that the addition of PCC improved gloss due to the increase of refraction and smoothness, until 30% addition but after 40% addition of PCC, the gloss decreased. The results indicate that although the refractive index of the coating surface increased due to the addition of PCC, the surface roughness, microroughness, increased due to a change in packing order of the fumed silica and PCC. In the coating of fumed silica and UFGCC, the gloss decreased until a 30% addition of UFGCC was added. After this point, the gloss improved.

Figures 7 and 8 show how the permeability of the coating layers affected ink density.

The coating layers with a more open pore structure (higher permeability) absorb the ink vehicle more quickly and help to fix ink pigment or dye on the surface. Since inkjet inks have negative charge, it is preferred that the coating layer have a positive charge to fix the ink dye or pigment to the surface. However, addition of PCC and UFGCC caused the reduction of positive charge of coating layer in this experiment.

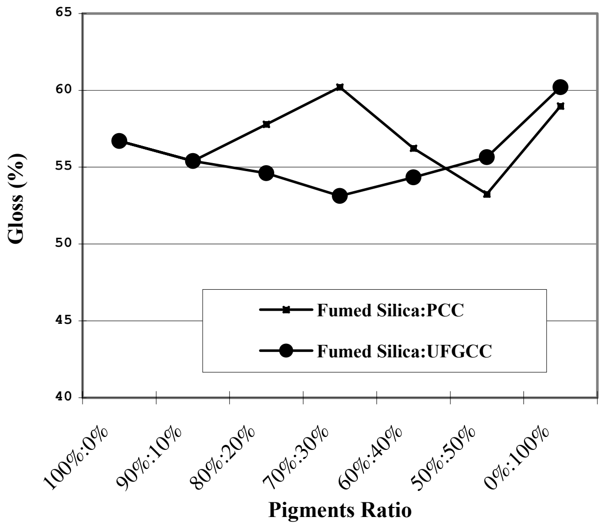

Figure 5. Gloss Comparison of PCC and UFGCC coatings (Coat weight:10 gsm, Calendered samples,Pigment:Binder=7:1)

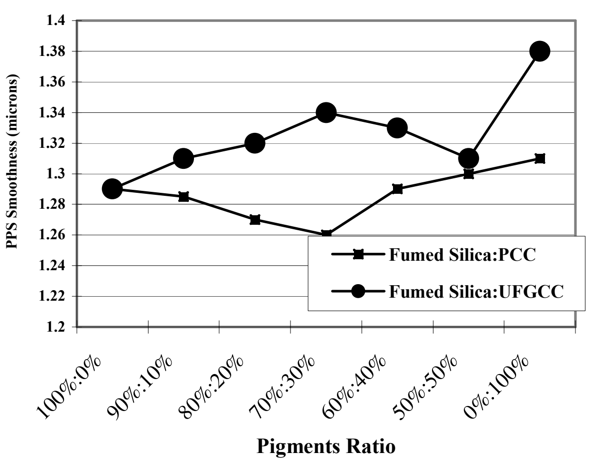

Figure 6. Roughness (PPS Smoothness) Comparison of PCC and UFGCC coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1)

Figures 7 and 8 show the relationship between ink gloss and paper gloss. Ink gloss decreased with paper gloss. Unlike the offset printing process, whereas the surface area of the substrate increases, ink gloss is reduced by the shrinkage of the ink film. For this printing process, the high surface energy of fumed silica does not cause shrinkage in the ink layer. Even the high surface energy of the coating layer can help ink dye to be fixed due to the fast-absorption of the ink vehicle.

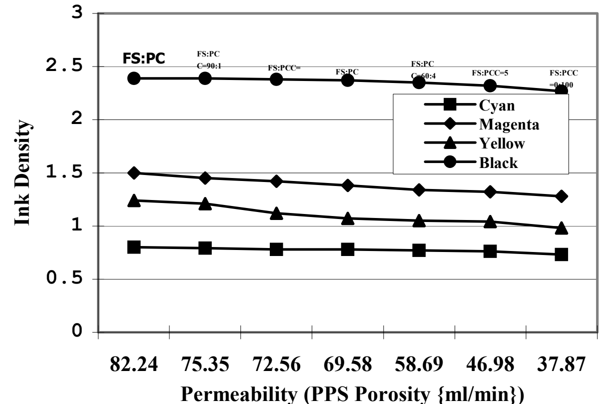

Figure 7. Ink Density vs Permeability (PPS Porosity) of Fumed Silica and PCC Coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1)

Figures 9 and 10 show the relationship between permeability and ink gloss. Permeability did not affect the change of ink gloss. In the conventional printed of coated paper, the pore structure of the coating layer is able to change ink gloss[25]. However, in this study, ink gloss was affected by paper gloss more than by permeability.

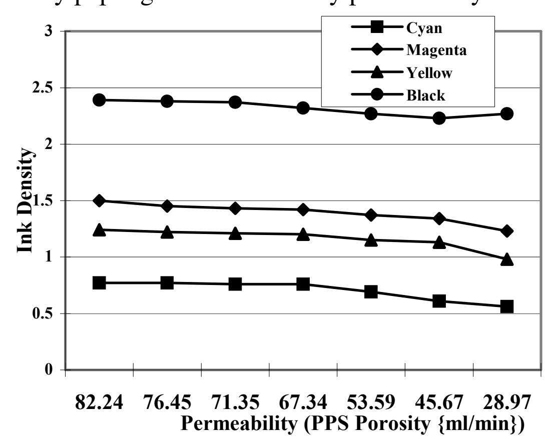

Figure 8. Ink Density vs. Permeability (PPS Porosity) of Fumed Silica and UFGCC Coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1).

Conclusion

This research indicates that brightness was influenced by the pigment type and the ratio of fumed silica and PCC and UFGCC. As the addition of PCC and UFGCC increased, the brightness value increased. The brightness value of fumed silica and PCC coating was higher than that of fumed silica and UFGCC.

The addition of PCC improved gloss until 30% of PCC was added. The results indicate that although the refractive index of the coating surface increased due to the addition of PCC, the surface roughness, microroughness, increased due to a change in packing order of the fumed silica and PCC. In the coating of fumed silica and UFGCC, the gloss decreased until a 30% addition of UFGCC was added. After this point, the gloss improved.

The addition of PCC and UFGCC decreased the ink density but there are not big differences in this research. Permeability did not influence ink gloss. In the conventional printed of coated paper, the pore structure of the coating layer is able to change ink gloss. However, in this study, ink gloss was affected by paper gloss more than permeability.

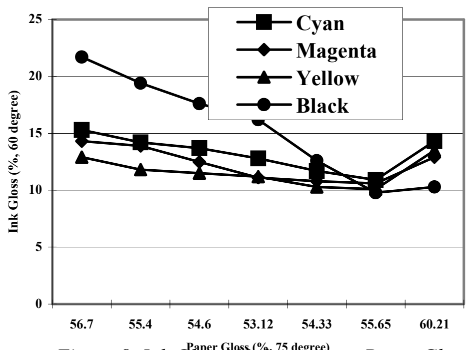

Figure 9. Ink Gloss(60 degree) vs. Paper Gloss (75 degree)of Fumed Silica and UFGCC Coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1).

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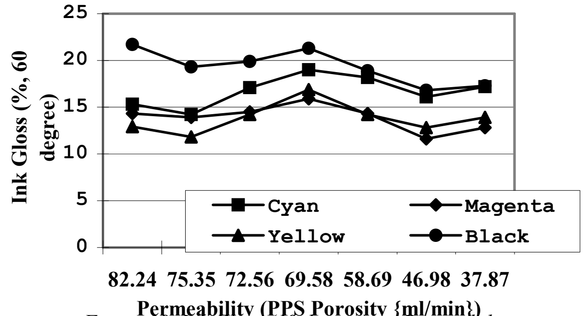

Figure 10. Ink Gloss (60 degree) vs. Permeability (PPS Porosity) of Fumed Silica and PCC Coatings (Coat weight:10 gsm, Calendered samples, Pigment:Binder=7:1).

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