Evaluation of Print Mottle of Double Coated Paper by Octave Band Filtering Technique

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The effects of the pre-coating color formulation, dwell time, and coat weights of pre- and top- coating layers on print mottle of double-coated paper were investigated using an octave band filtering image processing technique. To investigate the effect of pre-coating color formulations, six types of coating colors were used. Mottle index and coefficient of variance (COV) of double-coated paper increased as the size of pre-coating pigment particles decreased, which was attributed to the reduction in relative pore size of the pre-coating layer. Decreasing the dwell time decreased transfer time from the coating head to dryer of a Maiyoh coater. As the transfer time to dryer decreased, the mottle index and COV increased because of uneven distribution of latex within the coating layer. Pre-coat weight was found to have a greater impact on print mottle than the top-coat weight.

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INTRODUCTION

Print mottle, an irregular variation in ink density or color in printed materials, is a crucial quality factor for multicolor offset printing (Yamazaki *et al*. 1993; Lee 2008; Kasajová and Gigac 2013). Overcoming printing defects has become progressively challenging due to increased coating and printing speeds, coupled with a decrease in coat weight. Meanwhile, the demand for high-quality printed materials with special properties has become common.

Various types of print mottle, including back trap mottle, water-interference mottle, ink-trap mottle, *etc.* (Saini *et al*. 2016; Lee *et al*. 2021), can occur in offset printing. These print defects are linked to the unevenness of the coating layer and the printing processes such as ink formulation, printing sequence, ink volume, ink setting, *etc*.

The primary cause of print mottle is the uneven distribution of latex within the coating layer surface. This has been attributed to binder migration occurring during the coating process (Engström and Rigdahl 1992; Rajala *et al*. 2004). Binder migration mainly depends on changes in coating distribution and variations in the base paper absorbency (Gane 1989; Aslannejad *et al*. 2019). The variation in base paper absorbency leads to uneven distribution of latex on the coating surface, resulting in partial variations in dampening solution and ink density during printing, thereby causing the occurrence of uneven print density. Print mottle can be affected not only by the characteristics of the base paper but also by process conditions, such as drying conditions and dwell time. Dwell time is a crucial process factor that controls the penetration of water and binders into the paper. As the dwell time increases, there can be partial deviations in the migration of water into the base paper, which often leads to more severe print mottle (Fujiwara *et al*. 1991).

The advantage of double coating is that the pre-coating layer can cover the rough surface of the base paper first. Ideally, it will provide cost-effective coverage of the base paper, enabling application of a fine top-coat (Nutbeem *et al*. 2011). Coarse ground calcium carbonate is most widely used as precoating pigment (Findlay *et al*. 2017). Application of precoating reduces the partial concentration variations in surface binder, in comparison to relying upon a single coating (Nutbeem *et al*. 2011; Resch *et al*. 2010; Kim *et al*. 2016).

Recently, Youn and Lee (2022) studied the influence of pre-coating colors on the dynamic water penetration behavior of double-coated paper. They found that reducing water penetration into the base paper occurred when either a large amount of fine particle size pigment was used or the starch binder content in the coating color was increased. Thus, the higher water holding capacity of pre-coating color reduced binder migration into the base paper, while increasing latex content on the surface of double-coated paper. Additionally, Im *et al.* (2022) investigated the relationship between the dynamic water penetration behavior of top-coating color and double-coated paper properties. Results showed that as the number of pores per unit area at the surface of the pre-coating layer increased, the amount of water that penetrated the pre-coated paper increased, regardless of the low shear viscosity of the top coating color.

The most common method for evaluating print mottle is through visual inspection. However, this assessment is a subjective process typically assessed by a group of independent observers (Lindberg and Fachlcrantz 2005; Bhattacharya *et al.* 2015). For practical purposes, it is desirable to have a reliable way of establishing a mottle index through mathematical image analysis of digitized printed samples. In general, the printed image is converted into gray levels, and then the mottle index is defined as the ratio of standard deviation to the mean of gray levels. However, this coefficient of variance (COV) has been criticized for some reasons, including neglecting the spatial correlations between the converted pixel and disregarding individual visual perception (Dubé *et al.* 2005).

Image analysis by band-pass algorithms, which divides print mottle into different spatial wavelength bands, helps the analysis of the print mottle. Johansson (1994) has shown that the wavelength bands of 2 to 4 mm and 4 to 8 mm exhibit the strongest correlation with the visual evaluation of print mottle. In this mottle evaluation technique, calculating the mottle index involves dividing the standard deviation of the reflectance factor by the mean reflectance factor level of the analyzed point. Thus, it provides the amount of mottle in the printed paper. This study aimed to investigate the influence of the formulation of pre-coating color, dwell time, and pre- and top-coat weights on the print mottle of double coated papers using an octave bandpass filtering technique.

EXPERIMENTAL

Materials

Hankuk Paper Co. (Ulsan, Korea) provided commercially produced base paper with a grammage of 78 g/m^2. The fiber composition of the base paper consisted of 75% softwood bleached kraft pulp, 15% hardwood bleached kraft pulp, and 10% bleached chemithermomechanical pulp. To investigate the effect of pre-coating composition on the printing mottle, two types of ground calcium carbonates (GCC 60 and GCC 95) both from Omya Korea, and No. 2 clay (Imerys, Savannah, Ga, USA) were used as pre-coating pigments. For top coating, a mixture of GCC 95 and No.1 clay (Imerys, Savannah, Ga, USA) in a ratio of 4:6 was used. The particle sizes of these four pigments were analyzed using a particle size analyzer (Micromeritics, SediGraph 5100, Norcross, GA, USA), and the median sizes were found to be 1.15, 0.43, 0.45, and 0.29 μm, respectively. Acetate ester starch (Samyang Genex, Incheon, Korea) and styrene/butadiene (S/B) latex (LG Chemical Co., Yeochun, Korea) were used as binders. Small amounts of lubricant (Nopcote C 104, San Nopco Korea Ltd., Pyeongteak, Korea), insolubilizer (Neowet 101H), and thickener (JT-35B, Jeong Won Chemical Co., Ltd., Busan, Korea) were added as additives.

Pre- and Top-coating Color Formulation

As a follow-up study to previous papers (Im *et al*. 2022; Youn and Lee 2022), pre- and top-coating processes were performed similarly. Briefly, pre-coating colors were prepared using the two types of GCC and No. 2 clay, individually or in combination. Each pre-coating color was stirred in a high-speed stirrer, and starch, latex binder, and small amounts of additive were added. The solids content of the pre-coating color was adjusted to 63% using distilled water. The six types of pre-coating colors used in experiments are designated as P1, P2, P3, P4, P5, and P6, and the coating color formulations are presented in Table 1. For the top-coating color, GCC 95 and clay pigments were mixed in a ratio of 60:40, and binders were added in the same manner. The solids content of top-coating color was adjusted to 64% with distilled water and named T1 (Table 1). Table 2 shows the properties of pre- and top-coating colors. The solids content of the top-coating color was adjusted to 64% with distilled water, and it was named T1 (ÅA-GWR, Kaltec Scientific, USA). The principle of this method is based on the measurement of the amount of water passing through a cellulose ester membrane with a pore size of 0.2 μm (Advantec, Taiwan) to the absorbent paper. Water retention measurements for each coating color were performed under condition of 25°C and at a pressure of 1.5 bar for 120 s.

Table 1. Coating Color Formulation in Parts by Weight

Ingredients		Grade	P1	P2	P3	P4	P5	P6	T1
Pigment	GCC	GCC60	100	-	75	50	25	-	
		GCC 95	-	100	-	-	-	-	40
	Clay	No.2	-	-	25	50	75		
		No.1	-	-	-	-	-	100	60
Binder	Starch	Modified	8	8	8	8	8	8	4
	Latex	S/B	8	8	8	8	8	8	11

Table 2. Properties of Coating Color Depending on Pigment Formulation

Formulations		Solids Content (%)	Viscosity (cPs)	pH	Water Retention (g/m^2)
Pre- coating	P1	63.0	881	8.8	94.0
	P2	62.9	3,279	8.5	194.5
	P3	62.9	1,328	8.8	76.4
	P4	62.9	2,240	8.9	73.6
	P5	62.4	2,509	8.8	67.1
	P6	62.6	3,459	9.0	65.5
Top-coating	T1	63.8	2,609	9.3	97.0

Preparation of Double-Coated Paper

The coating process was performed using a laboratory sheet coater (Maiyoh coater, PM-9040, SMT, Japan) equipped with two coating heads, each designed to allow both rod and blade for doctoring purposes (Fig. 1).

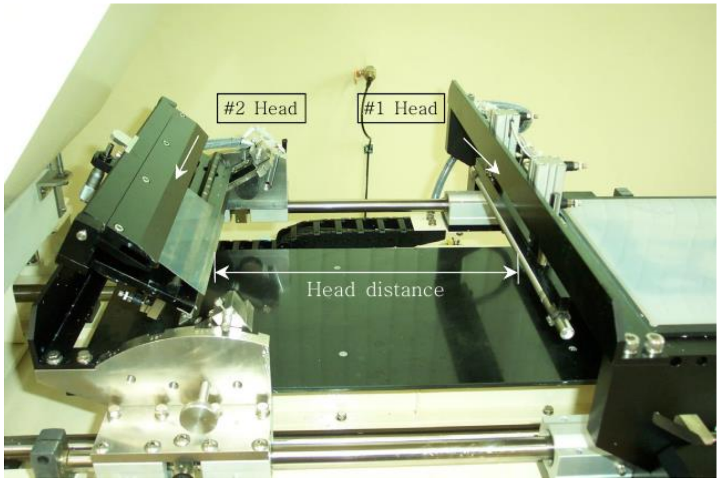

Fig. 1. Photograph of laboratory sheet coater

To prepare the pre-coated paper, the first head with a wire-wound steel rod was used to coat at approximately 100 g/m^2. The coating color was then doctored using a blade to achieve the desired coating weight. Top-coating color of 60 g/m^2 was applied onto the pre-coated paper using the selected coating rod and then metered with a doctor blade. The coating speed and head-to-head distance were adjusted to 150 m/min and 0.15 m, respectively, resulting in a dwell time of 0.06 s. Subsequently, the wet coating was dried using a hot air dryer at 150 °C. The double-coated paper was conditioned for more than 24 hours at a relative humidity of 50% and a temperature of 25 °C. After that, it was calendered twice using a laboratory calender at 0.9 kN/mm of line pressure and 45 °C of temperature.

Image Processing for Print Mottle Analysis

The analysis of printing mottle, depending on pigment types and printing process, was firstly conducted. Cyan ink (Deers I, Korea) was applied to the double-coated paper using a laboratory-scale printing tester (Prüfbau, München, Germany). To clearly distinguish the mottle level between the coated papers, an excessive amount of ink (0.7 mL) and a dampening solution (30 μL) were employed. Printing pressure, printing gap, and speed were adjusted to 20 kgf/cm^2, 7 s, and 96 m/min, respectively.

For the analysis of print mottle, an image analysis technique was used. After printing, test targets were scanned with a flatbed scanner (UMAX date systems, UMAX Astra 2400S, CA, USA), using 1200 dpi resolution in true color RGB mode. Brightness, contrast, intensity, and shading were kept uniform during the scanning process. Additionally, the gamma value was set to 1.0 to prevent internal color channel variations in RGB mode. Image samples were scaled at 1 × 1 inch and each image was saved as an RGB image using TIFF format. The saved images were then converted to grayscale with 8 bits per pixel for pixel-level analysis. After that, octave bandpass filtering was carried out for analysis of print mottle. The image processing for bandpass filtering and calculating the mottle index was conducted using the Image-Pro PLUS program (Media Cybernetics, MD, USA).

Analysis of Print Mottle Index

The mottle index was calculated using the coefficient of variation (COV) in each band-pass region of printed images subjected to octave band filtering, as described in Eq. 1:

$$\text{Mottle index} = ((COV_{(bp\ 0.25–0.5)} + COV_{(bp\ 0.5–1)} + COV_{(bp\ 1–2)} + COV_{(bp\ 2–4)})/4) \times 100 \quad (1)$$

The variations were evaluated at 4 different intervals (0.25 mm to 0.5 mm, 0.5 mm to 1.0 mm, 1.0 mm to 2 mm, and 2 mm to 4 mm). The variations below 0.25 mm were excluded from the calculation of the mottle index due to the difficulty in visual discernment. In addition, the variations beyond 4 mm were omitted from consideration as they rarely occur in actual printed images.

Surface Properties of Double-Coated Paper

Parker Print-Surf (PPS) roughness tester (Messmer Büchel, Gelderland Veenendaal, Netherlands) was used based on the TAPPI method (TAPPI T 555) to evaluate the roughness of double-coated paper under a pressure of 10 kgf/cm^2. Additionally, the surface roughness of the coated paper was also analyzed using a 3D noncontact surface profiling device (NanoScan E, NanoSystem & Technology, Daejeon, Korea).

RESULTS and DISCUSSION

Effect of the Type of Pre-coating Pigment on Print Mottle

The print mottle of double-coated paper with the same top-coating color applied to the pre-coating layer, which consisted of a single or mixture of pigments, was investigated. Table 3 shows the COV in each bandpass region on the printed surface and mottle index depending on the pigment composition of the pre-coating layer. It can be observed that the mottle index increased when the small size of pigments, such as GCC 95 (*i.e.*, P2T1) and No.2 clay (*i.e.*, P6T1), were used compared to when GCC 60 (*i.e.*, P1T1) was used. This trend was consistent across all ranges from 0.25 mm to 4 mm.

Moreover, in the case of P1T1, P3T1, P4T1, P5T1, and P6T1 with varying mixtures of GCC and No. 2 clay, an increase in the content of No. 2 clay led to an increase in the COV in all octave band regions, resulting in a deterioration in the mottle index (Table 3).

Previous research results indicate that as the particle size of pigments used in the pre-coating layer decreases, the pore size formed in the double coating layer decreases, while the number of pores per unit of surface area increases (Youn and Lee 2022). Furthermore, dynamic water absorption of the top-coating color increased with the increase of the number of pores in the pre-coated layer. Accordingly, the surface latex concentration of the top-coated layer decreased with the increases in the water penetration rate of the top-coating color into the pre-coated layer (Im *et al*. 2022). Thus, the increase in the mottle index of double-coated paper can be attributed to the uneven distribution of latex in the coated paper, depending on the type of pre-coating pigment used. The pore diameter in pigment coating layers typically ranges between 0.01 and 1.0 μm (Donigian *et al.* 1997; Johnson *et al.* 1999). It is important to determine the range of pore diameters that can be attributed to the coating layer because pores are present not only in the coating layer but also in the base paper.

Figures 2 and 3 show the correlation between the relative pore ratio in the coating layer and the mottle index. It can be seen that the uneven distribution of latex, which was caused by the increased number of pores per unit area at the surface in the pre-coating layer, resulted in the irregular penetration of the vehicle contained in the printing ink and increased print mottle of the double-coated paper. Similar results have been reported by Fujiwara *et al*. (1989). They studied the effect of latex concentration on the coating layer on offset mottling, and the results showed that the amount of water and latex that penetrated the pre-coating layer increased when the solids content of coating color is low, resulting in deteriorated offset print mottle.

Table 3. COV of Each Octave Band Width and Mottle Index Depending on Pigment Composition of Pre-Coated Layer

Sample	COV of Octave Band Width				Mottle Index
	0.25 to 0.5 mm	0.25 to 0.5 mm	1 to 2 mm	2 to 4 mm	
P1T1	0.240	0.158	0.115	0.086	14.99
P2T1	0.257	0.176	0.129	0.098	16.51
P3T1	0.238	0.168	0.127	0.094	15.67
P4T1	0.234	0.173	0.131	0.098	15.92
P5T1	0.261	0.174	0.127	0.097	16.48
P6T1	0.261	0.186	0.136	0.104	17.19

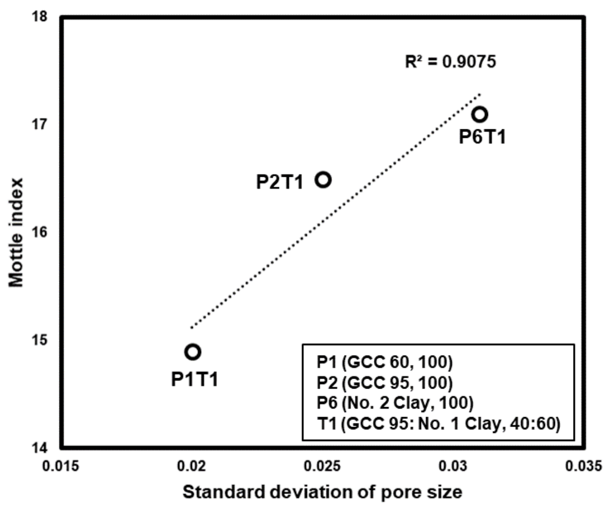

Fig. 2. Relationship between relative pore ratio and mottle index depending on pigment composition of precoated layer

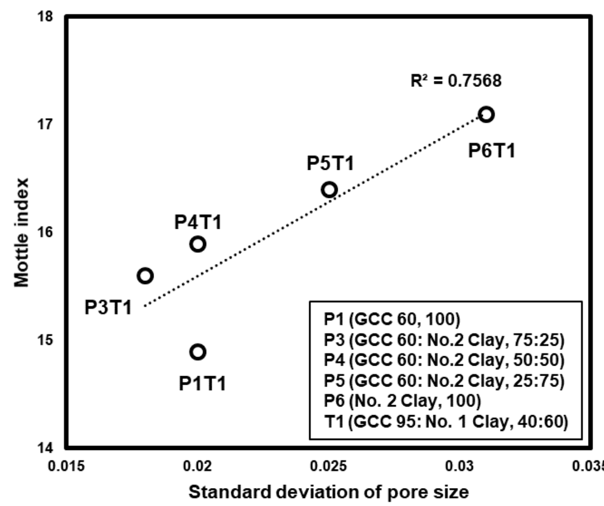

Fig. 3. Relationship between relative pore ratio and mottle index depending on pigment composition of precoated layer

Effect of the Dwell Time on Print Mottle

To investigate the effect of dwell time on print mottle, coating speed and the head-to-head distance were readjusted to 90 m/min and 0.45 m, respectively, which resulted in a dwell time of 0.30 s. Through controlling this adjustment, the time required to transfer the coated paper from the second 2 coating head to the dryer decreased.

In a previous study (Im *et al.* 2022), it was found that the number of pores per unit area at the surface of the coating layer was affected by the dwell time, which is the time between the rod and metering of the surplus coating color. Especially, the increase in dwell time during the pre-coating stage significantly increased the number of pores per unit area at the double-coated layer surface compared to the changes in dwell time during the top-coating. Furthermore, a shorter transfer time to the dryer caused a substantial amount of latex migration to the surface of the coating layer rather than penetrating the base paper or pre-coated layer.

Figure 4 indicates the COV at each octave band and mottle index. It can be seen that the mottle index and COV increased as the transfer time to the dryer decreased. The results suggest that the decreased transfer time to the dryer led to a rapid latex migration to the coating layer surface, which might cause an uneven distribution of latex on the coating layer. Uneven binder migration toward the coating surface may cause print mottle, because of nonuniformities in binder distribution at the coating surface may cause uneven ink transfer and nonuniform water and ink absorbency (Du *et al.* 2011).

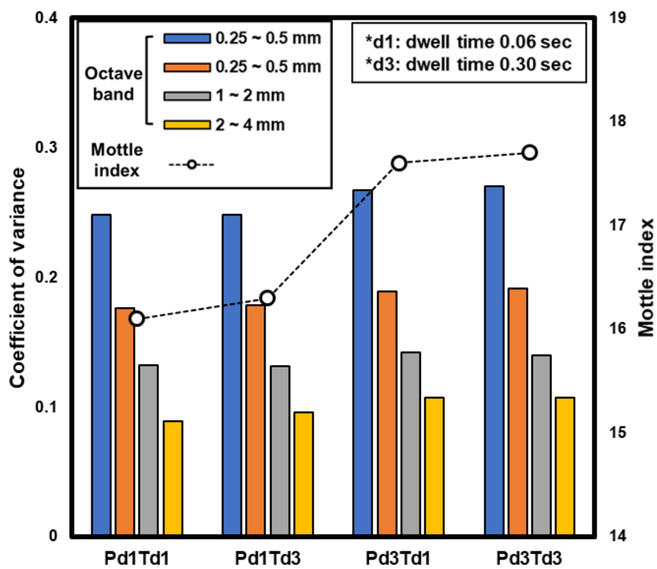

Fig. 4. Relationship between relative pore ratio and mottle index depending on pigment composition of pre-coated layer

Effect of Pre- and Top-coat Weights on Print Mottle

To evaluate the effects on print mottle, pre- and top-coat weights were adjusted under the conditions of P1T1. At a coat weight of 6.0 g/m^2 level of the pre-coating layer, the mottle index showed a notably high value of around 18, regardless of the coating amount of the top-coating layer.

Table 4. COV of Each Octave Band Width and Mottle Index Depending on Coat Weight

Sample	COV of octave band width				Mottle Index
	0.25 to 0.5 mm	0.25 to 0.5 mm	1 to 2 mm	2 to 4 mm	
$P_{6.0}T_{4.6}$ *1	0.287	0.192	0.135	0.097	17.8
$P_{6.0}T_{11.2}$ *2	0.271	0.190	0.140	0.109	17.8
$P_{9.3}T_{4.2}$ *3	0.231	0.165	0.121	0.092	15.2
$P_{9.3}T_{9.5}$ *4	0.225	0.160	0.117	0.093	14.9

*1: Coat weight: Pre 6.0 g/m^2, Top 4.6 g/m^2
*2: Coat weight: Pre 6.0 g/m^2, Top 11.2 g/m^2
*3: Coat weight: Pre 9.3 g/m^2, Top 4.2 g/m^2
*4: Coat weight: Pre 9.3 g/m^2, Top 9.5 g/m^2

However, the mottle index of double-coated paper decreased at a pre-coat weight of 9.3 g/m^2, irrespective of the coat weight of top-coating. Additionally, the increased coat weight of the top-coating layer did not affect any change in print mottle at the low coat weight of the pre-layer (*i.e.*, 6.0 g/m^2). As the coat weight of the pre-coating layer increased, an increase in the coat weight of the top layer resulted in a decrease in the mottle index (Table 4).

Figures 5 (a) and (b) show scanning electron microscopy (SEM) images of pre-coated paper with different coat weights. It can be observed that the surface of the base paper was not sufficiently covered at a pre-coating weight of 6.0 g/m^2. This tendency was also reflected in the surface roughness of the double-coated paper, as shown in Fig. 6. Furthermore,

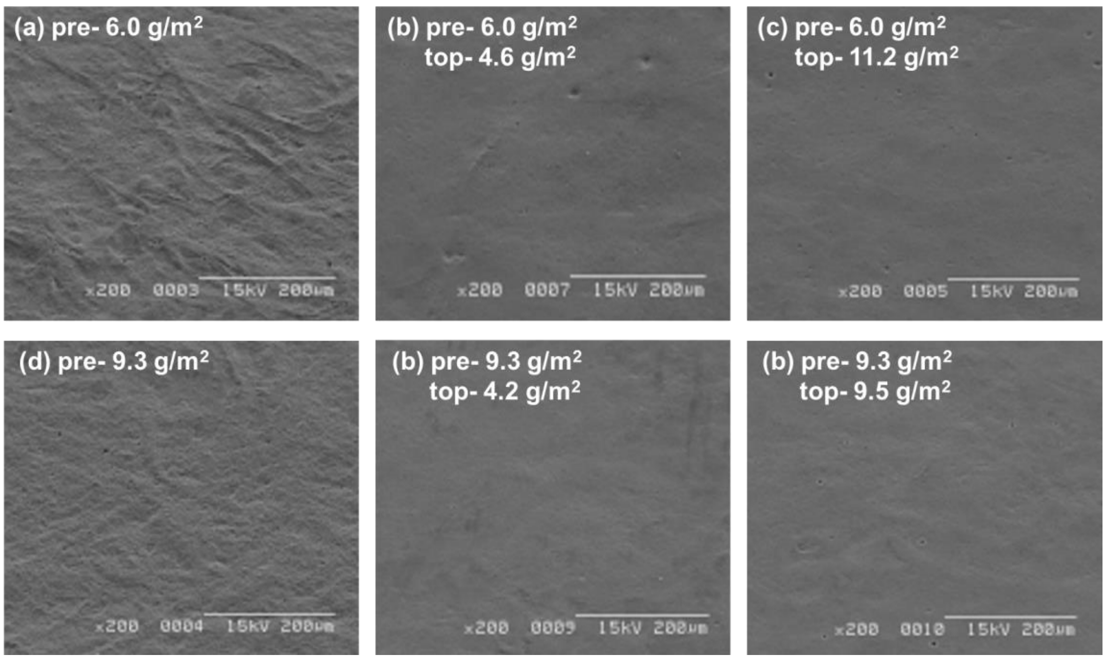

Fig. 5. SEM microphotographs depending on pre- and top-coating weights

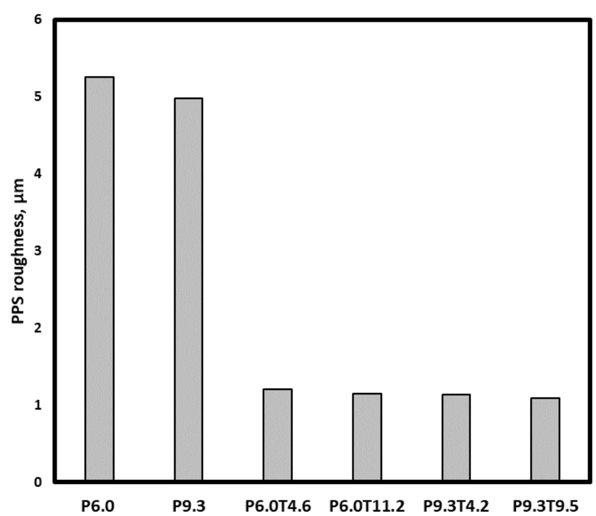

Fig. 6. Parker print-surf (PPS) roughness of pre- and double coated papers

Figure 7 represents a 3D view photograph of the coated paper, depending on pre- and top-coat weights. Similar to the roughness results, the actual surface roughness can be observed. It is worth noting that the coat weight of the pre-coating layer is a more crucial factor than the coating amount in the top-coating layer to enhance print mottle. Especially, it is suggested that increasing the coating amount of the pre-layer might be more effective in improving the print mottle compared to controlling the top-coating amount.

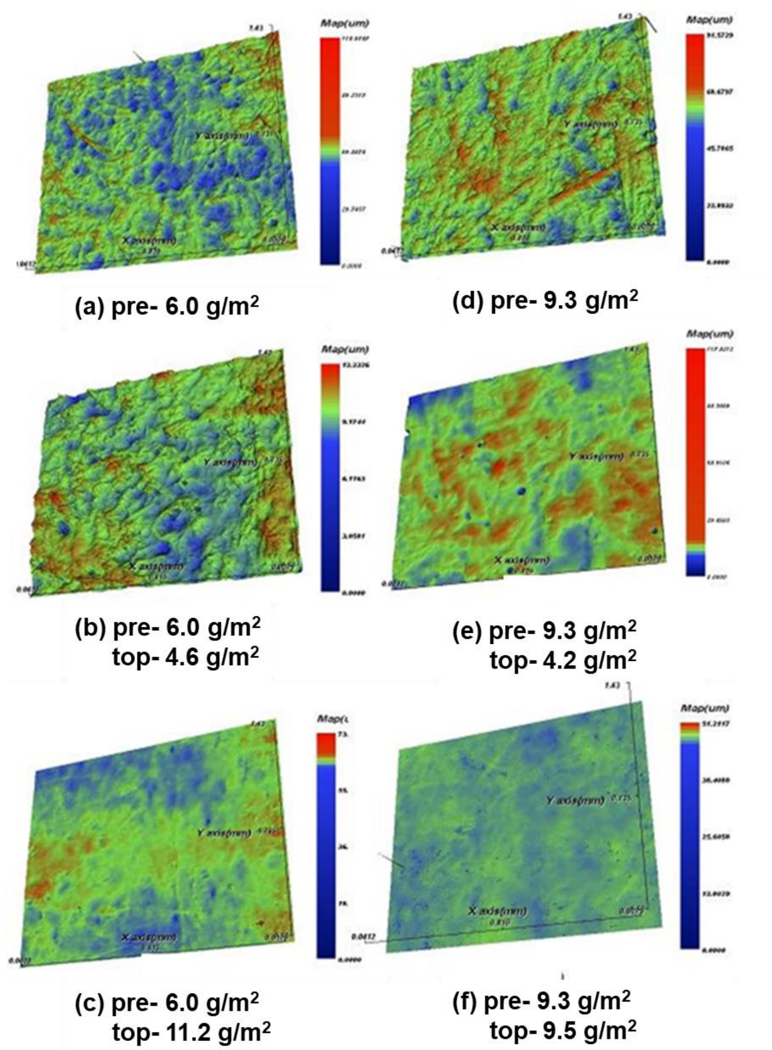

Fig. 7. 3D view photographs depending on pre- and top-coating weights

CONCLUSIONS

The influence of the pre-coating formulation, dwell time, and coat weight on print mottle was investigated. To evaluate the mottle index, an image analysis based on the octave band filtering technique was used. It was observed that the print quality of double-coated paper was significantly influenced by the pigment formulation of pre-coating color and the coat weight of pre-coating layer. Moreover, it is expected that enhancing the uniformity of printing can be achieved through rapid drying during the coating process.

1. The effect of pre-coating color formulation on the print mottle was investigated. The smaller size of pigment in the pre-coating color increased the number of pores per unit for surface area in the pre-coated layer, which is attributed to an increase of water penetration from top-coating color to the pre-coated layer. The latex distribution in the top coating layer becomes uneven due to the increased water penetration, leading to an increase in the mottle index.
2. Decreased transfer time to dryer by readjustment of coating speed and head-to-head distance causes rapid latex migration to the coating layer surface. Thus, uneven binder migration toward the coating surface increases the print mottle of double-coated paper.
3. Pre- and top-coat weights were adjusted to investigate effect of coat weight on print mottle and COV. Mottle index decreased with increased pre-coat weight, regardless of the top coat weight. This indicates that when the pre-coating layer dos not adequately cover to surface of the base paper, the print mottle was increased due to the roughness of base paper.

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