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Effect of coating pigment type on paper printability with water-based inks

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Abstract Nowadays, the need for papers and cardboards with improved surface properties is increasing with the development of the packaging industry. The improvements made are not only limited to the paper and paper surface, but also environmentalist approaches are exhibited in printing ink. For this purpose, the use of water-based ink tends to become widespread, especially in food packaging. In this study, five different paper coating formulations were prepared using different proportions of precipitated calcium carbonate (PCC) and kaolin pigments. Latex (Acronal S360D) was used as a binder. All prepared coating mixes were applied onto the base paper surface. All coated papers were also subjected to the calendering process. In addition, three different dyestuffs (14, 17, 20%) concentrations of water-based printing inks were prepared. Prints were made by the silk screen printing technique on the calendered-coated papers. The optical and physical properties (roughness, air permeance, paper gloss and printing gloss, contact angle and surface energy) of all papers were measured according to the standards. Paper gloss, print gloss, ΔE_{00} color differences, contact angles and surface energy measurements were made, and the effects of dye ratio on printing and gloss in water-based inks and coating type suitable for printability were investigated. As a result, papers can be coated and even calendered to achieve better paper surface properties and printability. In the case of printing with water-based inks, better printability properties are obtained by choosing PCC pigmented coated papers.

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Introduction

In the paper industry, it is important to produce high quality paper with low cost and to minimize environmental problems.[1] In the papermaking process, every process and component has an effect directly or indirectly on the surface properties.[2] In general, fillers are often used in papermaking to save energy, reduce the cost, improve optical and physical properties,[3] printability (ink absorption, smoothness and transparency) stability of dimension and the appearance of papers surface.[4] A paper coating formulation generally consists of low amounts of mineral pigments, polymeric thickeners, latex binders, and other additives (pH controllers, dispersants, dyes, biocides, and foam controllers, e.g.).[5–7] In a coating, pigments are used to provide optical properties, binders are used to provide the adhesion and strength properties of the pigment and coating layer, and polymer additives are used to adjust the runnability during the coating process and to make the paper properties more stable. The optical and physical properties of papers and cardboards affect their printability properties. One of the ways to obtain high quality printing is with good surface treatment on paper.[8–10]

In all printing systems, it is one of the best solutions to coating the surface of the base paper to form a good ink film layer on the paper surface to meet the requirements.[11,12] Surface treatments play a key role in ensuring adequate interaction of the paper surface with inks to achieve better print quality. The penetration of the ink can be controlled through the coating layer. It is ensured that the paper surface is sufficiently smooth to ensure a good printing and ink transfer. Surface coating processes also help to control and improve other paper properties such as tear resistance, tensile strength and other important properties such as air permeability and roughness.[13]

Traditional and frequently used inorganic fillers are natural ground calcium carbonate, kaolin clay and talc.[4] Kaolin is one of the main pigments used in paper coating applications.[14] Millions of tons of kaolin are used every year for paper coating, filler and different applications in the world.[15] Kaolin pigments are produced from the original mineral form through a series of extraction and refining processes. Kaolin is a valuable coating pigment due to its flat particle shape, good color (white or close to white), and ease of processing with particle size. Calcium carbonate ($CaCO_3$) is used both as a pigment and as a filler to improve the smoothness, gloss, and opacity of coated paper.[16]

Printing ink is an important consumable material for the printing industry. Printing inks are diversified depending on the binder (UV, solvent, oil or water based). Binders have an important role in ink as a type of adhesive that binds the pigment to the substrate. Volatile organic compounds (VOCs) used in the preparation and usage processes of traditional solvent-based inks and colorants, binders and additives used in other inks have several adverse effects on the environment and human health. Because of this reason new production methods and materials should be used to minimize dependence on petrochemical resources and negative environmental impacts.[17] Therefore, the development of environmentally friendly inks becomes more important.[18] The increase in the use of environmentally friendly inks will be effective in reducing the harmful environmental impacts caused by traditional inks.[19] Water-based inks are made with acrylic resins[20] and have a hydrophilic character.[21]

The water-based binder provides improved health and safety by eliminating the need for organic solvents in the manufacturing process.[22] The printing industry tends to be more sustainable, according to recent research. Water-based ink has been potentially considered as the most sustainable printing ink due to its environmentally friendly, high solids content and bright, low-viscosity advantages.[23]

In this study, two different pigments, kaolin and precipitated calcium carbonate (PCC) were used in coatings on the paper surface, and latex (Acronal) was used as a binder. Prints were made on the prepared coatings with water-based ink with three different dye ratios. Analyses were conducted to understand the effects of inks with different dye ratios such as ink acceptance, color intensity effect and printability on the paper surface coated with different pigments. This study aims to improve the printability performance of water-based inks with different pigment ratios on different coatings.

Materials and methods

Materials

Precipitated calcium carbonate (PCC) was provided from Omya, and kaolin was obtained from Imerys. Latex (Acronal S360D) as a binder and co-binder (Acrosol) were obtained from Celanese Corporation. 80 g/m^2 white office paper was provided by Antalis. Dyestuff was provided from TOYO Ink.

Paper coating formulations

For this study, five different paper coating formulations were prepared. All paper coating formulations were prepared in proportion as 60% solid using kaolin and precipitated calcium carbonate (PCC), latex and co-binder (Table 1). The pH values of coating formulations were adjusted and ranged from pH 8.5 and 9.5. The viscosities of the coating formulations were measured with Brookfield viscometer (spindle #2; at 100 rpm). The coatings were applied twice to a sized commercially produced base paper using a K- Control laboratory rod coater. After coating, the samples were air dried overnight under TAPPI conditions. The coated samples were then calendared at 150 PLI, 2-nips against a polished metal roll.

The pigment characteristics are given in Table 2. The binders' properties according to commercial firms are given in Table 3.

Optical and physical properties of papers

All the calendared-coated paper samples were conditioned for 24 h at 50 % RH and 23°C before testing. Paper roughness of all papers was carried out with Lorentzen & Wettre (L&W) in accordance with the standard ISO 8791-2:2013: Bendtsen method, and air permeance of all papers were carried out with L&W in accordance with the standard ISO 5636-3:2013.

Scanning electron microscope image of the obtained composites was recorded on Philips XL30 ESEM-FEG/EDAX. The hybrid composites were brought to

Table 1: Paper coating formulations

Coating	P0	P1	P2	P3	P4	P5
Ingredients	Base	Dry parts added				
Kaolin	–	–	30	50	70	100
PCC (Calcite)	–	100	70	50	30	–
Latex (Acronal S360D)	–	10	10	10	10	10
Co-binder (Acrosol)	–	0.25	0.3	0.3	0.3	0.3

Table 2: The characteristics of the mineral pigments

		Kaolin	PCC (Calcite)
Brightness	%	88	93
Particle size	wt% <2 μm	80	56
pH		8.50–9.50	8.50–9.50
Density	g/cm^3	2.55	2.7

Table 3: The characteristics of the binder (*Acronal S360D*)

Properties	Amount
Solid content	50 wt%
Average particle size	0.2 μm
Viscosity	375 mPa.s
Density	1050 kg m^{-3}
pH	8 ± 0.50

the solid phase with liquid nitrogen and then coated with platinum to prepare for SEM.

The gloss measurements of papers were carried out with BYK Gardner GmbH micro gloss 75º geometry in accordance with the standard ISO 8254-1:2009 and the gloss measurements of prints with BYK Gardner GmbH micro-Tri-gloss 60º geometry in accordance with ISO 2813:2014. And then, gloss difference of samples was calculated.

The contact angle and total surface energy measurements of the papers were performed by Pocket Goniometer PGX+ in accordance with the standard ASTM D5946.

Preparing water-based ink

Three different concentrations of water-based printing inks were prepared by mixing commercially available organic and water-soluble black dyestuff, water-based varnish and other ink components. High speed butterfly stirrer mixers were used for the ink preparation process. In addition, viscosity and pH values were fixed. The dyestuff ratios and general contents of the prepared inks are given in Table 4.

Printing conditions

In the study, prepared inks were used to make solid prints on base paper and coated papers in the squeegee printing parameters which have 77 tpc weaving density, 75° squeegee angle and 75 shore hardness by ARUS semi-automatic screen-printing machine.

Table 4: Formulations of water-based printing inks

Ingredients		Ink 1 (I1)	Ink 2 (I2)	Ink 3 (I3)
Dyestuff	%	14	17	20
Resin	%	20	20	20
Water	%	58	55	52
Additives	%	8	8	8

Color properties

The color properties of all papers and also printed papers were measured using CIEL*a*b* color values by using X-Rite eXact spectrophotometer according to ISO 13655:2017 standard. The measurement conditions of the spectrophotometer were determined as polarization filter with 0/45º geometry with 2º observer angle with D50 light source in the range of 400-700 nm. The difference between the colors of the different prints was calculated according to the CIE ΔE 2000 euclidean color-difference formula ISO 11664-6:2014. Calculations were made by taking the average of five measurements. ΔL^*, Δa^*, Δb^*: Difference in L^*, a^*, and b^* values between specimen color and target color. Lightness is represented by the L* axis which ranges from white to black. The red area is connected to the green by the a^* axis, while the b* axis runs from yellow to blue. The value of ΔE_{00} was calculated according to equation (1).

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C'}{k_C S_C}\frac{\Delta H'}{k_H S_H}} \quad (1)$$

where ΔL^*, ΔC^*, and ΔH^* are the CIEL*a*b* metric lightness, chroma, and hue differences, respectively, calculated between the standard and sample in a pair, and ΔR is an interactive term between chroma and hue differences. The S_L, S_C, and S_H are the weighting functions for the lightness, chroma, and hue components, respectively. The values calculated for these functions vary according to the positions of the sample pair being considered in CIEL*a*b* color space. The k_L, k_C, and k_H values are the parametric factors to be adjusted according to different viewing parameters such as textures, backgrounds, separations for the lightness, chroma, and hue components, respectively.[24] According to tolerance definition, $\Delta E_{00}^* \leq 2$ is classified as the very small noticeable difference for a standard observer, while $\Delta E_{00}^*=5$ is defined as a big noticeable difference in color that a standard observer can recognize.[25]

Results and discussion

Paper roughness and air permeance of all papers were carried out with L&W in accordance with the aforementioned standards. The measurement results are shown in Figs. 1 and 2.

When the roughness and air permeance values are examined, it is seen that in Figs. 1 and 2, the paper surface coating and then calendering process reduce the roughness and air permeability of the paper. The application made reduced the porosity by filling the pores of the paper, minimized the air permeability of the paper and improved printability by making the paper more compact. The results obtained are consistent with the literature.[26]

SEM images of all papers are given in Fig. 3. When the SEM images were examined, it was determined that there was a homogeneous distribution when

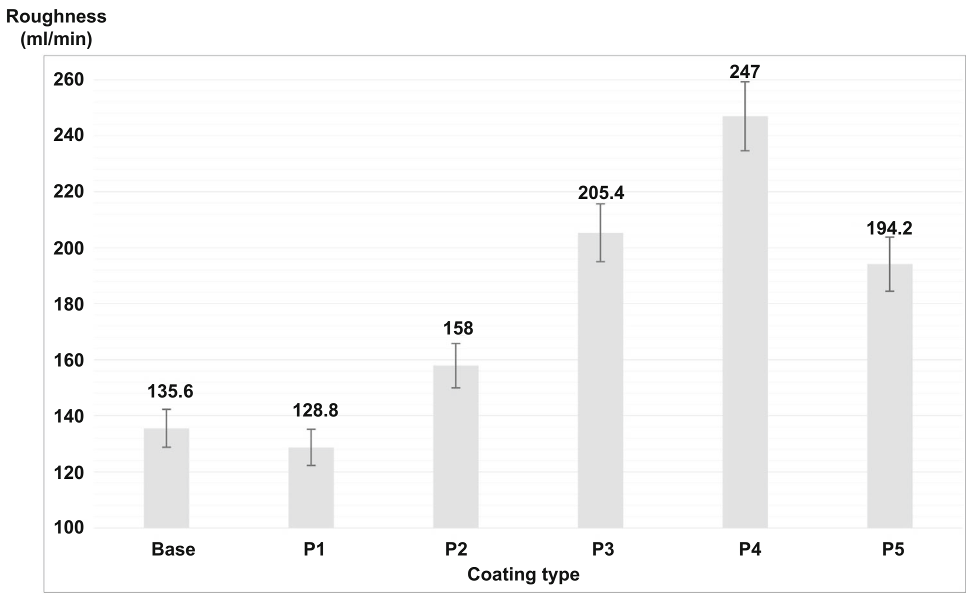


Fig. 1: The roughness of paper samples coated with formulations of different pigment composition (P1—100% PCC, P2—70% PCC and 30% Kaolin, P3—50% PCC and 50% Kaolin, P4—30% PCC and 70% Kaolin, P5—100% Kaolin)

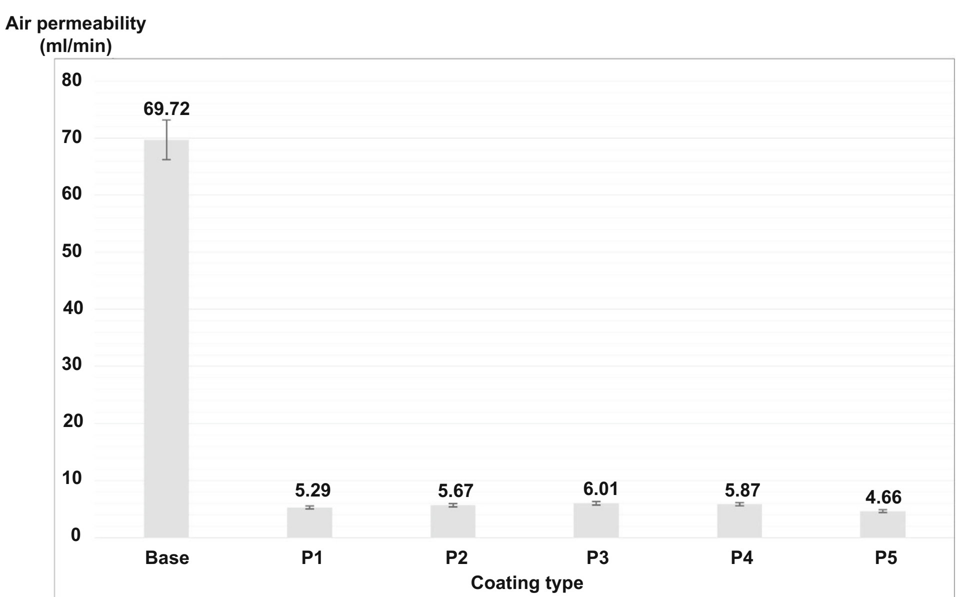


Fig. 2: Air permeability measurement of paper samples coated with formulations of different pigment composition

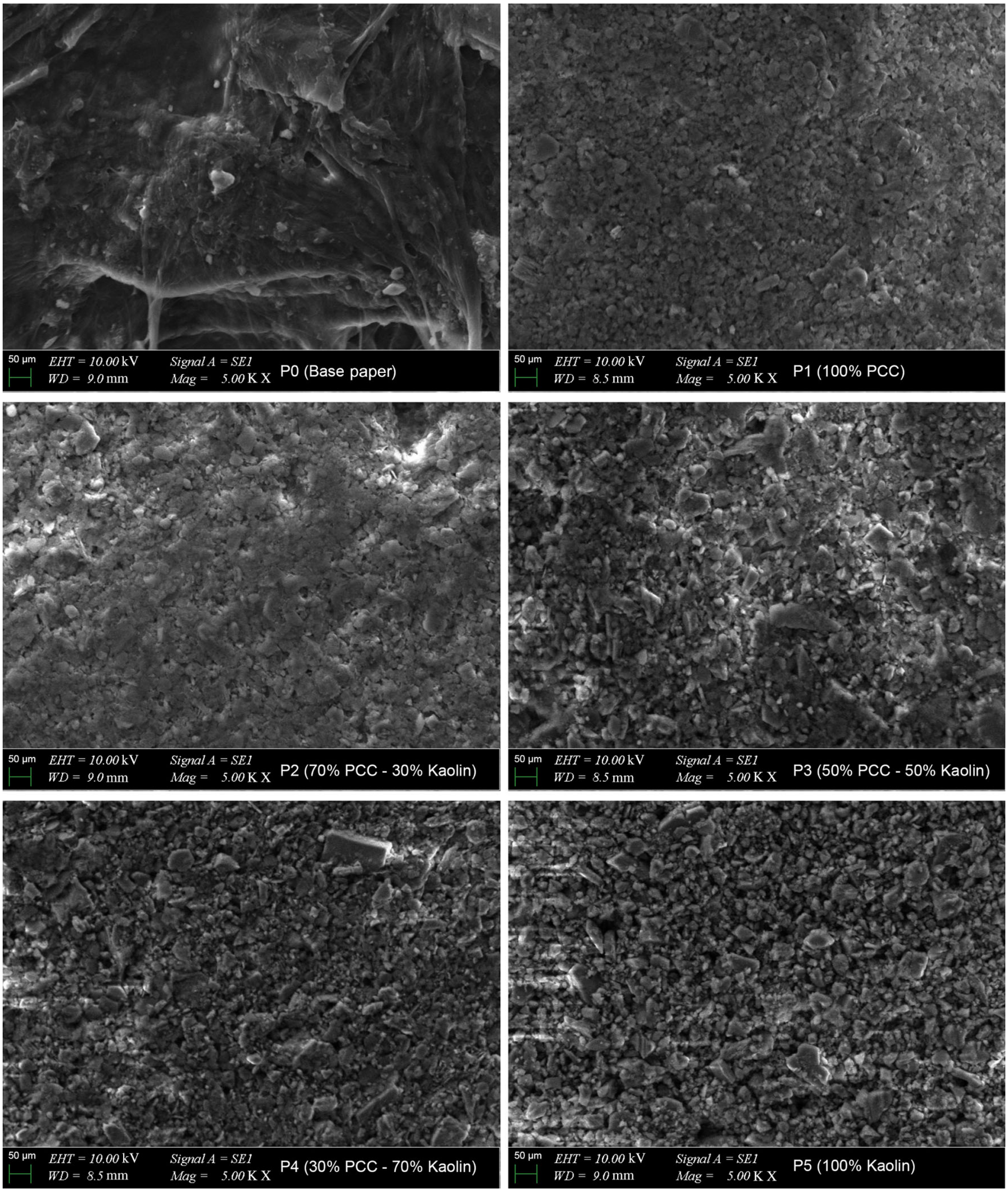


Fig. 3: SEM images of paper samples coated with formulations of different pigment composition

coatings were made on the base paper surface, and the particle sizes were obtained in accordance with Table 2 and there was no agglomeration. That is, the surface is homogeneously distributed and stable. In addition, the results of SEM-EDAX chemical content results are given in Table 5. When the table is examined, carbon (C) and oxygen (O) in the base paper content, and on the other hand, calcium (Ca) and aluminum (Al) used in surface coating processes were released. This is an expected result. The increase in calcium caused by $CaCO_3$ is seen in coatings made with PCC. The increase in the amount of calcium increases as the amount of PCC increases. In coatings containing kaolin, an increase was observed in the amount of silicon (Si) and aluminum (Al), and this increase varies in proportion to the percentages. That is, the chemical content of the coatings was confirmed by SEM-EDAX.

Gloss measurement results of base paper and coated papers are shown in Fig. 4. When the gloss values of the base paper and coated papers are examined, it is

Table 5: SEM-EDAX chemical content results of paper samples coated with formulations of different pigment composition

EDAX	Element	Wt %
P0 (Base Paper)	Carbon	27.2
	Oxygen	40.55
	Aluminum	4.27
	Calcium	27.97
P1 (100% PCC)	Carbon	16.21
	Oxygen	53.16
	Calcium	30.63
P2 (70% PCC and 30% Kaolin)	Carbon	16.79
	Oxygen	54.63
	Aluminum	10.02
	Silicon	7.25
	Calcium	11.32
P3 (50% PCC and 50% Kaolin)	Carbon	13.57
	Oxygen	53.55
	Aluminum	12.05
	Silicon	9.01
	Calcium	11.82
P4 (30% PCC and 70% Kaolin)	Carbon	10.64
	Oxygen	50.7
	Aluminum	15.82
	Silicon	13.37
	Calcium	9.46
P5 (100% Kaolin)	Carbon	7.19
	Oxygen	52.83
	Aluminum	21.54
	Silicon	18.44

seen that the gloss values increase in all coated papers. When the coated papers were examined, it was found that the gloss value was the highest in the papers using only PCC coating pigment. As the amount of PCC decreases and the amount of kaolin pigment increases, the gloss value decreases.

Gloss measurement results of water-based ink printed papers are shown in Fig. 5. Print gloss was higher on all coated papers compared to base papers. The highest print gloss value was obtained on papers using PCC pigment. The print gloss values decreased as the amount of PCC decreased or the kaolin increased. These results match up with the paper gloss values. When the water-based ink dyestuff ratios are considered, the print gloss values of the inks with the lowest dye ratio (I1, 14%) are higher than the others. The main factor of the affecting print gloss is the binder in the ink. The binder ratios of the inks used in the study were kept constant. The difference in the measurements is due to the difference in dye content. As the dyestuff ratios increased, the roughness on the paper surface increased and this caused a decrease in brightness.

Total surface energy values and contact angle measurements results are shown in Table 6. As the molecular structure of the PCC pigment differs from the kaolin, as the ratio of PCC pigment increases, the hydrophilic property increases, which means low contact angle and high surface energy. On the other hand, the kaolin pigment has reduced the hydrogen bonding capacity between water and cellulose fibers, thus increasing hydrophilicity and decreasing the surface energy as the contact angle increases. These results

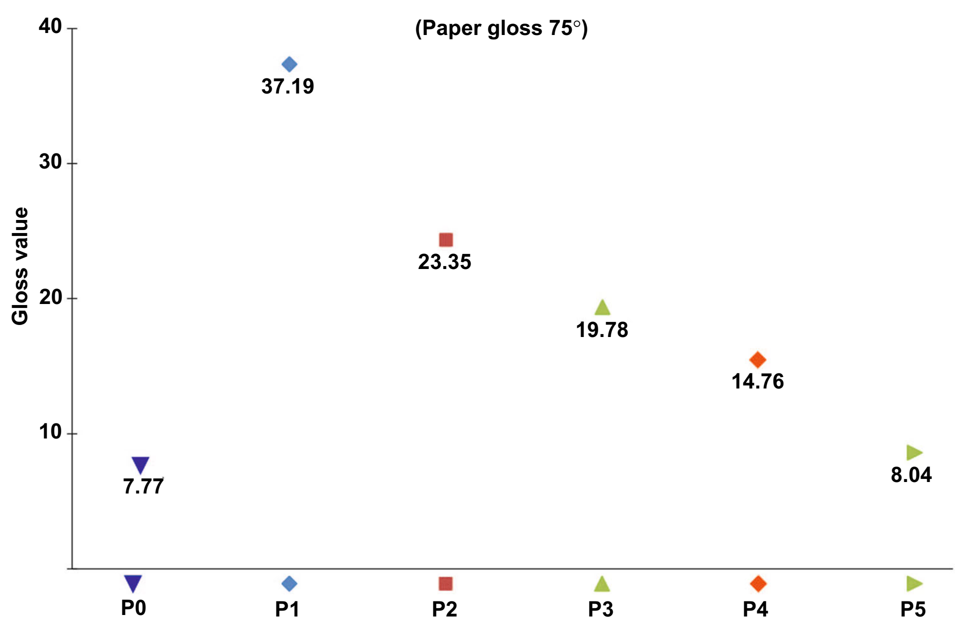


Fig. 4: Gloss values of papers according to ISO 8254-1:2009

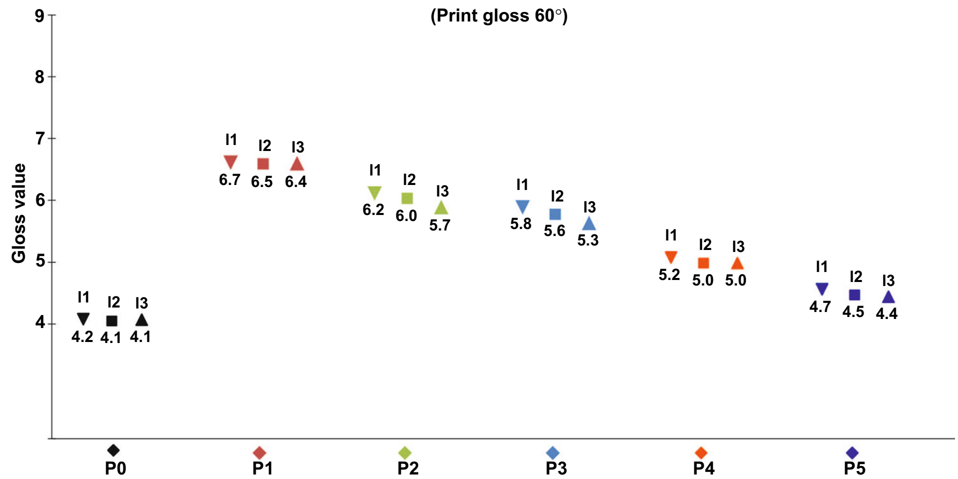


Fig. 5: Gloss values of printed papers according to ISO 8254-1:2009 (I1–14% Dyestuff, I2–17% Dyestuff, I3–20% Dyestuff)

Table 6: Total surface energy and contact angle values according to ASTM D5946 method

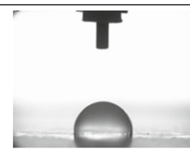
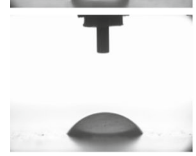
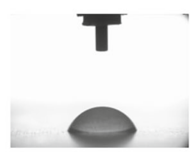
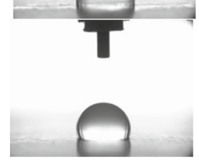
Coating type	Contact angle	Surface energy (mJ/m^2)	Contact angle images
P0	81.6	35.5	
P1	59.9	44.4	
P2	70	39.7	
P3	84.4	34.5	
P4	95.1	30.6	
P5	113.7	23.9	

Table 7: The difference between the color of the papers printed according to the CIE ΔE 2000 euclidean color-difference formula

	Ink 1 (I1) ΔE_{00}	Ink 2 (I2) ΔE_{00}	Ink 3 (I3) ΔE_{00}
P0	6.04	5.45	5.40
P1	4.04	3.00	2.08
P2	4.96	3.89	2.96
P3	4.80	3.94	3.78
P4	4.96	4.65	3.84
P5	4.48	3.94	3.96

mean better printability on coatings with PCC pigment or a high proportion of PCC pigment in prints made with water-based inks.

The values for the comparison of color difference ΔE_{00} of coated papers printed with water-based black ink according to ISO reference paper type 2 are given in Table 7. All prints were made with an equal amount of ink.

When the CIE ΔE 2000 color difference table of the prints made with water-based black ink on coated papers in Table 7 is examined according to the ISO reference paper type 2, the color differences in the coatings using PCC as the coating pigment are lower than the coatings using the kaolin pigment. The increase in color differences as the amount of kaolin in the coating increases shows the consistency of the coatings. The printing data for coatings using PCC were also consistent with the results of contact angle and surface energy measurements. When the inks were evaluated, the ΔE_{00} color difference was obtained less in the prints made with (I3) inks with high dye content. In the prints made with fixed viscosity, the increase in dye ratio increased the color intensity and better spectrophotometric results were obtained.

Conclusions

The paper surface coating and calendering processes reduce the roughness and air permeability of the paper. When the porous surface of the paper is coated with pigment, this decreases its air permeability.

The gloss values of all coated papers increase. A higher gloss value is obtained in coatings with PCC pigment compared to kaolin pigment.

In prints made with water-based inks containing different proportions of dyestuff, the print gloss decreases as the dye ratio increases. Considering the coated papers, a higher print gloss is obtained in the coatings using PCC pigment compared to the kaolin pigment.

The molecular structure of the PCC pigment is different from the kaolin, so as the proportion of PCC pigment increases, the hydrophilic property also increases, which means low contact angle and high surface energy. On the other hand, the kaolin pigment has reduced the hydrogen bonding capacity between water and cellulose fibers, thus increasing hydrophily and decreasing the surface energy as the contact angle increases. These results mean better printability on coatings with PCC pigment or a high proportion of PCC pigment in prints made with water-based inks.

In coatings using PCC as a coating pigment, CIE ΔE_{00} color differences are lower than coatings using kaolin pigment. When the L*a*b* values of the inks were measured, the ΔE_{00} color difference was obtained less in the prints made with (I3) inks with a high dye content. In the prints made with fixed viscosity, the increase in dye ratio increased the color intensity and better spectrophotometric results were obtained.

As a result, papers can be coated and even calendered to achieve better paper surface properties and printability. In the case of printing with water-based inks, better printability properties are obtained by choosing PCC pigmented coated papers.

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Human and animal rights This article does not contain any studies with animals performed by any of the authors.

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