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ORIGINAL ARTICLE

Coloration Technology

Usability of cellulose-based binder in water-based flexographic ink

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Abstract

Packaging must have a good commercial appearance and is generally obtained by ink transferred to its substrate. It is important that the ink used in packaging printing is produced from environmentally friendly and sustainable raw materials as well as being suitable for the printing system. The increasing demand in the field of printed packaging and the scarcity of resources to meet this demand have accelerated the search for new sources for inks. For this purpose, inks produced in the laboratory using a modified cellulose-based binder, a commercial acrylic resin and a commercial soybean protein were compared with a commercial ink. As a result of the study, it was determined that the printability properties of the ink obtained by using the modified cellulose-based binder were better than the ink obtained with commercial soybean protein. It was determined that it showed printability properties close to the ink produced with commercial acrylic binders. By using modified cellulose-based water-based flexographic ink instead of other commonly used binders, more environmentally friendly sustainable inks can be produced.

1 | INTRODUCTION

The use of printing inks for food packaging is regulated by the European Printing Ink Association (EuPIA) and studies on this subject are ongoing [1] and by the US Food and Drug Administration (FDA) in the United States [2]. In these regulations, it is emphasised that the raw materials used in food packaging production should not endanger human health, create an unacceptable change in the composition of the food or cause deterioration in organoleptic properties [3,4]. This has been required of the packaging industry in search of new resources, both in substrates and inks, to meet the requirements of food manufacturers and national regulations. It is of great importance that the newly developed packaging materials are inexpensive, sustainable, and environmentally friendly both for human health and for large-volume production [5].

In packaging, paper, cardboard, flexible materials, woods, glass, etc. and other substrates are being used. Of these materials, paper and cardboard packages are the most widely used and have the highest recycling rate [6,7]. Paper and cardboard materials are porous materials with a heterogeneous fibre network with large air voids [8]. For this reason, low molecular weight substances contained in printing inks, varnishes and adhesives that are not in direct contact with the food applied for the purpose of giving visuality to the packaging material may pass into the packaged food and create chemical reactions between the material components [9,10]. These may cause deterioration of packaged foodstuffs and, as a result, risks to consumer health [11,12]. For this reason, it is important that the ink or protective materials applied to the surface are in accordance with the national packaging regulations and are used as specified in the regulations [13,14].

Adding a better commercial appeal to the product stored in the packaging is achieved with the ink transferred onto the packaging material by using printing systems such as offset lithography, flexography, and rotogravure. Flexographic printing is one of the most widely used printing systems in packaging printing [15,16]. In this printing technique, the printing cylinder is covered with printing plates made of rubber or photopolymer material [17]. Ink is transferred onto the print roller by an anilox [18]. This transferred ink is applied to the substrate and the printing process is completed (Figure 1).

Printing inks consist of four main components: pigments or dyes, resins, solvents, and auxiliary substances such as additives [20]. Although the printed media market in publication sector is decreasing day by day, the demand for printed products in other areas of printing such as packaging is increasing. This demand causes the printing ink market to grow by 3% every year [21]. The scarcity of resources used in this growth production, increasing regulatory burden, and increasing environmental awareness are forcing ink manufacturers to improve on producing new sustainable inks [22].

In this study, the usability of modified cellulose-based binders [23], which were developed in the laboratory instead of traditional acrylic binders, were employed in ink formulation. For this purpose, water-based flexographic ink was produced using the modified cellulose-based binder developed in a laboratory environment, and its performance and suitability for packaging printing was evaluated by comparing it with a traditional acrylic binder-based ink and soy-protein ink developed in a laboratory environment [24,25].

2 | METHODOLOGY AND MATERIAL

Initially, SBS boards were conditioned for 24 h at 50% relative humidity and 23°C (73.4°F). Then, the Parker Print Printsurf Porosity was measured using a Parker Printsurf (PPS) tester at 1000 kPa clamping pressure with a soft backing. The thickness of the samples was measured using a TMI micrometer. After that, the air permeability values of the samples [26] were obtained from the PPS porosity. According to TAPPI T555-OM-99, the roughness value of the sample was measured using a PPS ME-90 (1000 kPa, soft backing). Using a Brightimeter Micro S-5, the brightness of the sample was measured according to TAPPI Standard T452-OM-98 (measured at 457 nm wavelength). Last, the sample gloss was measured at 75° using a Novo-Gloss™ Glossmeter based on TAPPI standard T480-OM-99 (Table 1).

A total of four inks were investigated in this study: commercial water-based ink, commercial acrylic resins, commercial soy protein and modified cellulose-based binder such as carboxymethyl cellulose (CMC) with glyoxalated polyacrylamide (GPAM). The ratio for CMC

TABLE 1 Optical and physical properties of a commercial SBS board

Properties	Average	Standard deviation
Brightness (%)	78.7	0.4
CIE L^*	94.5	0.2
CIE a^*	−0.2	0.1
CIE b^*	3.5	0.1
Specular gloss 75°	13.5	0.4
Roughness (μm)	5.9	0.2
PPS porosity (ml/min)	256.6	12.6
Thickness (μm)	353.6	6.1
Permeability (μm^2)	0.0044	2.7×10^{-3}
Tearing resistance (mN)	406.4	18.2
Bursting strength (kPa)	68.8	1.1
Tensile strength (kN/m)	30.1	3.0

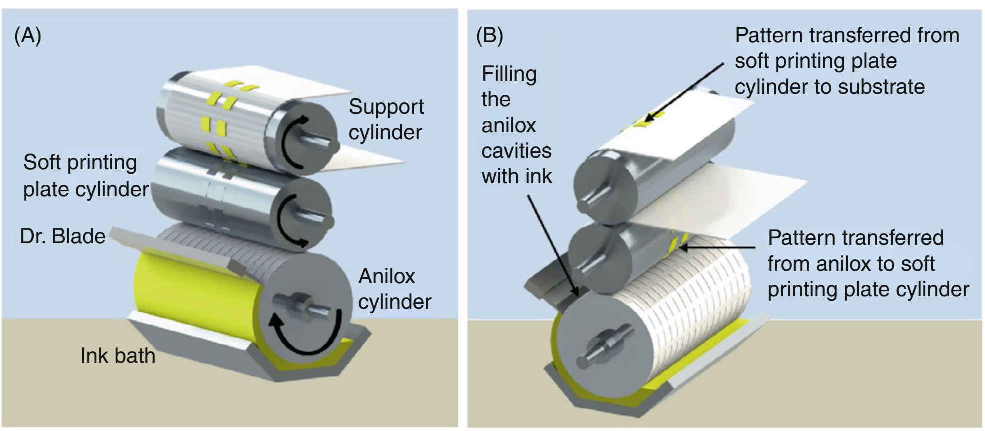

FIGURE 1 Flexography printing process. (A) Position of anilox, soft printing plate, and support cylinders. (B) Transferring of the ink to the substrate [19]

TABLE 2 Pigment dispersion physical and chemical properties

Appearance	Blue liquid
pH	9.0
Solubility in water	Miscible
Density (g/cm^3)	1.11
Viscosity (mPa s) – (centipoise)	20

TABLE 3 ProSoy powders physical and chemical properties

Soybean resins	CSR3
Dry appearance	Off white to tan granular powder
Solution colour	Opaque light brown
Bulk density	672 kg/m^3
Moisture	11.5% maximum
Solution solids	20%
Particle size	7% less than 841 nm

TABLE 4 Soy vehicle formulation

Material	Amount (%)
Water	80
ProSoy	15
Ammonia or amine	0.4 to 1.0
Isopropyl alcohol	4
Biocides[a]	Did not use
Antifoam[a]	Did not use

[a]They are very important for the long shelf life of the ink. Since we were using only a short-term test, we did not use.

to GPAM was 4:1 [23]. A cyan ink pigment dispersion (the colourant) was provided by American Inks & Technology Ltd company under the commercial name "PB15-44". Table 2 gives the physical and chemical properties of the pigment dispersion. Other common ink components, such as isopropyl alcohol, defoamer (FC-613), acrylic varnish, wax, and ammonia (NH_4OH) were also used.

Commercial soybean protein (CSP) was used in this study, provided by ARRO Corporation. Table 3 list its physical and chemical properties.

The preparation of a ProSoy water-based vehicle was done in an air mixer. Formulation of soy vehicle is given in Table 4. The water was heated to the desired cooking temperature, which was between 60 and 76°C. Then, a 5% concentration of ammonia solution was added to ensure that the pH range of the solution was 9.1. Agitation of suspension was done with a vortex mixer. Suspension was cooked for 40 min, and other components of the formulation were added to the protein solution while agitation continued [24] (Table 5).

TABLE 5 A standard water-based flexographic ink formulation

Material	Weight (g)
Pigment dispersion PB-15-44 (blue)	43.5
H_2O (deionised water)	7
Commercial acrylic varnish/soybean protein/ modified cellulose-based binder	48.1
Wax (AIT-PE-35)	1
Defoamer (FC-613)	0.4
Total weight	100

2.1 | Ink formulation

A water-based ink with a commercial acrylic resin (CAR), commercial soybean protein ink (CSP) and modified cellulose-based binder ink (MCB) [23] were prepared following the same formulation (Table 5). In all inks, pH was adjusted with 5% ammonia water to pH 9.1. Viscosity was measured as efflux time—on Zahn cup 2 at controlled temperature 25°C, the efflux time was adjusted to 25 s in all inks. A commercial cyan ink (CI) provided from Wikoff Colour Corporation was used as the primary reference, or the control.

2.2 | Printing conditions

The test ink samples, were printed on the SBS board using a Flexiproof 100 device, which is comparable to a single-colour printing press. A flexible photopolymer printing plate of size 260 mm × 90 mm with thickness value of 1.7 mm was used for printing. The printing speed is set at a constant speed 40 m/min. Pressure between anilox roller and plate cylinder was 45 units, and between plate cylinder and impression cylinder was 50 units. The screen frequency of plate was 39.37 L/cm (100 lpi). The screen frequency of anilox cylinder was 200.6 L/cm (510 lpi), and the capacity of its ink-cells was 5 cm^3/m^2 (μm) [27]. The ink used was a commercial cyan ink. After printing, print density, print contrast, dot gain and CIE $L^*a^*b^*$ values were measured using an X-rite eXact device, using M1 mode. These measurements were carried out using a D_{50} light source under an observation angle of 2°. The unprinted and printed gloss values was measured at 60° using a BYK micro-gloss meter based on TAPPI standard T480-OM-99. After that, delta gloss was calculated using these values [28].

FIGURE 2 Rub resistance testing instrument

Print topography and mottle were measured with Verity IA Print Target version 3 software with stochastic frequency distribution analysis (SFDA) algorithm on the solid patch [29] using the Epson Perfection V500 Photo scanner software at 600 ppi.

The rub resistance of printed test samples was carried out in accordance with BS 3110:1959 standard by Sutherland Ink Rub with a load of 4 lb weight on unprinted SBS/printed SBS and 60 strokes (Figure 2).

In the study, the abbreviation BP was used for a commercial SBS board, the abbreviation CI was used for a commercial ink, the abbreviation CARI was used for ink using a commercial acrylic resin, the abbreviation CSPI was used for ink using a commercial soybean protein and the abbreviation MCBI was used for ink using the modified cellulose-based binder.

3 | RESULTS AND DISCUSSION

3.1 | Print density

The print density was determined by calculating the amount of light reflected from the substrate and the ink printed on it by a densitometer. The measurement was carried out over the solid area [30]. The print density value obtained is important in terms of stabilising the ink film thickness applied during printing [31]. As the ink film thickness on the surface increases, less light will be reflected, and higher density values will be obtained. In other words, high-density prints will be achieved [32]. The decrease in print density after a certain number of prints will adversely affect the overall print quality of the image. For this reason, the printing density should be kept under control with the controls made at certain intervals during the printing process. When evaluating the print density, the porosity of the substrate, surface smoothness and surface tension should be considered [33,34]. On a highly porous and very rough surface, since the ink will flow and settle more in the pores, the print density will be decreased [35]. Another factor to consider is the ink selection. The correct choice of solvent, binder, pigment, and dyestuff that make up the ink is of great importance on the printing density [36]. The fact that the surface tension of the ink to be used in printing should be lower than the surface energy of the substrate will ensure a high printing density [37]. The print density was measured by an X-rite eXact device, using M1 mode in the cyan channel, as the commercial ink was labelled as a cyan ink.

Table 6 shows the change in print density values before and after rub resistance test (RT). Compared to CI, CARI has the highest compression density and CSPI the lowest. After the Rub test, while the compression density values of CI, and CARI were essentially unchanged, the density values of CSPI, and MCBI decreased.

The rub resistance index (RI) of inks were calculated using Equation (1).

$$\text{Rub resistance index(RI)} = \text{Density after rub}/\text{Density before rub} \times 100 \qquad (1)$$

3.2 | Print contrast

Print contrast has an important role for clear colours and easy-to-read text of any size in flexographic printing. Increasing print contrast is one of the most important variables in improving flexographic printing performance [38]. Print contrast is obtained by comparing solid ink density to 75% dot density [39]. It will result in better print contrast, improved tonal range, less mottle, brighter colours, and even sharper text.

The print contrast values of inks were calculated using Equation (2).

$$\text{Print contrast}(\%) = (D(s) - D(t))/(D(s)) \times 100 \qquad (2)$$

where $D(s)$ is the density of solid and $D(t)$ is the density of tint, typically 75%.

The print contrast values of the test samples measured at 75% screen density are given in Table 7. When the obtained values were compared with CI, the lowest compression contrast was obtained in CARI and CSPI. The print contrast of MCBI is slightly closer to CI. After RT, the print contrast of CI, CARI and CSPI was significantly reduced, while the loss of MCBI print contrast was lower.

TABLE 6 Print density of test samples

Inks	Print density before rub resistance test (RT)	Standard deviation	Print density after RT	Standard deviation	Percentage retained after RT
CI	1.28	0.01	1.27	0.01	99.2
CARI	1.27	0.01	1.26	0.01	99.2
CSPI	1.17	0.01	1.14	0.01	97.4
MCBI	1.21	0.01	1.18	0.02	97.5

TABLE 7 Print contrast of test samples

Inks	Print contrast before rub resistance test (RT)	Standard deviation	Print contrast after RT	Standard deviation
CI	9.65	0.49	5.90	0.49
CARI	4.74	0.03	3.59	0.58
CSPI	4.29	0.03	3.09	0.64
MCBI	7.45	0.95	6.84	0.76

TABLE 8 Specular gloss of test samples

Inks	Gloss before rub resistance test (RT)	Standard deviation	Gloss after RT	Standard deviation
BP	3.87	0.06	6.28	0.17
CI	3.92	0.18	4.06	0.15
CARI	3.57	0.12	3.90	0.14
CSPI	3.53	0.06	3.85	0.07
MCBI	2.04	0.13	2.96	0.18

3.3 | Specular gloss

The term gloss is a term used to express the degree of gloss of both an unprinted and a printed surface. Achieving a glossy print requires premature absorption of the ink to maximise its permanence. Otherwise, drying by evaporation will reduce the gloss of the ink [40].

Table 8 shows the change in print gloss values of BP and printed samples before and after RT. The data obtained show that the gloss value of all inks laboratory produced decreased after printing, and this decrease is most obvious in MCBI. After the rub test, it is seen that there is an increase in the gloss values of all inks due to friction, and this increase is also most obvious in MCBI.

3.4 | The CIE $L^*a^*b^*$ colour values and ΔE

The ΔE is a standard measurement that quantifies the difference between two colours that appear in additive colour on a display or subtractive colour on a natural, printed or painted surface. It was created by the Commission Internationale de l'Eclairage (CIE) (International Commission on Illumination) in 1971. The symbol ΔE_{76} has been used for this simple colour difference. Multiple improvements of this occurred, the most recent of these is denoted by ΔE_{00} [41], given in Equation (3). The obtained ΔE_{00} values give the difference between a displayed or printed colour and the original colour [42]. A ΔE_{00} value that is low indicates that the difference between the colours is low, and if it is too high indicates a large difference between the colours [43]. The ΔE is measured on a scale from 0 to above 100, the actual upper limit being determined by the reference illuminant. Here, values of $0 < 1$ or less represent colour difference that cannot be perceived by the human eye, values between 1 and 2 represent colour differences that can be perceived by close observation, between 2 and 10 represent perceptible colour difference at a glance, between 11 and 49 indicate that the colours have significantly different colour values, and > 100 indicates that the colours are nearly opposite to one another.

The CIE $L^*a^*b^*$ colour values (Figure 3) obtained before and after RI are given in Table 8. Keeping constant the CIE $L^*a^*b^*$ colour values of CI, the colour difference value (ΔE_{00}) of other inks were calculated using Equation (3).

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{K_L S_L}\right)^2 + \left(\frac{\Delta C'}{K_C S_C}\right)^2 + \left(\frac{\Delta H'}{K_H S_H}\right)^2 + R_T\left(\frac{\Delta C'}{K_C S_C}\right)\left(\frac{\Delta H'}{K_H S_H}\right)} \quad (3)$$

where R_T is a hue rotation term; K_L, K_C, and K_H are the parametric factors; L^*, C^* and H are the compensation for neutral colours; S_L is the compensation for lightness; S_C is the compensation for chroma; S_H is the compensation for hue.

Table 9 shows that before RT CARI's ΔE_{00} value is 0.4 and CI has the closest colour value. The MCBI's ΔE_{00} value for was 1.7, providing colour values slightly closer to CI than CSPI. The relatively small value of the difference between the formulated inks does not mean they are wrong, but the difference can be easily compensated in a colour managed workflow [45,46].

After RT there was a small increase in the ΔE_{00} value of all colours. Compared to other inks, the higher increase in the ΔE_{00} value of CSPI makes it further away from the CI colour value. When the L^* values are examined, the higher L^* value of CSPI does not explain the darkening in the colour.

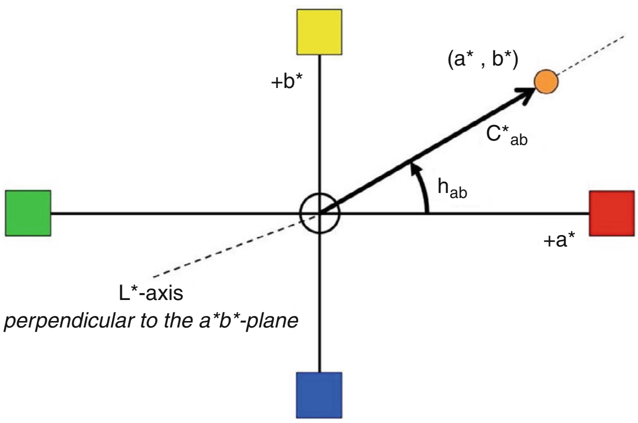

FIGURE 3 The CIE $L^*a^*b^*$ colour [44]

3.5 | Tone value increases (TVIs)

In multicolor prints, the tonal transitions of the colours are obtained with the screen dots. It is observed that the tone value obtained during proofing has increased slightly during printing [47]. This increase is due to the growth (increase in size) of the screen dot, which enables higher tone values to be obtained [48]. This increase is called tone value increase (TVI) in the printing industry. If the TVI is within certain limits, the expected colour values in the current proof can be kept within an acceptable range. Otherwise, it will cause unwanted chromatic aberration [49,50].

In Figures 4 and 5, the TVI values obtained as a result of the printing of the inks before and after RT can be seen. At 10% and 60% tone values, the lowest point gains are seen in MCBI and the closest TVI value to these values is seen in CARI. TVI values obtained in CI are higher than MCBI and CARI. TVI values closest to CI were obtained in CSPI. TVI values increased in all ink types after RT. This increase is a result of scattering in screen points caused by friction.

3.6 | Dot roundness

Roundness close to one gives an ideal dot shape. This value is an indication that the ink is spread evenly and provides a quality print. If the resulting value is 1.0, it means that the dot is a perfect circle, if it is less than 1.0, it means that the dot is not perfectly round and there are losses at the dot [51]. The points obtained without loss will ensure that the image has more precise lines, and will provide prints with higher definition and higher image quality [52].

The dot roundness results for 15% cyan dots are shown in Table 10. The dot roundness of CI, CSPI and MCBI are about the same, while CARI is slightly larger,

TABLE 9 The CIE $L^*a^*b^*$ colour values of test samples

	CIE $L^*a^*b^*$ before rub resistance test (RT)				CIE $L^*a^*b^*$ after RT			
Inks	L^*	a^*	b^*	$\Delta E_{00\ Print}$	L^*	a^*	b^*	$\Delta E_{00\ Print}$
CI	42.8	−12.5	−46.2	—	41.4	−10.8	−45.8	—
CARI	42.5	−12.8	−46.0	0.4	42.5	−12.2	−44.8	0.6
CSPI	44.5	−11.7	−43.6	1.8	44.8	−12.2	−43.5	2.3
MCBI	43.4	−11.0	−43.7	1.7	43.4	−11.3	−43.7	2.0

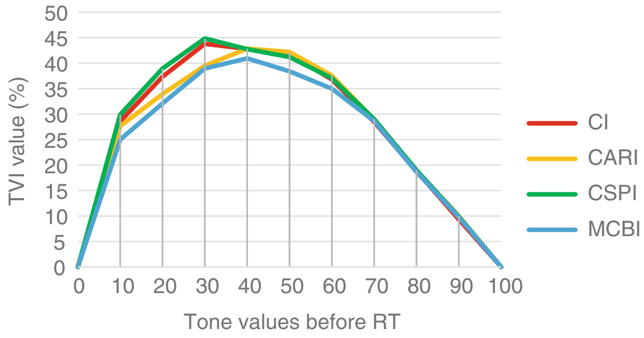

FIGURE 4 Tone increases values before rub resistance test (RT)

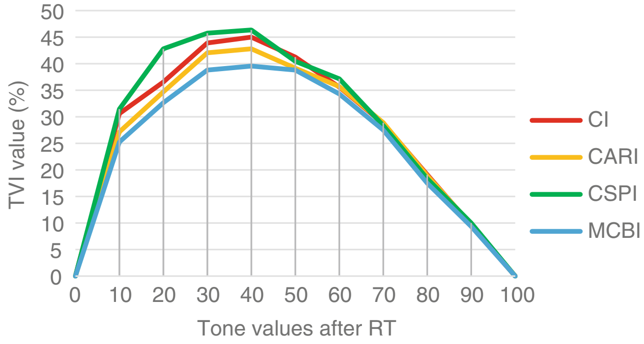

FIGURE 5 Tone increases values after rub resistance test (RT)

TABLE 10 Dot measurements of 15% screen dots

Inks	Dot roundness before rub resistance test (RT)		Dot roundness after RT	
	Average	Standard deviation	Average	Standard deviation
CI	0.95	0.14	0.96	0.13
CARI	0.96	0.11	0.97	0.11
CSPI	0.95	0.16	0.97	0.14
MCBI	0.95	0.15	0.96	0.16

although not significantly. In reality, the roundness values are about the same within the large variation reported.

3.7 | Print topography and mottle

If the ink transferred to the printing surface does not distribute evenly on the surface, it will cause different ink absorptions on the surface. As a result of this absorption, different amounts of ink/colourant will stay on the substrate surface. Density distributions that occur irregularly on the substrate material are called compression mottle [42]. Therefore, the surface properties of the substrate, the printing system and the properties of the ink used have a significant effect on the print mottle. In an ink produced with a lower viscosity (20 s), the ink will spread faster, causing unequal ink accumulations on the substrate surface, resulting in an increase in the print mottle value. Since the ink is of high viscosity (24 s) and the ink will be distributed evenly on the substrate, lower print mottle will be obtained. Thus, since the circularity of the dots obtained in printing will be better, sharper images can be obtained. In inks produced with viscosity values higher than 24 s, the print mottle value to be obtained will increase as irregular ink distributions will occur on the substrate due to the anilox cells clogging during printing [53].

Table 11 gives the print topography and mottle values before and after the rub test. When the print mottle values are compared, it is seen in Table 11 that CI has less increase in pressure mottle, while the next closest pressure mottle change value is obtained for MCBI and this value increases depending on friction in each ink type after the rub test. It has also increased the print topography of all ink types after printing. This increase was seen highest for CI, followed by CSPI, CAARI and MCBI.

In Figure 6, images obtained using the VERITY IA Target software are given. In this figure, cyan shows

TABLE 11 Print topography and mottle properties of test samples

Test samples	Print topography		Print mottle			Resolution
	Before RT	After RT	Before RT	After RT	Area of interest (cm^2)	(ppi)
CI	5.30	6.30	1.25	1.35	13.25	600
CARI	12.40	13.10	3.65	4.00	13.25	600
CSPI	21.30	22.05	4.60	5.05	13.25	600
MCBI	10.70	11.00	2.95	3.15	13.25	600

Abbreviation: RT, rub resistance test.

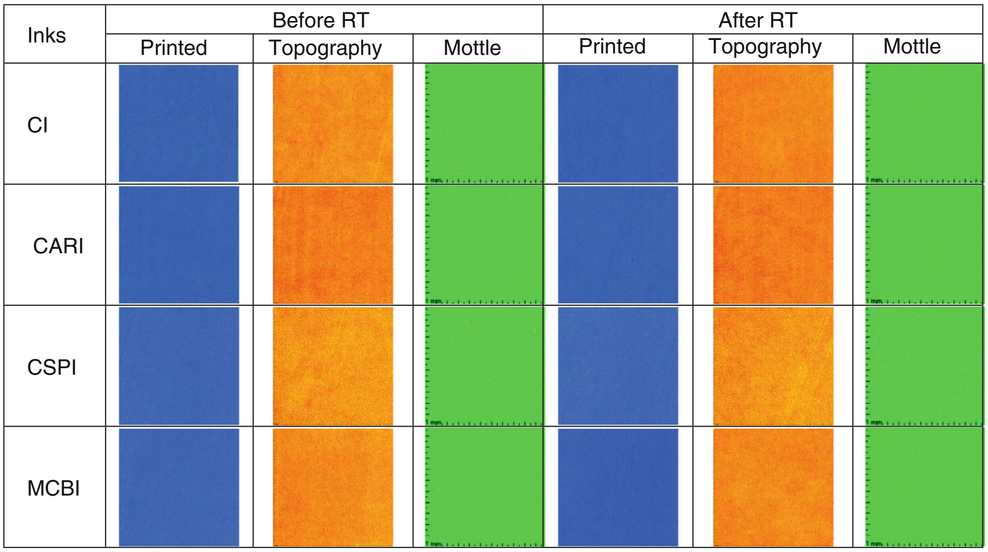

FIGURE 6 Print topography and mottle images of test samples

printed image, orange shows print topography, and green shows print mottle.

4 | CONCLUSION

Print quality values were reported for four water based cyan flexographic inks. The values are all similar to one another. The important thing is that the sustainable inks CSPI and MCBI provide acceptable substitutes for the petroleum based acrylic inks CI and CARI. The MCBI ink shows slightly better performance than the CSPI, but both are more than acceptable, especially since they are made from renewable resources. Going forward in the twenty-first century this is important since petroleum-based products come from depletable resources and many of these produce greenhouse gases that contribute to climate change.

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