

# A SURVEY ON THE EFFECTS OF ENVIRONMENTALLY FRIENDLY SOY PROTEIN INKS ON FLEXOGRAPHY PRINT PARAMETERS IN THE PACKAGING INDUSTRY

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The synthetic resins in printing inks are harmful to the environment because they are not biodegradable and emit volatile organic compounds. The printing industry has sought alternative printing inks to solve these issues. The aim of the work is to explore sustainable and environmentally friendly inks, without scarifying ink performance. To determine their performance and effect on print quality, soy proteins were investigated. Also, commercial and formulated acrylic inks were used as reference inks. From the results, the print density of soy protein water-based inks is higher than that of the reference inks. Moreover, they obtained a higher print gloss than the reference inks. The printing contrast values of soy protein water-based inks are relatively low. There was no considerable difference found in TVI values. Overall, the soy protein water-based inks produced a very competitive result in printability and presented high potential for replace synthetic components in current commercial inks.

**Keywords:** soybean, soy protein, resin, acrylic vehicle, ink, water based, flexographic, packaging

## INTRODUCTION

Printing inks are composed of resins, solvents and colorants. The widely applied inks for conventional printing, such as lithographic offset printing, gravure and flexography, are considered not environmentally friendly. For example, most applied acrylic resins are made from fossil oils, they provide strong bond strength for printing, however, they are not biodegradable by current criteria. The researchers have investigated biodegradable and sustainable resin alternatives to replace the acrylic resins.<sup>1</sup> The solvent in the inks, a carrier that provides the desirable fluid properties for printing processes, emits volatile organic compounds (VOCs) into the environment, which is considered as a safety hazard, being also harmful to our environment. Indeed, reducing the VOC emissions has been a topic of research

interest lately.<sup>2-4</sup> In order to reduce the VOC emissions, water-based inks have been widely used in both conventional and digital printing technology, rotogravure, flexography, screen printing and digital printing processes. Water-based inks have achieved comparable print quality to that of solvent-based inks. Today, water-based inks contain 90% to 95% less VOCs than solvent-based inks.<sup>5-6</sup> The ingredients of most commercial water-based inks include synthetic colorant, acrylic resin, water and additives. Acrylic resin-based inks yield better color reproduction and brightness – the main reason for which they are widely applied in the printing industry.<sup>7-8</sup> However, the acrylic resin is neither sustainable, nor biodegradable. The

industry needs to find an alternative to replace the acrylic resin in the future.

Soy is a major crop and its production is sustainable. A resin made of soybean oil is a feasible alternative to replace acrylic resin in water-based inks.<sup>9</sup> Indeed, soybean oil-based inks have been successfully used in lithographic offset printing and letterpress printing inks.<sup>10</sup>

Soybeans contain about 35% protein, 19% fat, 17% dietary fiber, 11% carbohydrates, 13% water and 5% other. Soybeans contain three natural surfactants, which are soy protein, soy lecithin and soy saponin. Soy protein is a by-product, obtained through the extraction of soybean oil (Fig. 1).<sup>11</sup> The soy protein concentrate contains 65-72% protein, which is obtained by removing the aqueous liquid portion of soybean. The soy protein isolate, the most refined form of soy protein, contains 90% protein and is obtained from defatted soy flour by removing the carbohydrates from the beans.<sup>12-13</sup> Soy protein is often seen in a variety of foods, such as salad dressings, frozen desserts, breads, and breakfast cereals.<sup>14</sup> Other industrial products containing soy protein include resins, inks, paints, adhesives, plastics, polyesters, cleaning materials, asphalt additives, cosmetics, and textile fibers. Industrial soy protein, which is preferred in the paper industry, is also widely used as a natural binder, especially in paper pigment coatings.<sup>15</sup>

There are three basic forms of soy protein: soy powder, soy protein concentrates and soy protein

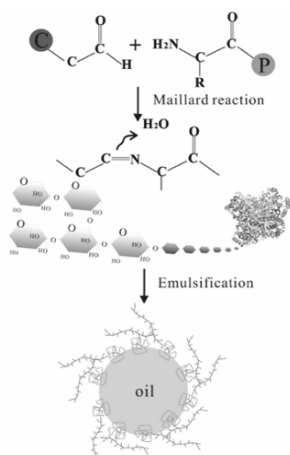


Figure 1: Formation of the soy protein-polysaccharide complex for emulsion stabilization<sup>11</sup>

## EXPERIMENTAL

### Materials and methods

In this study, a commercial solid bleached sulfate (SBS) board was used as a substrate for printing tests.

isolates. Proteins formed as a result of the condensation reaction of amino acid monomers form peptide bonds.<sup>11,16</sup> Water molecules emerge as a result of condensation between amino acids (Fig. 2).<sup>17</sup> Soy protein contains 19 different amino acids in a helical structure with peptide bonds and has a complex 3D shape. Proteins consist of positive and negative functional groups. It forms functional groups of soy protein from amino, carboxyl, hydroxyl, phenyl and sulfhydryl.<sup>18-19</sup>

Acrylic emulsion polymers and their various copolymers are widely used in water-based ink formulations. Water-based flexographic inks are formulated with a variety of acrylic polymers and copolymers that serve as emulsion resins to disperse pigments using deionized ultra-filtered water. It forms ink films when ink is curing (drying) and provides bond strength to hold colorant pigment onto the substrates. It does impact ink properties, such as rheology, adhesion or friction resistance.<sup>21</sup>

Flexography is a major printing method for packaging materials. The goal of this study has been to develop an environmentally friendly, sustainable water-based flexographic ink containing a resin made of soybean. A comprehensive evaluation has been conducted to investigate the performance of three soybean-based flexographic inks, compared to a commercial ink (the control) and a lab prepared water-based ink formula (second reference), both containing acrylic resin.

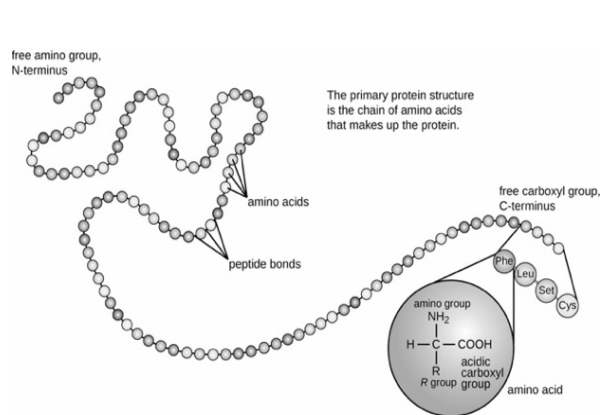


Figure 2: Primary protein structure<sup>20</sup>

The substrate was conditioned for 24 h at 50% relative humidity and 23 °C (73.4 °F), and tested for PPS porosity, thickness, roughness, gloss, CIE whiteness and brightness. Porosity was measured using a Parker

Print-Surf (PPS) tester at 1000 kPa clamping pressure (CP) with a soft backing and used to calculate air permeability.<sup>22</sup> The thickness of the SBS board was measured using a TMI Micrometer. Its roughness was measured using a PPS ME-90 (1000kPa, soft backing) based on TAPPI T555-OM-99. The brightness of the SBS board was measured with a Technidyne Brightimeter Micro S-5 based on TAPPI Standard T452-OM-98 (457 nm light). The SBS board gloss was measured at 75° using a Novo-Gloss™ Glossmeter based on TAPPI standard T480-OM-99. The optical and physical properties of this substrate are given Table 1.

A total of five inks were investigated in this study: three of them containing soy protein and the other two

using acrylic resin. Three different commercial soybean resins (CSR) were used in this study, provided by ARRO Corporation, under the brand of ProSoy, and named as CSR1, CSR2 and CSR3, respectively. Table 2 lists their physical and chemical properties.

A cyan ink pigment dispersion (the colorant) was provided by American Inks & Technology Ltd. Company, under the commercial name “PB15-44”. Table 3 gives the physical and chemical properties of the pigment dispersions. Other common ink components, such as isopropyl alcohol, defoamer (FC-613), acrylic varnish, wax and ammonia (NH<sub>4</sub>OH), were also used.

Table 1  
Optical and physical properties of a commercial SBS board

Properties	Average	Std. dev.
Brightness (%)	78.67	0.40
CIE L*	94.53	0.18
CIE a*	-0.21	0.05
CIE b*	3.52	0.10
Specular gloss 75°	23.30	0.40
Roughness (µm)	5.92	0.23
PPS Porosity (ml/min)	256.60	12.63
Thickness (µm)	353.60	6.10
Permeability (µm <sup>2</sup> )	0.00443	2.73x10 <sup>-3</sup>
Tearing resistance (mN)	406.40	18.24
Bursting strength (kPa)	68.80	1.10
Tensile strength (kN/m)	30.10	3.28

Table 2  
Physical and chemical properties of ProSoy powders

Soybean resins	CSR1	CSR2	CSR3
Dry appearance	Dark brown granular	Light brown granular	Off white to tan granular powder
Solution color	Brown	Opaque light brown	Opaque light brown
Bulk density	672 kg/m <sup>3</sup>	672 kg/m <sup>3</sup>	672 kg/m <sup>3</sup>
Moisture	11.5% Maximum	11.5% Maximum	11.5% Maximum
Solution solids	20%	20%	20%
Particle size	7% less than 841 nm	7% less than 841 nm	7% less than 841 nm

Table 3  
Physical and chemical properties of pigment dispersion

Appearance	“Blue” liquid
pH	9
Solubility in water	Miscible
Mass density (g/cm <sup>3</sup> )	1.11
Viscosity (mPa-s) - (centipoise)	20

### Three water-based inks with soy protein

The preparation of a ProSoy water-based vehicle was done in an air mixer. The formulation of the soy vehicle is given in Table 4. The water was heated to the desired cooking temperature, which was on average

60 to 76 °C. Then, a 5% concentration of ammonia solution was added to ensure that the pH range of the solution was ~9.1. During agitation with a vortex mixer, the solution was cooked for 40 minutes. Other components of the formulation were added to the

protein solution while agitation was continued (Table 5).<sup>23</sup>

Three water-based inks with soybean resin and a water-based ink with a commercial acrylic resin were prepared, with a standard formulation given in Table 5. Soybean resin inks were named as CSRI1, CSRI2 and CSRI3, respectively, according to the resins. Acrylic ink was named AI. The pH was adjusted with 5%

ammonia water to pH 9.1. Viscosity was measured as efflux time – on a Zahn cup 2 at a controlled temperature of 250 °C, the efflux time was read as 25 seconds. A commercial water-based cyan ink (CI), provided from Wikoff Color Corporation, was applied as the primary reference, or the control.

Table 4  
Soy vehicle formulation

Material	Amount (%)
Water	80
ProSoy	15
Ammonia or amine	0.4 to 1.0
Isopropyl alcohol	4
Biocides	As needed
Antifoam	As needed

Table 5  
Water-based flexographic ink formulation

Material	Weight (g)
Pigment dispersion PB-15-44 (cyan)	43.5
H <sub>2</sub> O (DI water)	7
Soy protein/commercial acrylic varnish	48.1
Wax (AIT-PE-35)	1
Defoamer (FC-613)	0.4
Total weight	100

### Printing conditions

The test samples, the SBS board, were printed using a Flex Proof 100 device, which is comparable to a single-color printing press. A flexible photopolymer printing plate of size 260 x 90 mm, with a thickness value of 1.7 mm, was used for printing. The printing speed was set at a constant speed of 40 m/min. The pressure between the anilox roller and the plate cylinder was 45 units, while between the plate cylinder and the impression cylinder – it was 50 units. The screen frequency of the plate was 39.37 l/cm (100 lpi). The screen frequency of the anilox cylinder was 200.6 l/cm (510 lpi), and the capacity of its ink-cells was 5 cm<sup>3</sup>/m<sup>2</sup> (μm).<sup>24</sup> The ink used was a CI, AI and CSR inks. After printing, print density, print contrast, dot gain and CIE L\*a\*b\* values were measured using an X-rite eXact device, using the M1 mode. These measurements were carried out using a D/50 light source under an observation angle of 2°. The gloss of the unprinted and printed SBS board was measured at 60° using a BYK-Gardner Glossmeter based on ISO 2813 and at 75° using a Novo-Gloss™ Glossmeter based on TAPPI standard T480-OM-99. Delta gloss was calculated. Dot quality was determined using Paxit software.

## RESULTS AND DISCUSSION

### Print density

Print density refers to the ink color density value, a high value that indicates a saturated color. A printer often controls the color density to monitor the ink consumption.<sup>25-26</sup> Reflection density was measured on the solid ink area in the C channel and was calculated according to Equation (1):

$$\text{Print density} = \log_{10} \frac{1}{R} \quad (1)$$

where R = reflectance.

As we shall see later (Table 8), these prints differ from standard cyan, so the C channel is not strictly correct. A better estimate of the density of prints obtained here can be obtained by the Shendye-Fleming<sup>27</sup> density method, which involves measurements of reflection spectra. Figure 3 shows print density values of five flexographic inks. The print density values of the formulated CSR inks were apparently lower than those of CI and AI. The print density value of CSRI3 was slightly higher than those of CSRI1 and CSRI2. The lowest density was obtained for CSRI1. AI inks had nearly the same print density as the commercial ink, but slightly higher than those of CSRI3 inks. CSRI3 can reduce ink consumption for the same print density. Since ink represents nearly 20% of the overall material cost

for a printing plant, this soy protein water-based ink can reduce the operating cost.

### Print contrast

The ink density between the solid area (100% ink coverage) and the tint area (typically, 75% ink coverage) is called print contrast. A high-contrast print provides much detailed information in shadow areas, an important parameter for a high-quality print.<sup>28</sup> It is calculated according to Equation (2):

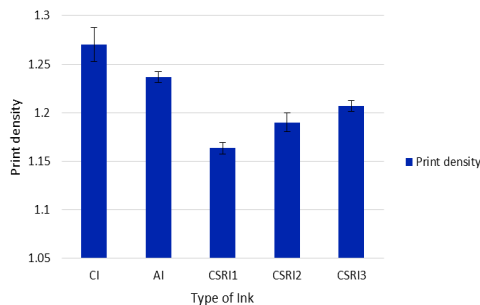


Figure 3: Apparent cyan print density values of formulated inks and commercial ink

### Tone value increase

The print image is composed of half-tone dots. There is an increase in the size of halftone dots before and during the printing process. This increase is called the tone value increase (TVI, dot gain). The TVI at mid-tone and the solid area is critical to achieve a good print, since a large TVI will eliminate the image details in dark areas. In order to obtain a good print quality, it is desirable that the TVI be a low value and the shape of the dots obtained should be a smooth round shape.<sup>29</sup> In flexographic printing, the TVI is affected by the structural properties of the anilox cylinder, the plate, ink properties and amount. In addition, the surface properties of the substrate used also have an effect on the TVI. Since TVI cannot be avoided, a small TVI, especially in the mid-range of tone, is desired.

The TVI of these five inks were evaluated based on Equations (3) and (4):

The Murray/Davies Dot Area Equation:

$$\text{Apparent dot area} = \frac{1 - 10^{-(D_t - D_p)}}{1 - 10^{-(D_s - D_p)}} \times 100 \quad (3)$$

where  $D_t$  = density of tint;  $D_s$  = density of solid;  $D_p$  = density of the paper/substrate;

The Yule-Nielson Dot Area Equation:

$$\text{Apparent dot area} = \frac{1 - 10^{-(D_t - D_p)/n}}{1 - 10^{-(D_s - D_p)/n}} \times 100 \quad (4)$$

$$\text{Print contrast (\%)} = \frac{D_s - D_t}{D_s} \times 100 \quad (2)$$

where  $D_s$  = density of solid,  $D_t$  = density of tint (typically 75%).

Figure 4 shows clearly that the CSRI3 had the highest print contrast values. The high print contrast revealed by the CSRI3 formulation shows that details in shadow areas are positively affected. CSRI1 had lower print contrast values. Meanwhile, AI ink had the same print contrast as CI, following after CSRI3, with a slightly lower value.

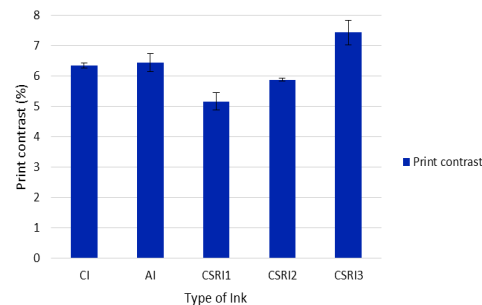


Figure 4: Print contrast values of formulated inks and commercial ink

where  $D_t$  = density of tint;  $D_s$  = density of solid;  $D_p$  = density of the paper/substrate;  $n$  = an empirically determined factor. The Murray/Davies equation is used to estimate the TVI on a physical point area. An empirically determined “ $n$ ” factor of the Yule-Nielson equation is included if these factors have an effect in the printing process, but we have a known “ $n$ ” value.

The TVI values in Figure 5 reveal the effect of ink difference on dot gains. The CI, AI and CSRI2 inks had similar TVI in the highlight area, or 10-20% screen dots, the other two inks, CSRI1 and CSRI3, had a relatively high TVI in the same dot range. Comparing the TVI in the mid-tone range, CSRI1 and CSRI3 obtained a higher value than the rest. It indicated that these two soy protein water-based inks did not have a good performance in TVI control. However, CSRI1 and CSRI3 yielded a high TVI at mid-tone, especially at 40% tone. A possible reason is that CSRI1 and CSRI3 might have a relatively low viscosity, which is hard to be detected by the Zahn cup measurement. The TVI in the dark areas showed no significant difference among the five inks.

Images of 15% screen dots were taken in Paxit (Table 6) to measure the half-tone dot roundness. Dot roundness and dot gain are the two key

parameters to have a good print, or preserve the image details in the middle and dark areas. A value of 1.00 indicates that the dot has an ideal roundness. Table 7 shows the dot roundness values for each ink. It was observed that the

values of the five inks were close to each other and their roundness was within the tolerance limits. Table 7 also shows that the CSR type does not have an effect on the dot area and roundness values.

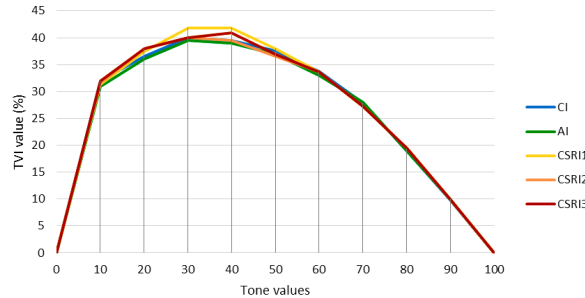


Figure 5: TVI values of formulated inks and commercial ink

Table 6  
Images of 15% screen dots

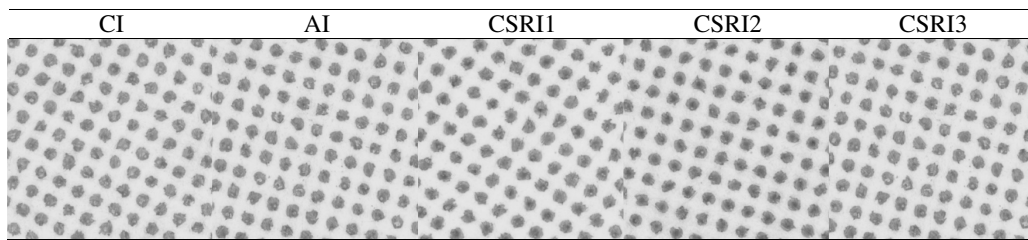


Table 7  
Dot measurements of 15% screen dots

Type of ink	Dot area (mm <sup>2</sup> )		Aspect ratio		Dot roundness (%)		Equivalent diameter (µm)	
	Ave.	Std. Dev.	Ave.	Std. Dev.	Ave.	Std. Dev.	Ave.	Std. Dev.
CI	0.023	0.004	0.99	0.13	83.00	6.67	170.39	22.79
AI	0.022	0.007	0.94	0.21	83.00	9.65	167.99	45.72
CSRI1	0.023	0.004	1.00	0.10	81.50	8.88	169.90	26.96
CSRI2	0.021	0.001	1.01	0.09	84.00	6.20	162.99	15.17
CSRI3	0.025	0.003	0.94	0.15	80.00	8.07	178.49	17.69

**Delta gloss**

Paper gloss was measured using a gloss meter, which compares the amount of light reflected from a paper surface from a light source. When light hits the surface of a paper, the direction of the reflected light rays determines the gloss of the paper.<sup>30</sup> Paper gloss depends on the paper surface, which is directly related to the paper coating and calendering process. The paperboard used in this study is a glossy sheet. Similarly, the ink gloss refers to the degree of gloss of a printed ink. The ink drying/curing method affects its print gloss. The faster the ink-holdout to the paper surface,

the higher the gloss. In some printing jobs, glossy papers are preferred to maximize ink brightness.<sup>31</sup>

Δ gloss measures the gloss difference between the print substrate and printing ink at the same measurement degree (60° is widely adopted in print industry, while 75° is used in the paper industry), and its unit is GU.<sup>32</sup> Following R. Xu *et al.*,<sup>33</sup> we measured the substrate and the print at both angles. These values unveil how the gloss changes after printing. Thus, Δ gloss was calculated according to Equations (5) and (6):

$$\Delta gloss 60^\circ = D_{IG} - D_{PG} \tag{5}$$

$$\Delta gloss 75^\circ = D_{PG} - D_{IG} \tag{6}$$

where D<sub>PG</sub> = gloss of paper; D<sub>IG</sub> = gloss of ink.

In this study, all  $\Delta$  gloss values are negative at 60° and positive at 75° because of the sign change in Equation (6), so that the inks reduced the substrate gloss. The CI performed slightly better than the other four inks, by roughly 0.5 GU. The AI and three CSR inks had the same delta gloss. The variance of all three CSR resins was negligible (Fig. 6) in this measurement.

### Print chroma

Similarly, the chromaticity of the area, which is considered as the ratio of the brightness of an illuminated area, is called “chroma”. The chroma value gives information about how colorful, clean, neutral, strong, weak or muddy a color looks.<sup>34</sup>

Metric Chroma  $C^*$  was calculated using Equation (7):

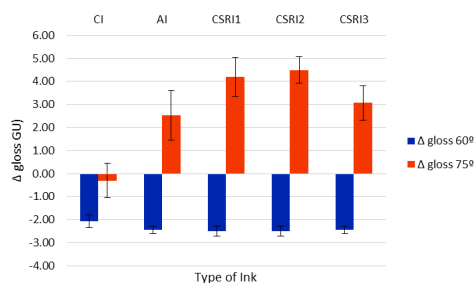


Figure 6:  $\Delta$  gloss 60° and 75° values of formulated inks and commercial ink

$$\text{Print chroma} = \sqrt{(a^*)^2 + (b^*)^2} \quad (7)$$

where  $a^*$ ,  $b^*$  are chromaticity coordinates in the  $L^*a^*b^*$  color space.

The print chroma of CI and AI ink is higher than that of CSR inks. The print chroma of CSRI3 is slightly higher than those of other CSR inks. CSRI1 and CSRI2 have close chroma values (Fig. 7). Note these chroma values are lower than the cyan standard, because  $a^*$  is too large (*i.e.*, not negative enough).

For the blue colorant used here, the actual chroma should correlate with Shendye-Fleming's<sup>27</sup> density function. The densities of the 100% apparent cyan inks studied here should be comparable to the “Bronze Blue” reported by Shendye – about 1.6.

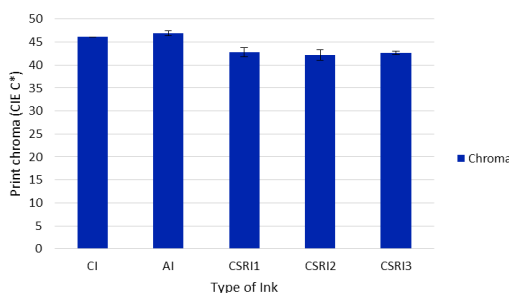


Figure 7: Print chroma values of formulated inks and commercial ink

Table 8  
CIE  $L^*a^*b^*$  values of formulated inks and commercial ink

Inks	CIE $L^*a^*b^*$			
	$L^*$	$a^*$	$b^*$	$\Delta E_{00 \text{ Print}}$
CI	41.45	-10.30	-44.96	-
AI	43.41	-12.95	-45.11	2.38
CSRI1	41.50	-7.26	-42.13	2.32
CSRI2	39.50	-5.21	-41.84	4.10
CSRI3	39.72	-6.31	-42.21	3.30

### Color differences ( $\Delta E_{00}$ )

The color difference, or  $\Delta E_{00}$ <sup>35-36</sup>, is a well-accepted color comparison that eliminates the influences from light sources and devices. The color measurement device converts the sample colors into an absolute color coordinate, and then compares with each other under the same conditions. It provides an effective and consistent color evaluation, regardless of any applied devices or substrates.

In the absolute color coordinate system, the  $L^*$  value indicates the lightness of the color, ranging

from 0 to 100. A positive  $a^*$  value indicates that the color shifts to red, the color shifts to green if  $a^*$  becomes negative. Also, a positive  $b^*$  value indicates that a color shifts to yellow, if it is negative, the color moves to blue.  $C^*$  shows the color intensity and  $H^*$  shows the difference between tonal angles.<sup>37</sup>

The measured CIE  $L^*a^*b^*$  values are given in Table 8. We see that the values are similar to one another, but are far from the standard cyan ink values. The ISO 12647 cyan  $L^*a^*b^*$  values are close to 55, -35 and 50.<sup>38</sup> The measured  $L^*$  and  $b^*$

values are not drastically different, but the  $a^*$  value is not negative enough. Because of this, these inks will produce a very weak green when overprinted with yellow. The insufficient  $a^*$  value is likely because of some degradation  $\beta$ -copper phthalocyanine blue to  $\alpha$ -copper phthalocyanine blue,<sup>39</sup> which shifts the  $a^*$  value towards red.

Color differences were calculated using  $\Delta E_{00}$ . If  $\Delta E$  between two colors is less than 1, it indicates that these colors are the same in the absolute color coordinate, or their difference cannot be noticed to the naked human eye. In fact, any  $\Delta E$  difference less than 3 is hard to distinguish by most observers. Hence, a  $\Delta E$  difference of 2–3 is a goal for the printing industry to manage color reproduction. Any  $\Delta E$  values beyond 3 indicate that the reproduced color does not match the original color value.<sup>40</sup>

Since the CI ink is the control, all the other four inks have been measured and then compared with it. The  $\Delta E_{00}$  values between all four formulated inks and the control were less than or approximately equal to 3, meaning these four formulated inks, including the three soybean-based inks, produced the same color as the control in accordance with the print industry standard. According to these calculations, the  $\Delta E_{00}$  value of AI ink was 2.38, that of CSRI1 ink was 2.32, that of CSRI2 ink – 4.10, and that of CSR3 ink – 3.30. Three of the four formulated inks performed well in this evaluation, while the  $\Delta E_{00}$  of CSR3 and the control are at the boundary, but still acceptable. Overall, all the colors produced with these five inks are close.

## CONCLUSION

This study aims to explore environmentally friendly resins to replace the acrylic ones in current water based flexographic inks. The alternative resins are made of soybean, which are sustainable and biodegradable. Five inks, including two containing acrylic resins and three containing soybean resins, were investigated based on their print performance.

Based on the results, the soy protein water-based inks present a great potential to replace the acrylic flexographic inks. One of the soy protein water-based inks, CSRI3, obtained high color density, exhibiting high potential for reducing ink consumption. CSR2 inks performed very well in controlling TVI (dot gain), in both highlight and mid-tone areas, indicating that they are able to produce a fine print, without scarifying image details. All five inks had comparable TVI in the

dark area, namely >75%. Meanwhile, the dot roundness analysis, another critical parameter for good flexographic print along with dot gain, also revealed that the soy protein water-based inks had a very competitive performance with that of the acrylic ones. The three soy protein water-based inks produced a comparable color to that of the commercial ink, which is the most critical evaluation. The  $\Delta E$  or color difference between CSR inks and commercial ink was within  $\pm 3$ , matching well the print industry standard. However, the print contrast of the soy protein water-based inks was slightly lower than those of acrylic inks. They also recorded slightly lower print gloss, compared to their counterpart.

Overall, the soy protein water-based inks performed very well compared to the acrylic inks. Considering the public pressure for sustainability and environmental protection, the print industry should pay more attention to soy protein water-based alternative inks, especially, since they have been accepted in other conventional printing methods.

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