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New Eco-friendly Coating Formulations for Recycled Paperboards: Effect on Print Quality and Ink Volume Consumption

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ABSTRACT

The coatings are commonly applied on paperboards in packaging to improve their strength and printability. This study focuses on the effect of new eco-friendly coating formulations on the colorimetric performance of recycled paperboards for packaging applications. China clay (F1) and montmorillonite clay (F2) based eco-friendly formulations were prepared and applied on selected recycled paperboards using a K-bar coater for three different coating thicknesses (4, 10, and 15 µm). The coated recycled paperboards were printed using an IGT printability tester, and the colorimetric performance was evaluated by comparing the color difference (ΔE) of the printed image with the ISO 12647-2 (2013) standard. The preliminary studies on coated recycled paperboards printed with black offset ink revealed that recycled paperboards coated with formulation F1 for 4 µm thickness showed better colorimetric performance compared to those coated with formulation F2. The China clay-based coating (F1) improved the surface structure by reducing the surface pores and roughness of the coated recycled paperboards. The improved surface structure, higher surface energy, and zeta potential of China-based formulation (F1) have resulted in better printability. Printing solid images using four-color offset ink with 1 and 2 mL ink volumes was used to test the colorimetric printability of coated recycled paperboards. Colorimetric printability was significantly improved using 1 mL of ink supply due to decreased ink absorption on coated recycled boards. The hypothesis test was carried out by performing paired t-test using IBM SPSS 20 software to verify the experimental results. Prog. Color Colorants Coat. 15 (2022), 175-189© Institute for Color Science and Technology.

1. Introduction

The packaging solution providers should consciously use environmentally sustainable alternatives such as recycled paperboards and eco-friendly coating material without reducing the quality of the packaging. Recycled paper has the tangible advantages of decreasing the use of trees in paper pulp processing, helping to build a world that is environmentally friendly [1, 2]. The coating used on paperboards improves their optical properties while also enhancing

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their visual appeal. Another significant advantage of coating paperboards is that it can increase the strength and printability of the paperboard [3]. Uncoated papers exhibit greater variations in printed color, owing to increased dot gain [4]. The absorbency of printing ink on coated paper increases with the increase in the porosity of the base paper. The non-uniformity of paper surfaces affects print uniformity [5]. The clay-based coating fills the surface cavities of the paper, lowering water absorption and improving printability [6]. Color reproduction is an important aspect of print quality. Smaller pores on the surface of the coated paper absorb ink slowly during the printing process. In comparison, larger pores can cause rapid absorption of the ink, resulting in a reduction in optical ink density [7]. Bio-based coatings derived from stearic acid in a hydrophobic starch matrix, when applied to the surface of the paperboard, exhibit increased resistance to water and grease absorption [8]. The solid print density, dot gain, and dot uniformity will influence the quality of conventional printing [9]. Due to the roughening of the coated surface caused by the addition of a large amount of latex binder, the specular gloss of coated carton is reduced [10]. Styrene-butadiene copolymer dispersion coatings improve the surface and barrier properties of paperboards [11]. The in-plane physical strength of the coated paperboard is higher when coated with formulations containing Kaolin clay and styrene-butadiene binder [12]. High-quality offset and flexographic color printing require effective ink trapping of primary color printing inks [13]. The amount of ink transferred on the substrate is determined by the nip pressure, ink rheology, and the amount of ink on the offset blanket [14]. The use of clay-based pigments combined with latex binder coatings for paper reduces porosity and ink absorption, resulting in increased print density [15]. Paperboards coated with organic nanoparticles synthesized by imidization of styrene-maleic anhydride copolymers demonstrated good offset printability due to better ink adhesion as compared to uncoated paperboards [16]. Latex-based coating formulations form a continuous film on the paperboard, improving its smoothness [17]. The recycled paper substrate can also be used to print a wide range of colors using an offset printing press [18]. The scanning electron microscopy (SEM) provides information about coated paper surface roughness and porosity [19]. The smoothness and absorbency of the paperboard surface will influence the colorimetric values (L*, a*, and b*) of the printed images. The optical ink density of the flexographic print increases with the increase in surface free energy of the biopolymer substrate [20, 21]. The higher optical ink density is the consequence of a wider coating network and increased bonding between printing ink and base paper [22]. The bleached kraft liner coated with nano-silica synthesized from rice husk (nano-RHS) improves the image clarity and color density of the printed image [23]. The objective of this research work is to evaluate the influence of two types of eco-friendly clay-based coating formulations, China clay (F1) and montmorillonite clay (F2), on the surface structure and colorimetric printability of nine types of recycled paperboards. The current work demonstrated a novel method for improving the colorimetric printability of recycled paperboards at the usage of low ink volumes by coating them with eco-friendly clay-based formulations. The coating formulations and coating thicknesses are also optimized based on colorimetric printability. The influence of ink volume on the colorimetric printability of coated recycled paperboards was evaluated by comparing with ISO 12647-2: 2013 standard [24]. The coated recycled paperboards printed using 1 mL of ink volume showed better colorimetric printability compared to those printed with 2 mL of ink.

2. Experimental

2.1. Materials

Nine types of imported recycled paperboards (Padmavati Fine papers, India) were used in this study. China clay (English Indian Clays Ltd, India), montmorillonite clay (Everest Starch, India, Pvt. Ltd), Sorbitol (Gulshan Polyols Limited, India), ammonia (Merck, India), Acronal (BASF, India), and four-color offset process inks (DIC (India) Limited, India) black (K), cyan (C), magenta (M), and yellow (Y) were purchased.

2.2. Categorization of the recycled paper boards

The brightness and colorimetric properties of the selected recycled paperboards suitable for Fast-moving consumer goods (FMCG) packaging applications were measured using Spectrodensitometer D530 (Xrite, USA). The instrument was calibrated using a standard white patch provided by Xrite. The colorimetric values

(L *, a * and b *) of recycled paperboard surfaces were measured by selecting the color measurement option. The brightness and colorimetric values of the paperboards were compared with ISO 12647-2: 2013 standard and categorized for further study.

2.3. Coating

The two types of eco-friendly clay-based coating formulations, F1 and F2, were prepared by mixing various ingredients using a magnetic stirrer for an hour to get a uniform coating formulation. The composition of the two coating formulations is shown in Table 1.

The particle size, conductivity, and Zeta potential of these coating formulations were measured using Mastersizer (Malvern Instruments Ltd, UK). The viscosity of the coating solutions at 30 °C was measured using Viscometer (Brookfield, USA) with spindle LV04 No. 64 at 60 RPM. The surface tension of the coating solutions was measured using a Surface Tensiometer (Jencon, India). The coating was carried out on nine types of recycled paperboards using K-Bar Coater (RK Print Coat Instruments, UK). The recycled paperboards were coated with formulations F1 and F2 each for three coating thicknesses of 4, 10, and 15 µm. The brightness and colorimetric properties of coated recycled boards were then measured using a spectrophotometer (Xrite, USA).

2.4. Printing

Both uncoated and coated recycled paperboards (coated with F1 and F2 each for 4, 10, and 15 µm thickness) were printed initially with 2 mL of black offset ink using Printability Tester (IGT Testing Systems, Netherlands) at 0.3 m/s printing speed and 400 N printing pressure. The L * a * and b * values of printed images were measured, and color deviations from ISO 12647-2: 2013 standard were calculated using the standard formula. The recycled paperboards coated with the best coating formulation for optimized coating thickness were chosen to print using four-color offset process inks. 1 and 2 mL of ink were then separately printed on recycled paperboards. The results were compared to the ISO 12647-2: 2013 standard to investigate the effect of ink volume on colorimetric printability [21].

Table 1: Composition of coating formulations F1 and F2.

Ingredients	Weight Percent for coating formulations	
	F1	F2
China clay	50	-
Montmorillonite clay	-	50
Acronal	30	30
Sorbitol	17	17
Ammonia	3	3

2.5. Microstructural surface study

The surface images of uncoated and coated Hammer white recycled paperboards were taken using Scanning Electron Microscopy (EVO 18, Zeiss, Germany). These images help to understand the changes in the surface structure of recycled paperboards after coating.

3. Results and Discussion

3.1. Properties of coating formulations

The properties of the two clay-based coating formulations F1 and F2, such as viscosity, surface tension, particle size, conductivity, and zeta potential, are presented in Table 2. The viscosity, surface tension, particle size, and zeta potential of coating formulation F1 are significantly higher than that of F2. The electrostatic attraction between molecules due to dispersion and adsorption of water-based binder on the China clay surfaces in formulation F1 has resulted in an intensely oriented surface structure, increasing the viscosity of the coating solution. The higher viscosity of the coating solution can offer a uniform coating coverage, and hence a smoother coated surface. The particle size of F2 is found to be finer than F1. Closely sized particles will result in a narrow particle size distribution. The larger size differences between the particles of F1 can lead to a broad particle size distribution of the coated surface. This type of arrangement in particle size distribution results in a closely packed surface structure [25]. The surface tension of the coating formulation will have an effect on the printability of the coated recycled paperboards. The increased surface tension of the coated surface leads to more ink transfer and higher optical ink density [22]. The higher Zeta potential of F1 indicates that it is more stable when compared to F2 [26].

Table 2: Properties of coating formulations F1 and F2.

Sl No	Coating Formulation	Viscosity (CP) at 60 RPM	Surface tension Dynes/ cm	Particle size (nm)			Conductivity (mS/cm)	Zeta Potential (mV)
				Largest	Smallest	Mean		
1	F1	4480	52.5	1300	155.9	185.2	0.172	-56.5
2	F2	1410	49	128.6	100.5	116.4	0.3	-28.7

3.2. Categorization of recycled paperboards as per ISO 12647-2: 2013

The standard characteristics of ISO paper types and their respective categories according to ISO 12647-2:2013 are shown in Table 3. The brightness and colorimetric values (L*, a*, and b*) of the recycled paperboards were measured using Xrite Spectrophotometer D530. The color difference (ΔE) between the selected recycled paperboards and the ISO standard was calculated using the formula (Eq. 1):

$$\Delta E = \sqrt{[\Delta L^2 + \Delta a^2 + \Delta b^2]} \quad (1)$$

Where ΔE is the color difference, ΔL, Δa, Δb is the deviation of L *, a *, and b * from the ISO standard measurements, respectively [25].

The ΔE values calculated in comparison with the ISO 12647-2 standard for uncoated recycled paperboards are recorded in Table 4. Then, the recycled paperboards are categorized according to ISO 12647-2: 2013 standard based on a minimum ΔE and brightness values.

Table 3: Standard L*, a*, b* and brightness of ISO 12647-2 paper type [25].

Paper type (ISO type)	L*	a*	b*	Brightness (%)
Gloss-coated, wood-free (1)	93	0	-3	89
Matte-coated, wood free (2)	92	0	-3	89
Gloss-coated, web (3)	87	-1	3	70
Uncoated, white (4)	92	0	-3	93
Uncoated,slightly yellowish (5)	88	0	6	73

Table 4: Classification of selected recycled paper boards into ISO paper type (n = 5).

Recycled paper boards	L*	a*	b*	Brightness (%)	ΔE	ISO type
Twill Bright White (I)	86.02 ± 3.45	5.90 ± 0.26	5.40 ± 0.25	72 ± 3.56	4.2 ± 0.94	5
Twill Avorio (II)	84.40 ± 3.68	1.88 ± 0.20	0.51 ± 0.15	90 ± 3.85	3.60 ± 1.05	4
Prisma White (III)	88.20 ± 3.78	7.10 ± 0.34	6.32 ± 0.28	71 ± 3.25	5.01 ± 1.34	5
Prisma Ivory (IV)	94.45 ± 3.98	6.50 ± 0.43	7.90 ± 0.38	71 ± 2.98	4.20 ± 1.29	4
Hammer White (V)	87.00 ± 3.47	4.80 ± 0.25	5.40 ± 0.23	92 ± 2.45	5.55 ± 1.45	4
Bianco Flash (VI)	89.90 ± 3.68	9.20 ± 0.38	8.00 ± 0.45	73 ± 2.10	9.95 ± 1.46	5
Contact laid Ivory (VII)	88.28 ± 3.43	5.65 ± 0.29	6.94 ± 0.42	92 ± 2.85	7.83 ± 1.35	4
Contact laid white (VIiI)	86.75 ± 3.18	3.75 ± 0.30	6.50 ± 0.36	75 ± 2.30	9.17 ± 1.58	5
Contact Natural white (IX)	89.48 ± 2.78	8.90 ± 0.45	9.44 ± 0.32	93 ± 3.46	9.90 ± 1.49	4

3.3. Optimization of coating formulation and thickness

All the selected recycled paperboards were coated separately with coating formulations F1 and F2 for three different coating thicknesses (4, 10, and 15 µm). The brightness and ΔE values for uncoated and coated recycled paperboards for three different thicknesses are presented in Tables 5 and 6, respectively. Table 6 shows that the ΔE values of the recycled paperboards coated with formulation F1 are much lower than those coated with formulation F2 for three coating thicknesses. Further, it is clear from Table 6 that the ΔE values of recycled paperboards coated with formulation F1 for 4 µm thickness showed minimum ΔE values.

Table 5: Brightness values of recycled paperboards before and after coating (n = 5).

Recycled paperboards	Uncoated	Brightness (%)					
		F1			F2		
		Coating thickness (µm)			Coating thickness (µm)		
		4	10	15	4	10	15
I	87.2 ± 3.36	86.02 ± 2.45	85.90 ± 2.20	85.40 ± 2.70	81.67 ± 2.34	81.00 ± 3.45	81.20 ± 3.80
II	69.2 ± 2.46	70.40 ± 2.67	71.88 ± 1.88	70.51 ± 1.84	67.42 ± 1.56	68.52 ± 2.34	68.66 ± 2.67
III	89.02 ± 2.56	88.20 ± 2.85	87.10 ± 2.44	86.32 ± 2.45	85.44 ± 3.57	84.76 ± 2.56	83.86 ± 3.58
IV	72.5 ± 2.28	74.45 ± 2.32	76.50 ± 2.14	77.90 ± 2.83	70.34 ± 3.18	69.78 ± 2.15	69.05 ± 3.26
V	90.2 ± 3.65	87.00 ± 3.23	84.80 ± 2.24	85.40 ± 3.4	82.18 ± 2.45	81.74 ± 3.58	81.26 ± 2.96
VI	91.8 ± 3.12	89.90 ± 3.57	89.20 ± 3.45	88.00 ± 3.26	83.18 ± 3.74	82.56 ± 3.26	81.78 ± 3.89
VII	71.2 ± 2.68	73.28 ± 2.12	75.65 ± 2.32	76.94 ± 3.23	70.85 ± 3.45	69.65 ± 3.25	69.80 ± 3.24
VIII	90.0 ± 3.45	86.75 ± 2.88	83.75 ± 2.52	86.50 ± 3.28	82.66 ± 3.12	81.98 ± 2.87	80.57 ± 3.12
IX	81.8 ± 1.89	79.48 ± 2.35	78.90 ± 2.15	79.44 ± 3.45	75.37 ± 3.41	75.05 ± 2.58	74.08 ± 3.69

[I] Twill Bright White, [II] Twill Avorio, [III] Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

Table 6: The ΔE values of recycled paperboards before and after coating (n = 5).

Recycled paperboards	Color difference (ΔE)						
	Uncoated	F1			F2		
		Coating thickness (µm)			Coating thickness (µm)		
		4	10	15	4	10	15
I	4.2 ± 0.94	4.2 ± 1.03	5.2 ± 1.36	5.7 ± 1.48	7.11 ± 1.95	7.34 ± 1.45	7.78 ± 1.26
II	3.60 ± 1.05	4.90 ± 1.34	4.60 ± 1.23	4.60 ± 1.05	6.55 ± 1.24	6.88 ± 1.23	7.15 ± 1.12
III	5.01 ± 1.34	5.81 ± 1.23	4.81 ± 1.42	4.81 ±1.08	6.45 ± 1.43	6.50 ±1.93	6.80 ±1.23
IV	4.20 ± 1.29	4.66 ± 0.95	4.60 ± 0.88	4.60 ± 0.98	7.18 ± 1.58	7.50 ±1.70	7.86 ±1.34
V	5.55 ± 1.45	6.55 ± 1.23	7.55 ± 1.07	7.55 ± 0.95	9.12 ± 1.98	9.45 ± 1.12	9.80 ± 1.23
VI	9.95 ± 1.46	7.85 ± 1.25	8.95 ± 1.27	8.95 ± 1.58	9.77 ± 1.56	9.80 ± 1.39	9.90 ±1.47
VII	7.83 ± 1.35	8.73 ± 1.26	8.83 ± 1.45	8.83 ± 1.25	11.12 ± 1.45	11.45 ± 1.23	12.30 ± 1.67
VIII	9.17 ± 1.58	8.87 ± 1.63	8.17 ± 1.89	8.17 ± 1.45	11.65 ± 1.38	11.80 ± 1.24	11.98 ± 1.45
IX	9.90 ± 1.49	8.70 ± 1.24	7.90 ± 1.85	7.90 ± 1.85	7.11 ± 1.25	7.45 ± 1.29	7.80 ± 1.56

[I] Twill Bright White, [II] Twill Avorio, [III] Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

The recycled paperboards coated with formulations F1 and F2 for 4, 10, and 15 μm thickness were printed using black offset ink at 400 N printing pressure. The optical ink densities and ΔE values of the black offset ink compared with ISO 12647-2:2013 are presented in Tables 7 and 8, respectively.

Table 7 shows that the optical densities of the printed black offset ink on the recycled paperboards increase after coating. The optical ink densities have increased with the increase in coating thicknesses. This may be because the clay pigment (formulation F1) covers the pores due to its closely packed surface structures, shape, layered characteristics, and greater aspect ratio [5]. Hence, the ink absorption of coated recycled paperboards decreases, leading to increased ink film thickness. Increased thickness of the clay-based coating will result in fewer surface pores, resulting in much lower ink absorption and higher optical densities.

Table 7: The optical density of black images printed on uncoated and coated recycled paperboards (n = 5)

Recycled paperboards	Optical density						
	Uncoated	F1			F2		
		Coating thickness (μm)			Coating thickness (μm)		
		4	10	15	4	10	15
I	0.75 ± 0.06	1.24 ± 0.08	1.54 ± 0.07	1.684 ± 0.04	0.73 ± 0.07	1.23 ± 0.02	1.52 ± 0.05
II	0.74 ± 0.05	1.30 ± 0.04	1.51 ± 0.03	1.76 ± 0.05	0.71 ± 0.04	1.29 ± 0.04	1.73 ± 0.05
III	0.78 ± 0.07	1.29 ± 0.03	1.54 ± 0.05	1.71 ± 0.06	0.76 ± 0.05	1.28 ± 0.05	1.69 ± 0.06
IV	0.79 ± 0.08	1.23 ± 0.08	1.53 ± 0.04	1.73 ± 0.08	0.75 ± 0.08	1.22 ± 0.07	1.71 ± 0.08
V	0.74 ± 0.04	1.25 ± 0.03	1.55 ± 0.06	1.72 ± 0.06	0.74 ± 0.04	1.24 ± 0.06	1.70 ± 0.07
VI	0.81 ± 0.04	1.23 ± 0.02	1.49 ± 0.07	1.70 ± 0.05	0.78 ± 0.03	1.22 ± 0.07	1.71 ± 0.05
VII	0.77 ± 0.02	1.24 ± 0.04	1.53 ± 0.05	1.73 ± 0.07	0.75 ± 0.05	1.22 ± 0.05	1.70 ± 0.05
VIII	0.80 ± 0.05	1.29 ± 0.05	1.50 ± 0.04	1.68 ± 0.03	0.78 ± 0.03	1.27 ± 0.07	1.62 ± 0.07
IX	0.76 ± 0.06	1.32 ± 0.06	1.56 ± 0.07	1.66 ± 0.05	0.75 ± 0.04	1.30 ± 0.04	1.63 ± 0.08

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Table 8: The ΔE values for a black image printed on uncoated and coated samples (n = 5).

Recycled paperboards	Color difference (ΔE)						
	F1				F2		
	Uncoated	Coating thickness (μm)			Coating thickness (μm)		
		4	10	15	4	10	15
I	11.40 ± 2.45	8.70 ± 2.56	13.50 ± 2.54	24.34 ± 2.78	10.50 ± 2.56	12.70 ± 1.67	24.34 ± 3.45
II	10.80 ± 265	8.80 ± 2.65	14.85 ± 2.78	25.39 ± 2.98	9.85 ± 2.65	12.80 ± 1.63	25.39 ± 3.45
III	14.50 ± 3.23	10.65 ± 2.98	17.68 ± 2.89	21.24 ± 2.65	14.68 ± 3.45	14.65 ± 1.43	21.24 ± 3.56
IV	12.10 ± 3.21	9.20 ± 3.25	19.58 ± 2.38	21.60 ± 2.67	11.58 ± 3.12	17.20 ± 1.46	21.60 ± 3.21
V	9.60 ± 2.56	6.35 ± 3.56	17.90 ± 2.94	23.71 ± 2.68	7.90 ± 2.67	15.35 ± 1.28	23.71 ± 2.98
VI	10.70 ± 2,78	6.95 ± 2.67	18.90 ± 2.76	24.42 ± 2.98	9.90 ± 2.87	16.95 ± 1.85	24.42 ± 2.59
VII	10.20 ± 2.54	8.40± 2.76	18.45 ± 2.34	21.09 ± 3.15	10.45 ± 2.69	16.40 ± 1.76	21.09 ± 3.23
VIII	11.80 ± 2.75	10.78± 3.24	19.90 ± 2.96	24.01 ± 3.65	12.90 ± 3.24	18.78 ± 1.72	24.01 ± 3.97
IX	17.50 ± 2.78	19.26 ± 3.24	19.50 ± 2.87	23.10 ± 2.56	20.50 ± 2.58	16.26 ± 2.09	23.10 ± 3.45

[I] Twill Bright White, [II]Twill Avorio, [III]Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

From Table 8, it is clear that the ΔE of the black printed image on the recycled paperboards coated with formulation F1 for a thickness of 4 μm shows the least color deviation from ISO 12647-2: 2013 standard. The ΔE has also increased with an increase in coating thickness for both the coating formulations. This is mainly due to the reduction in lightness values at higher coating thicknesses. The amount of ink transferred to the substrate, optical ink density, and lightness (L*) of the black image printed using black offset ink on uncoated and coated (with 4 μm thickness) recycled paperboards are listed in Table 9.

As shown in Table 9, the lightness (L *) value of black images printed on recycled paperboards with 2 mL of ink have decreased significantly after coating with formulation F1 for a coating thickness of 4 μm. Even though the amount of ink transferred is reduced, the optical ink densities of the black image are increased after coating. This is primarily due to the fact that printing ink has a lower absorption rate on coated surfaces, leading to lower lightness values (L*).

3.4. Microstructural surface analysis

Figure 1a, b, c shows SEM images of uncoated and coated (with F1 and F2 for 4µm thickness) hammer white recycled paperboard, respectively. The surface of the uncoated recycled paperboard is very porous and uneven (Figure 1a). The surface unevenness of the recycled paperboard is reduced after its coating. The coating of the recycled paperboards has resulted in the sealing of pores and gaps between the paperboard fibers. This may have resulted in the reduction of ink absorption on recycled paperboards. However, it is observed that the recycled paperboard coated with formulation F1 (Figure 1b) showed much-reduced surface unevenness when compared to that coated with F2 (Figure 1c).

3.5. Effect of ink volume on the color difference (ΔE)

The optical densities for black, cyan, magenta, yellow, red, green, and blue images printed using 2 and 1 mL of ink with formulation F1 for 4µm coating thickness are presented in Table 10.

Table 9: Ink transfer, optical ink density, lightness, and ΔE for the black image on uncoated and coated recycled paperboards.

Recycled paperboards	Uncoated				Coated (F1, 4 μm)			
	Ink transferred (g/m^2)	Optical density	Lightness (L*)	Color difference (ΔE)	Ink amount (g/m^2)	Optical density	Lightness (L*)	Color difference (ΔE)
I	1.22 ± 0.10	0.75 ± 0.06	39.8 ± 0.52	11.40 ± 2.45	0.90 ± 0.10	1.24 ± 0.08	14.56 ± 0.70	8.70 ± 2.56
II	1.32 ± 0.05	0.74 ± 0.05	39.23 ± 0.62	10.80 ± 2.65	0.83 ± 0.05	1.30 ± 0.04	12.86 ± 0.80	8.80 ± 2.65
III	1.30 ± 0.03	0.78 ± 0.07	39.53 ± 0.99	14.50 ± 3.23	0.94 ± 0.03	1.29 ± 0.03	12.45 ± 0.34	10.65 ± 2.98
IV	1.33 ± 0.04	0.79 ± 0.08	36.57 ± 0.88	12.10 ± 3.21	0.95 ± 0.02	1.23 ± 0.08	12.65 ± 0.54	9.20 ± 3.25
V	1.30 ± 0.03	0.74 ± 0.04	38.57 ± 0.92	9.60 ± 2.56	0.95 ± 0.04	1.25 ± 0.03	14.53 ± 0.73	6.35 ± 3.56
VI	1.32 ± 0.04	0.81 ± 0.04	37.22 ± 0.97	10.70 ± 2.78	0.93 ± 0.03	1.23 ± 0.02	14.75 ± 0.72	6.95 ± 2.67
VII	1.31 ± 0.02	0.77 ± 0.02	36.32 ± 0.87	10.20 ± 2.54	0.93 ± 0.02	1.24 ± 0.04	12.52 ± 0.73	8.40 ± 2.76
VIII	1.31 ± 0.03	0.80 ± 0.05	38.31 ± 0.76	11.80 ± 2.75	0.96 ± 0.04	1.29 ± 0.05	14.64 ± 0.72	10.78 ± 3.24
IX	1.25 ± 0.04	0.76 ± 0.06	39.06 ± 0.58	17.50 ± 2.78	0.93 ± 0.03	1.32 ± 0.06	15.54 ± 0.61	13.26 ± 3.24

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Table 10: Optical density for black (K), cyan (C), magenta (M), and yellow (Y) printed images.

Recycled paperboards	Optical Density							
	2 mL				1 Ml			
	K	C	M	Y	K	C	M	Y
I	1.24 ± 0.08	1.14 ± 0.05	1.13 ± 0.02	0.88 ± 0.03	0.96 ± 0.04	0.87 ± 0.03	0.87 ± 0.03	0.78 ± 0.04
II	1.30 ± 0.04	1.15 ± 0.03	1.14 ± 0.05	0.87 ± 0.05	0.86 ± 0.03	0.89 ± 0.04	0.88 ± 0.03	0.76 ± 0.04
III	1.29 ± 0.03	1.12 ± 0.04	1.13 ± 0.02	0.88 ± 0.03	0.95 ± 0.04	0.86 ± 0.04	0.87 ± 0.04	0.77 ± 0.02
IV	1.23 ± 0.08	1.14 ± 0.03	1.16 ± 0.03	0.88 ± 0.04	0.94 ± 0.03	0.85 ± 0.03	0.84 ± 0.03	0.76 ± 0.03
V	1.25 ± 0.03	1.15 ± 0.05	1.14 ± 0.03	0.90 ± 0.05	0.95 ± 0.04	0.86 ± 0.03	0.85 ± 0.02	0.75 ± 0.03
VI	1.23 ± 0.02	1.09 ± 0.02	1.07 ± 0.03	0.87 ± 0.02	0.93 ± 0.04	0.85 ± 0.04	0.84 ± 0.04	0.75 ± 0.02
VII	1.24 ± 0.04	1.10 ± 0.04	1.11 ± 0.04	0.85 ± 0.05	0.95 ± 0.03	0.84 ± 0.04	0.86 ± 0.03	0.76 ± 0.03
VIII	1.29 ± 0.05	1.13 ± 0.04	1.14 ± 0.04	0.87 ± 0.06	0.97 ± 0.03	0.87 ± 0.03	0.84 ± 0.04	0.78 ± 0.04
IX	1.32 ± 0.06	1.15 ± 0.04	1.15 ± 0.05	0.88 ± 0.05	0.94 ± 0.04	0.88 ± 0.02	0.87 ± 0.04	0.79 ± 0.02

[I] Twill Bright White, [II]Twill Avorio, [III]Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

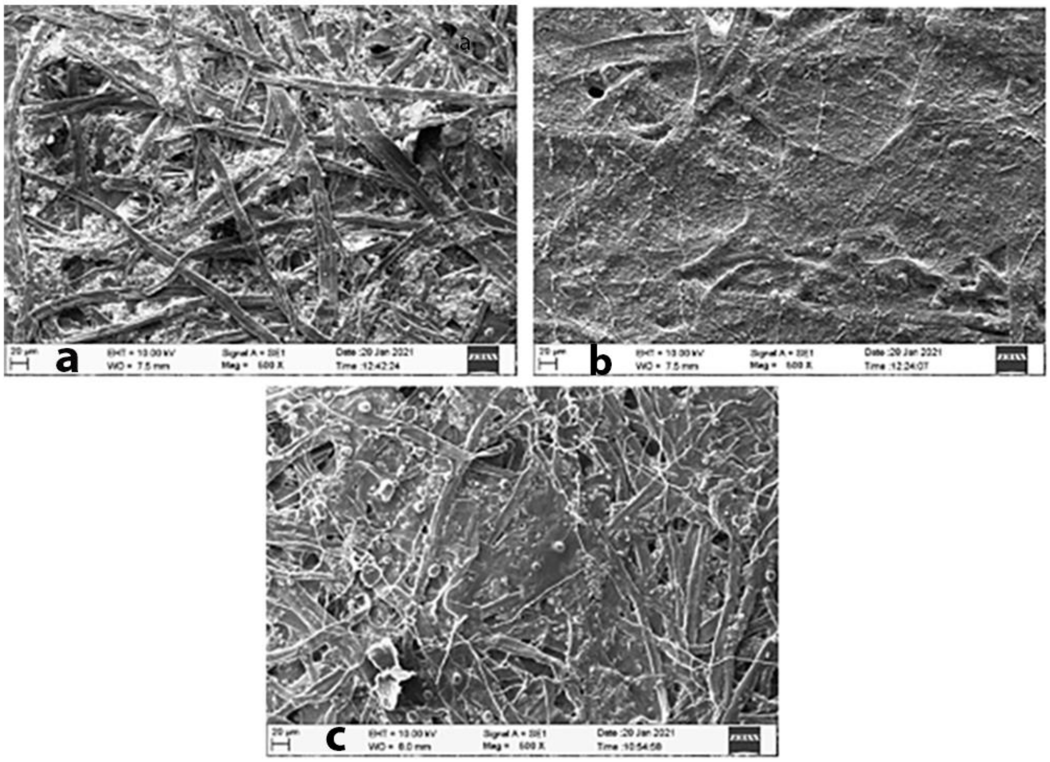

Figure 1: SEM images of recycled paper board (a) Uncoated, (b) Coated with F1, and (c) Coated with F2.

The L * a * b * values of black, cyan, magenta, yellow, red, green, and blue images printed with 2 mL of ink on an IGT printability tester at 400 N on coated recycled paperboards with F1 for 4μm thickness are measured. The ΔE values are then calculated in comparison with ISO 12647-2:2013 using the standard color difference formula and presented in Table 11.

The higher ΔE on coated recycled paperboards (Table 11) when compared with ISO 12647-2:2013 standard was mainly due to excessive ink transfer and reduced ink absorption by coated recycled paperboards. These results were the motivation to investigate the influence of reduced ink volume on the colorimetric printability of recycled paperboards. Therefore, the coated recycled paperboards are then printed using 1 mL of ink by keeping all other print settings constant. The ΔE between color images printed with 1 mL ink and ISO 12647-2 standard reference are presented in Table 12.

Table 11: Color difference (ΔE) for black (K), cyan (C), magenta (M), yellow (Y), red (R), green (G), and blue (B) for 2 mL ink supply.

Recycled paperboard	Color difference (ΔE) for 2 mL ink volume						
	K	C	M	Y	R	G	B
I	8.70 ± 2.56	6.87 ± 3.82	8.45 ± 3.65	12.97 ± 4.28	13.24 ± 3.39	17.27 ± 4.24	22.17 ± 5.27
II	8.80 ± 2.65	7.45 ± 3.32	8.78 ± 3.43	12.72 ± 4.56	12.38 ± 3.65	16.74 ± 4.32	23.07 ± 4.54
III	10.65 ± 2.98	7.04 ± 2.45	10.28 ± 3.54	12.79 ± 3.43	13.42 ± 4.32	16.45 ± 4.75	21.66 ± 4.54
IV	9.20 ± 3.25	7.51 ± 3.98	10.53 ± 3.45	11.41 ± 3.43	12.35 ± 2.56	15.12 ± 4.54	25.41 ± 5.65
V	6.35 ± 3.56	7.39 ± 2.53	12.81 ± 3.522	14.89 ± 4.43	14.28 ± 3.54	16.47 ± 4.32	20.03 ± 5.45
VI	6.95 ± 2.67	7.62 ± 1.85	10.89 ± 2.54	12.25 ± 2.34	13.63 ± 3.54	16.06 ± 4.68	23.44 ± 5.76
VII	8.40 ± 2.76	7.94 ± 2.54	10.83 ± 3.56	11.41 ± 3.43	12.99 ± 4.32	16.90 ± 3.56	22.17 ± 5.96
VIII	10.78 ± 3.24	7.34 ± 3.32	10.36 ± 2.72	12.06 ± 3.54	13.91 ± 3.86	16.57 ± 4.64	25.21 ± 4.65
IX	13.26 ± 3.24	7.72 ± 2.98	13.30 ± 3.42	12.62 ± 3.53	14.15 ± 2.68	15.65 ± 5.84	21.33 ± 5.76

[I] Twill Bright White, [II] Twill Avorio, [III] Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

Table 12: Color difference (ΔE) for black, cyan, magenta, yellow, red, green, and blue for 1 mL ink supply.

Recycled paperboard	Color difference (ΔE) for 1 mL ink volume						
	K	C	M	Y	R	G	B
I	7.26 ± 3.54	10.01 ± 4.35	8.06 ± 3.85	4.57 ± 4.43	5.45 ± 4.65	6.41 ± 3.49	12.59 ± 3.79
II	7.58 ± 3.65	10.19 ± 3.98	6.12 ± 3.75	4.18 ± 1.14	0.94 ± 0.24	5.67 ± 1.54	14.91 ± 4.43
III	6.34 ± 3.21	8.97 ± 3.87	5.33 ± 3.76	1.21 ± 0.56	2.78 ± 0.97	4.56 ± 1.42	12.73 ± 3.64
IV	6.45 ± 3.83	6.05 ± 3.57	6.47 ± 2.87	3.88 ± 1.58	1.91 ± 0.54	4.60 ± 1.63	13.08 ± 3.75
V	7.08 ± 3.92	9.94 ± 3.76	2.97 ± 1.43	3.58 ± 1.73	6.00 ± 1.24	5.14 ± 1.75	13.42 ± 3.54
VI	7.29 ± 3.64	9.69 ± 3.98	4.39 ± 3.21	2.36 ± 1.23	7.26 ± 1.42	4.43 ± 1.47	14.02 ± 65
VII	6.29 ± 3.52	9.04 ± 3.52	7.48 ± 4.54	4.53 ± 1.65	4.97 ± 1.53	4.78 ± 1.53	13.18 ± 4.54
VIII	7.17 ± 3.62	9.98 ± 3.67	5.72 ± 3.76	2.47 ± 1.24	3.68 ± 1.54	3.07 ± 1.64	13.00 ± 3.75
IX	6.47 ± 3.78	4.52 ± 4.65	7.84 ± 3.98	2.71 ± 1.23	4.10 ± 1.43	6.78 ± 1.53	13.08 ± 3.76

[I] Twill Bright White, [II] Twill Avorio, [III] Prisma White, [IV] Prisma Ivory, [V] Hammer White, [VI] Bianco Flash, [VII] Contact laid Ivory, [VIII] Contact laid white, [IX] Contact Natural white.

Figures 2-7 shows the visual comparison of ΔE between solid color images printed with 1 and 2 mL of ink volume on nine types of coated recycled paperboards, respectively.

It is evident from Figures 2-7 that the ΔE values for cyan, magenta, yellow, red, green, and blue ink images printed each with 1 mL ink volume on nine types of recycled paperboards have been reduced significantly compared to those printed with 2 mL ink. The ΔE of images printed with 2 mL of ink is higher due to the reduction of ink absorption on the surface of coated recycled paperboards. The improved colorimetric performance when printed with 1 mL of ink is due to the better transfer of ink on the surface of coated recycled boards. Further, this may be due to the improved surface energy of coated surface due to the higher zeta potential of China clay-based coating formulation. The printing on coated recycled paperboards with reduced ink volume has resulted in improved color quality and consistency. The reduction in ink supply offers several advantages, including increased ink mileage, reduced ink waste, and the easy cleaning of inking systems with a reduced amount of cleaning solvents. Thus, coating recycled paperboards with clay-based coatings will improve the sustainability of package printing.

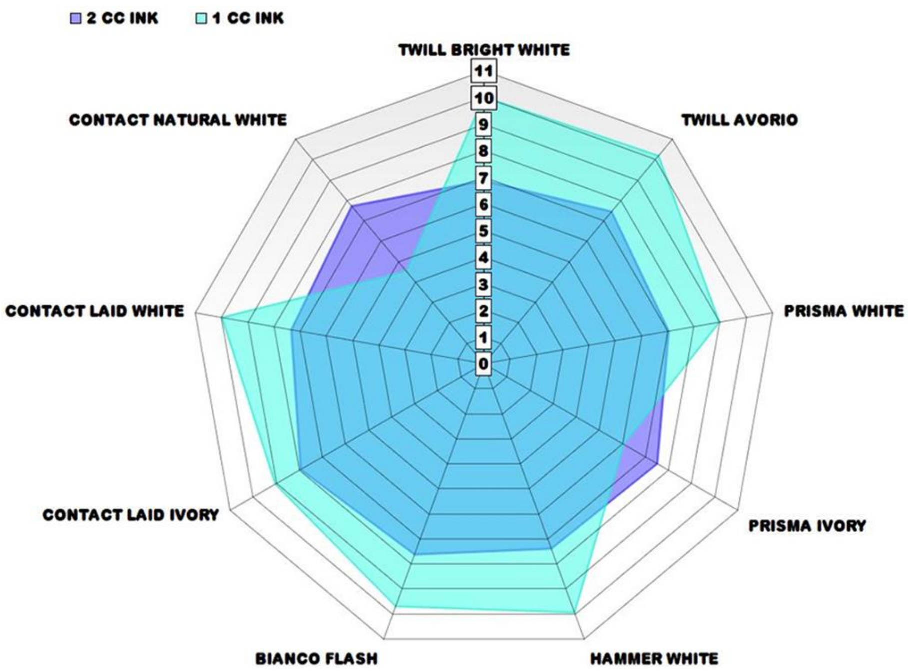

Figure 2: Comparison of color difference for cyan between 1 and 2 mL ink.

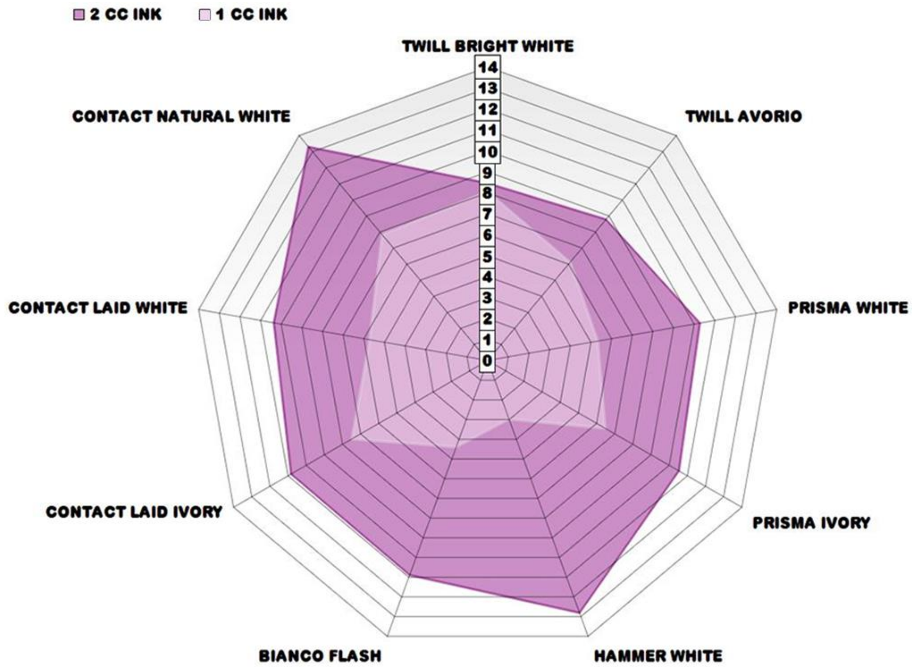

Figure 3: Comparison of color difference for magenta between 1 and 2 mL ink .

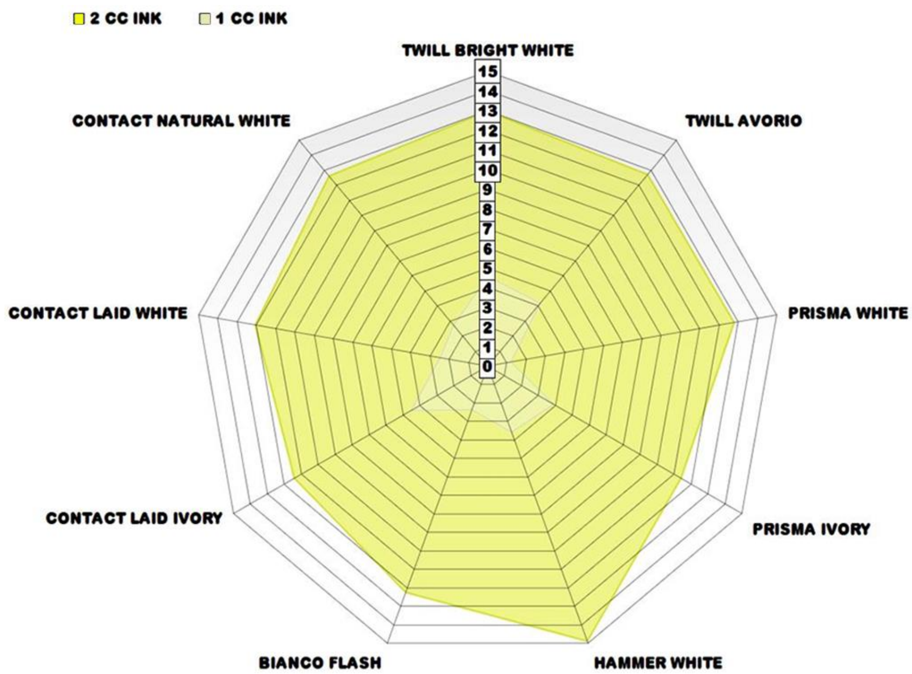

Figure 4: Comparison of color difference for yellow between 1 and 2 mL ink.

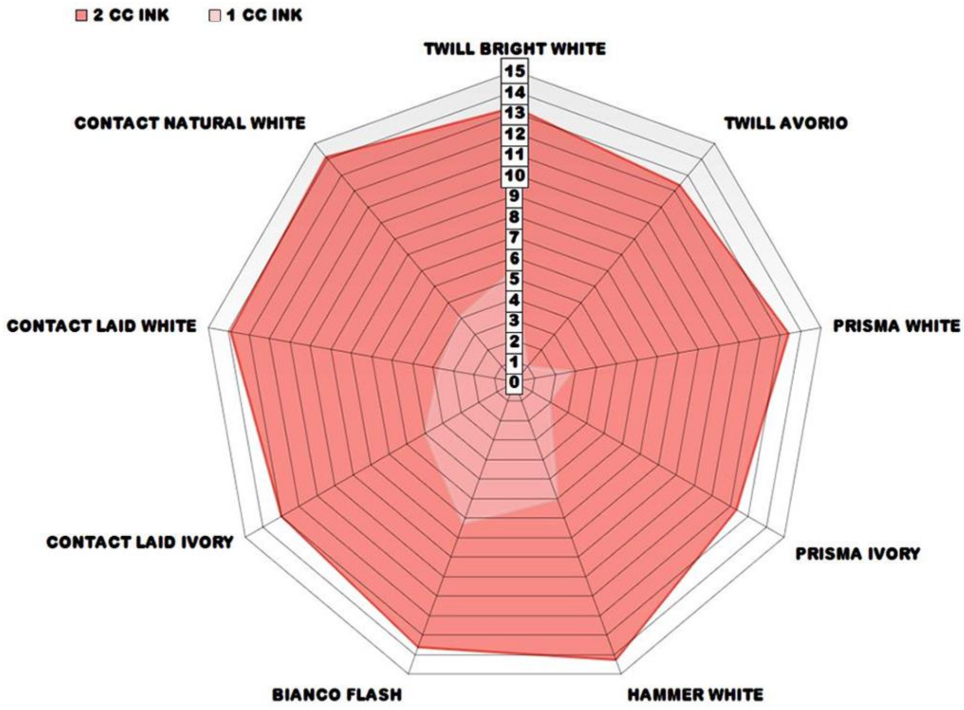

Figure 5: Comparison of color difference for red between 1 and 2 mL ink.

Figure 6: Comparison of color difference for green between 1 and 2 mL ink.

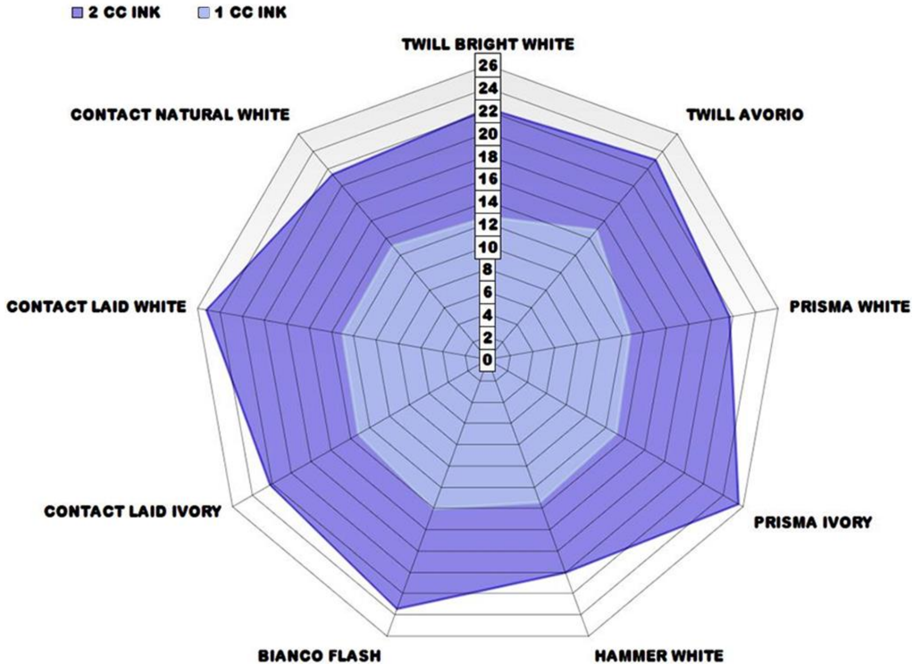

Figure 7: Comparison of color difference for blue between 1 and 2 mL ink.

3.6. Hypothesis testing for evaluating the influence of reduction in ink volume on colorimetric performance

The experimental results of ΔE values for coated recycled paperboards printed with 2 and 1 mL of ink were subjected to hypothesis testing at a 5 % significance level. The following hypothesis was tested by paired t-test [6].

H_a: There is a significant improvement in the ΔE values of black, cyan, magenta, yellow, red, green, and blue printed images after reducing the ink volume from 2 to 1 mL.

H_o: There is no significant improvement in the ΔE values of black, cyan, magenta, yellow, red, green, and blue printed images after reducing the ink volume from 2 to 1 mL. The results of paired t-test and correlation coefficients calculated using IBM SPSS 20 software are presented in Table 13.

Table 13: Statistical significance of coating on the ΔE values of coated recycled paperboards.

Colors	t	Significance value (2-tailed)	Result
K	7.74	0.004	Significant
C	1.87	0.080	Not significant
M	6.04	0.001	Significant
Y	17.96	0.001	Significant
R	12.93	0.001	Significant
G	26.1	0.000	Significant
B	14.76	0.000	Significant

Table 13 shows that the significance level calculated using IBM SPSS 20 is less than 0.05 for all the colors except cyan. Hence, the H_a is accepted. The ΔE of coated recycled paperboards have improved significantly after reducing the ink volume from 2 to 1 mL for magenta, yellow, red, green, and blue printed images [6]. Thus, the coating of recycled paperboards reduces lightness (L*) by lowering ink absorption, leading to a better colorimetric performance at 1 mL of ink while compared to those printed with 2 mL of ink.

4. Conclusions

Two clay-based eco-friendly coatings were developed using China clay (F1) and montmorillonite clay (F2) as the primary coating materials and coated separately on nine different types of recycled paperboards for three different coating thicknesses (4, 10, and 15 µm). The uncoated and coated samples (4, 10, and 15 µm) were printed initially with black color offset process ink using 2 mL of ink on an IGT printability tester at 400 N printing pressure. The optical densities of black color offset ink on the coated recycled paperboards increased due to decreased ink absorption. However, as coating thicknesses exceeded 4 µm, the color difference between printed images and the ISO 12647-2: 2013 standard increased. The high color deviation is primarily due to a reduction in lightness values caused by decreased ink absorption at higher coating thicknesses. The brightness and colorimetric printability of recycled paperboards coated with a China clay-based formulation (F1) at a thickness of 4 µm delivered optimal results. SEM was used to study the surface morphology of uncoated and coated (formulations F1 and F2) recycled paperboards with a thickness of 4 µm. The colorimetric printability of solid images printed with 1 and 2 mL of four-color offset inks was examined to investigate the effect of ink volume on colorimetric performance. The color deviation of most of the solid images from ISO 12647-2: 2013 standard was minimized when printed with 1 mL of offset ink on coated recycled paperboards compared to those printed with 2 mL ink. The hypothesis test results complemented the observation that reducing ink volume reduced color deviation in most cases.

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