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Effect of Ink Formulation and Paper Surface Morphology on Ink-jet Printing Properties

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ABSTRACT

The purpose of this study was to investigate the effect of different type of alcohols and humectants in inks formulation, as well as the influence of paper surface morphology on the ink-jet printing properties. In order to investigate the influence of alcohol and humectant types on printing properties, the optimum ink containing C.I. Reactive Blue 21 (Ink2) was formulated with different type of alcohols and humectants. The results of optical density and water fastness evaluations indicated that, the prints depend more on paper surface morphology rather than the type of alcohols and humectants in ink formulation. Optimum optical density and good water fastness properties were obtained on glossy coated ink-jet printed papers comprising optimum-diameter nano-porosities in their coating layer. Prog. Color Colorants Coat. 7(2014), 295-304 © Institute for Color Science and Technology.

1. Introduction

In the last two decades, drop-on-demand (DOD) ink-jet printing systems have grown to become a major topic in scientific research [1-6]. DOD ink-jet printers are widely used in small office and home office (soho) due to their low cost, low noise, full range color, low space demand, and ability to print on a variety of substrates, including plain papers, transparencies and papers for special printing. Three basic elements are important in ink-jet printing quality: the printing head used in the printer; the ink formulation and the substrate on which the ink droplets are deposited to form the image. The two most popular types of DOD ink-jet printers are thermal and piezoelectric. In this project, thermal ink-jet printers were used and the ink droplets are sputtered by the pressure brought about by selective evaporation of the ink that result from heating a thermocouple, which is integrated in the ink channel of the nozzle.

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The following properties are demanded from an ink formulation intended for office use; non-toxic for safety, good storage stability, resistance to heat, non-corrosive, neutral pH, optimum surface tension, low viscosity, capability to pass through the ink-jet nozzle without clogging, good light and water fastness on the substrate. Ink-jet printing inks are typically comprised of a liquid medium and colorant such as a pigment or dye. These inks, which have been developed for use in DOD ink-jet printing, are comprised of water and water-soluble organic solvent to prevent the ink from being deposited at the tip of the print head, co-solvents and humectant to limit evaporation, and a number of materials which act as penetrants and coloring agents. Water soluble dyes, such as basic dyes, acid dyes, direct dyes and reactive dyes were used in the first embodiment. Optically, dyes scatter little light and therefore provide highly saturated colors. However, the relatively large pigment sizes result in increased surface roughness of the ink layer, which causes light to scatter, thereby producing less saturated and duller prints. The flocculation and aggregation of the pigment dispersion in the ink tends to block the ink-jet printer nozzles and results in problems with reliably firing the ink from the nozzle head. In order to produce a high density color print, a water soluble dye was used as a coloring agent, which dissolves completely in water and does not proceed to clog the tip of the printer head. Therefore, the coloring agent employed for ink-jet printing is normally a water soluble dye rather than a pigment [7].

Since the print quality of plain papers need improvement in the ink-jet paper industry. Therefore, wide ranges of components are used to coat ink-jet papers [8]. Ink-jet coated papers can be classified into coated matte papers, and coated glossy papers. Glossy ink-jet papers are by far the best papers to use when photorealistic image are desired. They are currently made by different processes [9-10].The printing substrate must display adequate ink absorbency, drying speed, water fastness in order to create a superior color image. To achieve good water fastness, a critical balance of properties is required. For example, ink-jet printers need water soluble dye systems to prevent jet nozzle clogging, which can be obtained through dyes with greater solubility. Many researches have been carried out to improve the quality of ink-jet prints by improving individual elements such as developing improved water fast dye sets, [11-15] or with new materials. Novel materials, such as cationic polymers [16-21], mixed metal cations in the form of sulfate [22], cationic metal-organic charge complexes [23], or nano-particle metal oxides [21], can be integrated into paper coating formulations or some new materials such as nitrile (which has the quaternary or tertiary amine) can be used in the ink formulation [24] to improve the quality of ink-jet prints. One strategy to improve water fastness is chemically attaching dyes to the media surface during printing by coating the paper with silica pigments [25-27], poly vinyl alcohol binder (PVA) [28-30], nanocomposites [30-31], kaoline and a polymer [32-33], which are typically expensive [21, 34-42]. However, water and light fastness largely remain a concern for papers. Research in improving fastness properties for ink-jet prints has been overwhelmingly focused on paper coating formulations. In contrast, considerably less attention has been centered on the ink formulation itself.

Previous study was focused on the effect of reactive dye structure and penetrant types in ink formulation on the ink-jet printing properties of the printed objects on different kinds of papers. Ink-jet printing of three inks containing different dye structures on the substrate showed that the optical density, water fastness and light fastness of the printed objects were heavily dependent on the dye structure [43]. This study has focused on the effect of alcohol and humectant types in ink formulation on the colorimetric and fastness properties of the ink-jet printed objects on a range of substrates. This investigation was carried out using optimum reactive dyes from the preceding study [43], which were formulated with various humectants and alcohols differing in the number of alkyl groups in their structure. As the ink-substrate interactions are the most important mechanisms governing optical density values of the ink on to the substrate. Five different papers with the same weight and different textures or glosses were used in this study.

2. Experimental

2.1. Materials

The salt-free reactive dye (C.I. Reactive blue 21) with high purity was kindly provided by Alvan Sabet Company, Iran. diethylene glycol, triethylene glycol, tetra ethylene glycol and polyethylene glycol 200 provided by Merck Company, Germany, were used as humectant. Isopropyl alcohol, butandiol, pentandiol, hexandiol and 2-pyrrolidoneas co-solvents and diethylene glycol mono-butyl ether as penetrant were also received from Merck Company, Germany. Five different papers with the same weight and different textures and glosses provided from the market were used in this study (Table 1).

2.2. Equipment and instrumentation

The prepared inks were filtered through a 0.45 μm and 0.2 μm Sartorius Minisartfilter (Göttingen, Germany). The different papers were printed by a HP DeskJet 5150 printer. The reflectance measurements of printed images were determined using a Gretag Macbeth Spectrophotometer ColorEye7000A (New York, USA) with d/8° measurement geometry under the following conditions: measurement wavelength ranges from 400nm to 700 nm, measurement area of 10 mm in diameter, and the specular component included (SCI) measurement mode.

The CIEL*, a* and b* values were computed under a D65 illuminant and CIE 1964 (10°) standard observer. The gloss of each paper was measured using a Novo-Gloss IQ Goniophotometer manufactured by Rohpoint. The pH, surface tension and viscosity of inks were characterized using 827 pH Metrohm meters (Herisau/ Switzerland), Tensiometer K100MK2 Kruss (Hamburg, Germany) and Brookfield DVII (New Jersey, USA), respectively. The optical densities of printed objects were measured at four times with Spectrodensitometer Ihara S900, and the average value is reported. The ink-jet printed images color fastness to washing was determined by ASTM Test Method F 2292–03, in which the measurement repeated four times and the average value is reported. Static contact angle θ of the substrates were measured by a Kruss G40-type contact angle measuring system using distilled water as the probe liquid at temperature and humidity of 25°C and 30%, respectively.

To this end, a small drop of distilled water (2–3 μl) was exposed to the surface of the substrates. The test was done three times, and the average of the measurements was reported.

2.3. Printing of ink on substrate

The ink-jet printing was carried out with a HP DeskJet 5150 printer at 1200 dpi using the ink formulations shown in Table 2 on different papers. The inks pH was adjusted to 7-7.5 by McIlvaine buffer solution [44], which is composed of disodium hydrogen phosphate (82.35 cm^3, 0.2 mol) and citric acid (71.65 cm^3, 0.1 mol), to prevent print-head and cartridge damage. The prepared inks, Ink 1 to Ink 8 in Table 2, were filtered through a 0.45 μm filter and then through a 0.2 μm filter to prevent nozzles clogging. A solid single color area was printed on different white papers, which were subsequently used to evaluate the fastness and print quality. The viscosity values of ink-jet inks were between 3.4 and 5 cps, which are in the acceptable range for ink-jet printing inks [45]. The surface tension values of the formulated inks were in the range of 28-32 mN/m, which are also within the values of typical commercial printing ink-jet inks [46].

2.4. Optical density

The parameter used to quantify the darkness of prints is the optical density determined by $OD = -\log(I/I_0)$ where I_o is the intensity of incident light on a print, and I is the intensity of light reflected from a print. Alternatively, color coordinates such as *L**, *a* and b** can also be used to quantify color density, where *L**is the luminance, *a** represents the red/green balance of a color, and *b** defines the yellow/blue balance.

Table 1: Substrate used for printing

Substrate	Gloss/60°	Gloss/85 °	Finish	Paper	Compatibility
1	50.5	95	High Glossy	Photo Ink-jet Paper	All ink-jet printer
2	5.9	17.8	Wave	Premium Photo Paper	All ink-jet printer
3	9.3	17.6	Satin	Quality Photo Satin Paper Plus	All ink-jet printer
4	43.4	91.8	Glossy	Photo paper Premium Plus	All ink-jet printer
5	35.6	91.4	Glossy	Quality Photo Glossy Paper Plus	All ink-jet printer

Table 2: Ink formulations

	Ink composition	Ink 1	Ink 2	Ink 3	Ink 4	Ink 5	Ink 6	Ink 7	Ink 8
Dye	CI Reactive Blue 21	3%	3%	3%	3%	3%	3%	3%	3%
Penetrant	Diethylene glycol mono butyl ether	10%	10%	10%	10%	10%	10%	10%	10%
Alcohol	Isopropyl alcohol	4%	-	-	-	4%	4%	4%	4%
	Butandiol	-	4%	-	-	-	-	-	-
	Pentandiol	-	-	4%	-	-	-	-	-
	Hexandiol	-	-	-	4%	-	-	-	-
Humectant	Polyethylen glycol 200	15%	15%	15%	15%	-	-	-	-
	Ethylene glycol	-	-	-	-	15%	-	-	-
	Diethylene glycol	-	-	-	-	-	15%	-	-
	Triethylene glycol	-	-	-	-	-	-	15%	-
	Tetra ethylene glycol	-	-	-	-	-	-	-	15%
Co-solvent	2-pyrrolidone	7%	7%	7%	7%	7%	7%	7%	7%
water	De-ionized water	61%	61%	61%	61%	61%	61%	61%	61%

2.5. Water fastness test

The water fastness properties were assessed by using the ASTM /F2292-03 water fastness test. One out of the two prepared sample prints was submerged in distilled water at room temperature for 10 minutes. After taking out the sample from the water, the excess water was allowed to drain off. Then the print sample was placed flat and allowed to fully dry. The color difference in washed and unwashed samples which indicated the water fastness was computed by the CIELAB 1976 color difference formula.

3. Results and discussion

3.1. Morphology of selected ink-jet papers

In this study, five different commercial ink-jet papers with same weight (260 g/m^2) and distinct textures of satin, wave and glossy were selected (Substrates 1-5). The different ink formulations were printed on these substrates to produce an ink-jet image. Since all the commercial ink-jet papers were provided from the market, the formulations of the papers coating were not accessible. Comparing the surface morphology of the selected commercial ink-jet papers with the plain paper by scanning electron micrograph (SEM) proved the complete coating of the selected papers (Figure1).

The SEM image of the glossy papers surface morphology (Subtrates 1, 4 and 5) confirmed that these substrates had nano-porous coating layer with different structures. SEM images of wave and satin papers surface (Substrates 2 and 3) revealed the presence of coating cracks and micro pores, which may enhance the ink absorption behavior. The cracks in the coating layer detrimentally decrease the gloss of the paper [10]. Figure 1 clearly shows that Substrate 2 has a bi-modal pore size distribution with large pores (75 µm) between the particles and fine pores within the particles. Therefore, the porous diameter in coating layer is different and increased correspondingly from nanometer range in glossy papers to micrometer range in satin and wave papers, which may influence the ink absorption behavior.

To determine the absorption of ink in the commercial coated ink-jet papers, the water contact angles were measured. As seen in Table 3, the water contact angle is varied in different commercial coated ink-jet papers.

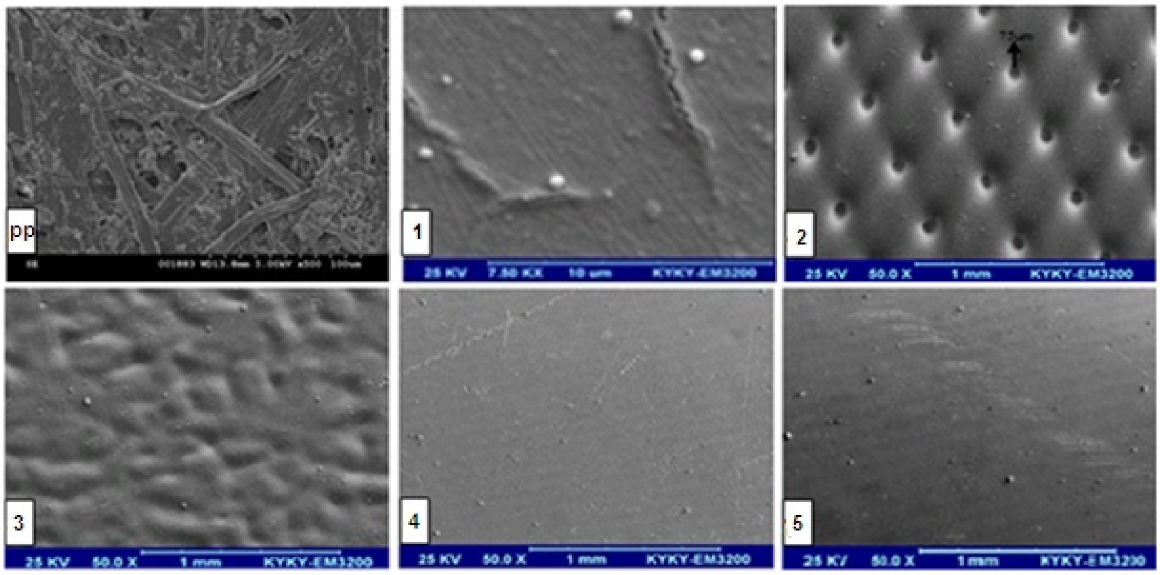

Figure 1: Scanning electron micrograph (SEM) of plain paper (pp) and Substrates 1 to 5.

According to Table 3, the water contact angles in all the coated glossy papers (Substrates 1, 4 and 5) are higher than the wave coated paper (Substrate 2) and satin coated paper (Substrate 3), which may be related to the wettability and surface free energy of the substrates. The presence of large cracks and pores can also be another reason for decreasing the contact angle in Substrates 2 and 3. The work of surface free energy values were calculated according to equations (1) and (2), which is known as the equations of state as proposed by Li and Neumann [47].

$$W_A = \gamma_{sv}\left(1+\cos\theta\right) \quad (1)$$

$$W_A = 2\left(\gamma_{lv}\gamma_{sv}\right)^{1/2}\exp\left[-\beta\left(\gamma_{lv}-\gamma_{sv}\right)^2\right] \quad (2)$$

Where γ_{lv} and γ_{sv} are surface tension of water and surface free energy of the substrate, respectively. θ is water contact angle and the value of β is 0.0001247 ±0.000010 $(mJ/m^2)^{-2}$. To calculate surface free energy, the work of adhesion (wA) was first calculated according to equation (1) (Young's equation). Surface free energy (γ_{sv}) was then calculated from equation (2). The results obtained are shown in Table3. According to Table 3, the surface free energy values of glossy papers (Substrate1, 4, 5) are lower than the wave paper (Substrate 2) and the satin paper (Substrate 3). The lower value of surface free energy of glossy papers is may be caused by the diminution of the polar properties of chemical components in coating layer. These can be decreased the wettability and increased water contact angle and surface hydrophobicity. The water contact angle of Substrate 4 is higher than other coated glossy papers (Substrate 1, 5) contact angles; this might be due to the surface free energy of substrate which is relative to nature of chemical components in nano-pores coating layer and makes the surface paper less hydrophilic. Consequently; by decreasing the water contact angles, the surface free energy of substrate increased and the substrate surface hydrophobicity decreased.

3.2. Effect of alcohol type on the properties of ink-jet printing objects

In order to gain some insight into the influence of different types of alcohol on the printing properties, optical density and water fastness, the optimum ink formulation, which contained C.I. Reactive Blue 21 (Ink 2) was formulated with various alcohols with different number of alkyl groups. So, isopropylalcohol (Ink 1), butandiol (Ink 2),pentandiol (Ink 3) and hexandiol (Ink 4) were tested. Figure 2 shows the optical density of Inks 1 to 4. On all substrates, as the alkyl groups in the alcohol molecules increased, the solubility of the ink in the paper faintly decreased. Due to this decrease, slightly higher optical density was observed.

Table 3: The water contact angle and surface free energy of the substrate.

Substrate	Contact angle	Surface Energy (mN/m)
1	26°	65
2	20°	68
3	25°	66
4	64°	45
5	30°	63

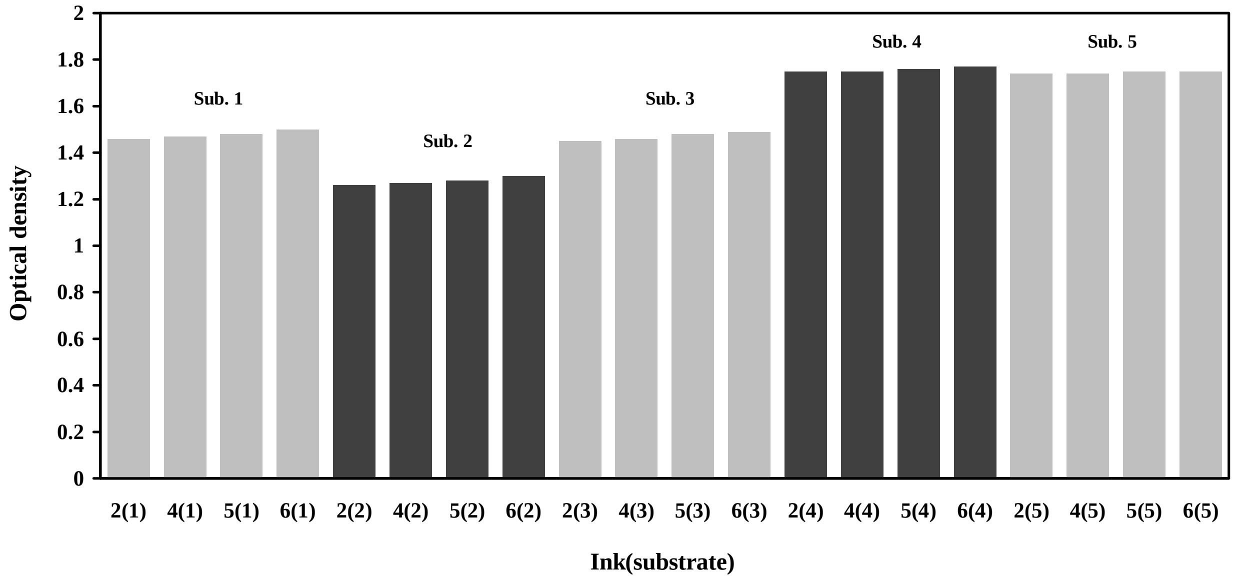

Figure 2: The optical density values for printed objects with Ink 1 to Ink 4 on different substrates (Substrates 1 to 5).

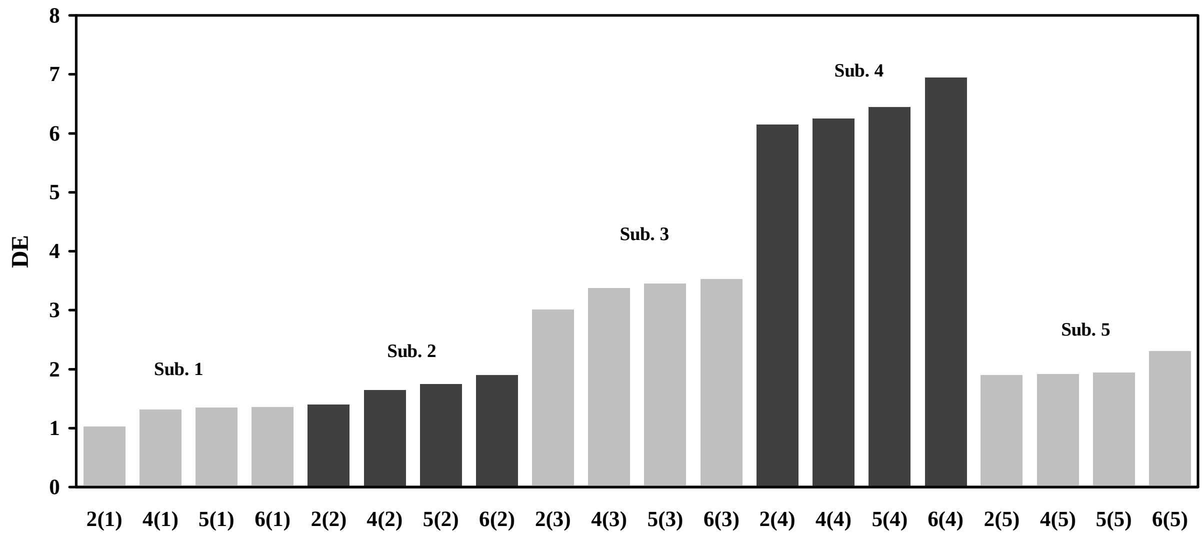

Figure 3: The water fastness results of printed objects with Ink 1 to Ink 4 on different substrates (Substrate 1 to 5).

The optical densities of printed objects are more dependent on the substrate than the alcohol.It can be seen from and Figure 3 that the water fastness results are more dependent on the substrates than the alcohol.

On all substrates, as the alkyl groups in the alcohol molecule increased, the solubility of the ink in the paper faintly decreased. Due to this decrease, the water fastness properties were slightly lower than those observed formulations containing fewer alkyl groups. Significant difference was observed in water fastness properties of Substrate 4 in comparison with other tested inks. This may be either due to the lower surface free energy or lack of chemical interaction between the paper and the ink. Therefore, the ink was not able to penetrate enough into small nano-porosities; hence most of the inks remain on paper surface and easily washed out by submerging in water.

3.3. Effect of humectant type on the properties of ink-jet printing objects

To investigate the influence of the various types of humectants on optical density and water fastness properties, the optimum ink formulation, which contained C.I. Reactive Blue 21 (Ink 1) was formulated with five different humectants with different number of alkyl groups. In this regard, ethylene glycol (Ink 5), di-ethylene glycol (Ink 6), tri-ethylene glycol (Ink 7), tetra-ethylene glycol (Ink 8), and polyethylene glycol 200 (Ink 1) were examined. Figure 4 shows the optical density of Ink1, and Inks 5 to 8. On all substrates, as the alkyl groups in humectant molecules increased, the solubility of the ink in the paper faintly decreased. Due to this decrease, slightly higher optical density of the printed object was observed. Subsequently, the optical density results appeared to be more dependent on the substrates than the types of humectants used in ink formulation.

It can be seen from Figure 5 that the water fastness is more dependent on the substrate than the humectants. On all substrates, as the alkyl groups in the humectant molecules increased, the solubility of the ink in the paper faintly decreased. Due to this decrease, the water fastness properties were slightly lower than those observed for ink formulations containing fewer alkyl groups. A significant difference was observed in water fastness properties of Substrate 4 compared with other tested inks. This may be either due to the lower surface free energy or lack of chemical interaction between paper and ink. Therefore, the ink is not able to penetrate enough into nano-porosities; hence most of the ink remains on the paper.

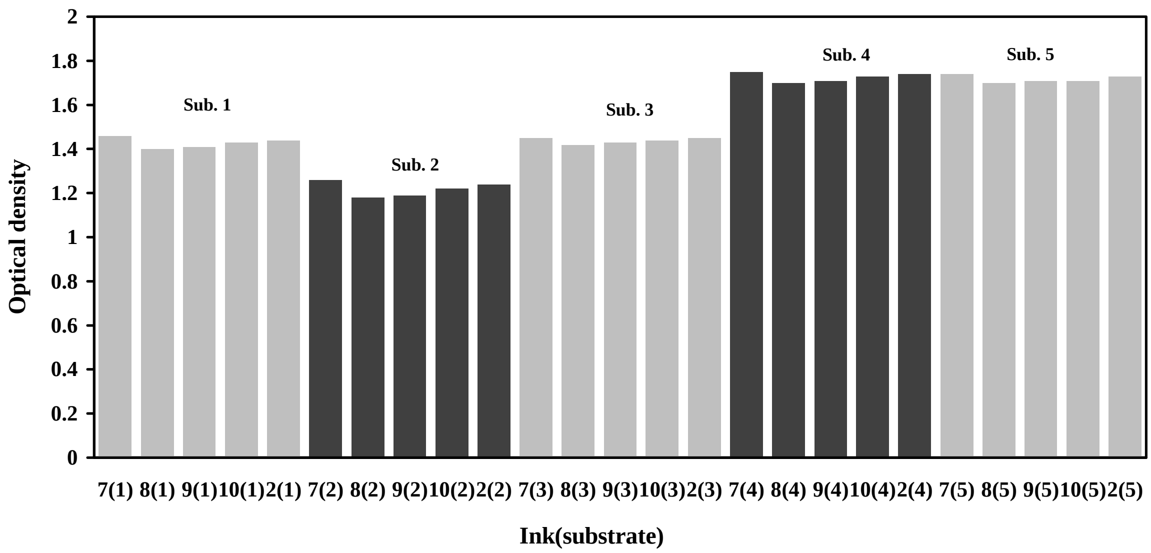

Figure 4: The optical density value of printed objects with Ink1, Ink5 to Ink8 on different substrates (Substrate 1 to Substrate 5).

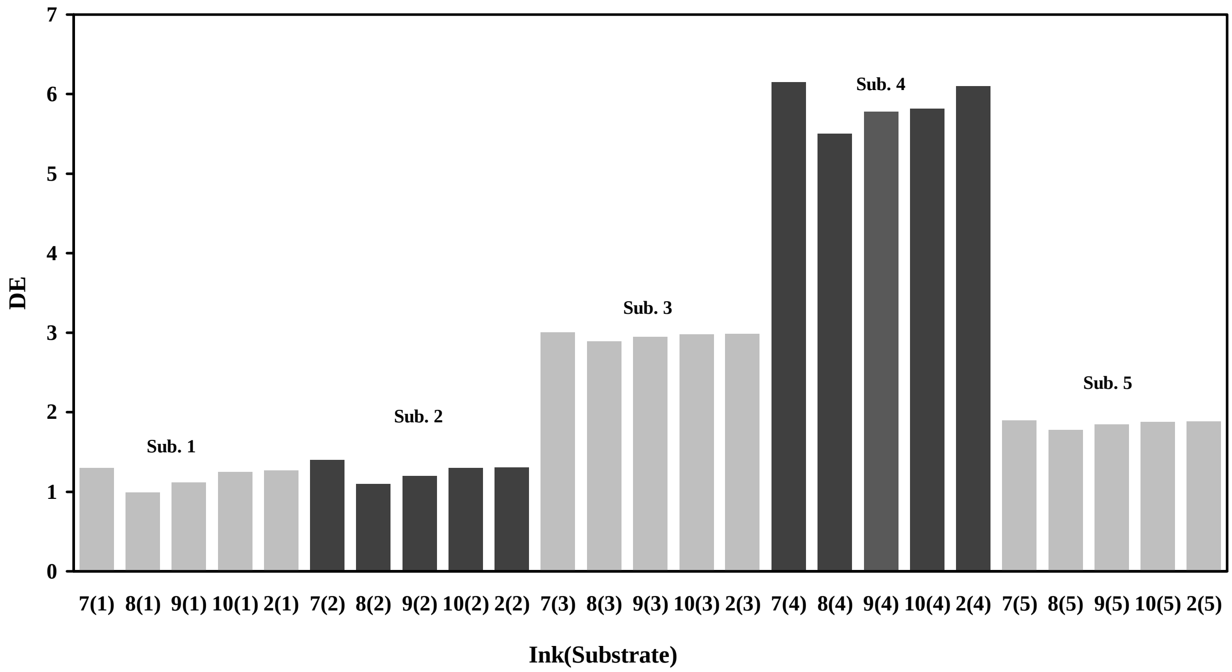

Figure 5: The water fastness results of printed objects with Ink 1, Ink 5 to Ink 8 on different substrates (Substrate 1 to Substrate 5).

4. Conclusions

The effect of ink formulation and paper surface morphology was investigated on the ink-jet printing properties. Five different papers (Substrate 1 to 5) with the same weight were printed on by eight different inks. To evaluate the effect of alcohol and humectant of the ink on the ink-jet prints, the optimum ink formulation containing C.I. Reactive Blue 21 was formulated with various alcohols and humectants by varying their number of alkyl groups. On all substrates, as the alkyl groups in the alcohol and humectant molecules increased, the solubility of the ink in the paper faintly decreased. Due to this decrease, slightly higher optical density of the printed object was observed and the water fastness properties were slightly lower than those observed for ink formulations containing fewer alkyl groups. Therefore, altering alcohols and humectants in the ink formulation did not show significant difference in optical density and water fastness properties of ink-jet prints. On the other hand, different substrates displayed various properties. The optical density and water fastness results depend on the chemical interaction between paper and ink, which can be governed by the surface free energy, the porosity and the pore size of the papers. Therefore, the type of paper played more important role in the ink jet printing properties compared to the ink formulation.

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