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PRINTABILITY AND QUALITY OF PAPERS COATED WITH DIFFERENT BINDERS

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Abstract: *Papers are the most commonly used substrates. A printable paper must have certain properties. These properties include surface smoothness, air permeability, surface energy, colour of the paper, opacity, whiteness, light fastness, gloss, and coefficient of elongation under force. In addition, a good printable paper should not allow ink to pass between its two surfaces. After the papers are produced, they are subjected to surface treatments in order to improve the above-mentioned properties and to provide extra specification. Surface treatments include calendering, surface sizing and coating processes. While filling the recesses and protrusions on the surface with the coating process, the paper's affinity for the ink is increased, and the penetration of the ink to the other surface of the paper is prevented. In the coating process basically, a filler is dispersed in a binder. In this study, it is aimed to obtain the highest quality and most printable paper by changing the type of binder used in the coating. In this study, equal amount (5% w/w) titanium dioxide filler was used in all coating formulations. In coating formulations; cationic starch, hydroxy ethyl cellulose and polyvinyl alcohol polymers were used as binders. Binder amounts are adjusted according to optimum viscosity. The obtained coating formulations were coated on the paper surface with a laboratory type coating device. Colour, gloss, surface smoothness, air permeability, surface energy and surface morphology of the coated papers were determined. It was printed with magenta offset printing ink on three different types of paper coated with different binders, using the IGT C1 offset printability test device, under 400 N/m^2 pressure printing conditions. Colour and gloss measurements of the prints were made. As a result; It was determined that three different binders improved the printability parameters.*

Keywords: printability, paper coating, binder, paper quality

1. INTRODUCTION

Paper and writing symbolize civilization and the future, and no matter how widespread digitalization is in our age, paper consumption will not decrease. Paper use has never decreased from the past to the present. Over time, there have been changes in paper usage preferences and the expected features of paper. Today, with the development of the packaging and label industry and the advertising industry, expectations from paper have also been important developments. Paper manufacturers and the printing industry have also focused on improving printability and many other paper properties to meet expectations. Developments in ink and paper, which are the basic raw materials of the printing industry, will also directly affect printability. Today, especially in the packaging and label industry, paper and cardboard are expected to have additional features. At this point, the simplest process is to improve the surface properties. The processes required to provide the desired properties from the paper are sizing, coating, and calendering.

There are two sizing processes in the paper industry. The first is the addition of internal sizing agents (wet end) to the pulp during production, and the second is surface sizing applied to the paper surface after production (Ginebreda et al., 2011). The sizing process mentioned here is applied to the surface. In the sizing process, the process of filling the pores on the surface is filled by covering the paper with a binder. Starch, polyvinyl alcohol and cellulose derivatives are commonly preferred in sizing processes. Starch is often used in situations in which smoothness and gloss are required for high quality prints. Starch is also used as an adhesive to bond surface fibres, increase paper strength and stiffness and improve offset printability and dimensional stability.

Although starch consumption by the paper industry is high, many synthetic materials can replace starch. The cost and performance characteristics of the substitutes to be used are considered, as well as their sensitivity to environmental factors. However, the use of starch has several advantages (Bamiller, 1997). Starch is a high molecular weight natural polymer that can be depolymerized to a large extent with control. It is a hydrophilic polymer that disperses in water and binds to cellulose fibres and pigments via hydrogen bonding. The key factors in choosing starch are the relatively low cost of the raw material, the fact that it is derived from a renewable resource, and its biodegradability (Bajpai, 2012; Maurer & Kearney, 1998).

PVA is a white granular synthetic polymer that is water soluble and is considered to be one of the strongest binders available in the paper industry. The PVA production process begins with polymerization of the vinyl acetate monomer through a free radical reaction to polyvinyl acetate. Polyvinyl acetate is then hydrolyzed to PVA via a base-catalysed saponification reaction (Gigac et al., 2016; Fatehi et al., 2009).

Cellulose is a widely used bio-based material in the paper industry and in various other industries (Abdel-Rahman et al., 2013; Hashem et al., 2011). Cellulose is the main material used in paper production. In addition to being the main materials, cellulose-based polymers are used as additives in both wet and dry surface applications in the paper industry. Thus, the ink to be printed on the paper was prevented from penetrating the paper. Thus, a higher printing brightness is achieved with less ink (Cao et al., 2009). Chemical modifications have been performed to increase the solubility of cellulose and its derivatives (El-Shafei et al., 2008). In addition to cellulose ether, hydroxyethyl cellulose (HEC) is frequently used in industry because of its biocompatibility, which improves the workability and adhesion of fresh material to the material (Azzaoui et al., 2015).

Hydroxyethyl cellulose (HEC) is a non-ionic water-soluble polymer. This polymer has excellent performance properties, such as the ability to thicken, bind, emulsify, suspend, disperse, stabilize, in addition to the ability to retain water and form film, which provides good protective effect. HEC dissolve easily in hot or cold water to produce solutions with a wide range of viscosities. Hydroxyethyl cellulose has applications in different industrial areas, such as thickening dyes (Dal-Bó et al., 2011), textile finishing (Gorgieva & Kokol, 2011), thickeners in cement mortar (Patural et al., 2011) and sizing agents in paper production (Kugge et al., 2004). HEC samples are often modified by ionic or hydrophobic groups to optimize their properties for various industrial applications. This modification causes an increase in the viscosity and is often thixotropic. Such systems can be further improved by adding small amounts of surfactants (Kästner et al., 1996)

In the coating process, a mixture of pigment, binder, and additives are coated on the paper surface. The main difference of the coating process as a mixture from the sizing process is that it contains pigment and therefore it is also referred to as pigment coating in some sources in the literature. Coating is the main industrial process used by the paper industry to improve the appearance and printability of paper, and accounts for 50% of the total chemical additive used worldwide (Ginebreda et al., 2011). Pigmented paper coating is usually accomplished using a high solids aqueous suspension of inorganic pigments and binders. In this process, basic materials other than the pigment, namely binders, are additives that aim to improve properties such as surface strength, gloss, waterproofing, print compatibility (Duan et al., 1999) and paper speed. As in the sizing process, the most common binders for paper coating are starch, polyvinyl alcohol, and cellulose. Binders can be used either alone or in combination with others such as starch.

Better printability properties are obtained using the paper surface coating process. These features can be counted as smoothness, opacity, surface smoothness, thickness, brightness, whiteness, non-tearing, and non-breaking (Ozcan & Zelzele, 2017). Coating formulations usually consist of inorganic pigments such as kaolin-clay, calcium carbonate, titanium dioxide, binders, dispersants and some other additives (Morsy et al., 2016; Ozcan et al., 2019). In addition to the above-mentioned pigments for coating, many different pigments are added to the formulation, and experiments are being carried out today (Mansour et al., 2000).

One or more pigments could be used in the coating process. The desired properties of pigments used in coatings include chemical resistance, compatibility with other components, appropriate particle size and shape distribution, purity, low density, gloss, opacity, good flow properties and low binder requirements. Binders are necessary for the pigment particles to fill the pores of the paper; that is, they act as a type of glue and determine the rheological properties of the coating. 5-20% binder by weight is used in the coating formulation (Lee et al., 2002). The properties required of binders are good adhesion, compatibility with other compounds, optical and mechanical properties, chemical and mechanical stability, and durability. Natural or synthetic polymers and latex derivatives are used the coating processes. Starch is one of the most commonly used binders owing to its easy supply and cost (Shogren, 1998). Polyvinyl alcohol is also one of the strongest binders that can be dissolved in water and used in the paper industry.

In this study, we aimed to obtain the highest quality and printability of the coated paper by changing the binder type used in the coating. In this study, equal amounts (5% w/w) of titanium dioxide pigment were used in all the coating formulations. Cationic starch, hydroxy ethyl cellulose, and polyvinyl alcohol polymers were used as binders in the coating formulations.

2. METHODS

The technical properties of the base paper used in this study are listed in Table 1. Cationic starch, hydroxyethyl cellulose (HEC) and polyvinyl alcohol (PVA) were purchased from Sigma-Aldrich and their properties are listed Table 2. TiO2 was purchased from Sigma-Aldrich. The Frimpeks UV Offset Process Magenta 22355 commercial offset printing ink was obtained from Frimpeks (İstanbul, Turkey).

Table 1: Technical properties of base paper used in the study

	Standard	Base paper
Grams per square meter (g/m^2)	ISO 536	70
Thickness (µm)	TAPPI T411	165
Whiteness (D65/10) (%)	ASTM E313	96
Gloss (75°)	ISO 8254-1	5.5
Yellowness	ASTM E313	0.06

Table 2: Technical properties of cationic starch, hydroxyethyl cellulose and polyvinyl alcohol used in the study

	Cationic starch	HEC	PVA
Appearance (color)	White	White to faint Beige	White to off-white
Appearance (form)	Powder	Powder	Powder
Viscosity (cps, 20°C)	120	80-125 (2% in H_2O)	120
Fineness %	>98	>98	>98
pH	6.0	6.0-8.5	6.0

2.1 Paper Sizing and Coating

Cationic starch, PVA, and HEC based surface sizing mixtures were applied to base paper. The sizing formulation applied consisted of 0.25% concentration of HEC binder, 7.5% cationic starch, 7.5% PVA in water which were heated up to 95 °C and stirred at 250 rpm with a mechanical stirrer for a while, and the resulting hot mixture surface sizing solution was cooled to 60 °C and then applied on to the paper surface using the Mayer rod with #2 in a laboratory-type paper coating machine. The same preparation process was repeated for the paper coating formulations, when the sizing mixtures cooled at 40 °C 5% TiO2 pigment was also added to the mixtures. The formulations were applied to the paper surface using a Mayer rod in a laboratory-type paper coating machine under laboratory conditions. All samples were conditioned at 50% RH and temperature of 23 °C for at least 48 h before characterization.

2.2 Contact Angle, surface energy, paper roughness and air permeance measurements

The contact angle and total surface energy measurements of the papers were performed using PocketGoniometer PGX+ in accordance with the ASTM D5946 standard. The paper roughness and air permeance of all papers were determined using Lorentzen & Wettre (L&W) in accordance with the ISO 8791-2:2013 - Part 2: Bendtsen method.

2.3 Printing and spectrophotometric properties

Base paper, sized papers and coated papers were printed using Frimpeks UV Offset Process Magenta 22355 commercial offset printing ink using an IGT CG1 offset printability test device under 400 N/m2 pressure printing conditions. All the samples were cured using an IGT UV curing device after printing. Then, the spectrophotometric properties of the papers were determined using CIEL*a*b* colour values by using an X-Rite eXact spectrophotometer according to the ISO 13655:2017 standard. The measurement conditions of the spectrophotometer were determined as polarization filter with 0/45° geometry with 2° observer angle and D50 light source in the range of 400-700 nm. The difference between the colours of the different prints was calculated according to the CIE ΔE 2000 colour-difference formula ISO 11664-6:2014. Calculations were performed by calculating the average of five measurements. ΔL*, Δa*, and Δb*: Difference in L*, a*, and b* values between the specimen and target colours, respectively. The lightness is represented by the L* axis, which ranges from white to black. The red area is connected to green by the a* axis, whereas the b* axis runs from yellow to blue.

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

where ΔL*, ΔC*, and ΔH* are the CIEL*a*b* metric lightness, chroma, and hue differences, respectively, calculated between the standard and sample in a pair, and ΔR is the interaction term between the chroma and hue differences. SL, SC, and SH are the weighting functions for lightness, chroma, and hue components, respectively. The values calculated for these functions vary according to the positions of the sample pair considered in the CIEL*a*b* colour space. The kL, kC, and kH values are the parametric factors to be adjusted according to different viewing parameters such as textures, backgrounds, and separations, for the lightness, chroma, and hue components, respectively (Bates et al., 2012).

2.4 Gloss

The gloss measurements of all papers were carried out with the BYK Gardner GmbH micro gloss 75° geometry in accordance with ISO 8254-1:2009, and the gloss measurements of prints with the BYK Gardner GmbH micro-Tri-gloss 60° geometry in accordance with ISO 2813:2014.

3. RESULTS

The contact angle and total surface energy measurements of the papers were performed using PocketGoniometer PGX+ in accordance with the ASTM D5946 standard. The measurement results are presented in Table 3. When the contact angle results are examined, the contact angle value of the base paper of 70.2°, which is compatible with the literature (Shen et al., 2000), has been reduced by all sizing and coating processes, that is, better wetting of the paper is ensured, and it is possible to print with less water (Crowe et al., 2021). When the sizing agents are evaluated among themselves, it has been determined that the contact angle increases with the order of HEC, cationic starch and PVA. This is due to the number of OH bonds. Polymers with more OH groups make more H bonds and lower the contact angle. The results are consistent with the literature (Ellison et al., 1953). Titanium dioxide in coating formulations has also provided a lower contact angle than you, which is lower than base paper. Because TiO2 has increased absorbency with its water-loving structure.

Table 3 (part 1): Total surface energy and contact angle values of all papers according to ASTM D5946 method

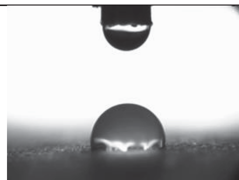
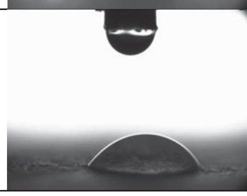
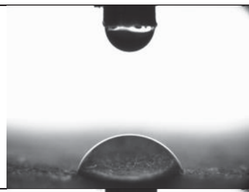
Paper samples	Total surface energy (mJ/m^2)	Contact Angle	**Images**
Base paper	39.6	70.2	
Cationic starch sized paper	47.5	48.5	
Cationic starch+TiO_2 coated paper	51.2	38.3	

Table 3 (part 2): Total surface energy and contact angle values of all papers according to ASTM D5946 method

PVA sized paper	42.3	62.8	
PVA+TiO_2 coated paper	43.9	58.6	
HEC sized paper	50.6	40.1	
HEC+TiO_2 coated paper	50.7	39.6	

The paper roughness and air permeance of all papers were determined using Lorentzen & Wettre (L&W) in accordance with the ISO 8791-2:2013 - Part 2: Bendtsen method. The measurement results are listed in Table 4. The surface roughness of the papers is the parameter that affects the print quality the most. Because ink is lost in rough papers and ink cannot reach thinner spaces. For this reason, it is one of the features that should be checked in papers. When Table 4 is examined, it has been determined that the roughness of the base paper decreases with the processes that have the highest surface roughness. The surface roughness of the coatings is higher than the size papers. This is due to the fact that the pigment creates a small roughness on the surface. The air permeability has been reduced during the processes and is in the same direction as the surface roughness. The results are in agreement with the literature (Karlović et al., 2010).

Table 4: Paper roughness and air permeance measurements for all papers

	Roughness (ml/min)	Air permeance (ml/min)
Base paper	225	821
Cationic starch sized paper	71	94
Cationic starch+TiO_2 coated paper	150	142
PVA sized paper	46	28
PVA+TiO_2 coated paper	58	34
HEC sized paper	55	65
HEC+TiO_2 coated paper	78	174

Paper coatings containing different binders were successfully prepared and coated on the paper surfaces. Then, magenta colour prints were obtained under 400 N/m2 pressure conditions using the IGT CG1 offset printability test device. All the samples were cured using IGT UV curing device after printing. Spectrophotometric colour measurements of all the papers were then performed, and the measurement values are listed in Table 5. When the results are examined, it is seen that all colour changes are below

140, that is, the two colours cannot be distinguished from each other with the eye. This occurs because the ink dries too quickly. Colour shifted to some yellow in all sizing and coating processes. It was concluded that the double bonds in these polymeric binders are due to the chromophore property. The relevant results are consistent with the literature (Ozcan et al., 2020). In addition, higher density was obtained than uncoated papers with equal amounts of ink. Because the polymeric film formed on the surface prevented the ink from entering the paper.

Table 5: Spectrophotometric colour and colour difference values of the papers used in the study

	Density	L*	a*	b*	ΔE00
Base paper	1.10	41.71	69.79	3.08	
Cationic starch sized paper	1.60	41.45	69.19	2.82	0.45
Cationic starch+TiO_2 coated paper	1.38	41.90	68.77	2.12	0.43
PVA sized paper	1.61	41.83	67.34	2.23	1.06
PVA+TiO_2 coated paper	1.48	41.94	67.14	2.17	0.77
HEC sized paper	1.60	42.07	67.48	2.65	1.04
HEC+TiO_2 coated paper	1.60	41.94	67.11	2.27	1.38

The gloss measurements of all papers were carried out with the BYK Gardner GmbH micro gloss 75° geometry in accordance with ISO 8254-1:2009, gloss measurement results of which are shown in Figure 1, and the gloss measurements of prints with the BYK Gardner GmbH micro-Tri-gloss 60° geometry in accordance with ISO 2813:2014. The gloss measurement results for magenta printed papers are shown in Figure 2. When the gloss values of the base and coated papers were examined, it was observed that the gloss values increased in all the sized coated papers. When the coated papers were examined, it was found that the gloss value of sized papers higher than coated papers. Because the pigment creates a slight roughness on the surface. This also reduces the gloss a bit. The increase in gloss in sizing and coating is due to the binders closing the gaps on the surface and reducing the surface indentations. The results are consistent with the literature (Elsayad et al., 2001). When the printing glosses are examined, the gloss decreased in all coated, uncoated and sized papers. Because the pigment in the ink created a slight roughness on the surface, which reduced the gloss.

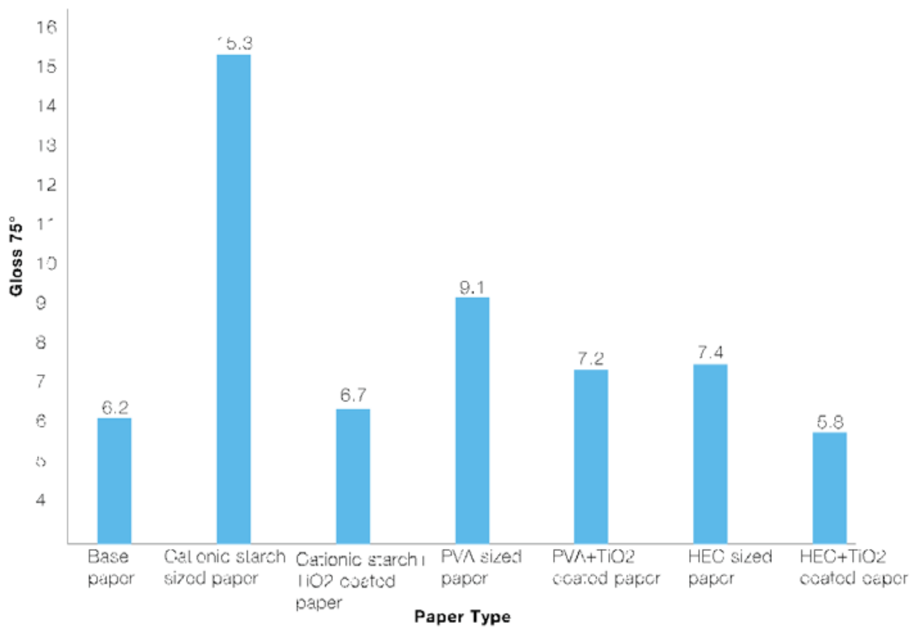


Figure 1: Gloss values of papers according to ISO 8254-1:2009

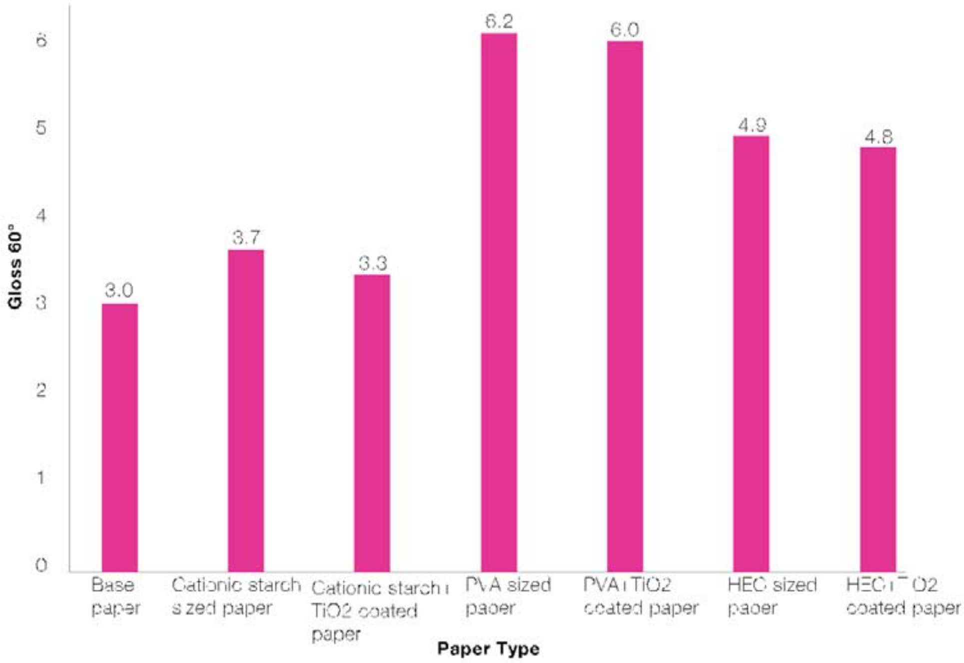


Figure 2: Gloss values of magenta printed papers according to ISO 8254-1:2009

4. CONCLUSIONS

In the paper industry, additives are extensively used for two main purposes. The first is to provide higher productivity by using less water and energy, thus contributing to minimizing the environmental impact of paper. The second is to provide the desired quality features to the final product. Paper is useful in many areas of daily live. Therefore, production and consumption are normal at a global scale. The additives used in the paper and cardboard industry should be made by considering the cost factor as well as the cost factor in the preference of binders and pigments. At this point, the choice of sustainable binders with biocompatibility, low cost, and high performance in addition to them is crucial.

As a result, it has been determined that better wetting is provided for more printing with the coatings made, the colour change is very low in the printing where the gloss is increased, and the surface roughness is reduced. Thus, the print quality parameters were increased and it was found that better quality prints could be obtained with all sizing and coatings. When the binders were compared among themselves, it was determined that the best results were obtained by cationic starch and HEC, but considering the cost, the best performance could be obtained from the cheaper cationic starch.

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