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Printing Effects of Different Natural Colourants on Cotton Fabric

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ABSTRACT
Natural colourants were widely employed in the ancient world to colour cotton garments. With the revolution of synthetic dyes, natural dyes have lost their position in respect of some performance. However, nowadays the demand for sustainable and eco-friendly products is increasing worldwide due to the awareness of ecological balance. The purpose of this study is to utilize natural colourants in printing on cotton fabric. In this regard, the colourants were extracted from myrobalan fruit, pomegranate peel and catechu, which were eventually applied to the fabric by screen printing technique. The print paste was prepared with carboxymethyl cellulose (CMC) as a natural thickener and polyvinyl alcohol (PVA) as a synthetic thickener. The stability of the print paste was studied with respect to time elapse by viscosity measurement. The depth of shade and colour fastness of the printed fabric were measured. K/S value was evaluated with respect to the stability of print paste. The viscosity was changed slightly in both the CMC and PVA thickener with respect to the stability of the print paste. The washing fastness grades for colour staining wash ranged from 4 to 5, rubbing fastness grades of dry rubbing also ranged from 4 to 5, wet rubbing fastness was 3/4 to 4/5 as well as perspiration fastness grades in alkaline, and in acidic it was 3/4 to 5. Thus, myrobalan fruit, pomegranate peel and catechu have shown excellent performance in printing on cotton textiles, which could be an alternative to the synthetic counterpart.

KEYWORDS
natural colourants, screen printing, viscosity, fastness, K/S value, cotton fibre

INTRODUCTION

Dye or colourant is a gorgeous material used for colouring fibres and fabrics that we use in our daily life. Natural and man-made dyes are the two main categories of dyes or colourants used in textile dyeing and printing (i.e., synthetic dyes). Natural dyes are derived from plant roots, fronds, flowers, fruits, animals, minerals, microbial, and fungal sources, and have been utilized in textiles since antiquity [1,2]. But it is very difficult to get the required shade by using natural colourants, which are showing poor colour fastness properties as well; at the same time, it is highly scarce and expensive. Moreover, only a small percentage of the population, particularly the aristocratic families, could afford it. To overcome these drawbacks, synthetic dye was introduced in 1856 and these dyes are mainly used to dye or print the fibre and fabric substances due to their superior properties to the naturally extracted dyes [3-5]. A huge number of synthetic dyes, which are capable to cover every possible shade and show superior fastness properties, are available on the market, and their subsequent commercialization had them replace natural dyes. As a result, natural dye use and application for textiles has substantially dropped. Synthetic dyes are made from a variety of chemicals and compounds that are readily available and inexpensive for commercial usage, but they are extremely hazardous to the environment and human health since they contain a large number of poisonous and allergenic components. As a result, environmental concerns are growing around the world. To escape the hazardous effects of synthetic dyes, at the present time researchers are trying to use natural colourants for coloration of textile substances, which are renewable, less dangerous to the environment, sustainable, non-toxic, hypoallergenic and eco-friendly [6-9].

Dyeing and printing are the two major colouration techniques for textiles. The exhaust dyeing of textiles is mainly characterized by the exploitation of a large amount of water with higher doses of chemicals to produce a single-colour effect. In this process, the textile material should exhibit a satisfactory level of absorbency, resulting in huge amount of toxic effluent generation. Printing, on the other hand, is the localized application of colour, with a view to obtain the multicolour effect in the fabric with less consumption of water. Consequently, the effluent generation in printing is low, creating less pollution load on environment. In addition, printing of textiles can be applied on a well prepared, grey or even previously coloured fabric. In exhaust dyeing, it is crucial to maintain the temperature and the chemical environment of the dyebath, whereas printing is easy and simple in operation [10,11,13].

Natural dye screen printing has become an efficient bridge between traditional printing processes and textile mass manufacturing [12]. As natural colourants, myrobalan fruit, pomegranate peel, and catechu, were employed in this study to print cotton fabric at optimal temperature, pH, and duration. Natural dyes or colourants have traditionally been preferred for their soothing colours, and it has been observed that screen printing on cotton fabrics with myrobalan fruit, pomegranate peel, and catechu has produced promising colours, making it a viable alternative to harmful synthetic dyes. The focus of this study is on the utilization of traditional patterns for screen printing with natural colourants. Because of new supplies of natural colourants and new trends in fashion application, this research could show a creative way of designing textile with screen printing, resulting in a global niche market for textile colouring. The major significance of this research is to increase the worth of textiles by increasing their aesthetic functions and commercial values, and the use of these approaches can be used as a new, full, or effective strategy for creating unique fabric designs. Very little research has been conducted on printing with natural colourants and this research contributes to the knowledge of plant-based colourants for screen printing. Printing with natural colourants is important in today's world for environmental reasons and the renovation of a cultural art form. It also supports sustainable practices, and printing on eco-friendly fabrics helps to understand the properties of these comparatively new fabrics, which may lead to increased demand. Therefore, the focus of this research was to develop a sustainable, eco-friendly way to screen printing on cotton fabrics with natural colourants.

EXPERIMENTAL

Materials

100% cotton (plain weave) fabric, thickeners such as carboxymethyl cellulose (CMC), polyvinyl alcohol (PVA) and raw materials of natural dyestuff, precisely myrobalan fruit (Terminalia chebula), pomegranate peel (Punica granatum) and catechu (Acacia catechu), were collected from Aarong Production Centre, Dhaka, Bangladesh.

Dyestuff Extraction and Printing Process

Aqueous method was used for dyes extraction published previously [1,6]. Briefly, the fruit (myrobalan)/peel (pomegranate)/wood (catechu) was grinded in the grinder machine. The dye solution was prepared with the required amount of colourants (powder form) and 200 ml of deionized water at boiling temperature for 30 min and the Ph was neutral. 5 gm, 10 gm and 15 gm of colourants was utilized for the light, medium and deep shades, respectively. In the printing paste, natural thickening agent (CMC gum powder) and synthetic thickening agent (PVA) were used. The printing was done by flat (hand) screen printing technique [12,13]. Following printing, the fabrics were dried in a laboratory drying machine at 100 °C for 10 min before being steam fixed at 150 °C for 15 min. Finally, soaping was used to remove unfixed/unreacted dyes and chemicals from the fabric's surface.

a) b) c) d) e) f)

Figure 1. Shade percentage variation for myrobalan fruit colourant with CMC and PVA thickener (a, d = 5 gm of dyes for a light shade), (b, e = 10 gm of dyes for a medium shade) and (c, f = 15 gm dyes for a dark shade)

a) b) c) d) e) f)

Figure 2. Shade percentage variation for pomegranate peel colourant with CMC & PVA thickener (a, d = 5 gm of dyes for a light shade), (b, e = 10 gm of dyes for a medium shade) and (c, f = 15 gm dyes for a dark shade)

a) b) c) d) e) f)

Figure 3. Shade percentage variation for catechu colourant with CMC & PVA thickener (a, d = 5 gm of dyes for a light shade), (b, e = 10 gm of dyes for a medium shade) and (c, f = 15 gm dyes for a dark shade)

Characterization

The viscosity of printing paste was measured by determining the velocity of flow through a capillary tube or Ostwald viscometer commonly known as U-tube viscometer where viscosity can be calculated through kinematic viscosity measurement equation (η_y=η_w d_y t_y / d_w t_w) [15]. Colour fastness to wash, rubbing, and perspiration were also observed according to ISO 105-C06, ISO 105×12, and ISO 105 E04 standard respectively [13,24]. The viscosity measurement printed samples were analysed to measure the K/S values by Data colour 850 (Reflectance spectrophotometer) where the measuring point of wavelength was 400 nm and the K/S value was calculated from reflectance *R*, commonly known as Kubelka-Munk equation (K/S = (1-R) 2/ 2R) [26].

RESULTS AND DISCUSSION

Viscosity of the printing paste

Viscosity is the measurement of a liquid's resistance to flow when forced. It is the material property which relates the viscous stresses in a substance to the rate of change of a deformation [13,17]. It is often measured by determining the velocity of flow through a capillary tube or an Ostwald viscometer, also known as a U-tube viscometer, which is then used to determine the viscosity of a liquid with a known density. However, this method is not appropriate for print pastes, and for this reason, a 1% stock solution was prepared for measuring the viscosity. The way of determining viscosity with this instrument entails measuring the time it takes for a known volume of liquid to flow through the capillary under gravity's influence, and the instrument must first be fine-tuned with materials of known viscosity, such as pure water and, this way, another value of viscosity can be calculated through kinematic viscosity measurement equation [15,16]. The viscosity of print paste changes as the rate of shear changes during shearing that occurs in the printing process, which determines the quality of the print. The force required to flow at a given velocity increases with the viscosity of the liquid, and a colloidal solution results in a significant rise in viscosity. The printing paste stability should be kept good because the thickener has a high molecular weight and, on standing, it may experience fractional crystallization, disrupting the flow property and concentration of the dye in the printing paste, interfering with the dye's regular distribution [13].

a) b) c) d)

Figure 4. Printing paste stability samples for the myrobalan fruit colourant with the CMC thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

a) b) c) d)

Figure 5. Printing paste stability samples for the myrobalan fruit colourant with the PVA thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

a) b) c) d)

Figure 6. Printing paste stability samples for the pomegranate peel colourant with the CMC thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

a) b) c) d)

Figure 7. Printing paste stability samples for the pomegranate peel colourant with the PVA thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

a) b) c) d)

Figure 8. Printing paste stability samples for the catechu colourant with the CMC thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

a) b) c) d)

Figure 9. Printing paste stability samples for the catechu colourant with the CMC thickener within one hour (a= sample after 15 min, b= sample after 30 min, c= sample after 45 min and d =sample after 60 min)

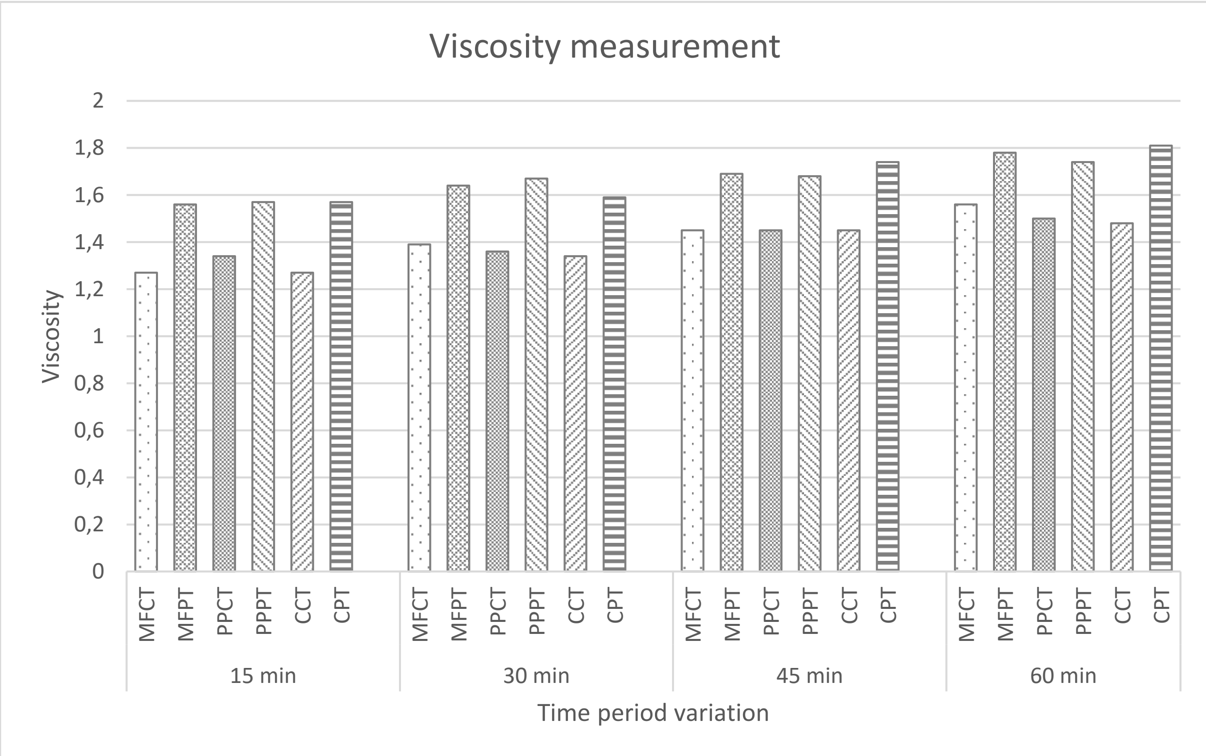

Figure 10. Viscosity of the printing paste for selected natural colorants with the CMC and PVA thickeners within one hour of standing (MFCT=Myrobalan fruit colorant with CMC thickener, MFPT= Myrobalan fruit colorant with PVA thickener, PPCT= Pomegranate peel colorant with CMC thickener, PPPT= Pomegranate peel colorant with PVA thickener, CCT= Catechu colorant with CMC thickener, CPT= Catechu colorant with PVA thickener)

In screen printing, the paste flows through the open pores of the screen onto the cloth, creating a hydrodynamic pressure between the blade and the screen. The created hydrodynamic pressure must be sufficient to fill the screen pores with paste, assisted by capillary forces drawing the paste, and to facilitate the transfer of a suitable volume of paste onto the fabric, with no paste passing through the screen before or after the actual printing transfer. This causes the creation and extension of numerous paste threads, which results in little mounds of colour on the film surface,

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resulting in a patchy look that impacts the fabric surface [13,18-20]. For this reason, the stability of the print paste of a thickener was studied with respect to time elapse by viscosity measurement. In the case of the printing paste on standing for one hour, the stability of the print paste was observed at different periods, specifically 15 min, 30 min, 45 min and 60 min and the concentration of dyes, thickeners and water for all the colourants was 10 gm, 5 gm (CMC) or 5 ml (PVA) and 100 ml, respectively.

In the case of the printing paste of myrobalan fruit colourant with the CMC thickener it was found that the maximum viscosity was 1.56 mm^2/s with the time of flow being 27 s at 60 min and the minimum viscosity was 1.27 mm^2/s with the time of flow being 22 s at 15 min. Therefore, when the timing distance of the print paste was increased, the viscosity was also increased and all the developed printing samples were darker on one side and slightly lighter on the other. On the other hand, in the case of the printing paste of the myrobalan fruit colourant with the PVA thickener it was found that the maximum viscosity was 1.78 mm^2/s and the time of flow was 28.5 s at 60 min and the minimum viscosity was 1.56 mm^2/s and the time of flow was 25 s at 15 min. Therefore, when the time duration of the print paste was increased, the viscosity was also increased and all the developed printing samples were darker on one side and slightly lighter on the other. The viscosity and times of flow of the printing paste of the myrobalan fruit colourant with the CMC thickener were comparatively lower than those of the printing paste of the myrobalan fruit colourant with the PVA thickener because the PVA thickener exhibits high density, 1.40 g/cm^3, compared to the density of the CMC thickener, which is 1.30 g/cm^3, and the PVA thickener has crosslinked copolymers with colloidal solution in their molecular structure which results in a substantial increase in the viscosity. The developed printing samples were found to be darker in the case of the printing paste of the myrobalan fruit with the CMC thickener compared to the PVA thickener because of the lower viscosity and longer time of flow.

In the case of the printing paste of the pomegranate peel and catechu colourants with the CMC and PVA thickeners, it was found that when the time duration of the print paste was increased, the viscosity was also increased and all the developed printing samples were found to be darker on one side and slightly lighter on the other. Among the three colourants, it was found that the maximum viscosity was 1.81 mm^2/s and the time of flow was 29 s at 60 min, which was the case with the catechu colourant with the PVA thickener and the minimum viscosity was 1.27 mm^2/s and the time of flow was 22 s at 15 min, which was the case with the myrobalan fruit colourant with the CMC thickener. Hence, it was found that the viscosity is changed in both the CMC and PVA thickeners for all the colourants.

Therefore, when the timing distance of the print paste was increased, the viscosity was also increased and all the developed printing samples were darker on one side and slightly lighter on the other both for the CMC and PVA thickeners for all the colourants as the printing paste was left to stand without stirring. However, generally, when the fabric is printed with the colour, the printing paste is not kept for a long time without stirring and the printing paste is mixed properly with a stirrer before being printed on the fabric.

Colour fastness properties

The stability of the colour in printing and its fastness is the most important issue for textile coloration. Colour fastness is commonly indicated as a loss of colour depth or colour shift in the original sample, or as a staining scale, which means that the adjoining material is stained by the colour of the original fabric [13,14,21]. The consumer's most essential characteristic is colour fastness, and most washing tests are conducted at low temperatures for short periods of time. According to the ISO 105-C06 standard, the printed sample was washed in a launder meter laboratory instrument. The sample was 100x40mm, the wash bath contained 4 g/l ECE phosphate reference detergent B, the bath volume was 150 ml, the bath temperature was 40 °C, and the washing period was 45 min. To accomplish washing, 10 stainless steel balls were introduced to each bath, equating to five domestic washings. The sample was washed twice with deionized water before being air dried at room temperature. The table shows the fastness ratings of natural colourants on cotton fabric with two different thickeners. These findings suggest that the washing fastness of the cotton fabric printed with the myrobalan fruit, pomegranate peel and catechu colourant was very good to excellent (4 to 5). The higher the wash fastness rating, the greater the colour depth and strength of the fabric. Washing fastness grades clearly reveal the slight colour staining. Hence, it was observed that the catechu-coloured printing fabrics had lower washing fastness compared to other coloured printing samples.

Table 1. Colour fastness properties for selected natural colourants

Name of natural colour with thickener	Depths of shade	Washing fastness (colour staining)	Perspiration fastness		Rubbing fastness	
			Alkaline	Acidic	Dry	Wet
Myrobalan fruit (CMC thickener)	Light	5	4-5	5	5	4-5
	Medium	5	4-5	4-5	5	4-5
	Dark	4-5	4-5	4-5	4-5	4
Myrobalan fruit (PVA thickener)	Light	5	4-5	5	5	4-5
	Medium	5	4-5	5	5	4-5
	Dark	4-5	4	4-5	4-5	4
Pomegranate peel (CMC thickener)	Light	5	4-5	5	5	4-5
	Medium	5	4-5	5	5	4-5
	Dark	4-5	4	4-5	4-5	4
Pomegranate peel (PVA thickener)	Light	5	4-5	5	5	4-5
	Medium	5	4-5	4-5	5	4-5
	Dark	4-5	4	4-5	4-5	4

Name of natural colour with thickener	Depths of shade	Washing fastness (colour staining)	Perspiration fastness		Rubbing fastness	
			Alkaline	Acidic	Dry	Wet
Catechu (CMC thickener)	Light	4-5	4	4	4-5	4
	Medium	4-5	4	4	4-5	4
	Dark	4	3-4	3-4	4	3-4
Catechu (PVA thickener)	Light	5	4-5	4-5	5	4-5
	Medium	4-5	4	4	4-5	4
	Dark	4	4	4	4	4

A fabric sample stitched between two layers of white cloth was treated with acid and alkaline solutions in order to determine its perspiration fastness [22,24]. The printed samples were perspired in the perspirometer following the ISO 105 E04 standard. The sample size was 100×20 mm and two artificial perspiration solutions (acid and alkaline) were prepared. The composite specimen was wetted in perspiration solution (alkaline) and was kept for 30 min at room temperature, 12.5 kPa pressure was applied for 4 h at room temperature (37 °C ± 2 °C). The pressure was removed and spearmen were separated and dried at a temperature not exceeding 60 °C. The changes of colour and staining and the assumed were measured with grey scale for each test. The same procedure was repeated with the acid solution. Perspiration fastness test was performed both in alkaline and acid conditions. The perspiration colour fastness indicates the stability of colour to perspirations of the fabric print under different conditions. The fastness to perspiration of the cotton fabrics printed with the myrobalan fruit, pomegranate peel and catechu colourants were found to be good to very good (3/4 to 4/5) in the alkaline medium and good to excellent (3/4 to 5) in the acidic medium. Hence, it was observed that the catechu-coloured printing samples had lower perspiration properties compared to other coloured printing samples. Just about all printed samples performed well in acidic and alkaline environments.

The rubbing or crocking fastness test is frequently performed on a range of fabrics to evaluate the transfer of surface colour from the test cloth to a piece of white cotton, which is known as rubbing [23,24]. According to the ISO 105×12 standard, the printed samples were rubbed in a tuber crock meter laboratory instrument. The sample was 100×20 mm in size, and the arm was weighted to provide a constant 9 N load on the sample at all times. A mechanical counter kept track of the completed 10 cycles. The samples were dried once the rubbing fastness tests were completed. Then it was compared to the AATCC grey scale for colour staining. Following the procedure for dry rubbing, one drop of water was added with the pipette to the rubbing cloth before placing it in the crock meter. Because many textiles transfer more colour when the dry rubbing fastness is slightly better than the wet rubbing fastness, moisture may impact the rubbing. Rubbing fastness was tested in both dry and wet settings, with higher crocking colour fastness values indicating greater colour depth and strength on the cloth.

Rubbing fastness of the cotton fabric printed with myrobalan fruit, pomegranate peel and catechu was found very good to excellent (4 to 5) in dry rubbing and wet rubbing fastness was recorded as good to very good (3/4 to 4/5). Hence, the rubbing fastness of catechu-coloured printing samples was found to be lower than that of other colour-printed samples. Nearly all printed samples showed superior results during dry and wet rubbing tests. According to the foregoing findings, printing on cotton fabric with natural colourants is effective, as the fastness tests show that cotton fabric is preferable.

K/S values with respect to the stability of the printing paste

Since prehistoric times, when people realized the need for cloth to cover their bodies, colour has played a significant role in human life. Colour is a visual component of perception that is determined by the spectral composition of radiant energy observed [13,25]. Colour is perceived as a characteristic of a surface. Hue, saturation and lightness are the three main attributes of a surface colour. Colour strength is the measure of the ability of a dye to impart colour to materials. Colour strength is evaluated by light absorption in the visible region of the spectrum. Relative colour strength can be defined as the ratio of K/S values for samples as compared to a standard at same wavelength, which is expressed as percentage. 'K' and 'S' are the absorption and scattering co-efficient of dyed sample and K/S is calculated from reflectance, R, using Kubelka-Munk equation [13,26,27]. The viscosity measurement of printed samples were analysed to measure the K/S values by Data colour 850 (spectrophotometer) and the measuring point of wavelength was 400 nm. After the completion of the printing process, the viscosity measurement of the printed samples was analysed by Data colour 850 (spectrophotometer). For sample analysis, in all cases a sample of 10 gm (15 min) was taken as the standard and we compared the colour strength with respect to such viscosity measurement of printed samples for each dye concentration.

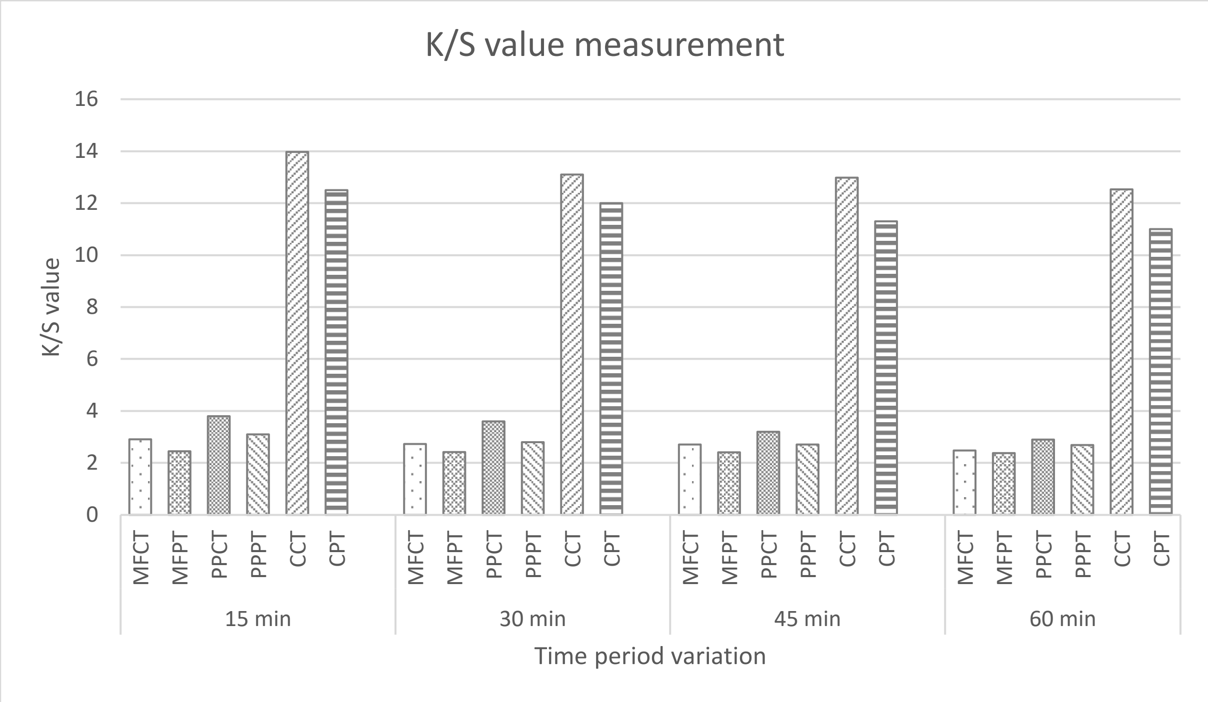

Figure 11. Colour strength of printed samples within one-hour printing paste stability for selected natural colorants with the CMC and PVA thickeners (MFCT=Myrobalan fruit colorant with CMC thickener, MFPT= Myrobalan fruit colorant with PVA thickener, PPCT= Pomegranate peel colorant with CMC thickener, PPPT= Pomegranate peel colorant with PVA thickener, CCT= Catechu colorant with CMC thickener, CPT= Catechu colorant with PVA thickener)

From the graph we can see that the colour strength was influenced by the printing paste stability. The K/S values decreased with the increase of the printing paste stability in all the colourants.

In the case of the myrobalan fruit colourant with the CMC thickener when printing paste stability was within 60 min, the K/S value was 2.48 and the viscosity was 1.56 mm^2/s and when the printing paste stability was within 15 min, the K/S value was 2.91 and the viscosity was 1.27 mm^2/s, which means that the K/S values were decreased with greater viscosity and the increase of printing paste stability. On the other hand, in the case of the myrobalan fruit colourant with the PVA thickener when printing paste stability was within 60 min, the K/S value was 2.38 and the viscosity was 1.78 mm^2/s and when the printing paste stability was within 15 min, the K/S value was 2.45 and the viscosity was 1.56 mm^2/s, which means that K/S values were decreased with greater viscosity and the increase of printing paste stability as it was with the myrobalan fruit colourant with the CMC thickener.

In the case of the pomegranate peel and catechu colourants with the CMC and PVA thickeners, it was also discovered that K/S values were decreased with greater viscosity and the increase of printing paste stability. Comparatively, among the three colourants in this research, it has been found that the highest K/S value was 13.97 with the viscosity of 1.27 mm^2/s at 15 min, which was found for the catechu colourant with the CMC thickener and the lowest K/S value was 2.38 with the viscosity of 1.78

mm^2/s at 60 min, which was found for the myrobalan fruit colourant with the PVA thickener. It can be seen from the above that the K/S values increase when the printing paste stability and viscosity decrease, indicating increased dye uptake with higher concentrations.

CONCLUSIONS

This study validates the utilization of natural colourants which were extracted from the myrobalan fruit, pomegranate peel, and catechu for screen printing on cotton fabric by using carboxymethyl cellulose (CMC) as a natural thickener and polyvinyl alcohol (PVA) as a synthetic thickener. The stability of the print paste with two different thickeners was examined with respect to time elapse by viscosity measurement, which found that when the timing distance of print paste was increased, the viscosity is also increased and all the developed printing samples are darker on one side and slightly lighter on the other. The colour fastness of the printed fabric was in an acceptable range. Hence, it was observed that the catechu-colour printed samples have lower fastness than other colour-printed samples. K/S value was evaluated with respect to the stability of the print paste, and it was revealed that as stability and the viscosity increased the K/S value decreased. The findings of this study may be useful to artisans in the development of their techniques, researchers in the production of additional dyes, and industry experts in the modification of their practices. A further contribution of this project was the advancement of the design scholarship. The research shows that by utilizing carboxymethyl cellulose (CMC) as a natural thickener and polyvinyl alcohol (PVA) as a synthetic thickener, cotton fabric may be successfully screen printed with natural colourants.

Author Contributions

Conceptualization – Rana B, and Bashar MM; methodology – Rana B; formal analysis – Rana B and Bashar MM; investigation – Rana B,; resources – Bashar MM and Rokonuzzaman, B; writing-original draft preparation – Rana B, Bashar MM; writing-review and editing – Hasan Z; Islam MA visualization – Rana B and Islam MA; supervision – Bashar MM. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

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