



Polyanion Biopolymers for Enhancing the Dyeability and Functional Performance of Different Textile Materials using Basic and Natural Dyes



Bouthaina M. Hegazy^a, Hanan A. Othman^a and Ahmed G. Hassabo^{b*}

^aTextile Printing, Dyeing and Finishing Department, Faculty of Applied Arts, Benha University, Benha, Egypt

^bNational Research Centre (Scopus affiliation ID 60014618), Textile Research and Technology Institute, Pre-treatment, and Finishing of Cellulose-based Textiles Department, 33 El-Behouth St. (former El-Tahrir str.), Dokki, P.O. 12622, Giza, Egypt

Abstract

Nowadays back to nature and using biopolymers in all fields especially in textiles are gaining attention to eliminating pollution. Because of the harmful effects of chemical dyeing and the continuing research of researchers in this field, naturally coloured textile fabrics are in high demand all over the world. Natural dyes have unique characteristics such as a soothing hue, biodegradability, non-toxicity, non-carcinogenicity, and antibacterial resistance. The current study aims to reduce chemicals used in textile dyeing and replace them with natural polymers via surface modification of different fabrics. The anionic surface modification aims to enhance the dyeability of various fabrics namely: cotton, wool and acrylic to basic and natural dyes and increase the colour strength and antimicrobial effect. The anionic fabric treatment has been done using sodium alginate (SA) and hydroxyethyl cellulose (HEC) then the treated fabrics were dyed using synthetic dye (basic dye) and natural dye (moringa). In the case of natural dye, crosslinkers have been used (citric acid (CA) with sodium hypophosphite (SHP)) to enhance the bonding forces between dye and fabrics. The optimum condition for polymers' concentration was 2% for the optimum dyeing conditions (45 min.; 75°C; dye conc. 2 g/L; pH 4). The dyeability of the treated fabrics with basic and natural dyes was significantly improved which leads to an increase in the colour strength of dyed fabrics. The results show an increase in fastness properties and antimicrobial effect for the treated fabrics than the untreated ones.

Keyword: Polyanion, Biopolymers, Basic dyes, Natural dyes, Moringa Oleifera

1. Introduction

Dyestuff is a coloured organic compound used for colouring different substrates. Textile is one of the materials that can be coloured by a lot of dyes. [1, 2] Dyes can be classified into two categories: Synthetics and Natural dyes. Cationic dyes (Basic dyes) are one of the Synthetics dyes that include a quaternary amino group that can make a connected system integral. [3, 4] Instead of nitrogen sometimes a positively loaded oxygen or Sulphur atom is observed. Electrostatic attraction links cationic dyes to the fibre and is not

readily migrated. [5]

Basic dyes are organic salts, water-soluble, colourful in solution which have a positive charge. These cations are attracted to substrates with a negative charge electrostatically. [6] Basic dyes can be applied to many substrates but its problem is sometimes it is difficult to achieve dark shades. [7] They are also commercial dyes whose price is low. They are still the brightest and most brilliant of all the synthetic dyes in terms of colour and saturation. Although Basic dyes are used in dyeing acrylic fabrics,

*Corresponding author e-mail: aga.hassabo@hotmail.com, Tel.: +20 110 22 555 13

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Unfortunately, they have quite low substantivity for wool fabrics and very low substantivity for cotton fabrics as they have no direct affinity for cellulose. [8, 9]

Nowadays, Natural dyes are gaining attention owing to their non-toxic and environmentally beneficial properties. [10, 11] Bio-colorants derived from natural sources are bio-resources that are both renewable and sustainable. [12, 13] *Moringa Oleifera* Lam., known as moringa, is considered to be one of the most popular and important trees in the world, and we can obtain natural dye from them. [14] *Moringa* is an excellent source of protein, vitamins and minerals, usually regarded as a miracle tree. All parts of the plant are excellent and consumable (leaves, flowers and roots) and regarded to be the 'natural tropic nourishment.' [15] Nearly majority of the plant was used in the traditional application of medicine. [16] It has so many medical effects such as (antibacterial, [17] wound healing activities, [16] anticancer, [15] antidiabetic, antioxidant and cholesterol-lowering activities). [15, 18] The plant can be also used as a natural dye in textile dyeing. The colouring compound can be extracted from the leaves. [19] To reduce chemicals used, or to have fewer hazards in the dyeing process some modifications can be applied to the fabric's surface.

Nowadays, the surface modification of textile fibres is considered the best method to obtain modern textile treatments. [20] In our work, we use anionic modification via using bio-polyanions (Sodium alginate (SA) – hydroxyethyl cellulose (HEC)). Using sodium alginate is one of the surface modifications to improve the colour strength of modified dyed fabrics. Because of the number of carboxylic groups found in alginates, this biopolymer can modify textile fibre surfaces. [21]

Alginic acid consists of brown seaweed and has a characteristic structure that consists of two uronic acids, β -D-mannuronic acid and α -L-guluronic acid as shown in **Figure 1**. Alginate is a stable gel formed in the presence of divalent cations through ionic interactions. Sodium Alginate can be used as a thickening and/or finishing agent in textile dyeing and printing, because of its low cost, easy availability, biocompatibility, ability to enhance wound healing, high moisture absorption, and strong ion-exchange capacity. [21-23]

Hydroxyethylcellulose (HEC) is often utilized as a water-binder and thickening agent in a variety of industries, including personal care products,

pharmaceutical formulations, construction materials, adhesives, and liquid soaps. [24, 25] It has hydroxyl groups as shown in its chemical structure in **Figure 1**. HEC polymer is a hydroxyethyl ether of cellulose formed by reacting cellulose with sodium hydroxide and ethylene oxide. They appear in the form of white free-flowing granular powders that dissolve easily in both cold and hot water to produce clear solutions with various viscosities depending on polymer concentration, type, and temperature. [24, 26, 27]

The purpose of the present work is to improve the dyeability of cotton, wool and acrylic fabrics to both basic dye (Methylene Blue) and natural dye (*Moringa Oleifera*), and use fewer chemicals in the dyeing process through pre-treatment by using anionic biopolymers like (sodium alginate (SA) or hydroxyethyl cellulose (HEC)) via a surface modification to enhance the colour strength, dye fixation, relative unevenness index (RUI) and antimicrobial effect of dyed fabrics.

In the present work, the natural surface anionic modification was performed on three fabrics under examination namely; cotton, wool and acrylic fabrics. Dyeing parameters were evaluated through colour strength (K/S); dye fixation; relative unevenness index (RUI) and the corresponding fastness properties were evaluated. Antimicrobial properties and also physical and mechanical properties were also evaluated. Optimization of dyeing conditions was performed to improve the performance of the dyeing process and antimicrobial effect.

2. Experimental

2.1. Materials

Bleached scoured cotton fabric (150 g/m²) supplied by Ghazel El-Mahala for Textile Industry Co., Egypt, scoured wool fabric (210 g/m²) supplied by Ghazel El-Mahala for Textile Industry Co., Egypt and bleached acrylic fabric (180 g/m²) supplied by GoldenTex for Textile Industry Co., Egypt, were used during this research. Sodium alginate (SA) low viscosity (C₆H₉NaO₇) purchased from ADVENT Co., hydroxyethylcellulose (HEC) medium viscosity (C₃₆H₇₀O₁₉) purchased from Fluka Co., citric acid (CA), were used as a crosslinker and sodium hypophosphite (SHP). Acetic acid (100%) and sodium carbonate (Na₂CO₃) were provided from Fluka to adjust the pH medium. Basic dye 9 (Methylene Blue) (C₁₆H₁₈ClN₃S) was purchased from Carl Roth GmbH Co. *Moringa Oleifera* tree leaves were supplied from

National Research Centre in Egypt. All the chemicals and reagents were used as received without purification.

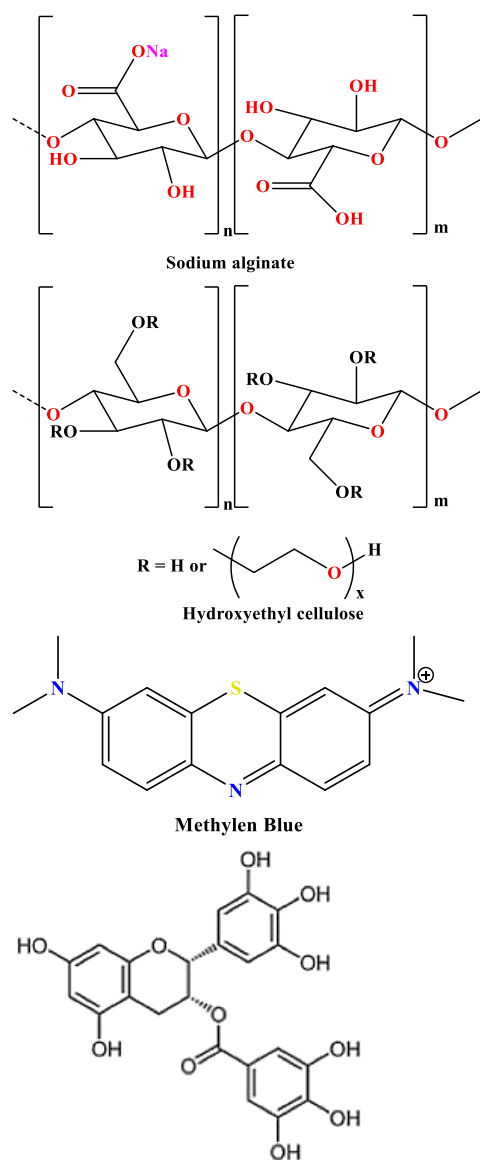


Figure 1: Chemical structure of sodium alginate, hydroxyethylcellulose, methylene blue and moringa

2.2. Methods

2.2.1. Extraction of Moringa Natural Dye

Moringa Oleifera leaves from the local market were washed and dried. Then they were crushed to turn into powder form. After that, we took 30 g of powder and boiled in one litre of distilled water for 24 h in Soxhlet system and then the solution

was filtered using filtration paper. The extracted colour was used as itris for further experiment.

2.2.2. The Pre-treatment of the fabric

Cotton, wool and acrylic fabrics were washed before treatment to remove the impurities from the outer surface of the fibres. The fabrics were washed with Na_2CO_3 solution 2 g/L, for 15 min at 40°C . Then fabrics were pre-treated using 10 g/L citric acid and 5 g/L sodium hypophosphite as crosslinkers. The fabrics were padded in the solution for 15 min in 50°C then squeezed with 100 % wet pickup, then dried at 80°C for 5 min.

2.2.3. Fabric treatment with polyanion compound

Sodium alginate (SA) and hydroxyethyl cellulose (HEC) solutions were prepared with different concentrations (0, 0.5, 1, 2 and 3%) in a 250 ml conical flask. Cotton, wool and acrylic fabrics were treated with polyanion compound using the pad-dry-cure method through immersing in the treated bath for 15 min at 50°C then squeezed with 100 % wet pickup, after that, the fabrics were dried at 100°C for 5 min.

2.2.4. Dyeing of Treated Fabrics with synthetic dye

Treated fabrics were dyed using 2 g/L Methylene Blue (Basic Dye 9) at different pH (4, 6 and 8), different times (30, 45 and 60 min), different temperatures (60, 75 and 90°C) then squeezed with 100 % wet pickup and dried at 100°C for 5 min then cured at 140°C for 3 min. The treated fabrics were used for further investigation.

2.2.5. Dyeing with natural dye

Treated fabrics were dyed using extract of moringa leaves as 30 g leaves /L water at different pH (4, 6 and 8), different times (30, 45 and 60 min), different temperatures (60, 75 and 90°C) then squeezed with 100 % wet pickup and dried at 100°C for 5 min then cured at 140°C for 3 min. The treated fabrics were used for further investigation.

2.3. Measurements

2.3.1. Determination of the carboxyl content

Carboxyl content of the treated fabrics according to the following method: weight about 0.25 g of the sample and put it a 250 ml stoppered conical flask, then 40 mL of 0.1 N NaOH was added. The conical flasks were left overnight with occasional shaking. Filter, then titrate the filtrate against HCl (0.05 N) with phenolphthalein as an indicator. Carboxyl content percent was calculated according to the following equation:

$$\text{Carboxyl contents \%} = \frac{(V_B - V_s) \times M_{\text{HCl}}}{W_t} \times 100$$

which V_b is the volume of HCl consumed by blank experiment, V_s is the volume of HCl consumed by sample, M_{HCl} = Molarity of HCl, and W is the weight of the sample.

2.3.2. Colour Measurements

A computer-based automated spectrometer (Data Colour Model 3890, Marl Co., Germany) was used to determine the colouration and spectrum reflection values of the treated dyed fabrics, as well as the colour strength K/S of dyed untreated and treated fabrics. Accordingly, the K/S values were calculated using the Kubelka-Munk equation. [28-35]

$$\frac{K}{S} = \frac{(1 - R)^2}{2R} - \frac{(1 - R_o)^2}{2R_o}$$

Where K stands for absorption coefficient, S for dispersion coefficient, R for colour reflection, and R_o for uncoloured (white) sample reflectance. The basic dye was measured at $\lambda = 610$ nm, whereas the natural dye was measured at $\lambda = 270$ nm.

2.3.3. Colourfastness properties

The AATCC test method 61-2013 was used to measure colourfastness to washing using a Launder-Ometer. [36] The AATCC test method 8-2016 was used to measure colourfastness to rubbing (dry and wet). [37] The AATCC test method 15-2013 was used to measure colourfastness to perspiration (acid and alkaline). [38] The AATCC test method 15-2013 was used to determine the colourfastness to light. [39] The grayscale colour change reference was used to evaluate washing, rubbing, and perspiration fastness

properties, while the blue scale colour change reference was utilized to evaluate light fastness properties for coloured materials.

2.3.4. Dye Fixation Measurement

To evaluate dye fixation, dyed fabrics were washed at 50°C for 30 minutes, then the colour strength values of the coloured fabric have been determined after and before washing. The dye fixation (%F) was calculated using the equation below

$$\% F = \frac{(K/S)_a}{(K/S)_b} \times 100$$

where $(K/S)_a$ is the dyed fabric's colour strength after washing and $(K/S)_b$ is the dyed fabric's colour strength before washing.

2.3.5. RUI Measurement

A reflectance spectrophotometer in the visible spectrum region was used to measure the reflectance values of 10 randomly selected places on the dyed sample. The relative unevenness index (RUI) was calculated using the equation below, where S is the standard deviation (equation below), R_m is the mean of reflectance values for n wavelengths, and V is the photopic relative luminous efficiency function. [40] For each wavelength, R_i is the reflectance value of the measurement number i .

$$S_\lambda = \sqrt{\frac{\sum_{i=1}^n (R_i - R_m)^2}{n-1}} \quad \text{RUI} = \sum_{\lambda=350}^{\lambda=700} \left(\frac{S_\lambda}{R_m} \times V_\lambda \right)$$

Excellent levelness was defined as $\text{RUI} < 0.2$, while good levelness was defined as $\text{RUI} < 0.49$. Poor levelness is defined as RUI values between 0.5 and 1, while bad levelness is defined as RUI values greater than 1. [41]

2.3.6. Mechanical properties

The AATCC Test Method 66 – 2014 was used to determine the dry crease recovery angle (CRA). [42] Fabric roughness was determined using a SE 1700 Surface Roughness Measuring Device and ASTM Test Method D 7127 – 13. [43] ASTM Test Method D 5035-2011 was used to determine tensile strength and elongation at break. [44] The average of three measurements was used to calculate the given results.

2.3.7. Antimicrobial Measurement

Gram-negative bacteria *Escherichia coli* (ATCC 25922), gram-positive bacteria *Staphylococcus aureus* (ATCC 6538), and pathogenic yeast *Candida albicans* (ATCC 10231) were utilized in this study. All bacteria were introduced via fresh overnight broth cultures that had been incubated at 37°C. [45]

The fabrics were applied to these tested microorganisms using the shake flask method to calculate antimicrobial activity as reduction (%) of the growth of these selected pathogenic strains was detected by optical density (OD) at 600 nm, and the antimicrobial activity was measured throughout the relative [OD (%)] reduction of these pathogenic strains after treated with the textile disc samples compared to the control of these pathogenic strains. [46-49] The following equation was used to characterize all of the results:

$$\text{OD Reduction (\%)} = \frac{A - B}{A} \times 100$$

Table 1 and **Figure 2**, it can be noticed that the colour strength, dye fixation and RUI values for all treated fabrics (cotton, wool and acrylic) showed higher colour strength than untreated ones. Increasing the polyanion concentration leads to increasing the colour strength, so we can say that the optimum concentration of treatment is 2% as after this concentration there is a slight increase in the results. Alginate has a carboxyl group (COOH) and hydroxyethylcellulose has a hydroxyl group (OH) as a functional group, so when the fabrics have been treated with these materials that lead to increasing the negative charge on the surface of the fabric. So, when the fabrics are steeled to dyeing with basic dyes (which has a positive charge, due to the presence of (N⁺) atom in its chemical structure), that leads to a chemical bond between the dye and the surface of the fabrics.

This chemical reaction enhances the dye fixation

Table 1 showed that the more concentration of alginate, the more carboxyl content is formed on the surface of Cotton, wool and acrylic fabrics. According to the degree of chemical activity, the carboxyl group

Table 1 represents the carboxyl content, colour strength (K/S), dye fixation and RUI values of pre-treated dyed fabrics with different concentrations (0.5, 1, 2 and 3 %) of polyanion compounds namely

where A represents the (OD) of a control flask that contains pathogenic strains and has not been treated, and B represents the (OD) of testing flasks after applying a disc sample that has been treated.

3. Results and Discussion

3.1. Optimization of the treatment condition

3.1.1. Effect of polyanion concentration

Four different treatment solutions based on polyanion compounds (alginate and hydroxyethylcellulose) have been prepared with different concentrations (0.5, 1, 2, and 3 %) to functionalize the surface of fabrics namely cotton, wool and acrylic fabrics. After modification of fabrics, the dyeing process has been done using both basic and natural dyes.

From

and RUI values of all dyed fabrics: cotton, wool and acrylic. The optimum concentration of treated fabric is 2%.

In the case of natural dye, moringa dye has a hydroxyl group (OH) so we need a cross-linker agent (citric acid with sodium hypophosphite) to make excellent attraction between the dye and different modified fabrics. [50-52]

Carboxyl content has been measured for blank and treated fabrics with alginate to know the number of carboxyl groups that have been attached to the treated fabrics. As known, alginate has (COOH) as its functional group so the more carboxyl groups have been coated on the surface of different fabrics, the more anionic charge will be on the surface due to the presence of (COO⁻). When the negative charge is increased that leads to more absorbance of basic dyes as its charge is positive.

of alginates is more active than the hydroxyl group of hydroxyl ethyl cellulose so that leads to higher K/S, higher dye fixation and higher RUI values

(Alginate and hydroxyethylcellulose) using basic and natural dyes.

Table 1: Effect of polyanion polymers concentration on the carboxyl content and colour performance

Fabric	Polyanion (%)	Carboxyl content %	Basic dye				Natural dye				
			K/S	Dye fixation	RUI		K/S	Dye fixation	RUI		
					value	rate			value	rate	
Cotton	Alg	0	0.11	13.18	58.50	0.53	P	9.41	41.77	0.38	G
		0.5	2.31	22.25	86.91	0.52	P	10.15	54.83	0.33	G
		1	3.07	23.52	91.35	0.42	G	10.83	57.04	0.26	G
		2	3.38	26.09	95.57	0.36	G	11.27	58.59	0.22	G
		3	3.54	26.49	97.25	0.37	G	11.59	59.47	0.26	G
	HEC	0	-	13.18	58.50	0.53	P	9.41	41.77	0.38	G
		0.5	-	17.72	72.70	0.44	G	10.04	48.12	0.29	G
		1	-	22.89	86.66	0.40	G	10.76	54.38	0.25	G
		2	-	24.81	97.41	0.37	G	11.39	60.24	0.23	G
		3	-	26.29	97.55	0.37	G	11.78	59.73	0.23	G
Wool	Alg	0	0.61	14.58	79.93	0.68	P	12.41	68.03	0.58	P
		0.5	3.31	19.63	89.76	0.69	P	12.73	70.82	0.54	P
		1	3.71	22.82	91.06	0.60	P	12.97	69.57	0.46	G
		2	4.11	23.29	93.60	0.47	G	13.54	71.22	0.36	G
		3	4.51	23.35	95.78	0.41	G	13.86	72.84	0.32	G
	HEC	0	-	14.58	79.93	0.68	P	12.41	68.03	0.58	P
		0.5	-	17.11	84.85	0.65	P	12.85	69.19	0.53	P
		1	-	21.23	86.66	0.57	P	12.98	67.21	0.44	G
		2	-	23.06	94.60	0.48	G	13.43	72.13	0.36	G
		3	-	23.32	94.69	0.47	G	13.97	72.03	0.36	G
Acrylic	Alg	0	0.56	3.69	78.51	0.49	G	5.17	79.94	0.69	P
		0.5	2.81	5.92	86.46	0.41	G	5.52	90.70	0.45	G
		1	3.39	8.27	92.87	0.36	G	6.21	93.65	0.33	G
		2	3.74	10.35	94.88	0.19	E	6.94	91.65	0.16	E
		3	4.02	10.64	95.53	0.23	G	7.11	91.38	0.19	E
	HEC	0	-	3.69	78.51	0.49	G	5.17	79.94	0.69	P
		0.5	-	4.81	82.46	0.39	G	5.45	86.50	0.48	G
		1	-	7.10	91.87	0.35	G	6.13	92.64	0.35	G
		2	-	9.31	96.88	0.19	E	6.78	93.58	0.17	E
		3	-	9.90	97.53	0.23	G	7.07	93.29	0.19	E

Treatment condition: polyanions (alginate (Alg) and hydroxyethyl cellulose (HEC)) (0.5, 1, 2 and 3 %), citric acid 10 g/L, sodium hypophosphite (SHP) 5 g/L, drying at 80°C for 5 min

Dyeing condition: dye conc.: 2 g/L (basic dye ($\square = 610$)) and extract of moringa oleifera leaves as 30 g leaves /L water ($\square = 270$), pH 4, dyeing time: 45 min, dyeing temperature: 75°C, drying at 100°C for 5 min, curing at 140°C for 3 min

RUI values range can be categorized into If $RUI < 0.2$ that considered as excellent levelness. (E). If $0.2 < RUI < 0.49$ that considered as good levelness. (G), If RUI is between 0.5 and 1 that means poor levelness. (P), If RUI values are greater than 1 that indicate bad levelness. (B)

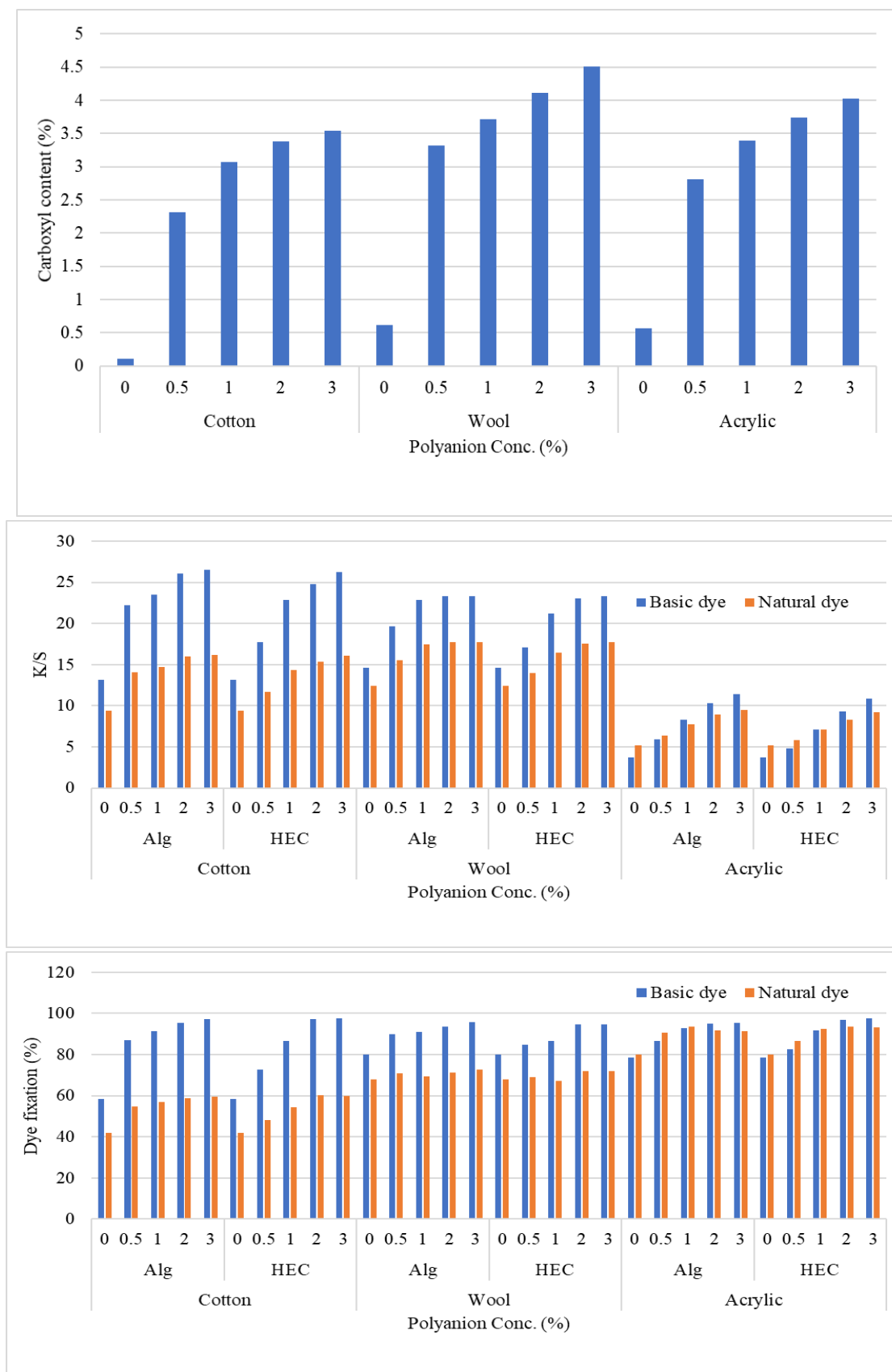


Figure 2: Effect of polyanion polymers concentration on the carboxyl content and colour performance

3.1.2. Effect of pH

One of the most important factors affecting dye adsorption onto textiles treated with polyanion compounds was the pH level of the dye bath. When the pH was between 4 and 8, changing the pH had a beneficial effect on the K/S ratio. However, as the pH value increased (higher than 4), the K/S ratio

Table 2 and **Figure 3** present the colour strength (K/S) values for the dyed treated fabrics with both polyanion polymers using basic and natural dyes at different pH values. The colour strength of cotton fabrics modified by SA and HEC provides increasing the K/S values as the dyeing pH is in an acid medium. In wool fabric modified by SA and HEC, there is an increase in the K/S values in the acid medium than in the alkali medium. The colour strength of acrylic fabrics modified by alginate provides increasing the K/S values as the dyeing pH decreases from pH 8 till pH 4. That means the higher K/S for acrylic fabrics using basic dyes is pH 4.

In basic and natural dyeing for cotton fabric, the optimum pH is 4 for both polyanions (Alg and HEC) as pH 4 gives higher color strength than pH 6 and pH 8. It shows in the case of basic dye K/S (26.09) with RUI (0.36) and natural dye K/S is (11.27) with RUI (0.22), which means a good rate of levelness of dyeing. And in the case of HEC for basic dye, the color strength is (24.81) for RUI value (0.37) and natural dye, K/S is (11.39) with RUI (0.23), which means a good rate of levelness of dyeing.

In the case of wool dyeing with basic and natural dyes, the optimum pH is 4 in the case of treated fabrics with both polyanions (Alg and HEC) as pH 4 gives higher color strength than pH 8 and pH 6. It shows in the case of fabric treated with alginate using basic dye, the color strength is (23.29) for RUI value (0.47) and in the case of natural dye, the K/S value is (13.54) for RUI (0.36), which means a good rate of levelness of dyeing. In HEC modification, the color strength of basic dye is (23.06) for RUI value (0.48) and in natural dyeing, the K/S value is (13.43) for RUI value (0.36), which means a good rate of levelness of dyeing.

In acrylic dyeing with basic dye and moringa natural dye, the optimum pH is 4 in the case of treated fabrics with both polyanions (Alg and HEC) as pH 4 gives higher color strength than pH 6 and pH 8. It shows in the case of basic dye with alginate polymer, that the color strength is (10.35) with RUI value (0.19) and in moringa natural dye the K/S is (6.94) with RUI

decreased. It might be related to the treated textiles' chemical structure according to coating surface material.

dye and (6.78) for moringa dyeing with RUI values (0.19) and (0.17) for basic and natural dyes respectively, which means excellent levelness of the fabric surface.

(0.16), which means an excellent rate of levelness of dyeing. In the case of HEC with a basic dye, the color strength is (9.31) with RUI value (0.19) and in moringa natural dyeing, the K/S is (6.78) for RUI value (0.17), which means an excellent rate of levelness of dyeing.

3.1.3. Effect of Dyeing Temperature

With the increase in the dyeing temperature, an overall characteristic of higher dyeability was observed. By increasing dyeing temperatures from 60 to 90°C, the colour strengths (K/S) of all fabrics modified by alginate and hydroxyethylcellulose increase (**Figure 4** and

Table 3). As more dyes molecules can be fixed in the fabrics, and transferred from the surface of fabrics to the non-crystalline parts that lead to more absorption of the dye solution. The optimum dyeing temperature is 75 minutes, based on the previous results because the percent increase in K/S as a result of increasing dyeing time from 75 to 90 minutes is quite small.

Table 3 and **Figure 4** present the colour strength (K/S) values for the dyed treated fabrics with polyanion using basic dye at different temperatures (60, 75 and 90°C).

In cotton fabric, treatment with alginate gives color strength higher than HEC as in 75°C, the K/S of cotton treated with Alg is (26.09) for basic dye and (11.27) for natural dye with RUI values (0.36) and (0.22) for basic and natural dyes respectively, that means good levelness of the fabric surface. In the case of cotton treated with HEC in 75°C, the K/S is (24.81) for basic dye and (11.39) for natural dyeing with RUI value (0.37) and (0.23) for basic and natural dyes respectively, which means good levelness of the fabric surface.

In wool fabric, treatment with alginate gives color strength higher than HEC as in 75°C, the K/S of cotton treated with Alg is (23.29) for basic dye and (13.54) for natural dye with RUI value (0.47) for basic dye and

(0.36) for natural dye, that means good levelness of the fabric surface. In the case of wool treated with HEC in 75°C, the K/S is (23.06) for basic dye and (13.43) for moringa natural dyeing with RUI values (0.48) and (0.36) for basic and natural dyes respectively, which means good levelness of the fabric surface.

In acrylic fabric, treatment with alginate gives colour strength higher than HEC as in 75°C, the K/S of acrylic treated with Alg is (10.35) for MB and (6.94)

for moringa dye with RUI value (0.18) for basic dye and (0.16) for moringa dye, that means excellent levelness of the fabric surface. In the case of acrylic treated with HEC in 75°C, the K/S is (9.31) for basic dye and (6.78) for moringa dyeing with RUI values (0.19) and (0.17) for basic and natural dyes respectively, which means excellent levelness of the fabric surface.

Table 2: Effect of pH on the colour strength

Fabric	Polyanion (2 %)	pH	Basic dye				Natural dye			
			K/S	Dye fixation	RUI		K/S	Dye fixation	RUI	
					value	rate			value	rate
Cotton	Alg	4	26.09	95.57	0.36	G	11.27	58.59	0.22	G
		6	25.05	96.49	0.43	G	10.84	57.64	0.26	G
		8	25.35	91.44	0.45	G	10.32	56.34	0.28	G
	HEC	4	24.81	97.41	0.37	G	11.39	60.24	0.23	G
		6	23.82	98.35	0.44	G	10.85	58.25	0.27	G
		8	24.10	93.20	0.46	G	10.51	57.92	0.29	G
Wool	Alg	4	23.29	93.60	0.47	G	13.54	71.22	0.36	G
		6	22.87	92.85	0.46	G	12.78	73.34	0.37	G
		8	22.29	92.60	0.47	G	12.31	71.22	0.36	G
	HEC	4	23.06	94.60	0.48	G	13.43	72.13	0.36	G
		6	22.82	98.89	0.45	G	12.74	74.28	0.38	G
		8	22.46	94.60	0.48	G	12.14	72.13	0.36	G
Acrylic	Alg	4	10.35	94.88	0.19	E	6.94	91.65	0.16	E
		6	9.87	93.37	0.17	E	6.61	89.49	0.15	E
		8	9.34	90.73	0.19	E	6.17	87.15	0.16	E
	HEC	4	9.31	96.88	0.19	E	6.78	93.58	0.17	E
		6	9.05	95.34	0.17	E	6.45	91.25	0.16	E
		8	8.86	92.64	0.19	E	6.34	89.65	0.17	E

Treatment condition: polyanions (alginate (Alg) or hydroxyethyl cellulose (HEC)) (2 %), citric acid 10 g/L, sodium hypophosphite (SHP), drying at 80°C for 5 min

Dyeing condition: dye conc.: 2 g/L (basic dye ($\square = 610$)) and extract of moringa oleifera leaves as 30 g leaves /L water ($\square = 270$), pH (4, 6, and 8), dyeing time: 45 min, dyeing temperature: 75°C, drying at 100°C for 5 min, curing at 140°C for 3 min

RUI values range can be categorized into If RUI<0.2 that considered as excellent levelness. (E). If 0.2<RUI<0.49 that considered as good levelness. (G), If RUI is between 0.5 and 1 that means poor levelness. (P), If RUI values are greater than 1 that indicate bad levelness. (B)

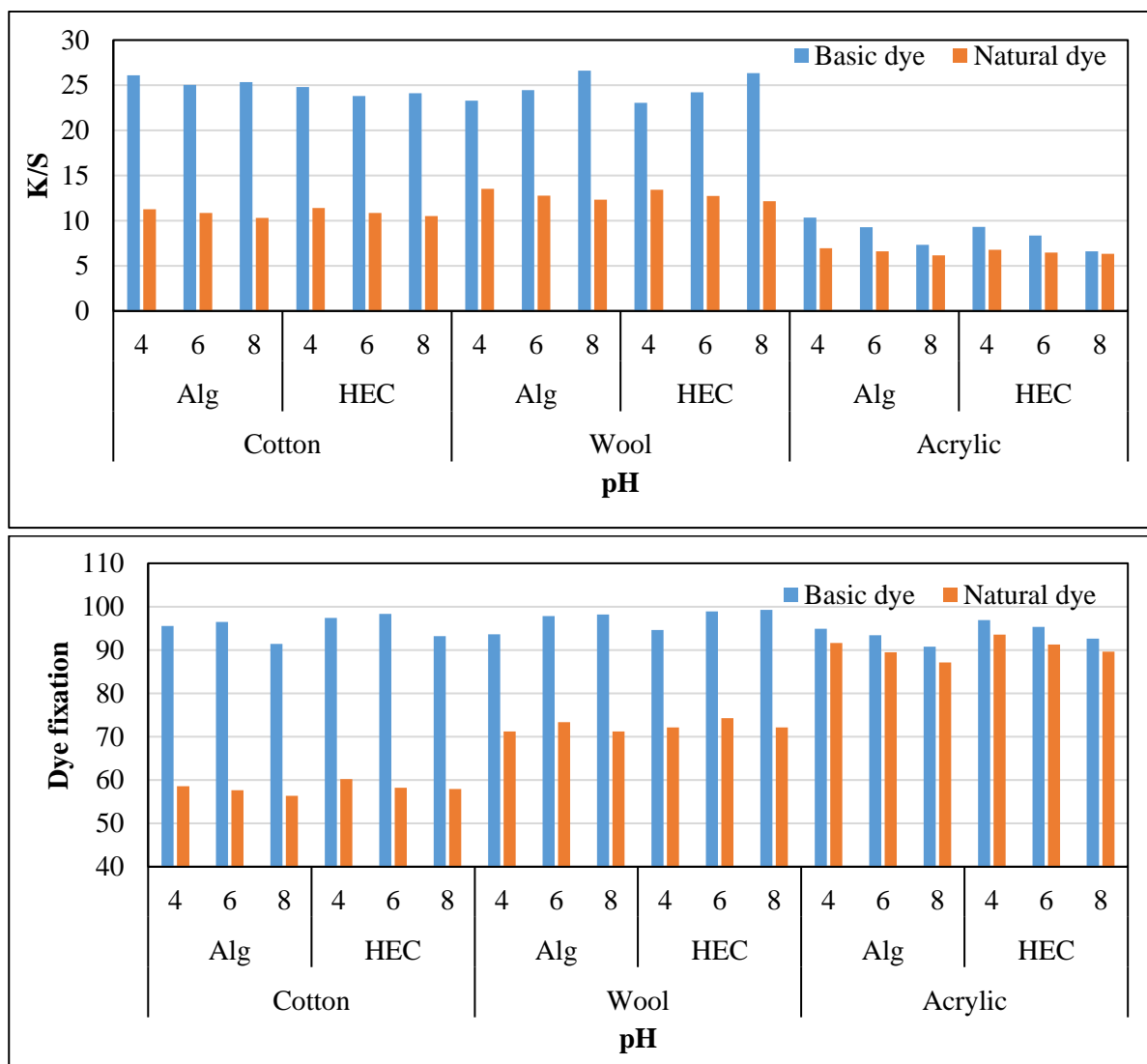


Figure 3: Effect of pH on the colour strength

Table 3: Effect of dyeing temperature on the colour strength

Fabric	Polyanion (2 %)	Dyeing temperature (°C)	Basic dye				Natural dye			
			K/S	Dye fixation	RUI		K/S	Dye fixation	RUI	
					value	rate			value	rate
Cotton	Alg	60	25.67	95.30	0.54	P	10.98	57.94	0.33	G
		75	26.09	95.57	0.36	G	11.27	58.59	0.22	G
		90	26.97	95.89	0.45	G	11.73	59.39	0.27	G
	HEC	60	24.17	97.15	0.55	P	10.85	59.42	0.34	G
		75	24.81	97.41	0.37	G	11.39	60.24	0.23	G
		90	25.24	98.08	0.46	G	11.94	62.06	0.28	G
Wool	Alg	60	22.91	93.32	0.47	G	12.98	71.08	0.39	G
		75	23.29	93.60	0.47	G	13.54	71.22	0.36	G
		90	23.81	94.08	0.29	G	13.73	72.04	0.32	G
	HEC	60	22.79	94.33	0.43	G	12.87	71.81	0.40	G
		75	23.06	94.60	0.48	G	13.43	72.13	0.36	G
		90	23.58	95.32	0.30	G	13.82	72.79	0.32	G

Acrylic	Alg	60	9.95	92.14	0.37	G	6.53	91.04	0.27	G
		75	10.35	94.88	0.19	E	6.94	91.65	0.16	E
		90	10.86	95.96	0.14	E	7.22	96.31	0.19	E
	HEC	60	8.86	95.98	0.23	G	6.84	93.02	0.21	G
		75	9.31	96.88	0.19	E	6.78	93.58	0.17	E
		90	9.95	97.93	0.12	E	7.16	94.34	0.13	E

Treatment condition: polyanions (alginate (Alg) or hydroxyethyl cellulose (HEC)) (2 %), citric acid 10 g/L, sodium hypophosphite (SHP), drying at 80°C for 5 min

Dyeing condition: dye conc.: 2 g/L (basic dye ($\square = 610$)) and extract of moringa oleifera leaves as 30 g leaves /L water ($\square = 270$), pH (4), dyeing time: 45 min, dyeing temperature: (60, 75, and 90°C), drying at 100°C for 5 min, curing at 140°C for 3 min

RUI values range can be categorized into If $RUI < 0.2$ that considered as excellent levelness. (E). If $0.2 < RUI < 0.49$ that considered as good levelness. (G), If RUI is between 0.5 and 1 that means poor levelness. (P), If RUI values are greater than 1 that indicate bad levelness. (B)

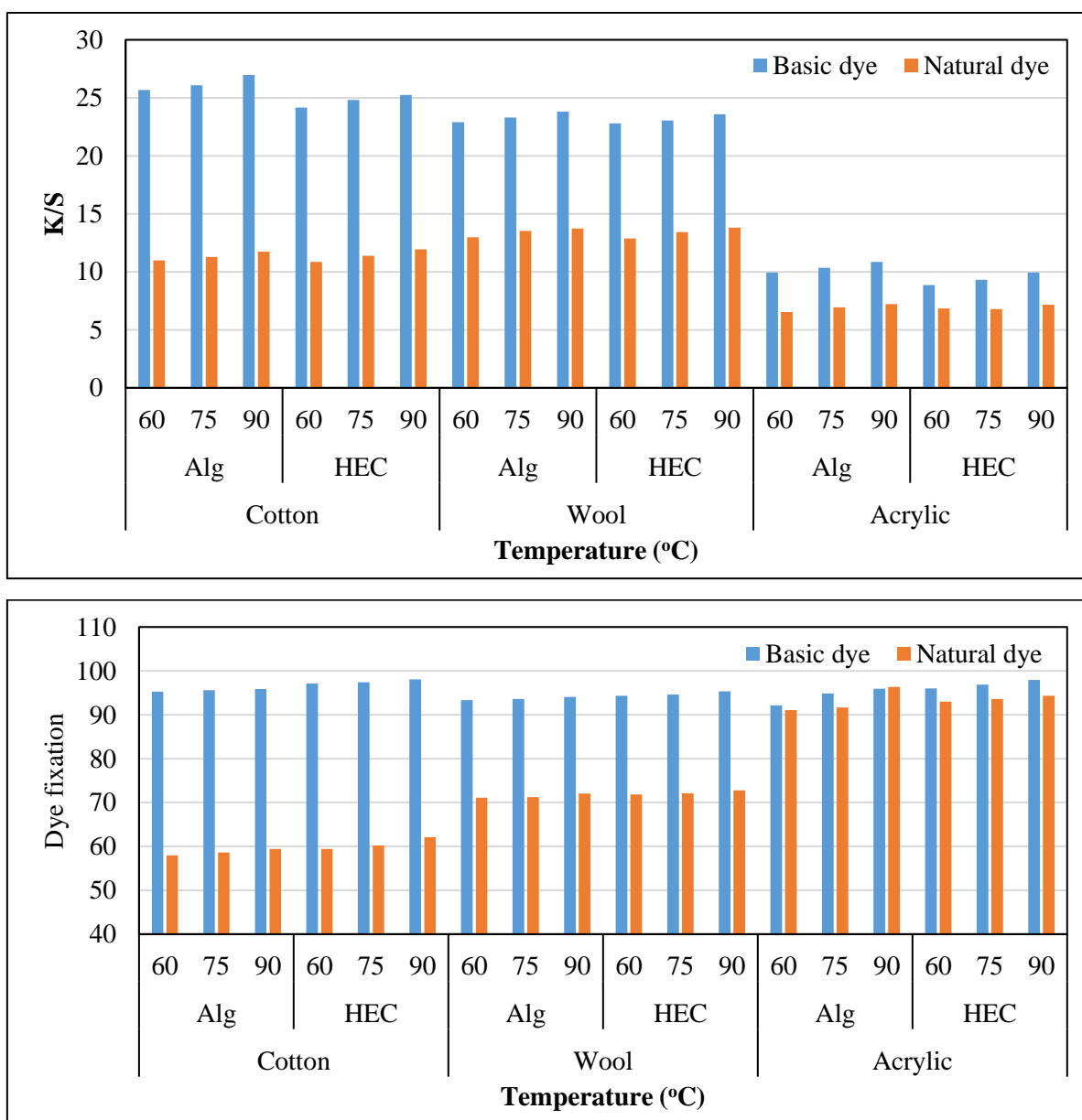


Figure 4: Effect of dyeing temperature on the colour strength

3.1.4. Effect of Dyeing Time

The colour strengths (K/S) of all fabrics increases with the increase of dyeing time. These are expected effects since the dye had more time to penetrate and spread throughout the fibres, allowing it to reach its maximum dye absorption capacity when it reached the dynamic dyeing equilibrium state. The optimum dyeing duration is 45 minutes, based on the previous results because the percent increase in K/S as a result of increasing dyeing time from 45 to 60 minutes is relatively tiny.

Table 4 and **Figure 5** present the colour strength (K/S) values for the dyed treated fabrics with polyanion using basic dye at a different time (30, 45 and 60 min.).

In cotton fabric, treatment with alginate gives color strength higher than HEC as in case of 45 min, the K/S of cotton treated with Alg is (26.09) for basic dye and (11.27) for natural dye with RUI value (0.36) for basic dye and (0.22) for natural dye, that means good levelness of the fabric surface. In the case of cotton treated with HEC in time 45, the K/S is (24.81) for basic dye and (11.39) for moringa natural dyeing with RUI value (0.37) for basic dye and (0.23) for natural dye, which means good levelness of the fabric surface.

In wool fabric, treatment with alginate gives color strength higher than HEC as in case of 45 min, the K/S of wool treated with Alg is (23.29) for basic dye and (13.54) for natural dye with RUI value (0.47) for basic dye and (0.36) for natural dye, that means good levelness of the fabric surface. In the case of wool treated with HEC in time 45, the K/S is (23.06) for basic dye and (13.43) for moringa natural dyeing with RUI value (0.48) for basic dye and (0.36) for natural dye, which means good levelness of the fabric surface.

In acrylic fabric, treatment with alginate gives color strength higher than HEC as in the case of 45 min., the K/S of acrylic treated with Alg is (10.35) for basic dye and (6.94) for natural dye with RUI value (0.19) for basic dye and (0.17) for moringa dye, that means excellent levelness of the fabric surface. In the case of acrylic treated with HEC in the case of 45 min., the K/S is (9.31) for basic dye and (6.78) for moringa natural dyeing with RUI value (0.19) for basic dye and (0.17) for natural dye, which means excellent levelness of the fabric surface.

3.2. Characterization of the dyed textile fabrics

3.2.1. Fastness properties

Table 5 shows the fastness properties of dyed treated fabrics (cotton, wool, and acrylic fabrics) with polyanion polymers (alginate and hydroxyethyl cellulose; 2%) using two different dye natures (basic dye (methylene blue) and nature dye (moringa extract)) at optimum dyeing conditions (45 min.; 75°C; dye conc. 2 g/L; pH 4).

Treated textiles showed superior washing, perspiration, light, and rubbing fastness properties than untreated materials. Because the polyanion treated textiles absorb more colour molecules, hydroxyethyl cellulose and alginate treatment significantly enhanced the washing and light fastness properties of coloured fabrics using both investigated dyes as compared to untreated materials. As the dye content in the fibre increased, the light-fastness properties of modified fabrics improved. The chemical connections created between polyanions-treated materials, crosslinker and dye molecules are responsible for this enhancement.

Furthermore, the dye molecules are bound within and on the textile surface due to enhanced hydrogen bonding between dye molecules, polyanion polymers, the fabric surface, and the crosslinker. In addition, dyeing homogeneity is increased when the fabric is treated with polyanion polymers because the fibres have a uniform thin coating of polyanion polymers on the surface, and the dye molecules are absorbed better and more uniformly.

3.2.2. Physical and mechanical properties

Tensile strength, elongation at break, roughness, and crease recovery angle of cotton, wool, and acrylic textiles in the warp and weft directions were measured before and after they were treated with 2% polyanion polymers, and the results are presented in **Table 6** and **Figure 6**. As the demonstrated presence of anion groups from both polyanion polymers have a beneficial effect on textile fabrics.

Table 6 and **Figure 6** shows that the presence of an anionic group in treated textile materials causes changes in physicomaterial properties. While the crease recovery angle was raised, fabric roughness, tensile strength, and elongation at the break all decreased. This indicates that the polyanion polymers under investigation were firmly embedded in the textile fabrics' microstructure, generating a thin coating layer on the fabric's surface that was

responsible for the observed changes. [53-56]

The impact of citric acid catalyzed by sodium hypophosphite (SHP) in the pre-crosslinking treatment of cotton, wool, and/or acrylic textiles in altering the performance of the aforementioned examined properties was also significant to emphasize. During this pre-treatment, covalent crosslinking connections between neighbouring cellulosic or proteinic chains would form, giving the cotton structure rigidity. Meanwhile, the citric acid and catalyst would damage the cotton structure chemically. Stiffness and chemical degradation were logically the causes of the reduction in tensile strength. The creation of an intensive network with a high degree of crosslinking via covalent chemical bonding was most likely responsible for the extraordinary increase in the crease recovery angle.

3.2.3. Antimicrobial properties

Table 7 shows the antimicrobial effects of cotton, wool, and acrylic fabrics treated with 2% polyanion polymer and dyed with two different dye natures (basic and natural (moringa oleifera extract)) using three different microorganisms (i) gram-positive (*Staphylococcus aureus*), (ii) gram-negative (*Escherichia coli*), and (iii) fungal (*Candida Albicans*).

The results show that both types of bacteria (gram-positive and negative bacteria), as well as a fungus (*Candida Albicans*), had a higher inhibitory influence on untreated dyed textile materials than blank material without dyeing. This action is caused by the presence of cationic groups in the basic dye, as well as phenolic acids and flavonoids in natural dye extract.

Because the cell walls of both the tested bacterial strains differ in composition, the treated fabrics are more efficient against gram-negative bacteria than gram-positive bacteria. The polycations in use also block ergosterol, a critical component of the fungal cell membrane. [34, 57-62]

Textiles treated with HEC have a stronger antibacterial impact than those treated with Alginate. This is owing to the presence of a significant number of hydroxyl groups (some carboxyl groups in alginate exist as sodium salts), which play a key role in disrupting microbial cell membranes while also having

a strong antibacterial impact. [52, 53]

Natural colouring extract (moringa oleifera extract) contains metals, phenolic acids, and flavonoids that have successfully interacted with the bacterium's cells. The cover's polyanion polymers spread and stabilize the extract dyes' components (metals, phenolic acids, and flavonoids) well on the fabrics surface, decreasing their potential to interact with bacterial cells. [50, 63-65]

Furthermore, treated and coloured textile materials have superior antibacterial characteristics before and after washing than untreated textile textiles. After washing, the treated materials' resilience gives an excellent antibacterial action against all pathogens tested.

4. Conclusion

This study highlights a simple green approach for dyeing bio-treated cotton, wool and acrylic fabrics using basic dye (methylene blue) and natural dye (moringa oleifera) aqueous extract to provide and achieve value-added characteristics dyed fabric based on sodium alginate (SA) and hydroxyethyl cellulose (HEC). Using anion biopolymers on various fabrics have enhanced the colour strength of the dyeing process with different dyes. Not only the colour strength of dyed treated fabrics (K/S) increase but also the antimicrobial effect of treated fabrics have been increased. The optimum polymers' concentration was 2%, for the optimum dyeing condition (45 minutes; 75°C; dye concentration 2 g/L; pH 4). The fastness properties of treated dyed fabrics show a higher result than untreated ones. Fabrics' surface modification is a trend that should be applied in the textile industry as this leads to fewer chemicals used and fewer hazards to the environment, especially if we use natural polymers as we have done in our work. The results of this study suggest that moringa oleifera dye aqueous extract might be used as a value-added sustainable colour in the eco-textile dyeing sector, where natural dye poses a significant challenge in the dyeing of textiles and greener textile colouration.

Overall bio-polyanion modification can be applied to different fabrics and achieve a great result in the dyeing process.

Table 4: Effect of dyeing time on the colour strength

Fabric	Polyanion (2 %)	Dyeing time (min.)	Basic dye				Natural dye			
			K/S	Dye fixation	RUI		K/S	Dye fixation	RUI	
					value	rate			value	rate
Cotton	Alg	25.95	92.85	0.54	P	10.94	57.53	0.35	G	25.95
		26.09	95.57	0.36	G	11.27	58.59	0.22	G	26.09
		26.83	96.32	0.37	G	11.71	59.98	0.24	G	26.83
	HEC	24.29	96.67	0.52	P	11.06	59.84	0.36	G	24.29
		24.81	97.41	0.37	G	11.39	60.24	0.23	G	24.81
		25.32	98.54	0.45	G	11.92	60.85	0.25	G	25.32
Wool	Alg	22.85	93.13	0.69	P	13.18	71.06	0.55	P	22.85
		23.29	93.60	0.47	G	13.54	71.22	0.36	G	23.29
		23.71	94.03	0.26	G	13.86	71.77	0.19	E	23.71
	HEC	22.66	92.71	0.54	P	13.16	71.89	0.55	P	22.66
		23.06	94.60	0.48	G	13.43	72.13	0.36	G	23.06
		23.84	95.07	0.28	G	13.97	72.68	0.20	E	23.84
Acrylic	Alg	6.95	88.31	0.37	G	6.87	91.01	0.38	G	6.95
		10.35	94.88	0.19	E	6.94	91.65	0.16	E	10.35
		10.76	96.16	0.23	G	7.29	92.36	0.19	E	10.76
	HEC	9.25	96.39	0.35	G	6.86	92.99	0.23	G	9.25
		9.31	96.88	0.19	E	6.78	93.58	0.17	E	9.31
		9.68	97.23	0.19	E	7.22	94.30	0.19	E	9.68

Treatment condition: polyanions (alginate (Alg) or hydroxyethyl cellulose (HEC)) (2 %), citric acid 10 g/L, sodium hypophosphite (SHP), drying at 80°C for 5 min

Dyeing condition: dye conc.: 2 g/L (basic dye ($\square = 610$)) and extract of moringa oleifera leaves as 30 g leaves /L water ($\square = 270$), pH (4), dyeing time: (30, 45, and 60 min), dyeing temperature: (75°C), drying at 100°C for 5 min, curing at 140°C for 3 min

RUI values range can be categorized into If $RUI < 0.2$ that considered as excellent levelness. (E). If $0.2 < RUI < 0.49$ that considered as good levelness. (G), If RUI is between 0.5 and 1 that means poor levelness. (P), If RUI values are greater than 1 that indicate bad levelness. (B)

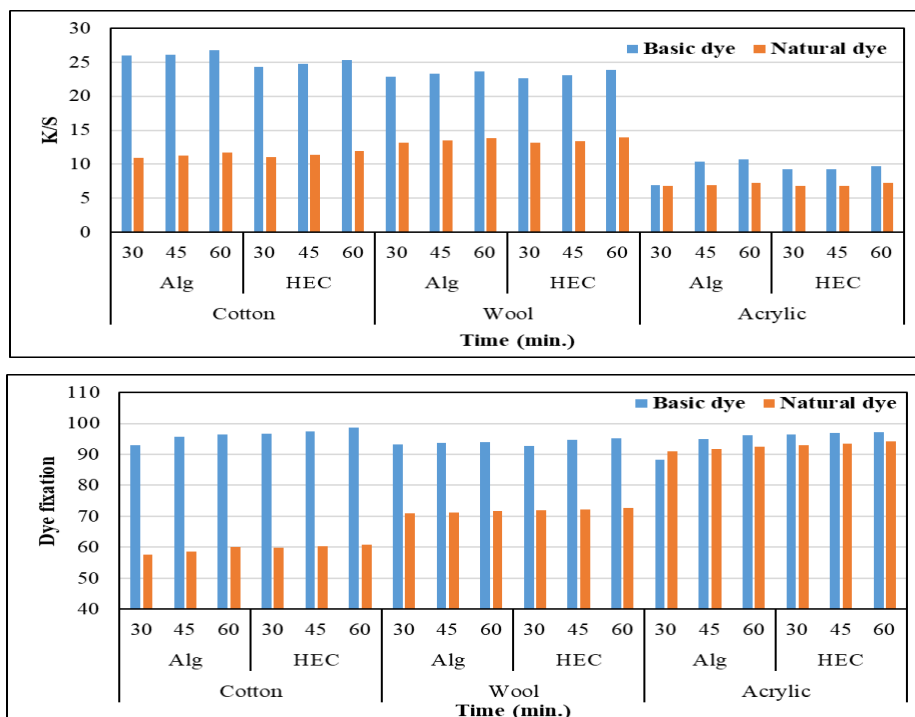
**Figure 5:** Effect of dyeing time on the colour strength

Table 5: Colour strength and fastness properties of dyed fabrics using basic and natural dyes

Fabric	Dye	Polyanion (2 %)	Fastness properties										Light	
			Washing		Rubbing				Perspiration					
			Alt	St	Dry		Wet		Acidic		Alkaline			
Cotton	Basic Dye	without	2	2	2	2	2-3	2-3	2	2	2	2	3-4	
		Alg	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4	4-5
		HEC	3-4	3	3	3-4	3-4	3	3	3	3	3	3	4
	Natural dye	without	2	2	2	2	2	2	2	2	2	2	2	2-3
		Alg	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4	4-5
		HEC	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4	4-5
Wool	Basic Dye	without	3	3	3	3	3	3	3	3	3	3	3-4	
		Alg	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4-5	
		HEC	3	3	4	4-5	3	3-4	3	3-4	3	3	3	4-5
	Natural dye	without	3	2-3	3	2-3	3	2-3	3	2-3	3	2-3	3	4
		Alg	4	4	3-4	3-4	3-4	4	3-4	3-4	3-4	3-4	3-4	5
		HEC	4	3-4	3-4	4	3-4	3-4	3-4	3-4	3	3	3	5
Acrylic	Basic Dye	without	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4-5	
		Alg	4	4	3-4	4	3-4	3-4	3-4	3-4	3	3	5	
		HEC	4	4	3-4	4	3-4	3-4	3-4	3-4	3	3	5	
	Natural dye	without	2-3	2-3	3	3	3	2-3	3	3	3	3	3	3
		Alg	4	4	3-4	3-4	3-4	3-4	3-4	4	4	4	4	4-5
		HEC	4	4	4	4	4	3-4	4	4	4	4	4	4-5

Table 6: physical and mechanical properties for treated dyed fabrics

Fabric	Dye	Polyanion (2 %)	Physical and Mechanical properties			
			Tensile Strength (N/mm ²)	Elongation at a break (%)	Crease Recovery Angle (warp + weft) (°)	Surface Roughness
Cotton	Basic dye	without	157.08	36.82	232.0	21.32
		Alg	141.38	32.25	226.9	20.46
		HEC	143.88	35.75	206.8	21.78
	Natural dye	without	149.23	34.98	220.4	20.25
		Alg	148.44	33.86	238.3	21.48
		HEC	151.07	37.54	217.1	22.87
Wool	Basic dye	without	161.16	37.74	232.0	21.83
		Alg	145.10	33.01	232.9	21.01
		HEC	147.63	36.63	212.2	22.44
	Natural dye	without	153.10	35.85	220.4	20.74
		Alg	152.25	34.65	244.5	22.06
		HEC	155.01	38.46	222.8	23.56
Acrylic	Basic dye	without	165.34	38.76	238.0	22.44
		Alg	148.75	33.88	238.8	21.45
		HEC	151.51	37.63	217.6	22.99
	Natural dye	without	157.08	36.82	226.1	21.32
		Alg	156.19	35.57	250.8	22.52
		HEC	159.08	39.51	228.5	24.14

R: Roughness, CRA (W+F): average Crease Recovery Angle in warp and weft directions

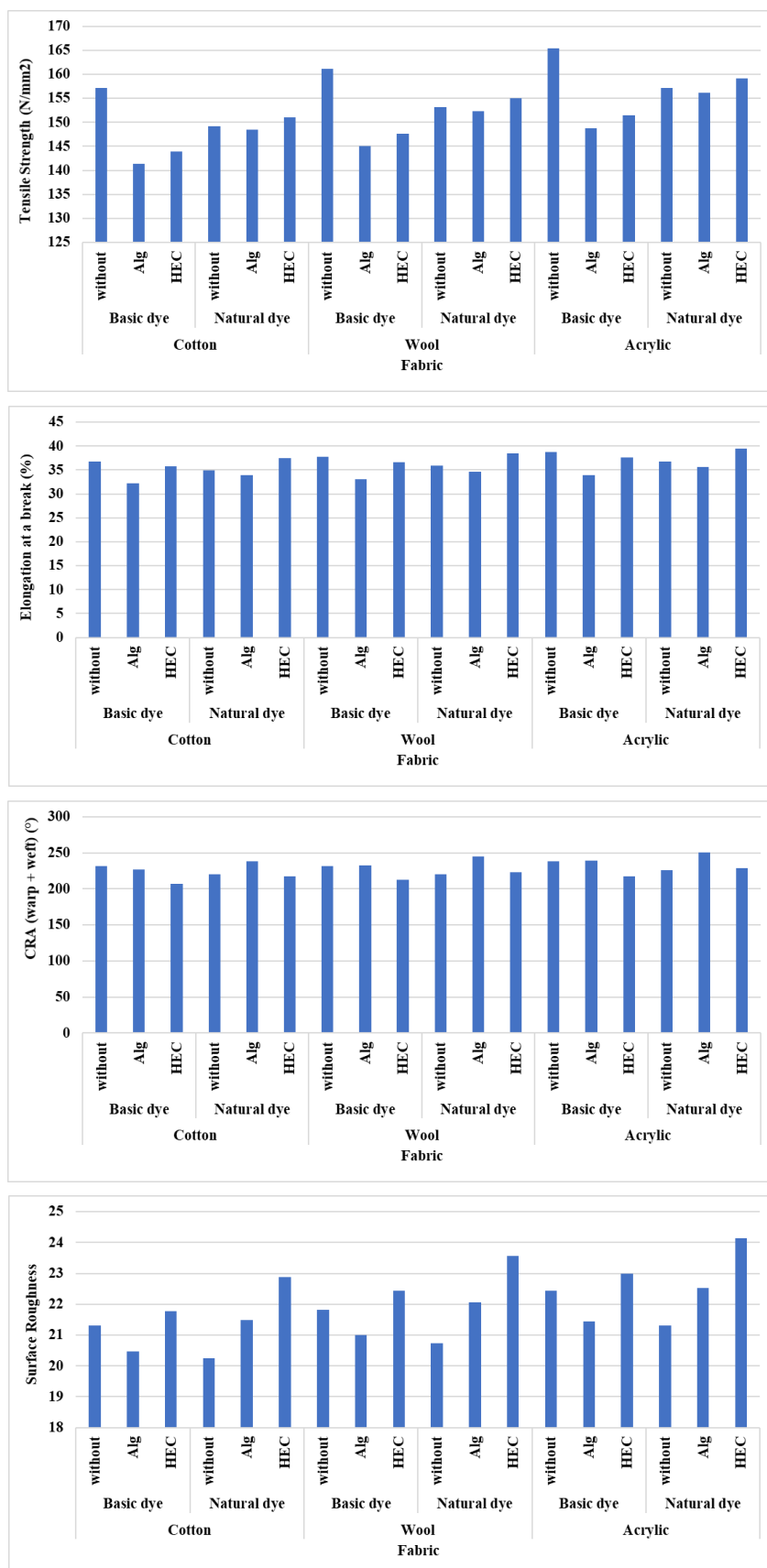


Figure 6: Physical and mechanical properties of dyed fabrics using different dyes

Table 7: CFU reduction (%) of different microbial strains for untreated and treated dyed fabrics before and after washing

Fabric	Dye	Polyanion (2 %)	Microbial Reduction %					
			E. coli (ATCC 25922)		S. Aureus (ATCC 29213)		C. Albicans (ATCC 10231)	
			before washing	after 10 washing cycles	before washing	after 10 washing cycles	before washing	after 10 washing cycles
Cotton	Blank		21.1	16.5	21.3	15.1	20.1	13.3
	Basic dye	without	23.21	18.25	25.14	17.88	24.11	16.23
		Alg	31.57	26.65	34.19	26.10	32.79	23.70
		HEC	42.93	36.24	46.50	35.50	44.59	32.23
	Natural dye	without	74.27	58.40	80.45	57.22	77.15	51.94
		Alg	88.38	85.26	95.73	83.54	91.81	75.83
HEC		98.74	94.22	99.97	92.31	95.88	87.01	
Wool	Blank		31.0	26.3	34.4	25.6	32.6	22.3
	Basic dye	without	36.44	32.30	39.47	31.65	37.85	28.73
		Alg	49.56	47.16	53.68	46.21	51.48	41.94
		HEC	67.40	60.52	73.00	59.29	70.01	53.82
	Natural dye	without	80.17	71.07	86.83	69.62	83.28	63.20
		Alg	89.20	84.89	91.25	83.17	92.66	75.49
HEC		94.36	84.72	94.90	83.00	98.02	75.35	
Acrylic	Blank		27.8	22.3	30.7	25.6	27.1	22.3
	Basic dye	without	32.30	31.80	34.44	31.65	33.03	28.73
		Alg	43.93	43.24	46.84	43.04	44.92	39.07
		HEC	68.97	67.89	73.54	67.57	70.53	61.34
	Natural dye	without	71.07	69.95	75.77	69.62	72.67	63.20
		Alg	79.08	77.84	79.63	77.47	80.86	70.32
HEC		96.56	95.05	95.60	94.60	98.74	85.87	

4. Conflict of interest

The authors declare that there is no conflict of interest

5. Acknowledgements

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