

Dyeability of Surface-Modified Cotton and Silk Fabrics with PAMAM Dendrimer Using *Sargassum* sp. Seaweed Extract

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Abstract

In this study, a PAMAM dendrimer was used as a surface-modifier to study the dyeability of cotton and silk fabrics using a sustainable dye generated from *Sargassum* sp. seaweed extracts. PAMAM dendrimer is a polymeric molecule characterized with multiple branched monomers containing numerous functional primary amino groups on the surface. These amine groups would readily act as a functional group similar to the structure of the natural protein fibers which is capable to improve the dyeability of textile fibers, especially with natural dyes. The crushed powder of dry *Sargassum* sp. was macerated for 48 h at 60 °C in methanol. The cotton and silk materials were pre-treated with a PAMAM dendrimer at concentrations that were determined in advance before the dyeing process began. The electrokinetic capabilities of these surface-modified fabrics were next investigated by measuring their zeta potential with a SurPASS Electrokinetic Analyzer. Exhaust dyeing and simultaneous mordanting with vinegar, alum and iron were done on both untreated and treated cotton and silk fabrics with a certain amount of PAMAM dendrimer. The dyeing process was carried out at 85 °C for intervals of 40, 60 and 80 min. It was discovered that surface-modified cotton and silk materials had a lower negative charge zeta potential, which enhanced their dyeability as well as the percent exhaustion and K/S values of the dyed fabrics. As a result of the surface modification, the zeta potential of the cotton fabric went from -20.40 to -14.63 mV. Similarly, the zeta potential of the surface-modified silk fabric, on the other hand, went from -36.80 to -26.85 mV. However, the percent exhaustion of surface-modified cotton and silk fabrics was increased to 21.17 and 39.99, respectively. Likewise, the K/S ratio of the surface-modified cotton fabric was raised from 0.06 to 0.20. Although the K/S value of the surface-modified silk fabric increased from 0.15 to 0.24. Additionally, the fastness properties of the dyed fabrics were graded as good to exceptional (between 4 and 5), except for lightfastness, which was classified as poor (rated between 3 to 5). Consequently, the sustainable dye from *Sargassum* sp. seaweed was successfully applied to cotton and silk fabrics.

Keywords: Dyeability, Maceration, PAMAM dendrimer, *Sargassum* sp., Simultaneous mordanting, Surface modification, Sustainable dye, Zeta potential

Introduction

In the textile business, synthetic dyes are widely utilized due to their availability, ease of production and the large range of colors. They also have a high color intensity and are easy to use. Due to the toxicity and detrimental effects of synthetic dyes on the environment and living organisms, the global demand for sustainable dyes derived from renewable and sustainable bio-resources is developing at an alarming rate [1,2]. In addition, sustainable dyes have grown in favor because they make textiles that are more original, unusual, soothing, soft tones and multi-hued than synthetic dyes, and they are also environmentally benign [3,4].

Seaweeds are multicellular, macroscopic, eukaryotic and autotrophic marine macroalgae. Taxonomically, they are divided into 3 large groups based on the color (pigment) of the thallus: Chlorophyta (green algae), Rhodophyta (red algae) and Phaeophyta (blue-green algae) (brown algae). They can be grown in a controlled environment or picked from the wild [5]. Most brown seaweeds contain fucoxanthin a carotenoid pigment as the primary coloring agent [6].

Sargassum species are brown seaweeds found in the subtidal and intertidal zones of tropical and subtropical waters [7]. It is the most aggressive marine intruder, having originated in South East Asia [8]. Seaweed is abundantly grown in Malaysia for carrageenan production, primarily in the state of Sabah,

specifically off the coasts of Semporna, Kunak, Lahad Datu, Tawau and Kudat [9]. Malaysia was the world's third-largest producer of dried seaweed in 2016, with a total production of 20,297 tonnes [10].

The dyeability and wet finishing of textile fibers depend on their chemical composition, hydrophilicity, density of cross-linking, roughness, crystallinity, conductivity and lubricity. For instance, the presence of specific carboxylic acid groups due to oxidation of the primary hydroxylic sites in natural cellulose fibers, such as cotton, generates negative surface charges (zeta potential) [11].

Additionally, as the pH of the dye bath rises over 8 the hydroxyl groups in the side chains of the hydroxymethyl groups may be ionized and promptly develop the zeta potential (negative surface charges) of the fiber [12]. As a result, anionic dyes are repulsed by the surface's negative charges, reducing the effectiveness of the dye exhaustion and therefore decreasing dye fixation in cellulose fibers [13].

However, the formation of negative surface charges of silk fiber is due to the preferential adsorption of hydroxide ions (OH⁻) from water and the establishment of negatively-charged carboxylate ions (COO⁻). Subsequently, in order to attract anionic dye molecules, the surface of the fiber must be positive in charge. Therefore, surface modification has a substantial influence on textile fiber processing and performance.

In recent years, surface modification of textile fibers has superseded conventional dyeing and printing as the most popular method for producing high-end textile treatments [14]. The treatment only modifies the surface of the fibers, not their bulk qualities [15], allowing the fibers to retain their original quality and appearance.

Dendrimers are a type of low-molecular-weight, nano-sized highly branched polymer and radially symmetric molecules with several functional groups comprising a central core and end groups that accelerate the interaction with other molecules [16]. The nature of the outer functional groups determines the dendrimers' reactivity and solubility. Due to the smaller molecular sizes, dendrimers have remarkable penetrability in fiber thus enhancing the exhaustion and fixing of reactive dyes on the modified cotton fabric [17] even in the absence of electrolytes and alkali [18]. Furthermore, Khakzar *et al.* [19] discovered that the color strength and fastness properties of direct dye on surface-modified cotton fabric with poly(propylene imine) dendrimer improved significantly.

Materials and methods

Seaweed and fabric preparation

The identified fresh brown seaweeds of the species *Sargassum* sp. were obtained from Perusahaan Rumpai Laut Juni in Semporna, which is located in the state of Sabah in East Malaysia. In order to prevent the seaweed from becoming deteriorated while being transported to Peninsular Malaysia, it was first exposed to the sun and allowed to dry up completely. Once obtained, the sun-dried *Sargassum* sp. seaweeds were soaked for 24 h in tap water to remove any remaining epiphytes, salt and sand. The seaweeds were then thoroughly washed, rinsed with tap water and drained before being dried in an oven for 36 h at 70 °C [20]. Prior to the color extraction procedures, the dried *Sargassum* sp. seaweeds were ground into a fine powder using a domestic blender.

Cotton and silk fabrics were chosen for this study because they are the most commonly used fibers in Malaysia's apparel and clothing industries, especially in the batik industry. The total numbers of samples studied are 16 samples involving 4 sets of samples which are untreated and treated with 0.5, 1.0 and 1.5 % PAMAM dendrimer. Each set contains 4 samples which are un-mordanted as well as mordanted with vinegar, alum and iron.

The fabric specifications are presented in **Table 1** and the established standard test methods are tabulated in **Table 2**.

Table 1 Fabrics specification.

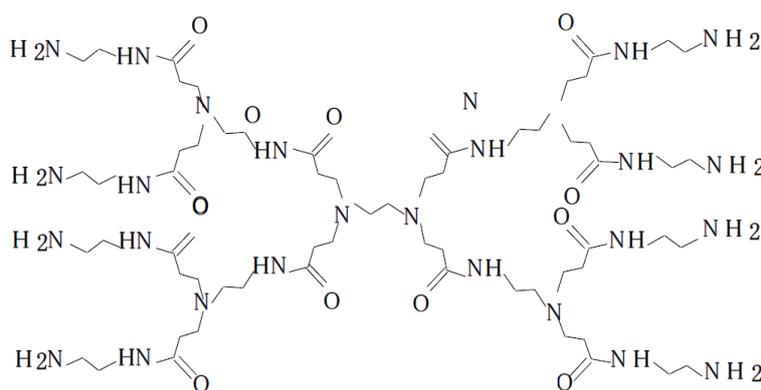
Type of fabric	Structure	Weight (g/m ²)	Thickness (mm)	Density (threads/cm)
100 % Cotton	Plain weave	111.50	0.37	Warp: 121 Weft: 60
100 % Silk	5 ends satin weave	66.30	0.34	Warp: 335 Weft: 123

Table 2 Standard tests methods for fabric measurements.

Fabric parameters	Standard test methods	Equipment
Weight	MS ISO 3801-2003	Weighing balance
Thickness	MS ISO 5084-2003	Thickness gauge
Density	MS ISO 7211/2-2003	Counting glass, pick counter, counterbalance and weighing balance

Chemical

The generation 1 poly(amidoamine) (PAMAM) dendrimer was purchased from Weihai CY Dendrimer Technology, Co. Ltd., China to be used as a surface modifier on the cotton and silk fabrics. This dendrimer contains 8 -NH₂ end groups as shown in **Figure 1**. Vinegar, aluminum potassium sulfate (alum) and ferrous sulfate (iron) were utilized as mordants in this study.

**Figure 1** Structure of PAMAM dendrimer (generation 1).

Extraction procedures

The maceration procedure was utilized to extract the colorant from the *Sargassum* sp. seaweed powder. The seaweed powder was placed in a conical flask containing methanol and the maceration procedure was performed in a water bath for 48 h at 60 °C. The conical flask was wrapped and covered with aluminum foil to prevent solvent loss due to evaporation during maceration. The mixture was then strained through a fine mesh to get the extracts. The extracted solution was vacuum evaporated using a rotary evaporator and then dried in an oven and a desiccator to acquire the crude colorant, which was then used in the dyeing process.

Surface modification of cotton and silk fabrics

Cotton and silk fabrics were treated at 70 °C for 45 min with PAMAM dendrimer at 3 different concentrations (0.5, 1.0 and 1.5 % based on the weight of fabric, owf) using the exhaustion method [21] keeping the liquor ratio at 1:20. After being treated, the fabrics were dried at 100 °C for 5 min and then cured at 150 °C for 3 min.

Zeta potential measurement

A SurPASS Electrokinetic Analyzer (Anton Paar GmbH, Graz, Austria) with a cylindrical measurement cell was used to measure the zeta potential of the fabrics. The measurements were carried out at the Melbourne Centre for Nanofabrication (ANFF - Victorian Node), an Australian National Fabrication Facility (ANFF) in Clayton, Victoria, Australia.

Zeta potentials of the untreated and treated fabrics with 0.5, 1.0 and 1.5 % of PAMAM dendrimer were measured at room temperature (~25 °C) in 1mM potassium chloride (KCl) solution and sample pH was set at 6.0. Prior to the zeta potential measurements, the pH and conductivity electrodes were first calibrated using standard calibration solutions. After preparation and during measurement, the electrolyte solution was immediately purged with nitrogen (N₂) to prevent the dissolution of CO₂ from the air into the electrolyte solution.

An approximately 50×50 mm² piece of the treated fabric was placed between 2 support discs in each cylindrical glass cell for each experiment. The sample was sandwiched between 2 porous discs and 2

measuring heads (electrodes) were inserted into the glass measurement cylinder from each side. Initially, the measuring cell was installed horizontally.

The sample was washed in an electrolyte solution for 300 s to remove any air bubbles before being squeezed with the knob to reduce the distance between the electrodes until the friction clutch prevented further compression of the sample. To determine the streaming potential of the fabric sample, an electrolyte solution was pumped through it at 200 mbar pressure.

Dyeing of cotton and silk fabrics

Cotton and silk fabrics were dyed with 2 % (owf) of colorant in crude form, maintaining a liquor ratio at 1:20. Simultaneously, 2 % of mordant (owf) was employed in the same dye bath to fix the colorant onto the cotton and silk fabrics. Exhaustion dyeing was carried out on both untreated and treated cotton and silk fabrics with PAMAM dendrimer for 40, 60 and 80 min at 85 °C [23]. The optimal dyeing time was determined to be used in subsequent research. Furthermore, the effects of PAMAM dendrimer on dyed cotton and silk fabrics were examined and evaluated.

Color assessments

The CIELAB color coordinates, colorimetric properties such as K/S (color strength) values and % exhaustion and fastness properties of the selected dyed cotton and silk fabrics (based on optimum dyeing time) were evaluated. The colors were measured using a HunterLab LabScan XE (LSXE) spectrophotometer and analyzed with HunterLab EasyMatchQC software.

The K/S values are attained directly from the instrument, which adheres to the Kubelka-Munk equation.

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

where K denotes the sample's absorption coefficient, S indicates the sample's scattering coefficient, and R is the degree of reflectance at the maximum absorption [24].

Eq. (2) is used to calculate the percentage exhaustion (also known as the percentage of dye bath exhaustion) based on the absorbance values recorded before and after dyeing.

$$\% E = \frac{A_0 - A_1}{A_0} \times 100 \quad (2)$$

where % E represents the dye exhaustion, A_0 represents the absorbance of the dye bath before dyeing and A_1 represents the absorbance of the dye bath after dyeing [25].

Fastness properties

The fastness properties toward washing, perspiration, rubbing/crocking and light of the dyed cotton and silk fabrics were evaluated in accordance with MS ISO standards, as tabulated in **Table 3**. The greyscale rating for the washing, perspiration and rubbing (crocking) ranged from 1 to 5 where 1 denotes the very worst possible performance and 5 denotes the best possible performance. The rating for lightfastness is ranged from 1 to 8 based on the Blue Wool Standard where 1 indicates a very poor rating and 8 indicates an excellent rating.

Table 3 Standard tests methods for fastness properties.

Colorfastness	Standard Methods	Equipment
Washing	MS ISO 105-C01-1966	Auto-wash
	MS ISO 105-A05-2003	Change in color
	MS ISO 105-A04-2003	Staining
Perspiration	MS ISO 105-E04-1996	Perspirometer
	MS ISO 105-A05-2003	Change in color
	MS ISO 105-A04-2003	Staining
Rubbing/Crocking	MS ISO 105-X12-2001	Crockmeter
	MS ISO 105-A04-2003	Staining
Light	MS ISO 105-B02-2001	Light fastness tester

Results and discussion

Zeta potential analysis

The zeta potential of untreated and treated cotton and silk fabrics with PAMAM dendrimer is depicted in **Figure 2**. Untreated cotton fabric has a zeta potential value of -20.40 mV. In contrast, the zeta potential value of untreated silk fabric was -36.80 mV. Cotton fibers have a negatively surface charge due to the presence of acidic carboxyl ($-\text{COOH}$), sulphonic acid ($-\text{SO}_3\text{H}$), phenolic ($-\text{C}_6\text{H}_5\text{OH}$) and hydroxyl ($-\text{OH}$) groups when brought in contact with water [26].

On the other hand, the presence of functional groups such as hydroxyl ($-\text{OH}$), sulphonyl acid ($-\text{SO}_3\text{H}$), acidic carboxyl ($-\text{COOH}$), phenolic ($-\text{C}_6\text{H}_5\text{OH}$), and amino ($-\text{NH}_2$) contributes to the surface charges of silk fabric [27]. In contrast, the zeta potential values of treated cotton fabrics with 0.5, 1.0 and 1.5 % PAMAM dendrimer were -18.23 , -15.48 and -14.50 mV, respectively. However, the zeta potential values of silk fabrics treated with 0.5, 1.0 and 1.5 % PAMAM dendrimer were -30.77 , -28.19 and -26.85 mV, respectively. It was found that the positive surface charge ($-\text{NH}_2$ groups) of cationic PAMAM dendrimers reduces the negative surface charges of those treated fabrics.

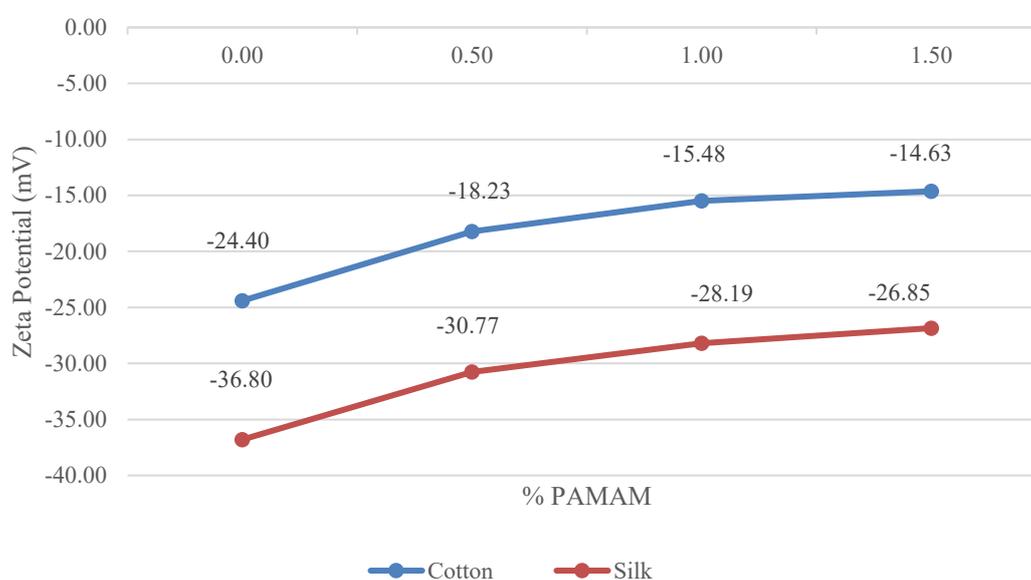


Figure 2 Zeta potential values of cotton and silk fabrics.

Effects of dyeing times on dyeing behavior of cotton and silk fabrics

Preliminary studies were undertaken on the dyeing behavior of untreated cotton and silk fabrics using seaweed extracts without the addition of a mordant (as controlled samples) at 85°C for 40, 60 and 80 min in order to determine the optimum dyeing time for subsequent studies. **Figures 3** and **4** show that the % exhaustion and K/S values of cotton and silk fabrics increased substantially when dyeing was carried out for 60 min as opposed to dyeing that was carried out for 40 or 80 min. At this optimum dyeing time, the % exhaustion of the dyed cotton was 17.39 and silk fabric was 23.87 %. Whereas the K/S values of the dyed cotton and silk fabric were 0.06 and 0.15, respectively.

Yet, when the dyeing time was raised from 60 to 80 min, both % exhaustion and K/S values of the fabrics were decreased. It seems that the equilibrium state of dyeing on cotton and silk fabrics was reached at 60 min, which may have resulted in slight desorption of dye. Similarly, the K/S values of the dyed cotton and silk fabrics reached their peak at 60 min of dyeing, indicating that the fabrics had attained maximum dye absorption. It was found that increasing the dyeing time would increase the K/S values.

However, once equilibrium was reached, the K/S values reduced even though the dyeing time increased further. Previously, Hasan *et al.* [28] observed that dyeing fabrics for a longer time decreased the K/S values. This over-dyeing process may cause the dye molecules to desorb out from the fibers into the dye bath. As a result, the optimum dyeing time (the time for maximum dye exhaustion) may be affected by a number of factors such as the percentage of shade and other dyeing conditions.

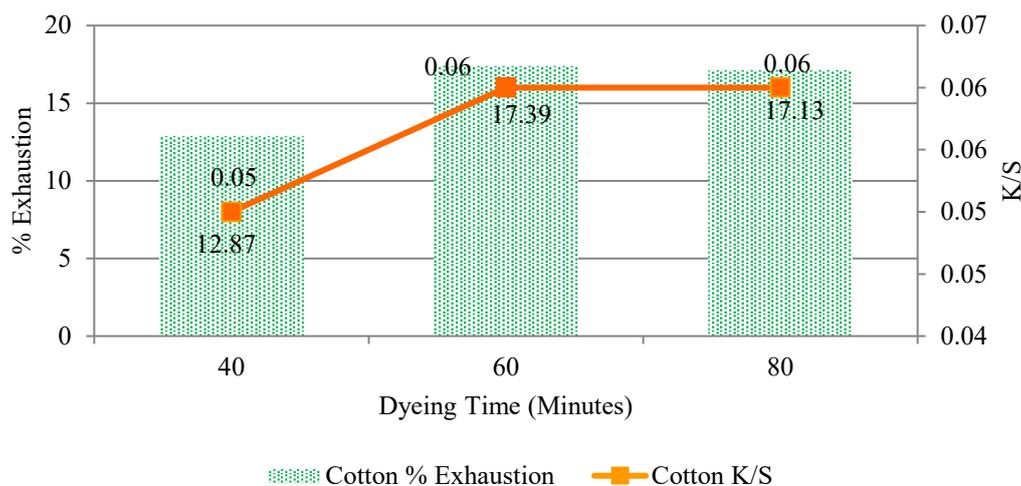


Figure 3 Relation between dyeing times, % exhaustion and K/S of untreated cotton fabric.

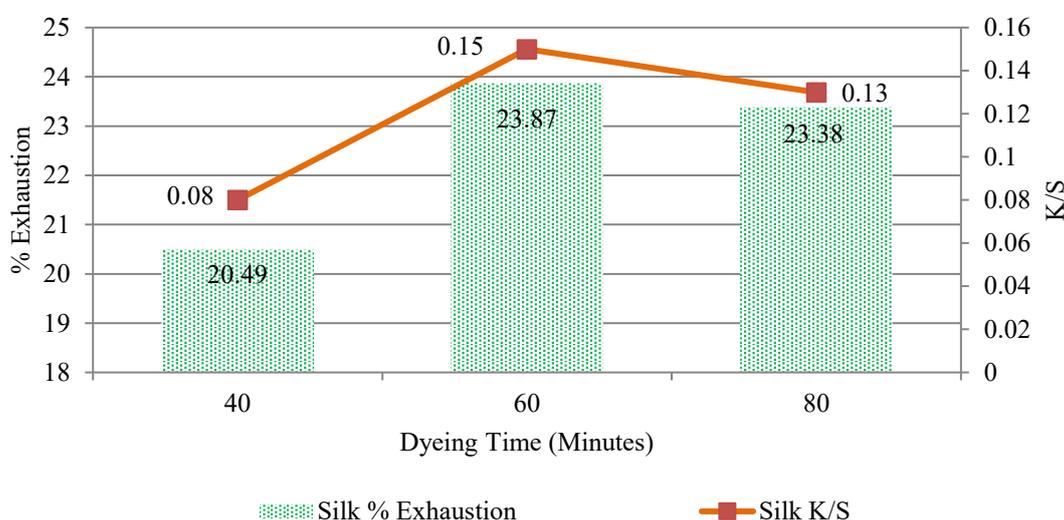


Figure 4 Relation between dyeing times, % exhaustion and K/S of untreated silk fabric.

Effects of zeta potential on dyeing behavior of surface-modified cotton and silk fabrics

Based on the optimum dyeing time, an application of PAMAM dendrimer as surface modifier would reduce the negative surface charges on the treated fabrics due to the addition of cationic charges on the fabric surface, which would result in more dye sites being created [29]. These results imply that the adherence of dye anions to fiber surfaces may increase, hence enhancing the dyeability of cotton and silk fabrics with sustainable dyes obtained from the seaweed *Sargassum* sp. This result was in agreement with studies done by Maiti *et al.* [18]; Khakzar *et al.* [19].

Surface treatments with PAMAM dendrimer improve both the % exhaustion and the K/S of the dyed fabrics. **Figure 5** depicts the effects of PAMAM dendrimer treatment on cotton fabrics. While the effects of PAMAM treatment on silk fabrics are depicted in **Figure 6**. The patterns of % exhaustion and K/S are comparable to those of the zeta potential values of pretreated fabrics with PAMAM dendrimer. In general, the highest values of % exhaustion and K/S were attained on 1.0 % PAMAM-treated cotton and silk fabrics. This is due to the structure of PAMAM dendrimer, which has a relatively high number of reactive primary amino groups ($-NH_2$) as a functional group and adds additional dye sites to cotton and silk fibers [17,18].

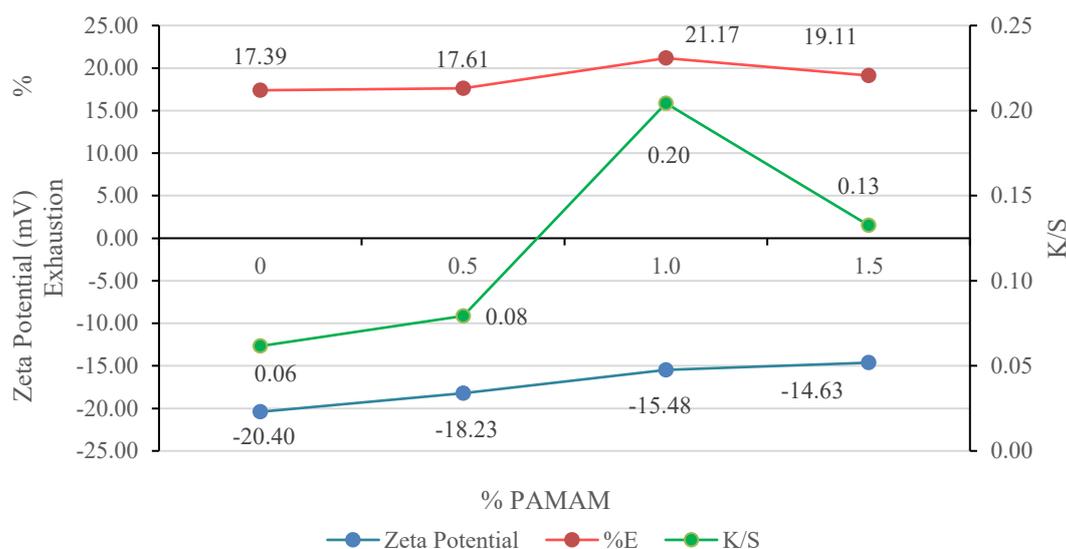


Figure 5 Effect of zeta potential on % exhaustion and K/S of treated cotton fabric with pamam dendrimer.

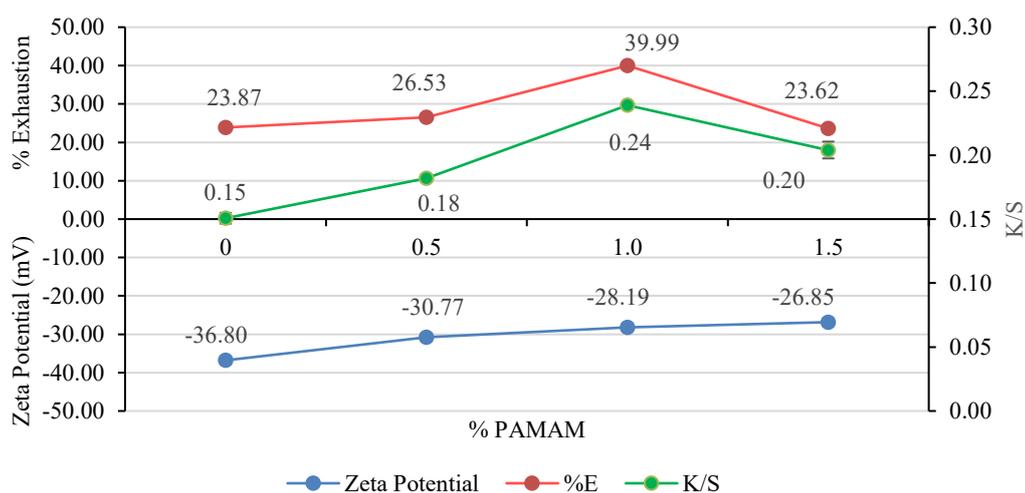


Figure 6 Effect of zeta potential on % exhaustion and K/S of treated silk fabrics with pamam dendrimer.

Overall, this study found that the % exhaustion and K/S values increase when the negative surface charge of pretreated and dyed fabrics is decreased. Additionally, the chemical composition of the fibers, surface polarity, microstructure, porosity, specific surface, fiber swelling capacity, the interaction energy of the fiber and solution, pH of the solution and the addition of electrolytes play an important role in the addition of positive charges (reduction of negative charges) on the surface of the fibers [30,31].

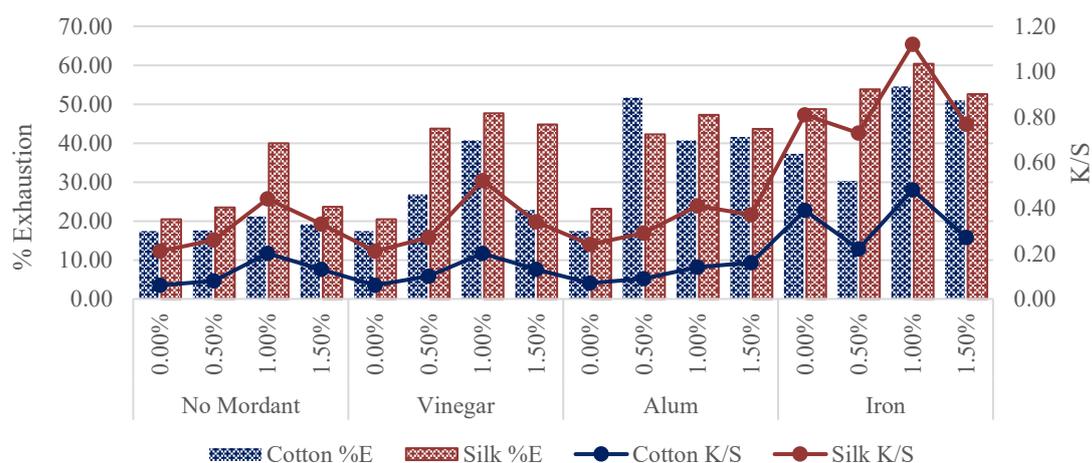
Effects of PAMAM dendrimer and mordants on dyeing behavior of surface-modified cotton and silk fabrics

The % exhaustion and K/S values of dyed cotton and silk fibers treated with PAMAM dendrimer and various mordants, including vinegar, alum, and iron are tabulated in **Table 4**. In general, the % exhaustion and K/S values rose together with the concentrations of PAMAM dendrimer. Treated cotton fabric with 1.0 % PAMAM dendrimer and iron-mordanted had the highest percentage of exhaustion i.e. 54.52 %. Apparently, similar treatment parameters as carried out on the cotton fabric gave the highest % exhaustion of treated silk fabric i.e. 60.41 %.

However, treatment with 1.5 % PAMAM reduced the percentage of dye exhaustion on both dyed cotton and silk fabrics. The best dye absorption was most likely observed on treated cotton and silk fabrics with 1 % PAMAM dendrimer. **Figure 7** illustrates the simplified readings.

Table 4 % Exhaustion and K/S values of surface-modified cotton and silk fabrics with different mordants.

Mordant	PAMAM dendrimer (%)	Cotton		Silk	
		% E	K/S	% E	K/S
No mordant	0.00 %	17.39	0.06	20.46	0.15
	0.50 %	17.61	0.08	23.46	0.18
	1.00 %	21.17	0.20	39.99	0.24
	1.50 %	19.11	0.13	23.62	0.20
Vinegar	0.00 %	17.39	0.06	20.46	0.15
	0.50 %	26.88	0.10	43.73	0.17
	1.00 %	40.73	0.20	47.66	0.32
	1.50 %	22.86	0.13	44.78	0.21
Alum	0.00 %	17.45	0.07	23.14	0.17
	0.50 %	51.70	0.09	42.29	0.20
	1.00 %	40.71	0.14	47.23	0.27
	1.50 %	41.57	0.16	43.68	0.21
Iron	0.00 %	37.29	0.39	48.73	0.42
	0.50 %	30.33	0.22	53.81	0.51
	1.00 %	54.52	0.48	60.41	0.64
	1.50 %	51.02	0.27	52.59	0.50

**Figure 7** Comparison of % exhaustion and K/S values of treated cotton and silk fabrics.

According to the data presented in **Table 4** and **Figure 7**, the treated and dyed fabrics exhibit substantial differences in their K/S values between one another. The highest K/S values for both treated cotton and silk fabrics were obtained from 1.0 % PAMAM dendrimer and mordanted with iron i.e. 0.48 and 0.64, respectively. The addition of mordant is another factor that contributes to the improvement in both the K/S values and % exhaustion.

CIELAB color coordinates

The CIELAB color coordinates (L^* , a^* and b^*) of the dyed cotton and silk fabrics treated with PAMAM dendrimer are shown in **Table 5**. Apparently, the effect of iron as a mordant was more prominent when compared to the effects of the other mordants as represented by L^* values. The L^* values represent perceived lightness or darkness, with 0 representing black and 100 representing white. The treatment with 1.0 % PAMAM dendrimer and iron as a mordant produced the darkest shades among all the dyed cotton and silk fabrics. These samples had the lowest L^* values, which were 70.89 and 68.94, respectively.

It was clear that the addition of mordants can alter the shades of the dyed fabrics as represented by the a^* and b^* values. The use of vinegar and alum as mordants on cotton and silk fabrics treated with PAMAM dendrimer resulted in negative a^* ($-a^*$) and positive b^* ($+b^*$) values, as shown in **Table 5**. The chromatic a^* axis stretches from green ($-a^*$) to red ($+a^*$), while the b^* axis stretches from blue ($-b^*$) to yellow ($+b^*$). The resulting shades were found in the green-yellow region, as indicated by the coordinates. While iron mordanted on treated cotton and silk fabrics with PAMAM dendrimer gave positive values for both a^* and b^* . These results show that the shades produced were established in the red-yellow spectrum. **Table 6** shows the swatches of the dyed cotton and silk fabrics. The resulting shades are greyish and brownish depending on the surface treatments employed and the mordants utilized in the dyeing procedure. Mordant appears to have a greater impact on the shades obtained. Surface treatment with PAMAM dendrimer, on the other hand, slightly changes the shade of the dyed fabrics.

Table 5 The CIELAB color coordinates for dyed cotton and silk fabrics treated with PAMAM dendrimer.

Mordant	PAMAM dendrimer (%)	Cotton			Silk		
		L^*	a^*	b^*	L^*	a^*	b^*
Undyed		91.14	1.31	-15.63	90.57	2.28	-5.92
No mordant	Untreated	88.50	-0.11	-1.87	82.86	-0.47	10.99
	0.50 %	88.15	0.31	0.12	80.92	-1.27	11.45
	1.00 %	77.42	-1.12	12.83	79.94	-0.64	11.92
	1.50 %	81.60	-0.84	11.31	80.77	0.12	10.73
Vinegar	Untreated	88.24	-0.32	-0.60	84.13	0.13	8.41
	0.50 %	85.79	-0.59	2.65	82.85	-0.61	10.05
	1.00 %	79.31	-1.44	11.09	80.48	-0.91	11.13
	1.50 %	83.74	-1.16	8.99	80.20	-0.41	9.94
Alum	Untreated	87.85	-0.68	0.48	83.67	0.40	7.96
	0.50 %	86.33	-0.60	2.57	82.55	-0.85	10.75
	1.00 %	83.48	-1.22	5.58	82.16	-0.77	9.51
	1.50 %	82.94	-1.32	8.42	80.77	-0.49	10.94
Iron	Untreated	75.50	4.47	18.46	71.60	4.34	21.28
	0.50 %	80.32	2.46	12.24	70.33	4.67	23.50
	1.00 %	70.89	4.13	22.19	68.94	5.97	26.86
	1.50 %	73.62	3.86	21.74	70.03	5.27	24.08

Table 6 Swatches of the dyed cotton and silk fabrics.

Mordants	Untreated	Treated with PAMAM dendrimer					
		Cotton			Silk		
		0.50 %	1.00 %	1.50 %	0.50 %	1.00 %	1.50 %
No mordant							
Vinegar							

Table 8 Fastness properties of treated silk fabric with PAMAM dendrimer.

Treatments	Mordants	Washing		Perspiration		Rubbing/ Crocking		Light		
		Change in color	Staining	Change in color	Staining	Dry	Wet			
			Cotton	Silk		Cotton	Silk			
Untreated	No mordant	4/5	5	5	4/5	4	4	4/5	4	3
	Vinegar	4/5	4/5	5	4/5	4/5	4/5	4/5	4/5	3
	Alum	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	3
	Iron	4/5	4	4/5	4	4	4/5	4	4	3
0.5 % PAMAM	No mordant	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	3
	Vinegar	4/5	5	5	4/5	5	5	5	4/5	5
	Alum	4/5	5	5	4/5	5	5	5	4/5	5
	Iron	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4
1.0 % PAMAM	No mordant	4/5	5	5	4/5	4/5	4/5	4/5	4/5	4
	Vinegar	5	5	5	4/5	4/5	4/5	4/5	4/5	4
	Alum	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4
	Iron	4/5	4/5	4/5	4/5	4/5	4	4/5	4/5	3
1.5 % PAMAM	No mordant	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4
	Vinegar	5	5	4/5	4/5	5	4/5	4/5	4/5	4
	Alum	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4
	Iron	4/5	4	4	4/5	4/5	4	4/5	4/5	4

Conclusions

The negative surface charges or zeta potential of the cotton and silk fabrics have been successfully decreased by surface modification using PAMAM dendrimer. The decrease in negative charges on the fabric surface diminishes the repulsion force between anionic dye molecules and the fabrics. Therefore, the possibility of anionic dye molecules being occupied by the fabric has increased.

Consequently, even without the presence of a mordant, the dyeability of surface-modified cotton and silk fabrics with PAMAM dendrimer was considerably enhanced. PAMAM dendrimer could boost the positive charges by introducing amino groups ($-NH_2$) to the surface of the fabric. The addition of positively charged amino groups increases the affinity between sustainable dyes and pretreated fabric. It can be concluded that the pretreated fabrics with PAMAM dendrimer were able to absorb more dyes and produce darker shades in comparison to the untreated ones.

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References

- [1] K Gong, Y Pan, LJ Rather, W Wang, Q Zhou, T Zhang and Q Li. Natural pigment during flora leaf senescence and its application in dyeing and UV protection finish of silk and wool - a case study of *Cinnamomum Camphora*. *Dyes Pigments* 2019; **166**, 114-21.
- [2] VN Swamy. Analysis and study of effect of natural dye from bombax malabarica on silk fabric. *Clothing Textil. Res. J.* 2020; **38**, 49-58.
- [3] J Arora, P Agarwal and G Gupta. Rainbow of natural dyes on textiles using plants extracts: Sustainable and eco-friendly processes. *Green Sustain. Chem.* 2017; **7**, 35-47.
- [4] AK Samanta. *Bio-dyes, bio-mordants and bio-finishes: Scientific analysis for their application on textiles*. In: AK Samanta, NS Awwad and HM Algarni (Eds.). Chemistry and technology of natural and synthetic dyes and pigments. IntechOpen, London, 2020.
- [5] J Venkatesan, B Lowe, S Anil, P Manivasagan, AA Al-Kheraif, K Kyong-Hwa and K Swe-Kwon. Seaweed polysaccharides and their potential biomedical applications. *Starch* 2015; **67**, 381-90.

- [6] S Méresse, M Fodil, F Fleury and B Chénais. Fucoxanthin, a marine-derived carotenoid from brown seaweeds and microalgae: A promising bioactive compound for cancer therapy. *Int. J. Mol. Sci.* 2020; **21**, 9273.
- [7] C Olabarria, IF Rodil, M Incera and JS Troncoso. Limited impact of *Sargassum muticum* on native algal assemblages from rocky intertidal shores. *Mar. Environ. Res.* 2009; **67**, 153-8.
- [8] T Wernberg, MS Thomson, PA Staehr and MF Pederson. Comparative phenology of *Sargassum muticum* and *Halidrys siliquosa* (Phaeophyceae: Fucales) in Limfjorden, Denmark. *Bot. Mar.* 2000; **43**, 31-9.
- [9] A Sade, I Ali and MRM Ariff. The seaweed industry in Sabah, East Malaysia. *JATI J. Southeast Asian Stud.* 2006; **11**, 97-107.
- [10] Department of Fisheries Malaysia. *Annual fisheries statistics 2016*. Department of Fisheries Malaysia, Putrajaya, Malaysia, 2016.
- [11] SM Phang. Seaweed resources in Malaysia: Current status and future prospects. *Aquat. Ecosys. Health Manag.* 2006; **9**, 185-202.
- [12] Department of Fisheries Malaysia. Perangkaan perikanan tahunan 2016 (*in Malay*), Available at: <https://www.dof.gov.my/index.php/pages/view/3754>, accessed October 2019.
- [13] DRD Eranza, A Bahron, J Alin, R Mahmud and SR Malusirang. On-going assessment of issues in the seaweed farming industry in Sabah, Malaysia. *J. Asian Acad. Appl. Bus.* 2017; **4**, 49-60.
- [14] K Stana-Kleinschek and V Ribitsch. Electrokinetic properties of processed cellulose fibres. *Colloid. Surface. Physicochem. Eng. Aspect.* 1998; **140**, 127-38.
- [15] CM Carr. *Chemistry of the textiles industry*. Chapman & Hall, Cambridge, United Kingdom, 1995.
- [16] PE Froehling. Dendrimers and dyes - a review. *Dyes Pigments* 2001; **48**, 187-95.
- [17] W Ma, S Du, S Yan, X Yu, Z Zhang and S Zhang. Salt-free dyeing of modified cotton through graft polymerization with highly enhanced dye fixation and good strength properties. *Polymers* 2020; **12**, 462-74.
- [18] S Maiti, G Mahajan, S Phadke and RV Adivarekar. Application of polyamidoamine dendrimer in reactive dyeing of cotton. *J. Textil. Inst.* 2017; **109**, 823-31.
- [19] BF Khakzar, RMA Malik and F Mazaheri. The effect of dendrimer on cotton dyeability with direct dyes. *Chem. Ind. Chem. Eng. Q.* 2014; **20**, 379-85.
- [20] PS Shelar, VK Reddy, GS Shelar, M Kavitha, GP Kumar and GVS Reddy. Medicinal value of seaweeds and its applications - a review. *Continent. J. Pharmacol. Toxicol. Res.* 2012; **5**, 1-22.
- [21] D Sun. *Surface modification of textiles for improved mechanical properties and energy saving of finishing processes through plasma treatment*. In: S Kalia (Ed.). *Biodegradable green composites*. Wiley, New Jersey, 2016, p. 23-40.
- [22] AA Elzaawely and S Tawata. Effect of extraction and drying methods on the contents of kava pyrones and phenolic compounds in *Alpinia zerumbet* leaves. *Asian J. Plant Sci.* 2011; **10**, 414-8.
- [23] SM Anisuzzaman, A Bono, D Krishnaiah, B Ariffin and R Lee. Effects of alkali treatment process parameters on semi refined carrageenan functional groups. *Indian J. Chem. Tech.* 2014; **21**, 386-91.
- [24] SM Burkinshaw. *Physico-chemical aspects of textile coloration*. Wiley, New Jersey, 2016.
- [25] DM Lewis and LTT Vo. Dyeing cotton with reactive dyes under neutral conditions. *Color. Tech.* 2007; **123**, 306-11.
- [26] M Gahlot, P Bhatt and J Joshi. Study on yield of plant extract using different solvents and methods. *Bull. Environ. Pharmacol. Life Sci.* 2018; **7**, 65-7.
- [27] MJ Jeyakodi, A Mariappan and K Vellingiri. A study on the functional properties of silk and polyester/lyocell mixed fabric. *Matéria* 2015; **20**, 924-35.
- [28] M Hasan, KA Nayem, AYMA Azim and NC Ghosh. Application of purified lawsone as natural dye on cotton and silk fabric. *J. Textil.* 2015; **2015**, 932627.
- [29] S Hemwimon, P Pavasant and A Shotipruk. Microwave-assisted extraction of antioxidative anthraquinones from roots of *Morinda Citrifolia*. *Separ. Purif. Tech.* 2007; **54**, 44-50.
- [30] C Bellmann, A Caspari, V Albrecht, TTL Doan, E Mader, T Luxbacher and R Kohl. Electrokinetic properties of natural fibres. *Colloid. Surface. Physicochem. Eng. Aspect.* 2005; **267**, 19-23.
- [31] HJ Jacobasch. Characterization of solid-surfaces by electrokinetic measurements. *Progr. Org. Coating.* 1989; **17**, 115-33.