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THE EFFECT OF CATIONIC SURFACTANT TREATMENT ON THE DYEABILITY OF COTTON AND SILK FABRICS WITH NATURAL DYE FROM BROWN SEAWEEDS *SARGASSUM* SP

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ABSTRACT

This study focuses on the surface modification of cotton and silk fabrics which might improve their dyeability to *Sargassum* sp. extracts. The extraction was performed by macerating the *Sargassum* sp. powder in methanol solution at 60^0C for 48 hours in dark room. The extracted colourant was measured with UV-vis Spectrophotometer to analyse the peak absorbance (λmax) as well as to estimate the pigments content in *Sargassum* sp. Exhaustion dyeing with simultaneous mordanting procedure were then performed at 85^0C for 60 minutes on treated and untreated cotton and silk fabrics with Cetyl Trimethyl Ammonium Bromide (CTAB) at different concentrations. The dyed samples were then analysed their colourimetrics and fastness properties in accordance to MS ISO standard. The results showed that the application of CTAB gave higher colour differences and colour strength values which indicate that the dyeability of those fabrics was increased significantly with out diminishing their fastness properties.

KEYWORDS: Colour Differences, Colour Strength, Dyeability, Natural Dyes, *Sargassum* Sp

INTRODUCTION

Natural dyes can be extracted from natural sources such as plants (e.g., indigo and saffron); insects (e.g., cochineal and lac); animals (e.g., some species of mollusks or shellfish); and minerals (e.g., ferrous sulfate, ochre, and clay) without any chemical treatment (Bechtold *et.al.* (2007); Chengaiah *et.al*. (2010); Manhita *et.al.* (2011)). However, Balagurunathan *et al*. (2011) and Nerurkar *et al*. (2013) claimed that currently, extensive researches have successfully extracted natural dyes from microorganisms such as algae, fungi, bacteria, and actinomycetes. Historically, natural dyes are purposely used for food colouring (Spears (1988)), painting (Barnett *et.al*. (2006)) and textile dyeing (Bechtold *et.al.* (2007)) ; Nagia *et.al.* (2007)). The application of natural dyes in textile dyeing is back in demand because they possess many excellent properties such as eco-friendly, less toxic, little side effect, biodegradable and sustainable as well as unique in shades depending on the mordants used (Siva (2007) ; Wan Yunus *et.al.* (2012)).

Yet, natural dyes suffer from certain inherent limitations such as tedious extraction procedures, low colour yield, limited dyeability towards textile fibres as well as poor fastness properties (Saxena *et.al.* (2007)). Thus, extensive researches are conducted from time to time with the aim to improve colour yield, explore new potential and sustainable sources, look for simpler extraction procedures as well as improve dyeability of the fibres.

Seaweeds are primitive photosynthetic multi cellular eukaryotic marine algae that grow almost exclusively in the shallow waters at the edge of the world’s oceans. Seaweeds can be classified into three broad groups based on their pigment constituents which are brown algae (Phaeophyta), red algae (Rhodophyta) and green algae (Chlorophyta) (Peter *et.al.* (2010)). Arad *et.al.* (1992) and Prasanna *et al.* (2007) stated that algae has a bulk of natural pigments like chlorophyll, carotenoids and phycobiliproteins, which exhibit colours ranging from green, yellow, brown and red. Algae pigments have great commercial value as natural colorants in nutraceutical, cosmetics and pharmaceutical in dustry as well as their health benefits (Paoline et.al. (2006) ; Indira *et.al.* (2012)).

MATERIALS AND METHODS

Materials and Chemical

Sargassum sp. was collected from Semporna, in the state of Sabah. The collected seaweeds were throughly washed and cleaned with fresh water to remove salty matters and debris. The cleaned *Sargassum* sp. was then dried in the oven at 70^0C for 36 hours. The dried *Sargassum* sp. was ground into powder form. Fabrics of 100% plain weave cotton and silk were used as the substrates. Two percents (2%) of metallic salts of ferrous sulphate (iron) and potassium aluminum sulphate (alum) as well as vinegar were used as mordants for each different dyebaths. Methanol (99.9% purity) was supplied by Merck. High purity grade of Cetyl Trimethyl Ammonium Bromide (CTAB) with 99.0% purity was supplied by Amresco.

EXPERIMENTAL METHODS

Extraction of Dyes

Maceration extraction method was performed using methanol as a solvent. *Sargassum* sp. powder was soaked in methanol and the extractions were performed at room temperature as well as heated in water bath at 60^0C for 48 hours accordingly. Both mixtures were placed in a dark room maintaining a liquor ratio of 1:20 (w/v). The crude form of natural dye was obtained by evaporating the extracted solution using rotary evaporator. Exactly 5 grams of crude dye were diluted in 100ml of distilled water to produce 5% dye stock solution.

Percentage Crude Yields

The crude yield produced by methanol extraction was expressed in percentage and calculated based on the stated formulation.

$$\%\ \text{Crude Yield} = \frac{\text{Weight of Crude Obtained (g)}}{\text{Weight of Seaweed Powder Used (g)}} \times 100 \qquad (1)$$

Surface Treatments

The treatment was performed with different concentrations of CTAB which were 0.5%, 1.0% and 1.5% o.w.f at 70^0C for 45 minutes (Baliarsingh *et.al.* (2013)) using exhaustion method keeping a liquor ratio of 1:20 (w/v). The treated fabrics were then squeezed and dried at 100^0C for 5 minutes followed by curing at 150^0C for 3 minutes.

Spectrophotometric Analysis

The extracted colorants from *Sargassum* sp. were analysed using Perkin Elmer UV-Vis Spectrophotometer Lambda 35 Series. The UV-Vis spectrums of the colouring matter from the extracted solution of *Sargassum* sp. was obtained in the visible range of 300 – 700 nm. Then the pigment compounds presence in the extracted solution was determined by measuring the absorbance value at the maximum wavelength or peak absorbance (λ_{max}). The pigments profiles extracted in methanol were estimated according to the formula of Lichtentaler and Wellburn (1985) as stated below:

$$\text{Chlorophyll a (mg/100g)} = 15.65\,A_{666} - 7.340\,A_{653}$$

$$\text{Chlorophyll b (mg/100g)} = 27.05\,A_{653} - 11.21\,A_{666}$$

$$\text{Carotenoids (mg/100g)} = 1000A_{470} - 2.860C_a - 129.2C_b/245$$

$$\text{Where: } C_a = \text{Chlorophyll a},\ C_b = \text{Chlorophyll b} \quad (2)$$

Dyeing of Cotton Nd Silk Fabrics

Cotton and silk fabrics were dyed with the extracted colorants in laboratory dyeing machine using exhaustion dyeing technique. Two percent (2%) dyeing of the dye stock solution was prepared based on weight of fabrics keeping up a liquor ratio of 1:20. Two percent (2%) of each mordant was used to fix the colorant onto the fabrics. Dyeing and mordanting were carried out simultaneously in one bath. The dyeing process was performed at 85^0C for 60 minutes (Muhammad Ismail *et.al.* (2013)). After the dyeing cycle was completed, the dyed fabrics were washed, rinsed with tap water and soap at boil for 20 minutes using 1 g/l standard soap followed by drying process under shade.

Fastness Properties

The dyed samples were evaluated in accordance to MS ISO standards as listed in Table 1. Colour fastness to washing, perspiration, rubbing/crocking and light were determined accordingly. The colour coordinates, colour differences (ΔE) and K/S values (colour strength) were measured using HunterLab LabScan XE (LSXE) spectrophotometer and analysed using HunterLab EasyMatchQC software under illuminant D65, 10^0 standard observer.

Table 1: Standard Methods Used for Colourfastness Assessments

Colourfastness	Standard Methods	Equipments
Washing	MS ISO 105-C01-1966 MS ISO 105-A05-2003 MS ISO 105-A04-2003	Auto-wash Change in Colour Staining
Perspiration	MS ISO 105-E04-1996 MS ISO 105-A05-2003 MS ISO 105-A04-2003	Perspirometer Change in Colour Staining
Rubbing/ Crocking	MS ISO 105-X12-2001 MS ISO 105-A04-2003	Crockmeter Staining
Light	MS ISO 105-B02-2001	Light Fastness Tester

Colour Coordinates and Colourimetrics Properties

The reflectance of the dyed fabrics was measured to visualize the colour coordinated values (CIE L* a* b*). The L* values indicate perceived lightness or darkness. Value of 0 indicates black and 100 indicates white. The values of a* indicate red (+a) and green (-a) while b* indicates yellow (+b) and blue (-b). The colour difference, (ΔE) was also calculated to investigate the apparent colours of the dyed fabrics by measuring the L*, a* and b* values for both undyed and dyed fabrics using the following equations (Kumar et.al. (2008)).

$$\Delta E = [(\Delta L^*) 2 + (\Delta a^*) 2 + (\Delta b^*) 2]1/2$$

Where: $\Delta L^* = L^*_{dyed} - L^*_{undyed}$, $\Delta a^* = a^*_{dyed} - a^*_{undyed}$, $\Delta b^* = b^*_{dyed} - b^*_{undyed}$ (3)

The colour strength (K/S) values were calculated in accordance to Kubelka-Monk equation to determine the dye uptake of each dyed fabric at maximum absorption wavelength from the reflectance value.

$$K/S = ([1-0.01R]2)/(2[0.01R]) \quad (4)$$

RESULTS AND DISCUSSIONS

Percentage Crude Yield And

Room temperature extraction produced 1.525% crude yield which is lesser in comparison with that produced at 60^0C which was 2.013%. The higher temperature might have softened the seaweed tissue as well as enhanced mass transfer and penetration of solvent into the seaweed tissue, thus accelerating the whole extraction (Hemwimon *et.al.* (2007) ; Al-Farsi *et.al.* (2008)).

Pigments Profiles

Two absorption peaks were shown at 405 nm and 664 nm in the UV-Vis spectrum as illustrated in Figure 2. Different pigments absorb light at different wavelengths. Mercadante (2008) found that most carotenoids absorb light in the region between 400-500 nm. Whilst, Marquez and Sinnecker (2008) stated that chlorophylls absorb light at the wavelength around 660 nm. The absorbance peaks from the UV-Vis spectrum showed that the presence of carotenoid which absorb light at 405 nm and chlorophyll absorb light at 664 nm in the extract solution. The absorbance peaks at those mentioned wavelengths are 0.42103 and 0.14577 respectively.

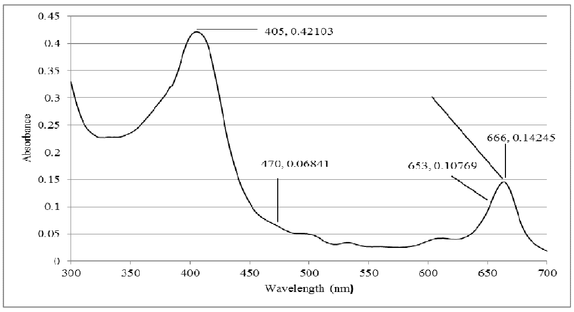

Figure 2: Absorbance Spectra of *Sargassum* Sp

Based on the formula of Lichtentaler and Wellburn and the absorbance curve in Figure 2, the estimated amount of pigments in the extracted *Sargassum* sp. seaweed is shown in Table 2.

Table 2: The Estimated Amount of Pigments in the Extracted *Sargassum* Sp

Types of Pigments	Amount (Mg/100g)
Chlorophyll a	1.4389
Chlorophyll b	1.3162
Carotenoids	63.6438

Colour Coordinates and Colourimetrics Analysis

The colour coordinates of the dyed cotton and silk fabrics are shown in Tables 3 and 4 accordingly. Generally, the effect of iron was more prominent in comparison with the rest of the other mordants. The darkest shade of dyed cotton fabric was obtained from treated with 1.0% CTAB and mordanted with iron was found to be 74.44 which is the lowest L* values found in all dyed cotton fabrics.

On the other hand, for the dyed silk fabric, the darkest shade was obtained from samples treated with 0.5% CTAB and mordanted with iron. The L* value was 70.85. Generally, the addition of mordants did not significantly contribute to the L* values. The mordants seem to be more significant in changing the shades.

Based on a* and b* values, the addition of iron as mordant increased the reddish-brown shades for both cotton and silk fabrics. Though, other mordants gave greenish shades as described by the values of a* and b*.

Table 3: Colour Coordinates of Dyed Cotton Fabric

Fabric	Treatments		L*	A*	B*
	Undyed		**91.14**	**1.31**	**-15.63**
Cotton	No Mordant	Untreated	88.50	-0.11	-1.87
		0.5% CTAB	85.85	-1.27	6.97
		1.0% CTAB	80.86	-1.97	12.66
		1.5% CTAB	86.81	-0.60	3.76
	Vinegar	Untreated	88.24	-0.32	-0.60
		0.5% CTAB	83.83	-2.00	9.99
		1.0% CTAB	82.14	-1.95	11.04
		1.5% CTAB	83.41	-0.89	7.25
	Alum	Untreated	87.85	-0.68	0.48
		0.5% CTAB	84.98	-1.75	7.73
		1.0% CTAB	81.04	-0.93	4.86
		1.5% CTAB	87.59	-1.44	9.59
	Iron	Untreated	75.50	4.47	18.46
		0.5% CTAB	78.62	2.31	17.49
		1.0% CTAB	74.44	5.11	25.21
		1.5% CTAB	77.70	2.28	15.43

Table 4: Colour Coordinates of Dyed Silk Fabric

Fabric	Treatments		L*	a*	b*
	Undyed		**90.57**	**2.28**	**-5.92**
Silk	No Mordant	Untreated	82.86	-0.47	10.99
		0.5% CTAB	82.56	-0.51	4.56
		1.0% CTAB	83.22	-0.08	4.27
		1.5% CTAB	82.82	-0.55	7.77

	Vinegar	Untreated	84.13	0.13	8.41
		0.5% CTAB	78.98	-0.87	9.37
		1.0% CTAB	85.44	0.48	4.12
		1.5% CTAB	80.78	-0.76	12.26
	Alum	Untreated	83.67	0.40	7.96
		0.5% CTAB	81.31	-0.30	7.86
		1.0% CTAB	86.33	-0.04	4.19
		1.5% CTAB	80.63	-0.66	11.68
	Iron	Untreated	71.60	4.34	21.28
		0.5% CTAB	70.85	3.39	20.36
		1.0% CTAB	75.41	4.77	20.90
		1.5% CTAB	72.82	3.84	21.11

Similarly, 1% CTAB and iron treatments on cotton fabric gave the highest values of the colour difference (ΔE) and colour strength (K/S) as 0.52 and 40.58 respectively. Whilst, the colour difference (ΔE) and colour strength (K/S) values for the silk fabric treated with 1% CTAB and iron were 0.65 and 31.48 respectively.

The summary of ΔE for the dyed cotton and silk fabrics are shown in Figures 3 and 4 respectively. The summary of K/S values for cotton and silk fabrics are simplified in Figures 5 and 6.

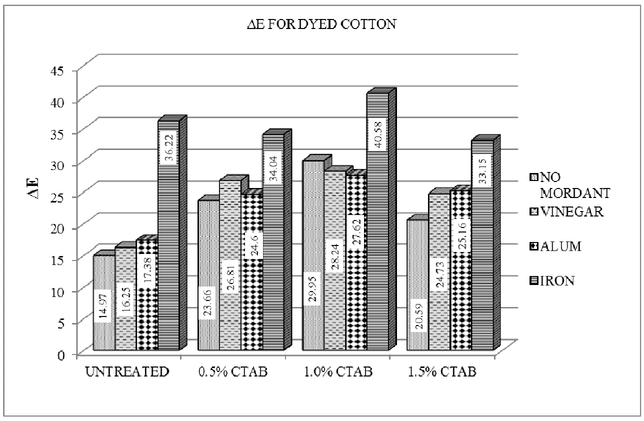

Figure 3: ΔE for Dyed Cotton Fabrics

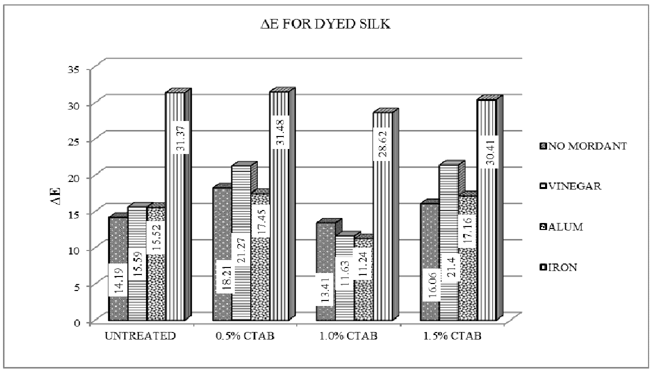

Figure 4: ΔE for Dyed Silk Fabrics

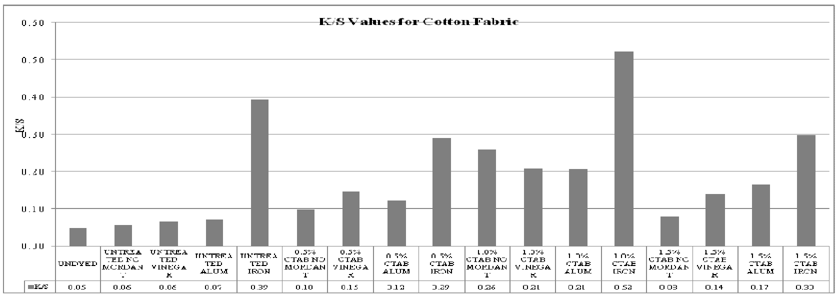

Figure 5: K/S for Dyed Cotton Fabrics

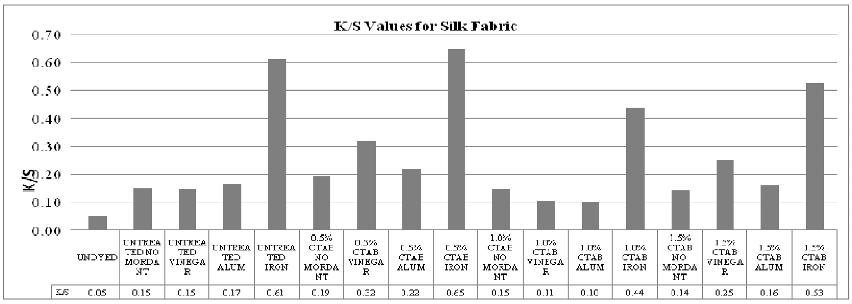

Figure 6: K/S for Dyed Silk Fabrics

Fastness Properties

The fastness properties of the dyed cotton and silk fabrics with natural dye extracted from *Sargassum* sp. seaweed are summarized in Tables 5 and 6. As for washing fastness properties, the colour change rates were excellent (4/5- 5) for both of them. Similarly, the rates of staining on cotton and silk composites for washing were also excellent (4/5- 5). The results for fastness properties to perspiration were found to be good to excellent rating (4 – 5). Rubbing/crocking evaluations in terms of dry and wet rubbing were graded as good to excellent (4 - 5). However, the fastness properties to light for both dyed fabrics were found to be poor, rated in the range of 3 – 5.

Table 5: Fastness Properties of Cotton Fabric

Treatments	Mordants	Washing			Perspiration			Rubbing/ Crocking		Light
		Change In Colour	Staining		Change In Colour	Staining		Dry	Wet	
			Cotton	Silk		Cotton	Silk			
Untreated	No Mordant	4/5	4/5	4/5	4/5	4	4	4/5	4	3
	Vinegar	4/5	5	5	4/5	4	4	4/5	4	3
	Alum	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4	3
	Iron	4	4/5	4/5	4	4/5	4/5	4	4	3
0.5% CTAB	No Mordant	4/5	4/5	4/5	4/5	4/5	4	4/5	4/5	4
	Vinegar	4/5	5	5	4/5	5	4	5	4/5	4
	Alum	4/5	5	5	4/5	5	4	5	4/5	4
	Iron	4	4/5	4/5	4	4/5	4	4	4	4
1.0% CTAB	No Mordant	4/5	5	5	4/5	5	4	4	4	4
	Vinegar	4/5	5	5	4/5	5	5	4/5	4/5	5
	Alum	4/5	5	5	4/5	5	5	4/5	4/5	5
	Iron	4/5	4/5	4/5	4/5	4/5	4	4/5	4/5	4
1.5%	No	4/5	5	5	4/5	5	4	4	4	4

CTAB	Mordant									
	Vinegar	4/5	5	5	4/5	5	5	4/5	4/5	5
	Alum	4/5	5	5	4/5	5	4	4/5	4/5	4
	Iron	4/5	4/5	4/5	4/5	4/5	4	4/5	4/5	4

Table 6: Fastness Properties of Silk Fabric

Treatments	**Mordants**	**Washing**			**Perspiration**			**Rubbing/ Crocking**		**Light**
		Change In Colour	**Staining**		**Change In Colour**	**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	5	5	4/5	4	4	4/5	4	3
	Alum	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4	3
	Iron	4/5	4/5	4/5	4	4/5	4/5	4	4	3
0.5% CTAB	No Mordant	4/5	5	5	4/5	4/5	4	4/5	4/5	3
	Vinegar	5	5	5	4/5	5	4	5	4/5	5
	Alum	4/5	5	5	4/5	5	4	5	4/5	5
	Iron	4/5	4/5	4/5	4	4/5	4/5	4	4	4
1.0% CTAB	No Mordant	4/5	5	5	4/5	4/5	4/5	4	4	4
	Vinegar	5	5	5	4/5	4/5	4/5	4/5	4/5	4
	Alum	4/5	5	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% CTAB	No Mordant	4/5	4/5	4/5	4/5	4/5	4/5	4	4	4
	Vinegar	4/5	4/5	4/5	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

Generally, the application of CTAB as surface modifier did not influence the fastness properties, but the results indicated that the application of mordants improved the fastness properties of the dyed fabrics by forming a chemical bond with fibre and natural dye.

CONCLUSIONS

Sargassum sp. seaweed can be exploited as a sustainable source of natural dye for textile coloration. It produces exciting and distinctive yellowish to brownish shades on cotton and silk fabrics with acceptable fastness properties even without mordant. The application of CTAB as surface modifier significantly enhances the dyeability of cotton and silk fabrics as the L*, ΔE and K/S values were obviously improve in comparison with untreated cotton and silk fabrics. This is due to the addition of the hydrophilicity and wettability properties of the fabrics with the application of CTAB (Marie *et.al.* (2015)). On the other hand, positive charges of amino group in CTAB could increase the positive charges on the surface of fabrics. Thus, enhancing the attraction between the fabrics and the dye (Marie *et.al.* (2015)) These properties could lead to more efficient dyeing.

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