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SUSTAINABLE WASTE UTILIZATION APPROACH FOR THE ECO-FRIENDLY COLOURATION OF ALPHITONIA EXCELSA LEAF-EXTRACT DYE ON NYLON SUBSTRATE

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ABSTRACT— *The utilization of synthetic dyestuff for the colouration of various textile fibres on commercial scale is quite efficacious due to their low cost, ease of production as well as application. However, the fact that synthetic dyes are carcinogenic in nature and are causing remarkable intimidation to the environment has led to the revitalization of the environmental friendly natural dyestuffs. Being eco-friendly, the natural dyes have a potential to gain importance commercially in the coming future. The present study deals with the application of a natural dye pertaining to the Rhamnaceae family. The natural colourant was extracted from the fallen, dried leaves of the Alphitonia excelsa tree and was applied onto the mordanted nylon substrate using exhaust dyeing technique. The mordant used for the study was a natural mordant, tannin, obtained from the used-up tea leaves and was applied by pre-, simultaneous- and post-mordanting techniques. The mordant plays an important role in attaining a preferred shade on a particular substrate. The colour strength of the dyed samples (both mordanted as well as unmordanted) was evaluated spectrophotometrically. The fastness properties of the dyed samples were found to be quite good. The washing fastness was rated as excellent for mordanted and dyed fabrics. The rubbing fastness, particularly dry rubbing fastness as well as the light fastness characteristics were found to be acceptable, from the dyer's point of view, for all the dyed fabrics. Utilization of waste fallen leaves of the tree for extracting the natural dye and waste used-up tea leaves for mordanting may be anticipated as an "environmental-friendly approach" and a feasible stride towards waste utilization.*

Keywords: Alphitonia excelsa leaves, nylon, exhaust dyeing, natural mordant, shades, waste utilization

1. INTRODUCTION

According to the world survey report by 'The Fibre Year' in 2016, the consumption of textiles and non-wovens all over the world is estimated to be about 101.4 million tons. The global dyes & pigments market size was valued at USD 30.42 billion in 2016, in terms of revenue. Increasing demand from various applications such as textiles, paints & coatings, construction, paints, and plastics are expected to drive the market growth [1]. The approximate consumption of dyes for textile colouration ranges from 20 to 25 lacks tons, which is being fulfilled by the utilization of synthetic dyes. However, the cognizance towards environment has modelled many limitations to the use of synthetic dyes. Synthetic dyes are not only harmful to the environment but also carcinogenic to humans and other living beings.

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The use of synthetic dyestuff, during their application in the dyeing and printing industries, has been criticized due to introduction of contamination into the environment 2 – 4]. An increasing realization that the intermediates and chemicals used in synthetic dyes are toxic and hazardous to human health as well as to the environment, has led to the revival of interest in the non-toxic eco-friendly colouring materials [5 – 8]. Recently, interest in the use of natural dyes has been growing rapidly due to the result of stringent environmental standards imposed by many countries in response to toxic and allergic reactions associated with synthetic dyes.

The application of natural dyes in textile wet processing is a step towards environmentally friendly process. Today, all over the world, people have come to accept the fact that the natural dyes are more traditional and hygienic. Natural dyes have been an integral part of human life since time immemorial. Egyptian mummies, documents of Mughal periods, etc. bear a testimony to the utilization of these dyes. In India, Rajasthan and Kutch still possess a rich tradition in the use of natural dyes for textile dyeing and printing. These natural dyes, obtained from natural resources, are non-pollutant, non-allergic, shade rich and warm. Vegetable dyes, in particular, are very good for skin and soothing to eyes and have been used for thousands of years by mankind [9]. With the advent of coal tar dyes (now synthetic dyes), the use of natural dyes declined tremendously because the existing natural dyes failed to fulfill the demand of the market. Moreover, synthetic dyes were cheaper, more readily synthesized, give better and more reproducible shades with better fastness properties to various agencies. However, many synthetic dyes, particularly dyes that are prepared from ∝-aryl amines have been found to be potentially carcinogenic. Certain chemicals used in the synthesis of dyestuffs are also regarded to be carcinogenic, mutagenic, as well as sensitizing or allergic [4, 10]. Thus, based on EC control of substance Hazardous to Health Act, 1989 a number of chemicals have been red-listed and have been banned in U S and Europe. German Government in an ordinance dated June 16, 1994 has also banned twenty amines from use in any garment or any other article that come in contact with the skin. Due to this, the revival of much less hazardous, eco-friendly natural dyes has been particularly realized by various textile manufacturers. The natural colourants are unsophisticated and harmonized with nature. They are obtained from renewable sources and their preparation involves a possibility of very little chemical reaction. Hence, they do not cause health hazards, but sometimes act as a health cure. Furthermore, the use of natural dyes offers no disposal problems [6]. However, the natural dyes have their own limitations, such as limited shade range owing to restricted number of suitable dyes available, colour yield and dyeing efficiency, cultivation proficiency, complications in shades reproducibility, complexity of dyeing process and ease of availability. Apart from these limitations, various technical shortcomings are also associated with the use of natural dyes [11 – 13], such as difficulty of blending dyes to produce compound shades, lack of standardization of dyeing and printing processes, difficulty in collection of the dyes, inadequate degree of fixation as well as fastness properties, and use of heavy metals and organic substances which may subsequently lead to water pollution. Dyers throughout the world are trying to overcome the limitations associated with the natural dyes to make their commercial acceptance possible once again.

The most common vegetable parts used for extracting dyes are seeds, flowers, leaves, berries, stems, barks and roots. Some parts may have more than one colour depending upon which part of the plant is used. The dyeing process based on vegetable resources include three major steps – first being the extraction of colouring matter from the plant part, second is creating a bond between the colouring matter and the fibre to be dyed and the last is the actual dyeing. The creation of a bond between the colouring matter and the fibre is called mordanting. A mordant is the chemical that when “cooked” with a fibre attaches itself to the fibre molecules. The dye molecule then attaches itself to the mordanted substrate. Vegetable dyes require mordants which are metallic salts of aluminium, iron, chromium, copper and

others, for ensuring the reasonable fastness of the colour to sunlight and also washing [14 – 19].

In the present study, one such natural vegetable dye has been extracted from the leaves of a medicinal tree *Alphitonia excelsa*. The dye has been applied on mordanted nylon substrate using exhaust dyeing method. The mordant used for the mordanting purpose was a natural tannin-rich mordant derived from tea leaves, which not only enhances the colour value but also produce rich and bright shades having different hues and tones. For making the process economic and environmental-friendly, the approach of waste utilization has been selected in which the dried and fallen leaves of the *Alphitonia excelsa* tree were used for extracting the colouring component as well as the used-up tea leaves were used for the mordanting purpose.

2. MATERIALS & EXPERIMENTAL PROCEDURES

2.1 Materials

2.1.1 Fabric

Nylon fabric (plain weave, 32 g/m^2) with 160 reeds/inch and 140 picks/inch was used for this study. Before dyeing, the fabric was first scoured to remove natural impurities so as to ensure uniform application of the color. The scouring bath was prepared with 5 g/l soda soda and 5 g/l non-ionic detergent (Lissapol N) using liquor ratio of 25:1. The treatment was given in the above bath at boil for 90 minutes. The fabric was then thoroughly washed and air-dried.

After scouring, the fabric was bleached to improve its whiteness. Although bleaching is not an essential operation for dyed fabrics, but the dyeing with natural colourants may produce pale shades and hence bleaching process is implemented to get better dyeing performance, particularly for light and medium shades. Bleaching was carried out in a bath containing

2-3 %	Hydrogen peroxide (30 volume)
5 cc	Sodium silicate
3 %	Soda ash

using liquor ratio of 25:1. The bleaching process was continued for 60 minutes at 80^o C, after which the fabric was thoroughly washed and air-dried

2.1.2 Natural dyestuff

The natural dye selected for the present work was extracted from the dried and fallen leaves of *Alphitonia excelsa* tree pertaining to *Rhamnaceae* family [Figure 1a]. The plant is commonly known as Red almond, Red ash, soap tree, etc. For medicinal applications, the crushed leaves are converted into a paste, mixed that with water and applied as a head bath to reduce headaches and treat sore eyes. The leaves contain saponin, and so when crushed can be lathered to produce a bush soap. In the present investigation, the fallen leaves of the Red almond tree were collected and further dried in shade for 4 to 5 days.

(a) (b)

Figure 1. (a) Leaves of Red Almond tree (b) Waste used-up tea leaves

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To prepare the dye solution, 20 gm of *Alphitonia excelsa* leaves were soaked in 500 cc boiling water for 10 - 15 minutes. 25 cc of glacial acetic acid was added to the liquor and the whole mixture was kept overnight in dark. Then the volume was adjusted to 1 litre by adding remaining water. The liquor was filtered and the solution was directly used for the colouration of nylon fabrics.

2.2.3 Mordant

The tea leaves, after preparation of tea, were collected from tea stall [Figure 1b] and the mordant was extracted from the leaves after preliminary washing the leaves with cold water. The wet used leaves (20 g) were weighed and soaked in 1 liter cold water at room temperature for about 15 min. and the contents were boiled for 60 minutes to extract the mordant and the colouring component present in the tea leaves. After boiling, the whole mixture was stored overnight in dark. On the next day, the contents were filtered through Whatmann Filter paper No. 1; the filtrate adjusted to 1 liter volume with cold water and used directly for mordanting purpose.

The tannin predominantly present in the leaves of green and black tea is Theaflavin (Figure 2); however, since the present study deals with the utilization of waste tea leaves as mordant to produce different shades on the nylon fabric dyed with the natural vegetable dye, the amount of Theaflavin present in the used-up tea leaves was not assessed and only practical application aspects of visualizing the effect of mordanting with tea leaves on dyeing of nylon with natural vegetable dye have been undertaken in the present investigation. The research based on the characterization and analytical determination of the tannin (theaflavin) content in the utilized waste tea leaves is under consideration and is ongoing.

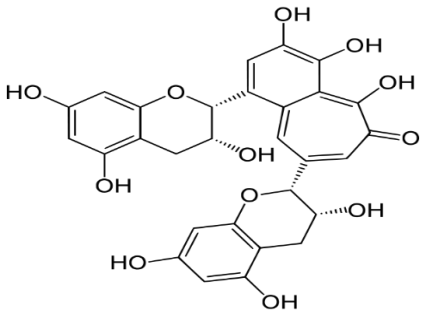

Figure 2. Chemical structure of tannin (theaflavin) present in green and black tea

2.2 Experimental methods

Following procedures were adopted for the application of *Alphitonia excelsa* dye on nylon substrates –

1. Pre-mordanting technique – mordanting followed by dyeing
2. Meta-mordanting technique – simultaneous dyeing and mordanting
3. Post-mordanting technique – dyeing followed by mordanting

2.2.1 Mordanting

The mordanting bath was prepared using 2 % mordant (1:40, on the weight of the fabric) using a liquor ratio of 40:1. The treatment was given at 60 - 70^{o} C for 45 minutes. The procedure was repeated using 5 % and 10 % concentrations of the mordant (on the weight of the fabric) to visualize the effect of mordant on the dyeing performance of the natural dye.

2.2.2 Dyeing

The exhaust dyeing of nylon fabric was performed on High temperature, high pressure dyeing machine. Glauber's salt has been used as an exhausting agent while acetic acid was used for maintaining the acidic conditions during application of natural dye on nylon substrate. The liquor ratio of 30:1 was utilized for the preparation of the dyebath. The dyebath was prepared as follows –

2 % *Alphitonia excelsa* dye solution (*owf*)
10 % Glauber's salt (1:10)
5 % Acetic acid (1:25)
MLR: 1:30
Temperature: 130^{o} C
Dyeing Time: 60 minutes

The nylon fabric (mordant/unmordanted) was the impregnated in the respective red almond leave's dye solutions at room temperature. The dyeing cylinders were closed and entered in the HTHP dyeing machine; the temperature of the dyebath was set at 130° C. The temperature of the dyebath was gradually raised to 130° C within 15 minutes and dyeing continued at this temperature for another 45 minutes.

2.2.3 After-treatments

After dyeing, all the dyed samples were thoroughly rinsed, soaped at 60° C for 15 min using 2 g/l detergent solution, washed thoroughly and dried at ambient temperature.

2.3 Testing and Analysis

2.3.1 Evaluation of dyed samples

Dyeing performance of various dyed samples was assessed by measuring the relative colour strength (*K/S* value) spectrophotometrically. These values are computer calculated from reflectance data according to Kubelka-Munk equation [20].

2.3.2 Assessment of fastness properties

The washing, light and rubbing fastness properties of various dyed samples were evaluated according to standard methods [21].

- *Fastness to washing:* Wash fastness of different dyed samples was assessed on Launder-o-meter using ISO standard Test No. 3. The change in shade was visualized using Grey scale and graded from 1 to 5; where 1 indicates poor and 5 excellent fastness to washing.
- *Fastness to Light:* Colour fastness to light was evaluated by exposing the dyed samples to sunlight according to AATCC test method 16B-1977. They were graded from 1 to 8; where 1 indicates poor and 8 excellent fastness to light.
- *Fastness to Rubbing:* The rub fastness of dyed samples was tested on Crockmeter. The specimen to be tested was rubbed against perfectly scoured and bleached cloth of dimension not less than 22 cm x 5 cm. The white rubbing cloth was placed over the end of the finger of the testing device. In the dry rubbing test, the cloth to be tested was rubbed 10 times in 10 sec in dry state; while in the wet rubbing test, the procedure was same, except that the rubbing cloth was wetted out and squeezed to 100 % expression. The grading was given by taking into consideration the intensity of stain obtained on white fabric as well as lowering in the depth of the rubbed sample. The staining on the rubbing cloth was assessed with the Grey Scale and grades awarded from 1 to 5, where 1 stands for poor and 5 for excellent fastness to rubbing.

2.3.3 Determination of Shades/Hues of the Dyed Samples
The shades / hues of various dyed samples were judged by visual assessments from various shade cards (Pantone color code, Asian paint, Dulux, Berger, and Nerolac) available in the Indian market. The final shade was given from the judgment of 5 different viewers.

3. RESULTS & DISCUSSION

In the present study, a natural dye, extracted from the dried leaves of Red almond tree (*Alphitonia excelsa*), has been selected for the application of vegetable natural dye on nylon. During trials, the dye extracted from the leaves of Red almond has given good performance on nylon substrate. This dye is then applied on nylon fabrics using exhaust dyeing method, in the presence and absence of tannin (present in tea leaves) mordant, using different mordanting techniques. All the dyeings are compared with those obtained on unmordanted (control) samples dyed under similar conditions. The dyed samples are evaluated for colour strength (*K/S* values), shades and hues obtained with different mordants and various fastness properties.

3.1 Dyeing performance of Alphitonia excelsa natural dye using tannin-based mordant

Table 1 represent the comparison of dyeing performance of the dye extracted from the waste leaves of Red almond tree on nylon substrate using exhaust dyeing and various mordanting techniques. The nylon fabric gave appreciable dyeing performances with *Alphitonia excelsa* dye. Excellent results have been obtained by all the three mordanting techniques. Moreover, the mordant used in the present study have shown excellent performance in terms of colour strength values. The effectiveness of the mordant used for the application of these dyes on nylon fabric increases with the increase in the concentration of the mordant during its application. From the results obtained, it can be depicted that the depth of the shade can be enhanced with the increase in the tannin concentration during mordanting. In case of single stage dyeing and mordanting, there has been an enhancement of about 173 % and 314 % when 5 % and 10 % of tea (tannin) mordant was used respectively in the bath along with *Alphitonia excelsa* natural dye extracted from its leaves.

Furthermore, on comparing the dyeing performance on the basis of the mordanting technique employed, it can be visualized from the Table 1 that the best performance has been achieved when simultaneous mordanting and dyeing operations were performed in the same bath. This was followed by the performance of *Alphitonia excelsa* dye on the samples pre-mordanted with tea leaves. The overall dyeing performance results can be summarized as –

Simultaneous mordanting and dyeing > Mordanting followed by dyeing > Dyeing followed by mordanting

It could also be noted from the colour strength values of the dyed samples that when the mordanting was done with lower concentrations of the tea mordant, a slight lower depth of colour value is obtained as compared to the control (unmordanted and dyed) sample, particularly in pre-mordanting and post-mordanting techniques. This may be due to insufficient amount of tannin mordant available in the waste (used-up) tea leaves at lower concentration of the mordant. The results might have been better if fresh tea leaves have been utilized for mordanting purpose. However, since the main objective of the present investigation was based on waste consumption approach, hence use of fresh tea leaves for mordanting was ignored.

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Table 1. Colour strength (in terms of *K/S* values) for nylon fabric dyed with 2 % *Alphitonia excelsa* dye

Application technique	Concentration of tannin-based tea mordant	*K/S* value
Control	–	6.43
Pre-mordanting	2%	5.28 (− 17.88)
	5%	10.67 (+ 65.94)
	10%	17.89 (+ 178.23)
Simultaneous dyeing and mordanting	2%	8.68 (+ 34.99)
	5%	17.56 (+ 173.09)
	10%	26.66 (+ 314.62)
Post-mordanting	2%	3.79 (− 41.06)
	5%	8.84 (+ 37.48)
	10%	14.86 (+ 131.10)

Note: Figures in parenthesis indicate percent loss/gain over control sample

3.2 Effect of tannin mordant on shade/hue of dyed nylon samples

Mordants are substances which improve the affinity of a substrate for a particular dye. In case of natural dyes, various metallic mordants are usually employed to improve the fastness property of dyed materials as well as to obtain a variety of shades with different tones and hues. From Table 2, it can be seen that when nylon was dyed with *Alphitonia excelsa* dye by exhaust dyeing method using tannin-rich tea mordant, various shades ranging from pale yellow through light brown to chocolate brown are obtained with different concentrations of the tea used for mordanting by different techniques. The shades are bright and even throughout the sample.

3.3 Effect of mordants on fastness properties of natural colour dyed nylon samples

Washing, light and rubbing (dry and wet) fastness properties of various samples, dyed with *Alphitonia excelsa* dye on unmordanted as well as mordanted nylon substrate, were examined and compared with each other. The fastness grades for various dyed samples are represented in Table 3. From the table, it can be clearly seen that the fastness properties of various mordanted and dyes samples are better than those of unmordanted and dyed sample. The fastness grades were found to be excellent for simultaneous application of the mordants during exhaust dyeing.

Table 2. Shades obtained for nylon fabric dyed with 2 % *Alphitonia excelsa* dye

Application technique	Concentration of tannin-based tea mordant	Shade obtained	Shade
Control	–	Buff	
Pre-mordanting	2%	Naples yellow	
	5%	Butterscotch	
	10%	Brick red	

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Table 2 Contd.

Simultaneous dyeing and mordanting	2%	Ecru	
	5%	Bistre	
	10%	Chocolate brown	
Post-mordanting	2%	Straw	
	5%	Sunglow	
	10%	Bright medallion	

Table 3. Fastness grades for nylon fabric dyed with 2 % *Alphitonia excelsa* dye

Application technique	**Concentration of tannin-based tea mordant**	**Fastness properties of dyed nylon samples**			
		Wash	**Light**	**Rub**	
				Dry	**Wet**
Control	–	4	6-7	4-5	4
Pre-mordanting	2%	4-5	6-7	5	4-5
	5%	4-5	6	4-5	4-5
	10%	4-5	6	4-5	4-5
Simultaneous dyeing and mordanting	2%	5	6-7	4-5	4
	5%	5	7	5	4-5
	10%	5	7	5	5
Post-mordanting	2%	4-5	7	4-5	5
	5%	4	6-7	4-5	4
	10%	4	6-7	4	4-5

4. CONCLUSIONS

Various ecological aspects such as eco-friendliness, eco-conservation and eco-protection are of great prominence in today's scenario. Everyone is highly apprehensive with not only protection of the environment but also of human health. Commercial synthetic dyes may be toxic/carcinogenic to human being as well as aquatic life/organisms and may also disturb the ecological balance. Thus, in the present study an attempt has been made to utilize a natural dye, derived from vegetable resource, instead of hazardous synthetic dyes. The natural dye, extracted from the leaves of *Alphitonia excelsa* tree, was selected for the work to obtain different shades with different hues and tones on nylon substrate. Mordants play an important role in obtaining a desired shade on a particular fabric. Using this concept, a

variety of shades was achieved with this dye with the help of tea (tannin) mordant. The shades having yellows and browns tones and hues have been obtained with different concentrations of the mordant. Various fastness properties of the dyed samples were found to be adequate and comparable with the control (unmordanted and dyed) sample. Utilization of waste products for mordanting and dyeing purpose may be regarded as an "eco-friendly and green technological approach" and should be adopted commercially to meet with the increasing demand of coloration of natural dyes on various textiles.

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