Title	Study on the Extraction and Utilization of Natural Dye from Tamarind Seed Testa
Author	Dr. Aye Aye Mar
Issue Date	

Study on the Extraction and Utilization of Natural Dye from Tamarind Seed Testa

Aye Aye Mar[1], Ar Kar Chan Myae Thu[2]

Abstract

The present research work is emphasized on the preparation of natural dye (concentrated solution) from tamarind seed testa. Tamarind seed contains nearly 30 percent of testa and 70 percent of kernel. The testa was removed from the seeds by roasting and subsequent pounding. The testa contains a dyestuff which was used for dyeing of cotton fabrics. The physico-chemical properties such as pH, acidity, moisture content, ash content and total solids content of tamarind seed testa were determined by Association of Official Analytical Chemists (AOAC) method. The chemical compounds present in tamarind seed testa were investigated by phytochemical tests. An attempt was made on the dyeing of cotton fabrics using of natural dye (concentrated solution) from tamarind seed testa with different mordants such as zinc sulphate, potassium dichromate, ferrous sulphate and three different dyeing methods such as pre-mordanting, post-mordanting and simultaneous mordanting and dyeing method. Moreover, fastness for rubbing and washing of dyed cotton fabrics was determined. Staining and colour change of dyed cotton fabrics were assessed by using standard Grey scale. The dye extraction was carried out at testa to water ratio of 1:40 and the resulting natural dye (concentrated solution) was employed for dyeing of cotton fabrics. Dyeing at 90ºC for 45 min followed by post-mordanting with potassium dichromate enhanced the highest amount of dye adsorbed on cotton fabrics and good fastness properties.

Key words: natural dye, mordant, phytochemical tests, cotton fabric, dyeing, Grey scale, fastness

Introduction

Dyes and textile are an important part of our everyday life. In the production of clothes, dyeing is a very important process. Main sources of natural dyes are vegetable, animals and minerals. They can be obtained from any part of plants viz. leaves, fruits, seeds, flowers, barks and roots. Natural dyes are derived from natural sources without chemical processing. They can be biodegradable and do not pose a problem of pollution for the waste. After the dyeing process, they are easily absorbed by the soil. In textile dyeing industry, most of the dyes employed are the synthetic azo dyes of which the azo bond cleavage brings about aromatic amines that may be harmful to human health and environment.

Nowadays, a great awareness on the impacts of toxic chemicals on the environment and human health has turned down the use of synthetic chemicals and in the circumstance; a higher demand is put towards the greener alternative substance. For this reason, natural dyes are among the promising options for developing a greener textile dyeing process. However, the major drawback of the natural dyed-textiles is their poor colour fastness to light and washing. Incorporation of the mordants in natural dyeing can help to improve such inferior properties and at the

[1] Associate Professor, Dr., Department of Industrial Chemistry, University of Mandalay

[2] MSc Student, Department of Industrial Chemistry, University of Yangon

same time, provides a wide variety of shades of natural dyes obtained on the same textile substrates. (Marjo Moeye, 1993)

Tamarind (*Tamarindus indica* L.) is classified as an important economical plant being widely grown in Myanmar. Tamarind is classified into two major kinds according to the providing taste of the tamarind fruits, namely sour tamarind and sweet tamarind. Tamarind seed testa contains 40 percent of water-extractable matter of which 80 percent is the dyestuff. The latter comprises about 35 percent of pholobatannin, 55 percent of depside of tannins and 10 percent of uncharacteristized material. Tannins could improve the colour yield and colour fastness properties on the mordant-dyed fabrics.

Extraction refers to separating the desired colour component by physical or chemical means with the aid of a solvent. Natural dyes of different origin can be extracted using aqueous method i.e. by using water for the extraction with or without addition of salt or acid or alkali or alcohol. In the extraction bath-alcoholic or organic solvent extraction by using relevant extracting equipment and finally to filtrate, evaporate and to dry using rotary vacuum pump. The aim of this research work was to produce the natural dye (liquid) from tamarind seed testa and to utilize it in the dyeing of cotton fabrics. And then, to evaluate the colour fastness properties of dyed cotton fabrics using washing and rubbing fastness tests.

Materials and Methods

Materials

In this research work, tamarind seeds were collected from North Ywar Ma Village, Seikphyu Township, Magway Region.

95% ethanol, zinc sulphate, potassium dichromate, ferrous sulphate, ferric chloride, potassium ferrocyanide, 95% sulphuric acid, sodium chloride, sodium hydroxide and benedict's solution (analar grade, British Drug House Co. Ltd., England) were used. Magnesium turnings, alum, sodium chloride and vinegar (Myanmar vinegar) were also used.

Methods

Preliminary Preparation of Tamarind Seed Testa

The tamarind seeds were washed with water to remove the adhering dirt and impurities. They were dried at room temperature and weighed. The tamarind seeds were roasted at 100ºC for 10 min on electric stove. Then they were pounded, ground and sieved through the screen of (-42) mesh size. The powder tamarind seed testa was stored at room temperature for further work.

Physico-chemical Characterization of Tamarind Seed Testa

The physico-chemical characteristics such as moisture content, ash content, total solids content, acidity and pH of tamarind seed testa were determined by Association of Official Analytical Chemists (AOAC) method.

Phytochemical Characterization of Tamarind Seed Testa

The main compounds which are commonly found in tamarind seed testa are polyphenols, phenolic compound, sugar, tannins and saponins. Phytochemical examination of tamarind seed testa was performed according to the methods and procedures expressed in the Phytochemical Bulletin of Botanical Society of America

Preparation of Natural Dye (Concentrated Solution) from Tamarind Seed Testa

In the first extraction of tamarind seed testa dye, 10 g of tamarind seed testa and 100 ml of clean water were placed in a conical flask and heated at 60ºC for 20 min using an agitation speed of 500 rpm. The tamarind seed testa extract was filtered and collected in a glass bottle. The second, third and fourth extractions of tamarind seed testa dye were conducted using the same tamarind seed testa residue with 100ml of water for each extraction using the above procedure. Then, the natural dye (concentrated solution) was obtained from the first, second, third and fourth tamarind seed testa extract were mixed and evaporated at 90ºC for 40 min in a water bath. Then the natural dye (concentrated solution) was weighed and stored in a freezer at 4ºC for dyeing of cotton fabrics.

Physico-chemical Characterization of Natural Dye (Concentrated Solution) from Tamarind Seed Testa

The physico-chemical characteristics such as pH, acidity and total soluble solids content of the natural dye (concentrated solution) from tamarind seed testa were determined by Association of Official Analytical Chemists (AOAC) method. Then the absorbance value of the natural dye (concentrated solution) from tamarind seed testa was determined at the wavelength of 425nm by UV-Vis spectrophotometer (TRILP, TRSP-722).

Absorbance Value of Mordanted Natural Dye (Concentrated Solution) from Tamarind Seed Testa

(100) ml of natural dye (concentrated solution) from tamarind seed testa was mixed with (1) g of zinc sulphate and heated at 60ºC for (10) min. Then it was cooled and the absorbance value was measured by UV-Vis spectrophotometer. The same procedure was conducted using the different amounts such as (3) g and (5) g of zinc sulphate mordant and different mordants such as potassium dichromate and ferrous sulphate ranging from (1) g to (5) g. The respective absorbance values were recorded.

Mercerizing of Cotton Fabrics

Firstly, (2) g of caustic soda was dissolved in 100 ml of distilled water before adding to the cotton fabrics. Cotton fabrics were simmered in caustic soda solution at 90ºC for 10 min. The simmered cotton fabrics were washed with water to remove excess caustic soda. Any remaining alkali was neutralized by treating the cotton fabrics with vinegar solution (5 ml vinegar to 100 ml of water) for 30 minutes. To prepare cleaned cotton fabrics, the cotton fabrics were rinsed with water and then dried at room temperature. After that, the cotton fabrics were tested with a few drops of iodine. The cotton fabrics were cleaned if purple black colour did not appear.

Mordanting of Cotton Fabrics

In this method, mordant solution was prepared by heating 3 g of zinc sulphate with 100 ml of water. The zinc sulphate mordant solution and 1 g of cotton fabric (8"x 8") was heated at 90˚C for 10 minutes. During mordanting, the cotton fabric was frequently stirred to obtain good penetration of mordant into the cotton fabric. After that, mordant solution was removed and the mordanted cotton fabric was rinsed with water and then dried at room temperature. The same procedure was carried out with different mordants such as 3 g of potassium dichromate and 3 g of ferrous sulphate to obtain the mordanted cotton fabrics.

Dyeing of Cotton Fabrics

For direct dyeing of cleaned cotton fabrics, 1 g of cleaned cotton fabric (8"x 8") and 100 ml of prepared natural dye (concentrated solution) from tamarind seed testa were added to a stainless steel pot and simmered at 90˚C for 30 min. After dyeing, the cotton fabric was rinsed with water to remove unfixed dye and subjected to air drying. The amount of dye on cotton fabric was determined and colour development on dyed cotton fabric was observed. In this research, pre-mordanting method, post-mordanting method and simultaneous mordanting and dyeing method were used. In pre-mordanting method, mordanting was done before dyeing. In post-mordanting method, mordanting was done after dyeing. In simultaneous mordanting and dyeing method, mordant, dye and cotton fabric were added in a pot and treated together.

Effect of Dyeing Time on the Amount of Dye on Cotton Fabrics

The effect of dyeing time on the amount of dye on cotton fabrics was determined by using the same dyeing processes such as pre-mordanting, post-mordanting and simultaneous mordanting and dyeing methods at 90˚C for various dyeing time ranging from (30) minutes to (60) minutes. The amounts of dye on cotton fabrics were determined and colours development on dyed cotton fabrics were observed.

Testing the Colour Fastness of Dyed Cotton Fabrics

Fastness to Washing

In this experiment, Launder Meter washing machine (Model No. L-4), containing 4-rack test bottle was used and the operation was carried out with Test No.3 of ISO 105 to assess the colour fastness to washing. Dyed cotton fabric was cut into (10 cm × 10 cm) and was sandwiched between two adjacent white bleached cotton fabrics having the same dimension with the test specimen. Four sides of the composite specimen were sewn at each corner by using a white thread.

The required soap solution was prepared with (5) g of soap per liter. After that, the soap solution was added to the jar and preheated at 60˚C. Then the composite sample was placed in the jar and tested at 60˚C for 30 minutes. After washing, the specimen was rinsed with water for (5) times and dried at room temperature. The change in colour of the specimen was assessed by comparing with the original dyed cotton fabric and the rating is defined by using Grey scale for colour change and for staining.

Fastness to Rubbing

The rubbing fastness of dyed cotton fabric was determined by Rubbing Tester machine (JIS STDL-0241). The test specimen of (25 cm x 25 cm) and a piece of white bleached cotton fabric about 5 cm were cut parallel to warp direction for rubbing fastness test. In rubbing fastness test, the test specimen was mounted on the white bleached cotton fabric and placed on the rubbing tester. The rubbing distance was 100 mm. The test specimen was rubbed 100 times with (500) g load at the speed of 30 circles per minute. The white bleached cotton fabric and the test specimen were rubbed together on the tester by the aid of motor. Six different materials were tested simultaneously on the machine. In this study, both of dry and wet rubbing fastness tests were carried out. In the wet rubbing fastness test, the rubbed cotton fabric was wetted with water and the amount of water was two times of the fabric weight. The fabric was finally dried at room temperature.

Results and Discussion

The physico-chemical characteristics such as pH, acidity, moisture content, ash content, total solids content of tamarind seed testa were determined. The results in Table (1) indicate that pH 6.79, acidity 0.167 %v/w, moisture content 12.0 %w/w, ash content 1.30 % w/w and total solids content 88.0 % w/w. According to the results in Table (2), it was found that the colour compounds such as polyphenols, phenolics, sugar, tannins and saponin were present in tamarind seed testa. From the results in Table (3), it was observed that the physico-chemical characteristics of natural dye (concentrated solution) from tamarind seed testa were pH 6.75, acidity 0.168 %v/w, total soluble solids 2ºBrix and absorbance value 1.780.

The absorbance values of natural dye (concentrated solution) with different mordants such as zinc sulphate, potassium dichromate, ferrous sulphate and the different amounts (1, 3, 5) g were measured by UV-VIS spectrophotometer. From the results in Table (4), it was noted that mordanted natural dye solutions containing (3) g of different mordants enhancing the highest absorbance value than the other amounts.

The effect of dyeing time on the amount of dye on cotton fabrics was determined and the results are shown in Tables (5) to (8). According to the results in Tables (5) to (8), dyeing with post-mordanting method and potassium dichromate mordant gave 391 mg/g of dye on cotton fabrics at 90ºC for 45minutes. Therefore, (45) minutes is a suitable dyeing time which enhanced comparatively highest amount of dye on cotton fabrics.

Washing and rubbing fastness properties of dyed cotton fabrics were tested for each group of mordants and three dyeing processes are shown in Table (9). In washing fastness, the change of shade ratings showed fair in fastness whereas staining on cotton and polyester had good fastness grade. Dyeing of cotton fabrics using post-mordanting method with potassium dichromate mordant was suitable because it has a change in shade 3 to 4 (fair to good).

In rubbing fastness, good to excellent (4 to 5) dry rubbing fastness properties and good wet rubbing fastness properties of dyed cotton fabrics were observed by using post-mordanting method with potassium dichromate mordant. Rubbing fastness results of dyed cotton fabrics indicated that all dyed cotton fabrics samples were found to be good to excellent in dry state and fair to good in wet state.

Table (1) Physico-chemical Properties of Tamarind Seed Testa Powder

Sr. No.	Properties	Experimental Value
1	pH	6.79
2	Acidity, %v/w	0.167
3	Moisture content, %w/w	12.00
4	Ash content, %w/w	1.30
5	Total solids content, %w/w	88.00

Table (2) Phytochemical Investigation for Tamarind Seed Testa Powder

Sr. No.	Tests	Solvent	Reagents	Observation (colour)	Inference
1	Polyphenols	EtOH	1%$FeCl_3$ + 1%$K_3\{Fe(CN)_6\}$	Greenish blue	+
2	Flavonoids	EtOH	H_2SO_4(conc:) + Mg	-	-
3	Glycosides	H_2O	10% $FeCl_3$	-	-
4	Phenolics	H_2O	10% $FeCl_3$	Blue-black	+
5	Sugar	H_2O	Benedict's Sol^n	Red ppt	+
6	Tannins	H_2O	2% NaCl+1% $FeCl_3$	Deep blue ppt	+
7	Saponin	H_2O	1% $NaHCO_3$	Froth	+

Note; + = Present, - = Absent

Table (3) Physico-chemical Properties of Natural Dye (Concentrated Solution) from Tamarind Seed Testa

Weight of tamarind seed testa = 10g
Weight of water = 100 ml × 4 times = 400 ml
Yield of natural dye = 100 ml
Extraction condition = 60ºC, 500 rpm, 20 min × 4 times

Sr. No.	Properties	Experimental Value
1	pH	6.75
2	Acidity, %v/w	0.168
3	Total soluble solids, ºBrix	2.00
4	Absorbance value at 425nm	1.780

Table (4) Effect of Mordant Concentration on the Absorbance Value of Natural Dye (Concentrated Solution) from Tamarind Seed Testa

Sr. No.	Tamarind Seed Testa Dye (ml)	Weight of Mordant (g)	Absorbance Values of Natural Dye with Different Mordants at 425nm		
			Zinc sulphate	Potassium dichromate	Ferrous sulphate
1	100	1	1.774	1.791	1.693
2	100	3*	1.803	1.822	1.829
3	100	5	1.804	1.821	1.805

*Suitable amount of mordant

Table (5) Effect of Dyeing Time on the Colour of Dyed Cotton Fabric using Direct Dyeing Method without Mordant

Weight of mercerized cotton fabrics = 1g

Sr. No.	Tamarind Seed Testa Dye (ml)	Dyeing		Dye on Cotton Fabrics		Colour Developed on Dyed Cotton Fabrics
		Temp. (ºC)	Time (min)	(mg/g)	(%w/w)	
1	100	90	30	260	26.0	Pale brown
2	100	90	45*	304	30.4	Pale brown
3	100	90	60	304	30.4	Pale brown

*Suitable dyeing time

Table (6) Effect of Dyeing Time on the Amount of Dye on Cotton Fabrics using Pre-mordanting Method

Amount of mordant = 3 g

Weight of mercerized cotton fabrics = 1 g

Sr. No.	Tamarind Seed Testa Dye (ml)	Dyeing		Dye on Cotton Fabrics (mg/g)		
		Temp. (ºC)	Time (min)	Zinc sulphate	Potassium dichromate	Ferrous sulphate
1	100	90	30	239	260	260
2	100	90	45*	304	347	304
3	100	90	60	304	347	304

*Suitable dyeing time

Table (7) Effect of Dyeing Time on the Amount of Dye on Cotton Fabrics using Post-mordanting Method

Amount of mordant = 3 g

Weight of mercerized cotton fabrics = 1 g

Sr. No.	Tamarind Seed Testa Dye (ml)	Dyeing		Dye on Cotton Fabrics (mg/g)		
		Temp. (ºC)	Time (min)	Zinc sulphate	Potassium dichromate	Ferrous sulphate
1	100	90	30	304	304	304
2	100	90	45*	347	391	347
3	100	90	60	347	391	347

*Suitable dyeing time

Table (8) Effect of Dyeing Time on the Amount of Dye on Cotton Fabrics using Simultaneous Mordanting and Dyeing Method

Amount of mordant = 3g
Weight of mercerized cotton fabrics = 1g

Sr. No.	Tamarind Seed Testa Dye (ml)	Dyeing		Dye on Cotton Fabrics (mg/g)		
		Temp. (ºC)	Time (min)	Zinc sulphate	Potassium dichromate	Ferrous sulphate
1	100	90	30	217	260	260
2	100	90	45*	304	347	304
3	100	90	60	304	347	304

*Suitable dyeing time

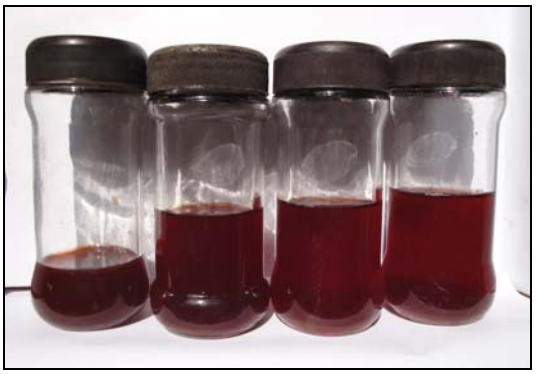
Table (9) Colour Fastness Properties of Dyed Cotton Fabrics using Various Methods and Mordants

Methods	Mordants	Rubbing Fastness		Washing Fastness	
		Dry Mark Code (1-5)	Wet Mark Code (1-5)	Change of Shade	Staining on Cotton Fabric
Pre-mordanting method	$ZnSO_4.7H_2O$	4-5	3-4	2	4
	$K_2Cr_2O_7$*	4-5	3-4	2-3	4
	$FeSO_4.7H_2O$	4-5	4	2	4
Post-mordanting method**	$ZnSO_4.7H_2O$	4-5	4	2	4
	$K_2Cr_2O_7$*	4-5	4	3-4	4
	$FeSO_4.7H_2O$	4	4	2	4
Simultaneous mordanting and dyeing method	$ZnSO_4.7H_2O$	4-5	3-4	2	4
	$K_2Cr_2O_7$*	4-5	3-4	3	4
	$FeSO_4.7H_2O$	4	3	2	4

* Suitable mordant
** Suitable dyeing method
Note: 5 = excellent, 4 = good, 3 = fair, 2 = poor, 1 = very poor
The experiments were conducted at No. (1), Development Centre for Textile Technology, Textile Industry, Mayangone Township, Yangon Region.

Figure (1) Tamarind Seeds

Figure (2) Tamarind Seeds Testa



I II III IV

I = Tamarind seed testa dye extract from 1st extraction
II = Tamarind seed testa dye extract from 2nd extraction
III = Tamarind seed testa dye extract from 3rd extraction
IV = Tamarind seed testa dye extract from 4th extraction

Figure (3) Natural Dye Extract from Tamarind Seed Testa

Figure (4) Natural Dye (Concentrated Solution) from Tamarind Seed Testa

Dyeing Time (min)	Direct Dyeing Method	Pre-mordanting Method	Post-mordanting Method	Simultaneous Mordanting and Dyeing Method
30				
45				
60				

Figure (5) Colour Development on Dyed Cotton Fabrics using Zinc Sulphate Mordant

Dyeing Time (min)	Direct Dyeing Method	Pre-mordanting Method	Post-mordanting Method	Simultaneous Mordanting and Dyeing Method
30				
45				
60				

Figure (6) Colour Development on Dyed Cotton Fabrics using Potassium Dichromate Mordant

Dyeing Time (min)	Direct Dyeing Method	Pre-mordanting Method	Post-mordanting Method	Simultaneous Mordanting and Dyeing Method
30				
45				
60				

Figure (7) Colour Development on Dyed Cotton Fabrics using Ferrous Sulphate Mordant

Conclusions

By simple boiling of tamarind seed testa in water, the water soluble dye could be isolated and later used to dye the cotton fabrics. Natural dye extracted from tamarind seeds testa is a type of adjective dye or mordant dye because it has no direct affinity for unmordanted material and it has to be applied by the use of a mordant. Mordants or metal salts used as a fixing agent for dyeing affected the shade and colour fastness properties of the dyes. On comparing with the unmordanted dyed cotton fabrics, the cotton fabrics dyed with post-mordanting for natural dye (concentrated solution) from tamarind seed testa exhibited attractive colours and acceptable fastness on cotton fabric especially with potassium dichromate.

As the mordant is a metal salt compound, it can attract the dye molecules and form a complex structure with the dyes. As a result, the dyes are generally more strongly fixed on the cotton fabrics. Therefore, the dyed cotton fabrics which have gone through the mordanting process normally have better fixation on the cotton fabrics. However, as the molecular structure of the tamarind seed testa dye is rather unstable, the washing process may cause a shade variation more or less depending on the types of mordant employed in dyeing process.

The results generated in this research show a promising potential of the tamarind seed testa dye to be applied practically in the textile industry where the uniqueness of the products is exploited. For the preparation of natural dye (concentrated solution) from tamarind seed testa, the suitable extraction conditions are tamarind seed testa powder to water ratio 1:40, extraction temperature 60ºC for 4 times of 20 min and evaporation temperature 90ºC for 40 min.

Acknowledgements

The author thanks Dr Thida Win, Rector, University of Mandalay and Dr Nu Nu Yi, Pro-rector, University of Mandalay for their kind permission to submit this article. The author would like to acknowledge Professor Dr Khin Thet Ni, Head of the Department of Industrial Chemistry, University of Yangon, for giving permission to perform this research and for her valuable suggestions and advice throughout this research work.

References

Dange, S., (1996), A Study of Some Natural Dyes for Their Application.

De Lumen, (1990), Legumes and a Cereal with high methionine/cyctine contents, Journal of Agricultural and Food Chemistry, 34: 361- 364.

Harborne, J.B., (1973), Phytochemical Methods, Chapman and Hall Ltd., New York.

Lewis, (2005), Legumes of the World. Royal Botnaic Gardens, Kew.

Lyle, (1977), Performance of Textiles, United States of America.

Marjo Moeye, (1993), Natural Dyeing in Thailand, White Lotus Co., Ltd., Bangkok.

Max S. Peters ,(1958), Plant Design and Economic for Chemical Engineers.Page 108-109

Morton, J. (1987), Fruits of Warm Climates, Miami FL: 115-121.

New Delhi, (1953), the Wealth of India, A Dictionary of Indian Raw Material and Industrial Products, (Part II D- E) Council of Scientific & Industrial Research Page…7 dye stuff 2.2

P.S.Rao,(1947), The Colouring Matter of the Tamarind Seed Testa, Forest Research Institute, Dehra Dun.

Paison Kongkachuichag *et al*, (2002), Studies on Dyeing of Silk Yarn with Lac Dyes: Effcets of Mordants and Dyeing Conditions, Sciences Asian Research Article.

Pearson, D., (1970), The Chemical Analysis of Food, Churchill Livingstone, 6th Edition, London and New York.

Seiko Akiyama, (1996), the Dye Plants of AWA; the Colour of Teats. Dai-ichi Co., Inc., Tokushima, Japan.

Sone, Y. and Sato, K., (1994), Measurement of Oligosaccharides Derived from Tamarind Xyloglucan by Competitive ELISA assay. Bioscience Biotechnology andBiochemistry, 58: 2295-2296.

Websites

http://en.wikipedia.org/wiki/ saponins

http://en.wikipedia.org .

http://en.wikipedia.org/polyphenol

http://en.wikipedia.org/wiki/Colour_fastness

http://en.wikipedia.org/wiki/condensed tannins

http://en.wikipedia-org/wiki/chemicals used in dyeing

http://oecotextiles.wordpress.com/category/ textile/mercerization

http://www.thesmarttime.com/testing/testing-procedures-34.html

http:/www.exoticfruitx.com/2011/tamarind.

http;//www. dsir.gov.in/ reports/tmrpes/vegdye.pdf.

www.tannicacid.info.com

www.whfoods.com/genpage.php?tname-nutrient & dbid=11g

www.wikipedia.org/phenol chemical structure

www.wisegeek.com/what are mordant.htm-42k

www.wisegeek.com/what is a glycoside.html

www.dharmatrading.com/info/mercerization/html