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## Effect of Catechu Natural Dye Extracts on Coloration and Antibacterial Protection Factor for Different Cellulosic Fabrics

H. M. Ibrahim<sup>\*</sup>, Hamada M. Mashaly, Nancy S. El-Hawary, Mona M. Kamel & Ekhlas A. El-Alfy

Textile Research Division, National Research Centre, 33 El Bohouth st. (Former El Tahrir St.), Dokki, Giza, Egypt, P.O.12622

### ABSTRACT

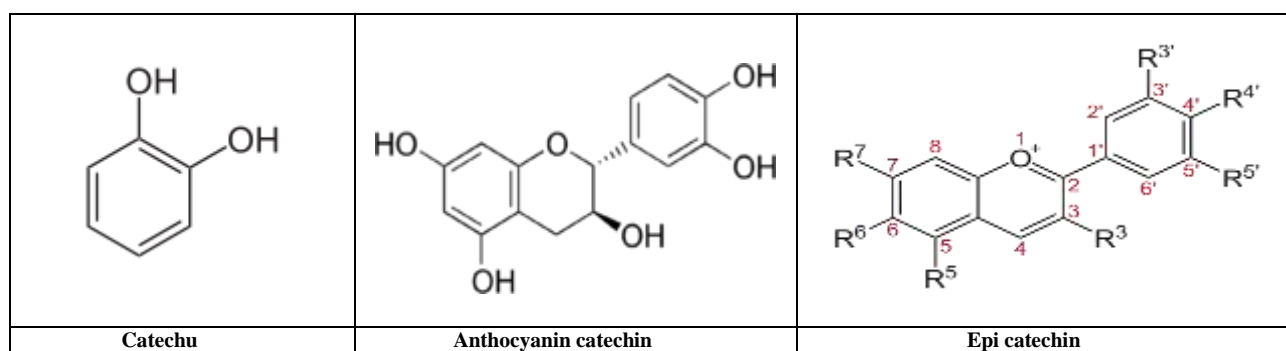
This work aims to prepare and evaluate a natural dye named catechu as a dye and antibacterial substance. Cotton and viscose fabrics were dyed with catechu natural dye at different conditions such as pH, dyeing time and temperature. The dyed samples were characterized. In addition, the antibacterial properties of the dyed fabrics were evaluated towards Gram-positive and Gram-negative bacteria. Results showed that the dyed fabrics have higher antibacterial activity so that it can be used as promising bactericidal material at the same time it showed very good durability to washing up to 25 times wash cycles.

**Keywords:** Natural dye, Viscous, Cotton fabric, Antibacterial

### INTRODUCTION

Nowadays natural dyes have received a great interest by scientists all over the world. Research works in dyeing using natural dyes for textiles are done, applied and compared to the synthetic ones. Natural dyes are biodegradable, and eco- friendly in nature with a variety of shades. Natural dyes are obtained from different sources like plants, insects/animals and minerals. Multifunctionalization of fabrics is of great interest, beside the environmental benefits of using natural dyes. A number of researches are now available for the multifunctional properties of natural dyes such as antimicrobial.

**Catechu:** Chemical structure is Catechu composed of Catechol and Catechin:  $C_6H_4(OH)_2$  dihydroxy benzene (Scheme 1):



Scheme 1: Chemical structure of catechu

Catechu, it is a vegetable dye used for tanning and dyeing wool, silk and cotton to give a yellow brown colour. It gives a gray brown with iron mordant and olive brown with copper mordant. Catechu has excellent light and washing fastness properties [1,2].

Catechu (or Cutch/ Katha) is a brown natural dye obtained chiefly from the heartwood of *Acacia catechu*, found in most of the Indian sub-Himalayas. The chief coloring component present in the catechu is catechin having molecular formula  $C_{15}H_{14}O_6$  [3]. The content of catechin in catechu varies from 4-7% [4]. This dye is being used extensively in dyeing of textile materials. Pharmacological studies have demonstrated that catechu is used in traditional medicines, show anti-inflammatory and anticancer activities. The powder is an extract prepared by steeping the

wood in boiling water until a syrupy liquid is formed and evaporating, drying and ground into powder. Catechu is common to most parts of India, Indonesia and Peru.

#### Taxonomy of Catechu

Plant	Botanic name	Components of colors	Family	$\lambda_{max}$ (nm)
Acaci catechu or dark catechump 104°C soluble in water, combustible	Catechu	Tannins and dyes	Catechol and catechin	385

#### MATERIALS AND METHODS

##### Materials and fabrics

Chemical reagents: are catechu and glacial acetic. Two bacterial strains from Bacterial Lab, Botany Department, the Faculty of women for Art, Science & Education, Ain Shams University, Cairo, Egypt were employed. They include *Staphylococcus aureus* as Gram-positive bacteria and *Escherichia coli* as Gram-negative bacteria. *S. aureus* and *E. coli* were selected as test cells because they are the most frequent bacteria in the wound infection and represent Gram-positive and Gram-negative bacteria, respectively. Fresh inoculants for antibacterial assessment were prepared on nutrient broth at 37°C for 24 h. Different two fabrics were used (cotton, viscose), Cotton fabric was purchased from Misr Spinning & Weaving Co. (Mehalla), El-Mehala El-kubra, Egypt. Viscose fabric was purchased from El- Chorbagy Co.

##### Methods

###### Extraction method via conventional extraction

Conventional extraction was carried out in 100 ml at different temperatures degrees (60-100°C) in distilled water using different amounts of the dye materials (1-5%) for different time intervals (20-120 min) at pH (2-11). After filtration the solution was used in dyeing of the fabrics.

##### Dyeing procedure

The dye bath containing 4% Catechu dye with liquor ratio 40:1, the fabrics were dyed using conventional heating at different pH values (2-11) for different durations (20-120 min) and at different temperatures (60-100°C).

##### Fastness properties

###### Washing fastness

The colour fastness to washing was determined according to ISO 105-CO2:1989 test method [5]. The washing fastness tests were conducted in a launder meter (ATLAS–Germany) using 5 g/l nonionic detergent at 50°C for 45 min. the liquor ratio was 1:50, then the specimen rinse with running tap water, squeeze and dry in air. The test specimen and the two adjacent fabrics (cotton and wool) were compared using the gray scale.

###### Light fastness

This test was evaluated according to ISO 105-B02: 1988 test method [6] using a carbon arc lamp. Samples were exposed to a continuous light for 35 h in order to determine the degree of colour resistance to light photo- degradation.

###### Rubbing fastness

Rubbing fastness was determined according to test method ISO 105-X12: 1987 [7] using a crock-meter under conditions for determining dry and wet fastness.

##### Testing

###### Color measurements of the dyed fabrics

Colour-difference formula= $\Delta E$  CIE ( $L^*$ ,  $a^*$ ,  $b^*$ )

The total difference= $\Delta E$  CIE ( $L^*$ ,  $a^*$ ,  $b^*$ ) was measured using the Hunter-Lab spectrophotometer (model: Hunter Lab DP-9000)

The total difference= $\Delta E$  CIE ( $L^*$ ,  $a^*$ ,  $b^*$ ) between two colours, each given in terms of  $L^*$ ,  $a^*$ ,  $b^*$  is calculated from:

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

Where,  $\Delta E^*$  value: is a measure of the perceived colour size of the colour difference between standard and sample and cannot indicate the nature of that difference,  $\Delta L^*$  value: indicates any difference in lightness, (+) if sample is lighter than standard, (-) if darker,  $\Delta a^*$  and  $\Delta b^*$  values: indicate the relative positions in CIELAB space of the sample and the standard, from which some indication of the nature of the difference can be seen.

##### Evaluation of antibacterial activity *in vitro*

The antibacterial activity of treated and dyed samples was determined against the test bacteria by disk diffusion method on an agar plate [8,9]. Briefly, 1 cm diameter blended film samples were cut and put into 10 ml of nutrient agar, to which 10  $\mu$ l of microbe culture was inoculated, after the solidification. The plates were incubated at 37°C for 24 h, after which the diameter of inhibition zone were measured and recorded.

#### RESULTS AND DISCUSSION

##### Conventional extraction of dye

###### Effect of dye concentrations on colour strength

The extraction of dry berry leaves was carried out at 100°C for 60 min and at different concentrations (20-100 g/l). Table 1 shows that as the dye

amount % (Amount of dye used per 100 ml distilled water) increases, the K/S of the dye extract why increases. The maximum extractability was attained with 8 g/100 ml water.

**Table 1: Effect of concentrations of plane extraction on colour strength**

Effect of concentration (%)	K/S of cotton fabric	$\Delta E$ of cotton fabric	K/S of viscose fabric	$\Delta E$ of viscose fabric
10%	2.40	33.99	5.5	36.44
20%	2.89	41.18	6.38	44.63
30%	4.04	40.48	6.73	44.12
40%	3.74	41.60	7.12	44.95
50%	3.12	40.42	6.75	41.21

Condition of extraction (Time 60 min; Temp. 100; L:R 1:40)

#### Effect of temperature of plane extraction

The extraction of dye was carried out at dye concentration 80 g/l for 60 min for viscose fabric and 30 g/l for cotton and at different temperatures (60-100°C). Table 2 shows that as the temperature increases, the K/S of the dye extract also increases when using either conventional or microwave. The maximum extractability was attained with 100°C.

**Table 2: Effect of temperature of plane extraction on colour strength**

Effect of temperature	K/S of cotton fabric	$\Delta E$ of cotton fabric	K/S of viscose fabric	$\Delta E$ of viscose fabric
60°C	1.67	28.18	6.74	31.06
70°C	1.84	29.58	6.33	32.62
80°C	1.97	31.44	7.41	35.65
90°C	1.86	30.92	4.47	33.11
100°C	1.88	31.50	4.58	34.58

Condition of extraction (Conc. 40%; Time 60 min; L:R 1:40; pH 4)

#### Effect of time of plane extraction

The extraction of dye was carried out at dye concentration 80 g/l for 60 min for viscose fabric and 60 g/l for cotton at 80°C for different times (20-120 min). Table 3 shows that as the time increases, the K/S of the dye extract also increases when using either conventional. The maximum extractability was attained 100 min.

**Table 3: Effect of time of plane extraction**

Effect of time	K/S of cotton fabric	$\Delta E$ of cotton fabric	K/S of viscose fabric	$\Delta E$ of viscose fabric
20min.	2.71	32.23	3.30	23.09
40min.	3.59	26.46	4.70	24.06
60min.	4.76	29.85	6.27	27.80
80min.	3.90	30.15	7.49	32.91
100min.	4.01	28.65	5.53	26.83

Condition of extraction (Conc. 40%; Temp. 80°C; L:R 1:40)

#### Factors affecting on dyeing process

##### Effect of dye bath pH

Table 4 show that the pH values of the dye bath have considerable effect on the dyeability of the fabric with the dye under conventional. It is clear that the dyeability of the fabrics at pH 2 and 4 values but started to decline sharply as the pH increases with rather lower dyeability. The effect of dye bath pH can be attributed to the correlation between dye structure and the fabric structure.

**Table 4: Effect of pH of dye bath**

Effect of pH	K/S of cotton fabric	$\Delta E$ of cotton fabric	K/S of viscose fabric	$\Delta E$ of viscose fabric
2	4.10	41.92	8.49	48.27
4.	3.61	40.90	9.45	48.53
6	3.90	42.51	6.89	40.79
8	2.50	35.78	5.78	42.32
9	3.00	39.55	6.72	43.50
10	2.26	33.22	7.38	37.93

Condition of extraction (Conc. 40%; Temp. 80°C; L:R 1:40; Time. 60 min)

#### Effect of temperature of dyeing

The effect of temperature on the dyeability of the fabrics with the dye was conducted at conventional conditions at different temperatures. As shown in Table 5 it is clear that the colour strength increases with the dyeing temperature increase in conventional.

**Table 5: Effect of dyeing temperature**

Effect of temperature	K/S of cotton fabric	$\Delta E$ of cotton fabric	K/S of viscose fabric	$\Delta E$ of viscose fabric
60 °C	4.24	34.91	10.16	44.85
70 °C	3.73	34.76	9.67	44.60
80 °C	3.77	33.62	9.15	44.48
90 °C	3.75	34.52	8.92	45.01

100 °C	3.65	34.38	8.66	43.95
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Condition of dyeing (Conc. 40%; Time 60 min; L:R 1:40, pH 4)

### Effect of dyeing time

The effect of dyeing time was conducted at a concentration of the dye 8 g/100 ml water for viscose and 6 g/100 ml water for cotton fabric. As shown in Table 6, the colour strength K/S obtained was increased as the time increases in conventional dyeing.

**Table 6: Effect of dyeing time**

Effect of Time	K/S of cotton fabric	ΔE of cotton fabric	K/S of Viscose fabric	ΔE of Viscose fabric
20min.	2.75	34.11	4.43	43.35
40min.	2.45	34.63	7.22	50.80
60min.	2.37	34.96	6.78	50.51
80min.	3.52	40.97	8.92	54.32
100min.	3.96	41.89	8.98	50.78

Condition of dyeing (Conc. 40%; Time 60 min; L:R 1:40; Temp.90°C; pH 4)

### Antibacterial activities of natural dye

The extracted dye was tested for their antibacterial activity *in vitro* through disk inhibition zone method by using two bacterial strains, *S. aureus* as Gram-positive bacteria and *E. coli* as Gram-negative bacteria, taking tetracycline and ciprofloxacin as standard drugs. Table 7 shows the antibacterial activity of the extracted dye with different dye concentrations from 1-6% (wt/v).

**Table 7: Results of antibacterial test for the extracted**

Dye concentration, % (wt./v)	Zone of inhibition (diameter in mm)	
	<i>S. aureus</i>	<i>E. coli</i>
1%	0	0
2%	6.0	4.5
3%	13	10.5
4%	20	17.5
5%	20	18
6%	20	19
ciprofloxacin	22	19
tetracycline	20	18.5

The effect of dye concentration on antibacterial activity was studied and the results summarized in Table 7. The inhibition zone in each case was recorded the results of undyed samples shows that they don't have bacterial activity. Table 7 shows that the antibacterial activity of the dye increased as its concentration increase from 1% to 4% (wt/v) then its antibacterial activity remains constant. So that we used this concentration (4%) in dyes processes.

Table 7 also shows that this dye showed as excellent antibacterial activity towards *S. aureus* and very good towards *E. coli* with respect to standard drugs so that this dye is bactericidal not bacteriostatic agent. The antibacterial activity of Epicatechin due to its cationic nature of the dye, this positive charges allow dye molecule to be adsorbed readily onto bacterial surface, and then penetrate its cell membrane, followed by destruction of cell membrane and leakage of cell inclusion body causing bacterial death. The dye showed better efficiency against *S. aureus* as Gram-positive bacteria than *E. coli* as negative bacteria because of the structure of its cell wall [10,11].

### Effect of washing durability of dye on antibacterial activity

Table 8 shows that effect of washing durability of dyed fabrics on its antibacterial activity after twenty five times wash fastness. The durability of washing of these antibacterial functions depends on bonding between dyes and fabrics.

**Table 8 Effect of washing durability of dyed samples on antibacterial activity**

Fabrics	Wash cycle	Zone of inhibition (diameter in mm)	
		<i>S. aureus</i>	<i>E. coli</i>
Cotton	0	16.5	15
	1	15	12.5
	5	14	12
	10	14	11.5
	15	10.5	8.5
	20	7.5	6.5
	25	5	1.5
Viscose	0	18.5	17
	1	17	15.5
	5	14.5	11.5
	10	14	11.5
	15	12.5	9
	20	9.5	7.5
	25	6.5	3.5

Table 8 show that dyed fabrics are durable to wash fastness after 25 times it still have antibacterial activity for both types. Since the bonds between fibers and dyes through chemical bonding that cause wash fastness of its antibacterial functions. In addition, it was clear that from that from Table 8 that antibacterial activity of all viscose dyes samples shows higher than that for cotton dyed samples due to fabric nature and

structure.

#### CONCLUSION

It can be concluded that cotton and viscose fabrics can be dyed with catechu natural dye extracts. The optimum conditions for dyeing process are at pH 4 and 80°C temperature. The dyed fabrics show higher antibacterial activity towards both Gram-positive and Gram-negative bacteria so that they can be used as wound dressing material. In addition, dyed fabrics are durable towards washing up to 25 wash cycles.

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