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Improvement of dyeability and antibacterial properties of gelatin treated cotton fabrics with synthesized reactive dye

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Cotton fabrics were treated with gelatin polymer to enhance its dyeability, color strength using synthesized reactive dyes and antibacterial properties. Pad- dry- cue method was used for the treatment process. The prepared treatment pad- bath was contained dimethylol dihydroxy ethylene urea (DMDHEU), ammonium dihydrogen phosphate catalyst and the gelatin at concentration (5 %, w/w) and with a non-ionic wetting agent. Then The fabrics were dyed with synthesized reactive dyes. The dyed samples were assessment towards both dyeing behavior and antibacterial activity. These results indicating high quality dyeing properties, However, monochlorotriazine reactive dyes showed higher exhaustion and fixation values, calorimetric data (CIE L*a*b*C*h^0) and fastness properties of the dyed cotton fabrics. In addition, these reactive dyes have higher antibacterial activity toward Gram negative and Gram positive compared with two widely used antibiotics named ciprofloxacin and tetracycline.

Keywords: Gelatin; Cotton; Antibacterial activity; reactive dyes; Dyeing; Gram negative; Gram positive.

INTRODUCTION

Collagen is a structural protein which by degradation can produce a natural biopolymer with low cost; gelatin (Crespo et al., 1995). Gelatin contains 18 different amino-acids with various concentrations. It has a high level of prolin, hydroxyproline and glycine. The main structure is -Ala-Gly-Pro-Arg-Gly-Glu-4Hyp-Gly-Pro-. These amino-acids are connected with each to form polypeptide chains containing more than a thousand amino-acids. Gelatin is widely used in the photographic, food and pharmaceutical industries (Digenis et al., 1994).

Gelatin can be used in bone surgery as well as in trauma. It is used as screws, plates, and rods because the biopolymer is nontoxic, biodegradable and inexpensive. Bone-fixing screws prepared from gelatin were obtained by Parkany and Horvathas early as the 1970s (Zeeshan et al., 2015, Wei et al., 2015)

Several studies have elucidated that mechanical properties of gelatin can be improved and Adjusted (Wan et al., 2000, Wan et al., 2000, Khan et al., 2010, Li et al., 2015). Recently, composites composed by gelatin and inorganic materials, such as tricalcium phosphate and hydroxyapatite, have been (Lin et al., 1998).

Gelatin microcapsules containing vitamin C were prepared using emulsion hardening technique. Both the optical microscopy and scanning electron microscopy demonstrated that the newly developed microcapsules were in the form of core-shell spheres with relatively smooth surface (Shuk et al., 2009)

Other work was studied the effect of cellulose grafting to create dimethyl amino ethyl methacrylate groups to improve dye ability of cellulose towards both reactive and direct dyes. On the other hand, these created groups realized dyeing properties of the acid dyes (Mohamed et al., 2016).

The aim of our work is to improve the dye ability of cotton fabric toward reactive dyes by treatment this fabric by gelatin natural substance with dimethylol dihydroxy ethelyen urea (DMDHEU) and dyeing this cotton fabric under the typical exhaust dyeing conditions with synthesized reactive dyes. Also, we measured the antibacterial activity toward Gram negative and Gram positive.

MATERIALS AND METHODS

Fabrics

Mill-scoured and bleached cotton fabric, 130 g/m^2 was obtained from El-Mahala Co., Egypt. Befor dyeing, the fabric was treated with a solution containing 3 g/l non-ionic detergent (Hostapal CV, Hoechst) and 5g/l sodium carbonate at the boil for one hand a liquor ratio 50:1(LR) thoroughly then washed thoroughly in water and dried at room temperature.

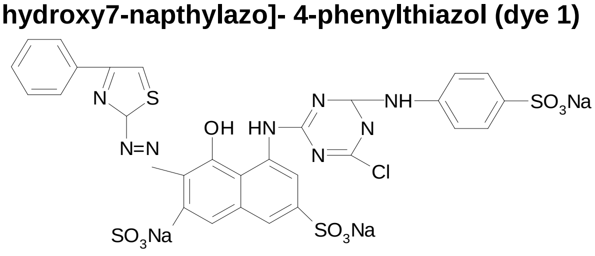
Chemicals

Gelatin, ammonium dihydrogen phosphate and dimethylol dihydroxyethelyenurea (DMDHEU). Cyanuric chloride (98 %) were brought from Merk-Schuchardt. Thiourea, 4-aminobenzene-sulfonic acid and H-acid were obtained from FlukaChemite AG. All other chemicals used in our work were found in laboratory.

Synthesis

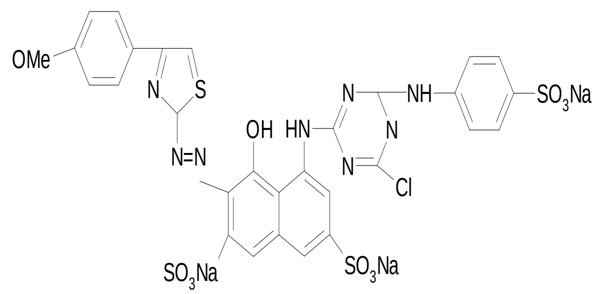
We synthesized reactive dyes according to previous methods (Mohamed et al., 2018).

[(p-sodium sulphonato-4-phenylamino)-1,3,5-triazine-2-ylamino]-3,6-disodiumsulphonato-8-hydroxy7-napthylazo]- 4-phenylthiazol (dye 1)



Dye 1: Blue, m.p.> 300˚C, yeild 89 per cent, λ_{max} (H_2O) 568 nm.

[(p-sodiumsulphonato-4-phenylamino)-1,3,5-triazine-2-ylamino]-3,6-disodiumsulphonato-8-hydroxy7-napthylazo]-3-methoxy-4-phenylthiazol (dye 2)



Dye 2: Blue, m.p.> 300˚C, yeild 91 per cent, λ_{max} (H_2O) 570nm.

Preparation of treated fabric

The fabric treated for finishing through the pad-dry-cure method. The pad-bath was containing gelatin at concentration (5 % w/w), ammonium dihydrogen phosphate catalyst (1-3 g/L) as catalyst, dimethylol dihydroxy ethelyen urea (DMDHEU) (20 g/L) as binder and a non-ionic wetting agent. The fabrics were padded to 100% wet pick up, then fixed in pin frames, dried at 80 ^{0}C for 5 min and cured at 140 ^{0}C for 3 min. All cured samples were washed at 50 ^{0}C for 5 min and conditioned before testing and analysis.

Dyeing procedures

In the dyeing process, synthesized dyes 1 and 2 were applied 1% , 2% and 3% of owf in an Ahiba dyeing machine in the conventional dyeing, all conditions used were recommended by the dye manufacturer with the liquor ratio 40∶1 and anhydrous sodium sulphate (60 g/l), which was added twice within 30 min in exhaust dyeing. Then the temperature was increased to the fixation temperature with the heating-rate of 2 °C·min^{-1}, followed by addition of 10 g/l of sodium carbonate for dye fixation. The fixation temperature was that recommended for conventional dyeing 80 °C (Karapinar et al., 2007). In the salt-free dyeing, a piece of the cotton pretreated with D.M.A.E.M was dyed at 40°C, over 30 min. with the liquor ratio of 40∶1. Then, the cotton was dried in air.

Measurements and Testing

Dye Exhaustion

Uptake of the reactive dyes by the cotton pretreated with different concentration of gelatin and dimethylol dihydroxy ethelyen urea (DMDHEU) was measured by sampling the dyebath before and after dyeing on a Shimadzu UV-2401PC UV/V is spectrophotometer at the λ_{max} value using a calibration curve previously obtained using known dye concentrations (g/l). The percentage of dyebath exhaustion (%*E*) was calculated using Eq. 1.

$$\%E = \left[1 - \frac{C_2}{C_1}\right] X \; 100 \longrightarrow (1)$$

Where C_1 and C_2 are the dye concentrations in the dyebath before and after dyeing, respectively.

Colour measurements

The colour parameters of the un treated and pretreated with gelatin and dimethylol dihydroxy ethelyen urea (DMDHEU) of dyed cotton fabric were determined using an Ultra Scan PRO spectrophotometer (Hunter Lab) with a D65 illuminant and 10° standard observer (Hu et al., 1987, Savarino et al., 1989) The corresponding *K*/*S* values were calculated from the reflectance data at λ_{max} of the dyeing.

Fastness Testing

Dyed the pretreated cotton fabric with gelatine and dimethylol dihydroxy ethelyen urea (DMDHEU) samples, after washing-off using 2 g/L nonionic detergent at 80°C for 15 min, were tested by standard ISO methods. Wash fastness (ISO 105-C02 (1989), crock fastness (ISO 105-X12 (1987), and fastness to perspiration (ISO 105-E04 (1989) were evaluated using the visual ISO Gray Scale for both color change (AATCC Evaluation Procedure (EP) 1-similar to ISO 105-A02) and color staining (AATCC EP 2—same as ISO 105-A03). Light fastness (carbon arc) was evaluated using ISO 105-B02.

Evaluation of Antibacterial in vitro:

Two bacterial strains from Bacterial Lab, Botany Department, the faculty of woman for Art, Science& Education, Ain Shams University, cairo, Egypt, were employed. They include Staphylococcus aureus (*S.aureus*) as Gram-positive (G+ve) bacteria and Escherichia coli (*E. coli*) as Gram-negative (G-ve) bacteria. Fresh inoculants for antibacterial assessment were prepared on nutrient broth at 37 ˚C for 24 h. We were selected these species as test cells because they are the most frequent bacteria in the wounded infection.

The antibacterial activity of treated and dyed samples was determined against the test bacteria by disk diffusion method on an agar plate as reported method (Zhang et al., 2005, Seyam et al., 2012).

RESULTS AND DISCUSSION

The cotton fabrics were pre-treated with gelatin in the presence of di methylol di hydroxy ethylene urea (DMDHEU) by pad-dry-cure method and then the fabrics can have treated cotton fabrics can absorb the reactive dyes via the electrostatic interaction in the dye bath (Ma et al., 2003). The treatment of cotton fabrics with gelatin has direct effect of absorption capacity of gelatin on the fabrics which cause an increase of the positive charge density on the cotton fabrics to increase the cationic properties of the cotton fabrics and further affect the dye ability of the treated fabrics.

In this study, dye ability of the pretreated cotton fabrics was used to estimate the pretreatment conditions to obtain the optimum condition.

Colorimetric properties (Colour strength)

The colorimetric data CIE lab coordinates (L* a* b*) and color strength (K/S) of the dyed cotton fabrics with the two reactive dyes 1, 2 were measured at different concentrations of dyes and shade depth from 1-3% owf. The data represented in tables 1,2 and 3.

Table 1 shows colour measurements for untreated cotton fabric while tables 2 and 3 show these measurements for treated fabric with gelatin. Tables 2 and 3 show that the values of K/S increased in treated cotton fabrics more than that for the untreated fabrics in table 1 and the values increased as the concentration of the dye increased.

CIE lab system and modified CIE $L^*C^*h^0$ (D65/10) system were used to evaluate the colour parameters of the dyed fabrics. Digital CIE lab system measurs the following colour parameters for each dyed samples: L* for lightness, a* for redness (+ coordinate) or greenness (- coordinate). b* for yellowness (+ coordinate) or blueness (- coordinate), C* for chromaticity and h for the colour hue.

Fastness Properties

The pretreated dyed samples were washed-off using 2 g/l non-ionic detergent at 100 for 15 min, then fastness properties were measured as mentioned in the experimental part.

Table 1: Colorimetric data of the untreated dyed cotton fabrics using dyes 1 and 2

No.of dye	Conc.of dye (%)	K/S	L*	a*	b*
1	1%	25.12	50.43	6.09	-22.53
	2%	26.37	46.39	5.48	-25.16
	3%	28.06	42.93	6.84	-26.28
2	1%	20.06	23.01	11.77	-28.03
	2%	24.62	23.71	11.94	-26.63
	3%	25.32	29.22	9.00	-22.44

Table 2: Colorimetric data of the dyed pre-treated cotton fabric using dye 1

Concentration of dye 1	K/S	L*	a*	b*	▲E	h
1%	28.16	42.81	6.72	-26.00	72.07	285.32
2%	29.21	38.72	5.93	-24.53	73.92	283.41
3%	30.82	35.81	6.99	-22.12	74.13	284.90

Table 3: Colorimetric data of the dyed pre-treated cotton fabric using dye 2

Concentration of dye 2	K/S	L*	a*	b*	▲E	h
1%	25.21	29.31	9.30	-25.54	68.09	285.3
2%	27.32	26.92	10.01	-18.82	70.01	282.8
3%	28.34	25.81	11.11	-15.00	72.2	285.75

Table 4: Fastness properties of dyed pre-treated cotton fabric using dyes 1 and 2

Dye No.	Fabric	Fastness to rubbing		Wash fastness**			Fastness to perspiration**						Light
							Alkaline			Acidic			
		Dry	Wet	Alt	SC	SW	Alt	SC	SW	Alt	SC	SW	
1	C	4-5 4-5	4 4	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5
2	C	4-5 4-5	4 4	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	4-5 4-5	5

* C, Cotton **Alt = alteration; SC = staining on cotton; SW = staining on wool

Table 4 shows that the fastness properties to both rubbing, washing and perspiration of the gelatin treated cotton fabrics were excellent to very good values in comparison with that for the untreated cotton fabrics.

Antibacterial activity of the reactive dyes under investigation:

Tables 5 and 6 show the antibacterial activity of the synthesized reactive dyes, evaluated invitro via disk diffusion method, by using *S. aureus* as Gram-positve bacteria and *E. coli* as Gram-negative bacteria.

The antibacterial of the dyes were evaluated with comparison two widely used antibiotics names, ciprofloxacin and terracycline to show the efficincy of these two dyes as antibacterial agents towars both Gram positive and Gram negative bacteria at the sam concentration of 20 µg/ml as shown in the Table 5. So that we can use these dyes to impart the cotton fabric antibacterial activity in addition dyeing propertes discosed before.

Table 6 shows that the antibacterial activity of pretreated dyed cotton fabrics expressed in inhibition zone increases as the dye concentration increase (from 1-3% w/v) for both dyes 1 and 2 .In addition, dye 1 show higher antibacterial activity than dye 2 at all concentration. All reactive dyes have antimicrobial activity. It was attributed to contain Thiazole and NH group in molecular structure which adsorb onto bacterial surface, to penetrate cell membrane, finally destruct cell membrane causing bacteria death. These dyes showed higher antibacterial activity toward S. aureus as Gram positive bacteria more than E. coli as Gram negative bacteria (E. coli) due to their cell wall structure (Ma and Sun, 2005, Liu et al., 2007, Liu and Sun 2008).

Table 5: Antibacterial activity of synthesized reactive dyes vs. ciprofloxacin and tetracycline antibiotics (all at 20 µg/ml concentration):

Dye powder	Zone of inhibition (diameter in mm)	
	Staphylococcus aureus	*Escherichia coli*
Dye 1	22.0	18.0
Dye 2	23.5	19.0
Ciprofloxacin	24.0	22.5
Tetracycline	21.0	20.0

Table 6: The effect of dye concentration for pre-treated dyed cotton fabric on antibacterial activity.

Dye	Concentration, %	Zone of inhibition (diameter in mm)	
		Staphylococcus aureus	*Escherichia coli*
Dye 1	1	11	10.5
	2	16	15.5
	3	23	20
Dye 2	1	9.5	9.0
	2	14	11.5
	3	19	18.5

CONCLUSION

We successfully prepare mono-functional reactive dyes 1 and 2 previously based on thiazole moiety have antimicrobial activity toward Gram positive and Gram negative bacteria. Also, we can improve dyeability of cotton fabric by pretreatment with gelatin and (DMDHEU). In addition, we evaluate the efficiency of antibacterial activity of the dyed cotton fabrics towards both Gram-positive and Gram-negative bacteria. From our results we can conclude that the dyability and antibacterial activity of dyed fabrics enhanced so that it can be used in medical and pharmaceutical industries as medical textiles. Moreover, fastness to washing, rubbing and perspiration of all samples dyed were excellent to very good.

CONFLICT OF INTEREST

The authors declared that present study was performed in absence of any conflict of interest.

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