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Synthesis and Application of New Reactive Disperse Dyes Based on Isatin Derivatives and their Antibacterial Activity

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A NEW two reactive disperse dyes based on a hybrid structure of isatin and benzanthrone or anthraquinone via 1, 3, 5 triazine spacer were prepared. The prepared compounds were characterized by ^{1}H-NMR, mass spectrometry and elemental analysis. The UV/vis absorption spectra and emission spectra were measured in *N*, *N*-dimethylformamide (DMF) solution at room temperature, the effects of substituent's on the emission spectra of these compounds were interpreted. The dyeing application of the prepared dyes on wool and polyamide-6 fabrics at various concentrations of dye and different pH were investigated as well as the exhaustion and dye fixation were studied. The antibacterial activity of the prepared reactive disperse dyes were studied against different kind of bacteria such as *Staphylococcus aureus* (Gram-positive bacteria) and *Escherichia coli* (Gram-negative bacteria). The antibacterial affinity of the prepared dyes was exhibit a significant effect compared with selected antibiotics as reference standard. The fastness properties of the dyed fabrics were studied which showing excellent wash fastness, rubbing and perspiration fastness as well as high stability to light.

Keywords: Reactive disperse dyes, Dyeing, Polyamide, Wool, Antibacterial.

Introduction

Different classes of dyes according to their application on the fibers are known long time ago [1, 2], while reactive dyes for cotton dyeing and disperse dyes for synthetic fibers such as polyester fabric are the most common classes [3-8]. Although disperse dyes can be used for dyeing of nylon fiber, but the use of disperse dyes are not common and have some limitation in nylon dyeing because of their week build up and its low fastness characters. Thus introducing a reactive moiety in the disperse dye structure have a great advantage which improve the buildup of the dye in nylon fabrics throw its covalent bond with the fiber, this type of dyes known as reactive disperse dyes which have the favorable characteristics of both disperse and reactive dyes [9-11]. Triazinyl derivatives are very important compounds not only as dye intermediate but also in the area of preparing functionalized 1,3,5-Triazine derivatives which covering a wide area of application such as organic light emitting diodes (OLED's), medical, textile, plastic, and rubber industries [12-20].

On the other hand the import fluorescent character in the dye molecules is considered as add value of the dye character, which increase the brightness of the color with high visibility textile product [21]. Thus, the improvement of the dyes have tended to concerning on specific compounds introducing a new characters by build up a new functionalities for potential application in various fields such as medical applications, photodynamic therapy, lasers, reprographic technology, functional colorants, nonlinear optical systems and data storage applications [22,23].

Otherwise, 1H-indole-2, 3-dione (isatin) recently become one of the most interesting intermediate. Isatin derivatives are particularly

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interesting due to their biological and pharmacological properties [24]. Isatin was used for the synthesis of many different heterocyclic compounds as indoles, quinolones and in drug synthesis as starting material [25]. Recently isatin derivatives were used in modern application such as sensor and high performance disperse dyes for polyester dyeing [26-28].

In this work we are focus on the synthesis of new reactive disperse dyes based on two different chromophoric system (anthraquinone, benzanthrone) bearing isatin moiety via triazinyle spacer. The spectral study of the prepared dyes such as absorption and emission spectra and fluorescence quantum yield were studied in details as well as the dyeing applications of the prepared dyes on polyamide and wool fabric were investigated. Due to the antibacterial behavior of isatin moiety the antibacterial affinity of prepared dyes was also studied.

Experimental

Materials and Equipments

Materials

Isatin 99.0%, 2-aminoanthraqinone, 2-aminobenzanthrone and hydrazine hydrate were purchased from Sigma-Aldrich (Germany). All solvents used for the synthesis were as pure solvents and *N, N*-dimethylformamide was spectroscopic grade. All chemicals used without any further purifications. Mill-scoured, bleached wool (160 g/m^2) and nylon fabric (114 g/m^2) were obtained from El-Mahalla El-Kobra Company, Egypt. Thin-layer chromatography (TLC) was performed with a Kieselgel 60 F254 instrument (Merck, Germany).

Equipments

The melting points of the prepared compounds were determined on a Stuart (UK) melting point SMP 30 instrument and are uncorrected. Elemental analysis was conducted on an EA 1108 (Fisons Instruments, USA). H-NMR spectra were run at 300 MHz in deuterated dimethylsulphoxide (DMSO-d6). Mass spectra were measured on a Varian MAT CH-5 spectrometer (70 eV). UV/Visible spectra were measured on a Shimadzu UV-2401 PC UV/Vis spectrophotometer using *N, N*-dimethylformamide. The steady-state fluorescence spectra were measured using JASCO FP-8300 spectrophotometer. The instrument provides corrected excitation spectra directly; the fluorescence emission spectra were corrected for the characteristics of the emission monochromator and for the detection photomultiplier response. The fluorescence emission spectra were recorded by excitation at absorption maxima. The fluorescence quantum yields (Φ) were determined using the classical formula: $\Phi_x = (A_s \times F_x \times n^2_x \times \Phi_s)/(A_x \times F_s \times n^2_s)$ where A is absorbance at the excitation wavelength, F the area under the fluorescence curve, and n is the refractive index of the solvents used. Subscripts 's' and 'x' refer to the standard and to the sample of unknown quantum yield, respectively. Anthracene in ethanol (Φ= 0.27) was taken as the standard [29]. The visual color strength (K/S) of dyed fabric was measured on Data color International SF 600 plus.The reflectance values of the fabric were measured using a Data Color SF 600+. Relative color strengths (K/S values) were determined using the Kubelka-Munk equation. K/S = (1-R) 2 /2R

Synthesis of dyes

The synthetic procedure of the intermediate **1** (isatin hydrazide) and intermediate **2** (dichloro triazinyle derivative) as a key intermediate for the synthesis of dyes 1 and 2 were prepared as the published procedure, analytical data was matching the previously published method [30-32].

Synthesis of 3-(2-chloro-6-(7-oxo-7H-benzo [de]anthracen-3-ylamino)-1, 3, 5-triazin-2-yl) hydrazono) indolin-2-one (Dye 1)

To a mixture of intermediate **1** (0.5g, 1.6mmol) and sodium acetate (0.5g) in 30 ml acetic acid solution of 3-Aminobenzanthrone (0.4 g, 1.6 mmol) was added dropwise. The reaction mixture was stirred and heated at 50 ºC for 6 hours until the starting material disappeared. The precipitate was filtered and washed with cold water to obtain 0.72g (87% yield) of dye 1 as brown powder recrystallized from ethanol. m.p > 300 ºC.

1H-NMR: (DMSO-d6, 300 MHz): δ = 6.92-8.43(m, 13H, aromatic), 10.95 (s, 1H, NH), 11.26 (s, 1H, NH), 12.86 (s, 1H, NH).

Mass spectra: m/z (%): 519 ([M + H]$^+$, 100), (MW 518 g mol^{-1}). Anal. calcd. for $C_{28}H_{16}ClN_7O_2$:C (64.93%) H (3.11%) N (18.93%); found: C (65.04%) H (3.18%) N (18.99%).

Synthesis of 2-(4-chloro-6-[2-(2-oxo-1, 2-dihydro-3H-indol-3-ylidene) hydrazinyl]-1, 3, 5-triazin-2-yl) aminoanthracene-9, 10-dione (Dye 2)

To mixture of intermediate **1** (2.73g, 10 mmol) and sodium carbonate (0.5g) in 100 ml tetrahydrofuran a solution of 2-Aminoanthraquinone (2.23 g, 10 mmol) was added dropwise. The reaction mixture was stirred and heated at 50 ºC for 3 hours until starting material disappeared. The reaction mixture was filtered to remove the inorganic salt and dye 2 was precipitated from mother liquor by mixing with 150 ml cold water to obtain 3g (60% yield) of dye 2 as red powder recrystallized from ethanol, m.p 268 ºC.

^{1}H-NMR: (DMSO-d6, 300 MHz): δ = 7.1-8.21 (m, 11H, Aromatic proton), 9.2 (s, 1H, NH), 11.26 (s, 1H, NH), 12.86 (s, 1H, NH). Mass spectra: m/z (%): 497 ([M + H] $^+$, 80), (MW 496 g mol^{-1}).Anal. calcd. for $C_{25}H_{14}ClN_7O_3$: C (60.55%) H (2.85%) N (19.77%); found: C (61.1%) H (2.70%) N (19.80%).

Dyeing procedure

Effect of pH and dye concentration

The fabrics were scoured before application in an aqueous bath containing 2 g/L of nonionic detergent at 80 °C for 30 min to remove any processing oils and impurities. The dyes were applied at various pH (3-4-5-6-7) for wool and (4-6-8-10) for nylon using 2% owf dye concentration and liquor ratio 50:1. Each dyeing was performed at 40 °C, allowing the temperature of the dye bath to raise to the boiling over 30 min. The dyeing was continued at the boiling temperature 98 ºC for further 60 min. At the end of the dyeing process, the samples were thoroughly rinsed and air-dried. By following the same dyeing procedure the effect of dye concentration on the dyeing behavior was also investigated using different dye concentrations (1, 2, 3, 4 and 5 % owf) at pH 5 for wool and pH 8 for nylon using liquor ratio 50:1.

Exhaustion and fixation measurements

The dyeing application on both wool and nylon fabrics for studying the exhaustion and fixation yield was done at pH 5 for wool and pH 8 for nylon using liquor ratio 50:1 and 2% dye concentration. The dyeing process was started at 40°C, allowing the temperature of the dyebath to raise to the boil over 30 min. The dyeing was continued for a further 60 minutes and the dyed samples were thoroughly rinsed with water and dried.

Dye exhaustion

The dye exhaustion was measured by sampling the dyebath before and after dyeing. The dye concentration (g/l) of the dyebath was measured spectrophotometrically at λmax of each dye. The percentage of dye exhaustion (% E) was calculated using equation (1):

$$\%E = \frac{C_1 - C_2}{C_1} \times 100 \qquad (1)$$

where C_1 and C_2 are the concentrations of dye in the dye bath before and after dyeing, respectively.

Dye fixation

Dye fixation (%F) (percentage of the exhausted dye that chemically bound on the fiber) was measured by refluxing the dyed samples in 50% aqueous DMF (liquor ratio 20:1) for 15 min to extract the unfixed dye. This procedure was repeated until the extract was colorless. The concentration of the extract was then measured spectrophotometrically at λmax of each dye and the dye fixation ratio calculated using equation 2:

$$\%F = \frac{C_1 - C_2 - C_3}{C_1 - C_2} \times 100 \qquad (2)$$

Where, C_3 is the concentration of extracted dye.

From the dyebath exhaustion (%E) and dye fixation (%F), the total dye fixation (%T), which is the percentage of dye chemically bound relative to the total amount of dye used, was calculated for the prepared dyes using equation 3:

$$\%T = \frac{(\%E \times \%F)}{100} \qquad (3)$$

Dyeing measurements

The relative color strength (K/S) and CIELAB coordinates (L*a*b*ΔE) of the dyed fabrics were also measured using a Hunter lab's Ultra Scan PRO spectrophotometer (USA) under illuminant D65, 10°standard observer. The K/S value of dyed fabrics was measured by the light reflectance technique using Kubelka-Munk equation 4. The reflectance (R) of the dyed fabrics was measured according to the following equation.

$$\mathbf{K/S = (1-R)^2/ 2R} \qquad (4)$$

where,

R = Decimal fraction of the reflection of the dyed fabric

K = Absorption coefficient, and S = scattering coefficient

Fastness testing

Dyed nylon and wool samples with 2% shade (owf) after washing-off using 2 g/l nonionic detergents at 80 °C for 15 min were tested for their wash fastness, light fastness , rub fastness and perspiration fastness using AATCC test methods (AATCC technical manual 2000). The washing fastness, was evaluated by AATCC method 61(2A) using an Atlas-Launder Ometer. Fastness to light was evaluated by AATCC method 16 E using an Atlas CI 3000 + Xenon Weatherometer. The fastness to rubbing was also evaluated as per AATCC 116-1995 standards using crock meter and fastness to perspiration (AATCC Test Method 15) were evaluated using the visual ISO grey scale for both color change.

Antimicrobial activity test:

Materials

Two strains of bacteria Gram (+) bacteria as Staphylococcus aureus, Bacillus subtilis; Gram (-) bacteria as Escherichia coli, Pseudomonas aeuroginosa they were incubated at 37ºC. for 24 hours, then the diameters of the inhibition zones were measured in millimeters Standard discs of Ampicillin (Antibacterial agent),) served as positive controls for antimicrobial activity but filter discs impregnated with 10 µl of solvent (DMSO) were used as a negative control. The agar used is Meuller-Hinton agar that is rigorously tested for composition and pH. Further the depth of the agar in the plate is a factor to be considered in the disc diffusion method. This method is well documented and standard zones of inhibition have been determined for susceptible and resistant values [33].

Test Method:

The antibacterial activities of prepared dyes as well as the dyed fabrics were determined against the test bacteria by disk diffusion method on an agar plate [34]. Briefly, 1 cm diameter blended film samples were cut and put into 10 ml of nutrient agar, to which 10 µl of microbe culture was inoculated, after the solidification. The plates were incubated at 37°C for 24 hours, after which the diameter of inhibition zone were measured and recorded.

Result and Discussion

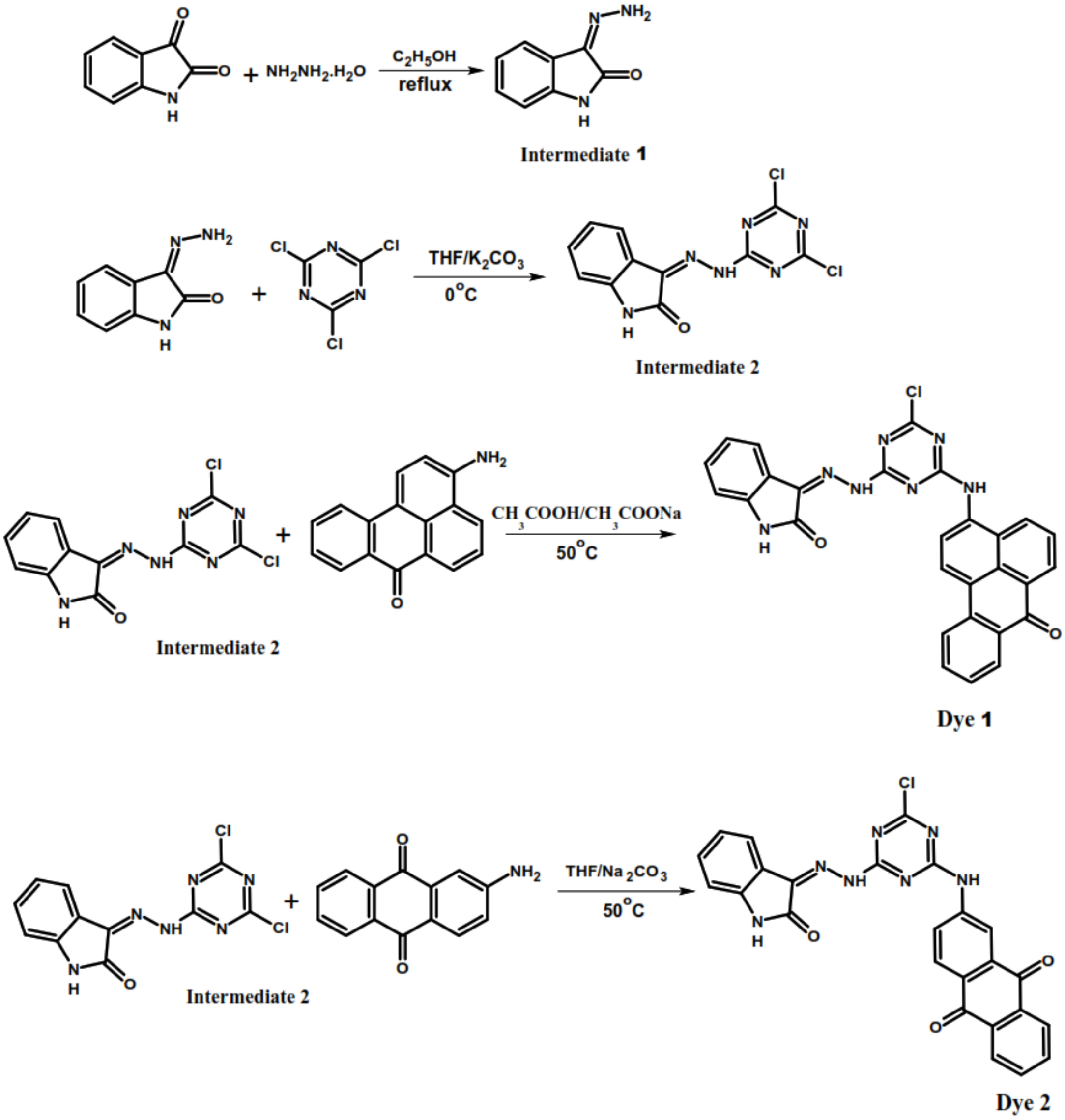
Synthesis

Starting intermediate 1 was prepared according to described method by the condensation of the isatin intermediate with hydrazine hydrate in ethanol which form a yellow crystal in very good yield [29, 30].While the dichloro triazinyle derivatives 2 were prepared by the electrophilic substitution reaction between the intermediate 1 and cyanuric chloride under controlled pH and temperature in dry THF in presence of potassium carbonate for adsorbing the acid formed during the reaction [31]. The dyes 1 and 2 were prepared by the nucleophilic substitution of second chlorine atom of compound 1 by molar equivalent of 3-aminobenzanthrone and 1-aminoanthraquinone in the presence of sodium acetate and sodium carbonate respectively at 50 °C as shown in Scheme 1. Dye 1 also can be prepared in tetrahydrofuran in presence of sodium carbonate but the reaction required more time (15h in tetrahydrofuran, 6h in acetic acid) and yield in tetrahydrofuran is lower than in acetic acid. The structure of prepared dyes was confirmed by NMR, mass spectra and elemental analysis.

Spectral properties of the prepared dyes

The spectral properties of the prepared dyes such as absorption and fluorescence spectra were measured in DMF at room temperature by preparing a solution (1×10^{-5} mol/L) of the dye concentration. Figures 1 and 2 were showing the shape and position of absorption and emission spectra of dye 1 and 2. The spectral shape of dye 1 and dye 2 showed a strong HOMO-LUMO transition band, dye 1 shows two absorption band at 415nm and 354 nm in additional to a shoulder band near 345 nm while dye 2 did not show any vibrionic band in absorption spectra as illustrated in Fig. 1, 2 which ascribe to the π-π* transition. Fluorescence quantum yield and molar absorptivity for the prepared dyes 1, 2 was calculated and summarized in Table 1. Dye **1** has higher molar absorptivity (20560) L.mol^{-1}. cm^{-1} than dye **2** (10170) L. mol^{-1}. cm^{-1}.

The absorption and emission spectra of dye 2 shows a significant bathochromic shift comparing with dye 1 as expected due to the higher conjugation and the effect of benzanthrone moiety. The fluorescence quantum yield was relatively low 0.040 and 0.023 for dye 1 and dye



Scheme 1. Rout for the preparation of dye 1 and dye 2

2 respectively. It is clear that by the nucleophilic substitution on amino group in position 2 of the started chromophore there was a significant quenching of the emission especially for dye 2.

The dyeing application on nylon and wool fabrics

Effect of pH in the dye uptake.

In this work we study the effect of the pH change on K/S value on wool and nylon 6 fabrics which were dyed with dye1 and dye2. The dyeing was carried out for 60 min, L: R (50:1), at 100 ^{0}C, 2 % (owf), and at pH (4-10) for nylon fabric and pH (3-7) for wool fabric. The K/S values for the dyed wool and nylon fabrics by dye 1 and dye 2 were recorded, in the case of wool fabric by increasing the pH from 3 to 7 the dye uptake was decreased, in the case of dye 1 the decrease in dye uptake by increasing pH was significantly observed comparing with the slightly decrease in case of dye 2 as shown in Fig. 3, this is may be ascribed to the high aggregation of the dye 1 due to its higher molecular weight which decrease of the penetration of the dye molecule to the fabrics . In case of nylon 6

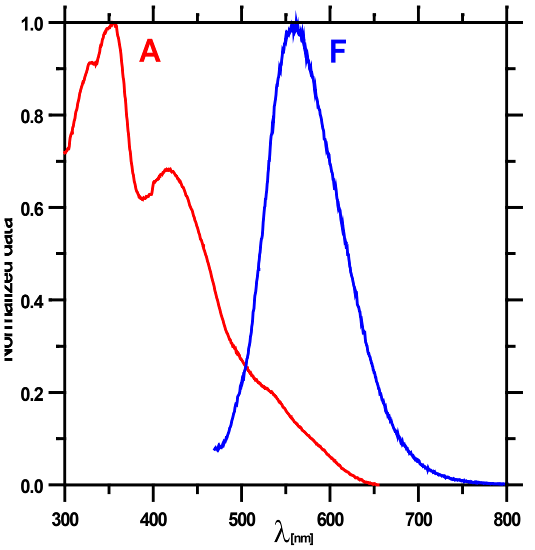


Fig. 1. Absorption (A) and fluorescence (F) spectra of dye 1 in DMF.

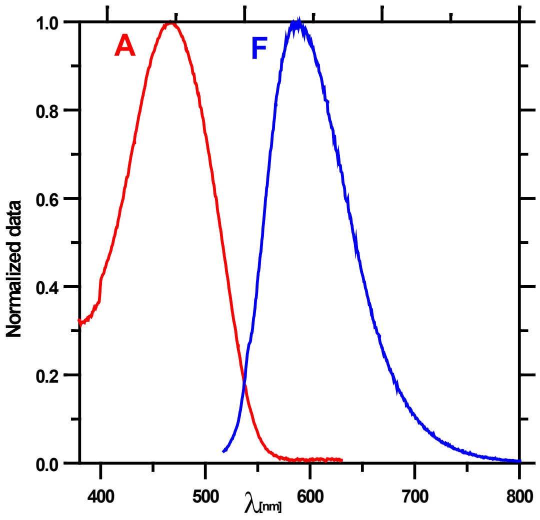


Fig. 2. Absorption (A) and fluorescence (F) spectra of dye 2 in DMF.

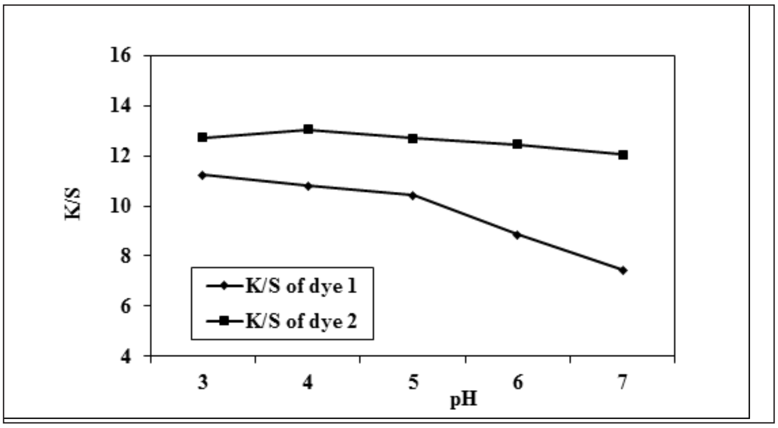


Fig. 3. Effect of pH in wool dyeing using dye 1 and dye 2

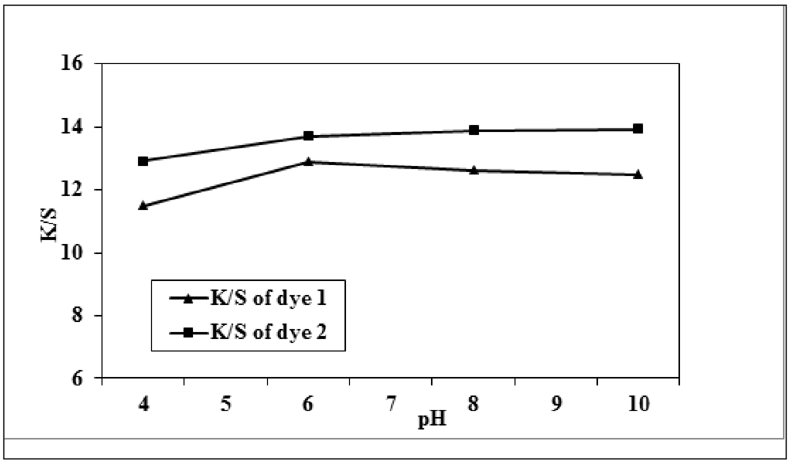


Fig. 4. Effect of pH in nylon 6 dyeing using dye 1 and dye 2

by increasing pH up to 10 the dye uptake was increased and the dyeing behavior in neutral and slightly basic showing higher buildup of the dye into the fabric as shown in Fig. 4, the explanation is due to the increase of the probability of the electrophilic substitution reaction between the chlorine atom in the dye molecule and the terminal amino group in the fabric structure at higher pH values.

Effect of dye concentration

The dyeing application of the prepared dye 1 and dye 2 on wool and nylon 6 using different concentration was shown in Fig. 5 and 6. The dyeing was continue for 60 min, at 100^{0}C, 2 % (owf), L.R (50:1), at pH5 for wool dyed with dye 1 and dye2 respectively, and at pH7 for nylon dyed with dye 1 and dye2at (1-5%) dye concentration. The dye uptake in both cases was increased by increasing the dye concentration. The using of dye 1 was showing a slightly a higher dye uptake comparing with dye 2. The results clarify that, with lower dye concentration lower value of K/S was recorded than the higher depth of shade.

Dyes exhaustion and fixation on wool and nylon fabrics

The dyeing of wool and nylon 6 fabric using the prepared dyes exhibited very good exhaustion and total fixation as well as the K/S values on the fiber at pH 5 for wool and nylon 6, From the results displayed in Fig. 7and 8, it can be seen that the extent of dye exhaustion and total fixation are extremely higher for dye 2 compared to those achieved using dye 1. As dye 1 has high molecular weight resulted in a lower affinity toward the fiber with reduced levels of exhaustion and total fixation comparing to the dye 2.

Color data

The dye uptake and color parameters L, a, b, and ΔE values of dyed wool and nylon fabric are summarized in Table 2.

The data recorded as shown in Table 2 showed that the dyed nylon and wool fabric with dye 1 and dye 2 shows L* value in the ranged of (59 to 68.10), (64.83 to 71.62) respectively from lightness or darkness, a* value ranged from (30.45 to 13.63), (23.85 to 10.94) respectively, and b* value ranged from (34.10 to 19.81), (36.68 to 25.99) orange color for dyed nylon and wool fabric.

According to the color data results in Table 2 we can conclude that, the dye uptake on the dyed nylon and wool fabrics at 2% owf and at optimum pH value with both dye derivatives (dye 1 and dye 2) not showing a significant difference. While the deepness of dye 2 showing a significant lower depth than dye 1.

Fastness properties

The fastness properties for washing, rubbing, perspiration and light of the dyed nylon and wool fabrics which dyed with dye 1 and dye 2 respectively are shown in Table 3. The results of fastness properties showed that dry crocking of dyed nylon with dye 1 give better results (4-5) than wet, while wet crocking of nylon dyed with both of dye 1 and dye 2 and wool dyed with dye 2 better than dry crocking. On the other hand washing fastness gives very good results ranged from (4-5). Also acidic and alkaline perspiration gives good fastness properties (4-5). Finally light fastness properties ranges from very good to excellent.

Antibacterial activities of the prepared dyes

The prepared dyes were tested against two strains of bacteria Gram (+) bacteria as *Staphylococcus aureus*, *Bacillus subtilis*; Gram (-) bacteria as *Escherichia coli*, *Pseudomonas aeuroginosa* using disc diffusion method and Ampicillin as anti-bacterial agent as shown in Table 4.

From the antibacterial results it was found that dye1 showing a slightly higher activity comparing with dye 2 due to the presence of amino benzanthrone moiety. The antibacterial affinity on dyed fabrics was studied and showed that in the case of both dyes the antibacterial affinity on dyed wool was always higher comparing in the case of nylon 6 as expected due to the antibacterial affinity of wool itself.

Conclusion

Two new reactive disperse dyes based on isatin derivatives were successfully prepared. The spectral behavior such as absorption and emission spectra in solution were investigated and showed a characteristic absorption and emission band at 354, 415 nm and 562 nm for benzanthrone based derivatives respectively while a maximum absorption at 467 nm, emission wavelength at 583 nm for anthraquinone based derivatives. The dyeing application on wool and nylon 6 fabrics were showing high affinity

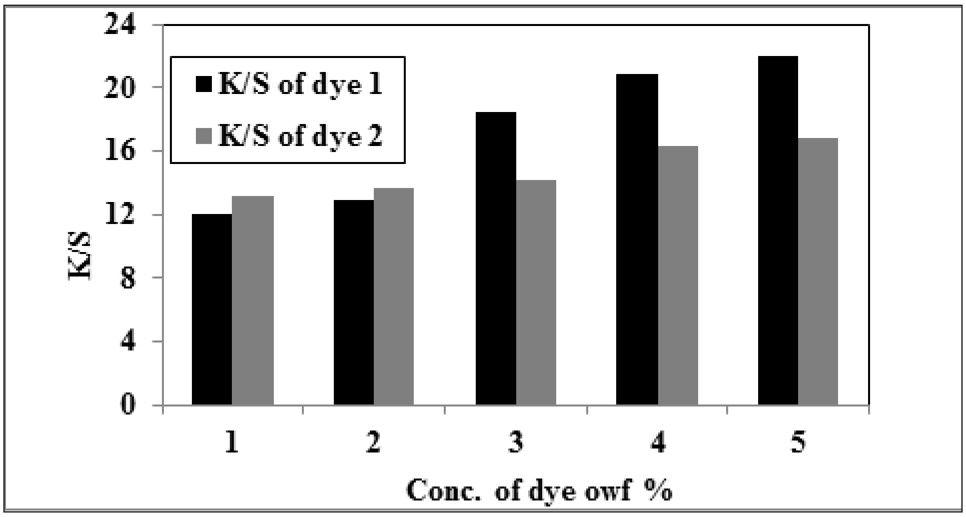


Fig. 5. Effect of dye concentration on wool dyeing

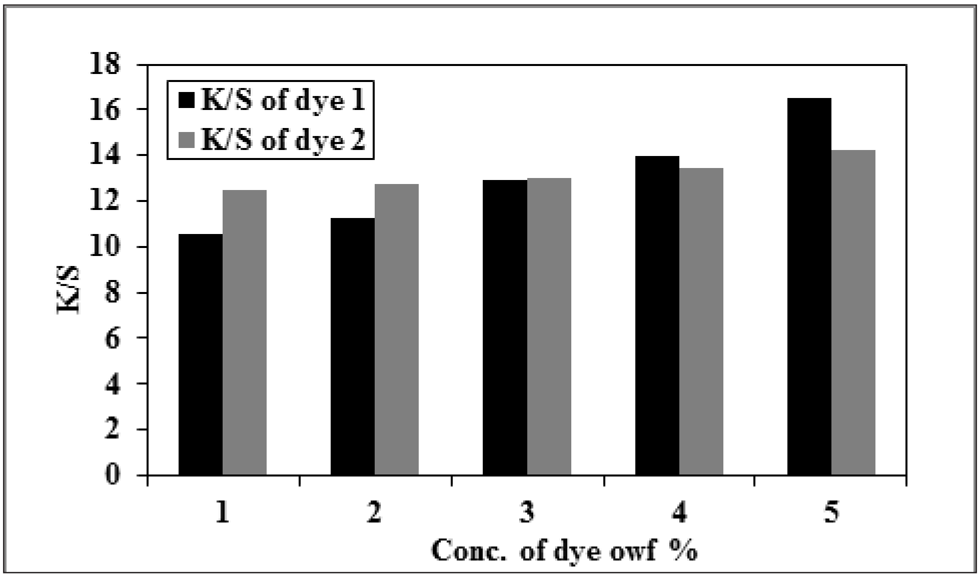


Fig. 6. Effect of dye concentration on nylon 6 dyeing

TABLE 1. Absorption (λA), fluorescence (λF) maxima (nm), fluorescence quantum yields (qF) and molar absorption coefficient (εmax) of all compounds in N,N-dimethylformamide (DMF).

Dye	Absorption λ_A(nm)	Emission λ_F(nm)	q_F	ε_{max} [L mol^{-1} cm^{-1}]
Dye 1	354, 415	562	0.040	20560
Dye 2	467	583	0.023	10170

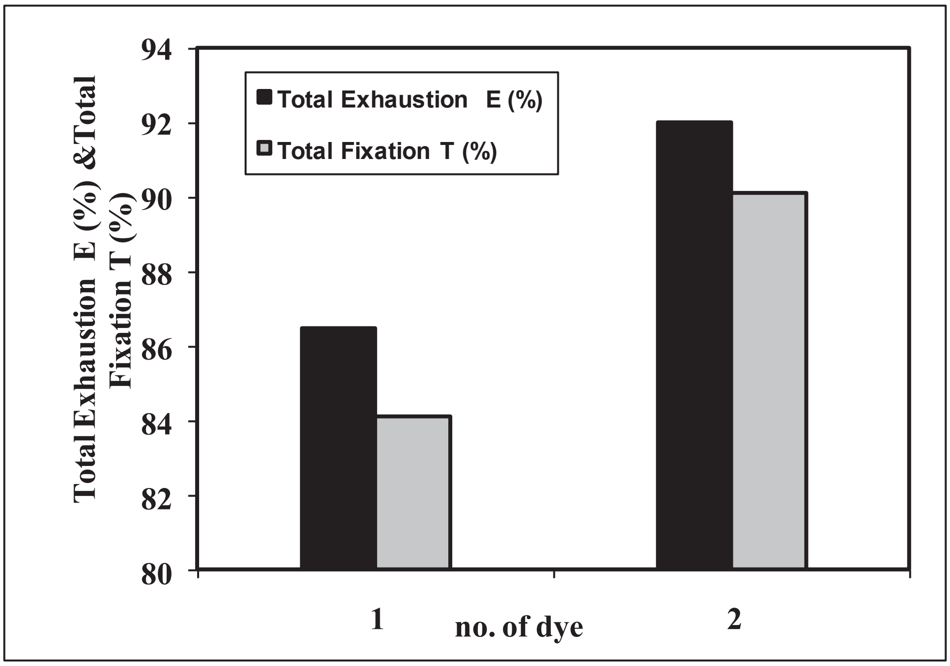


Fig. 7. Exhaustion and total fixation on wool at pH 5 for dye 1 and dye 2.

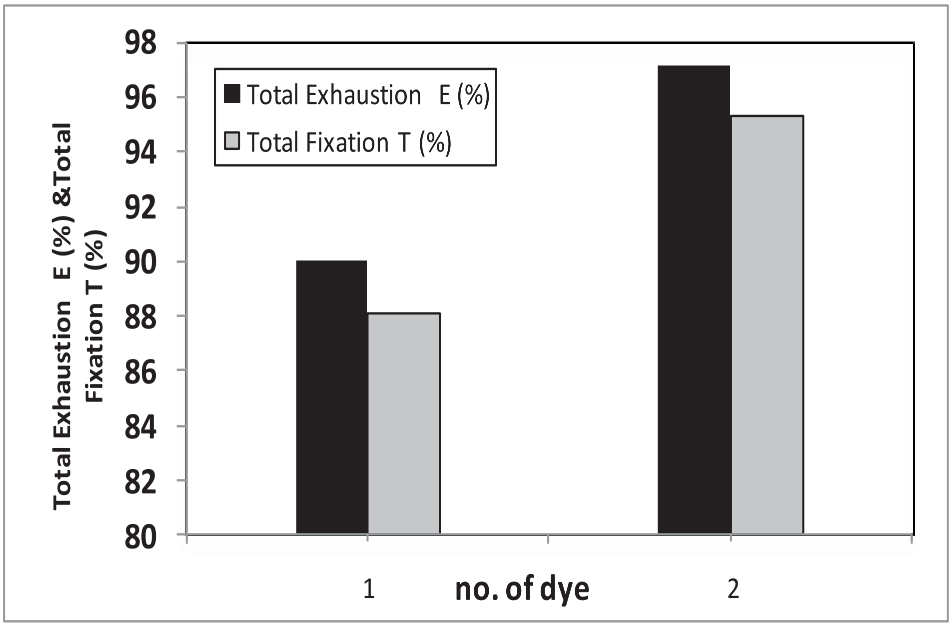


Fig. 8. Exhaustion and total fixation on nylon 6 at pH 8 for dye1 and dye 2.

TABLE 2. The color data of dyes 1,2 on nylon and wool at 2% concentration of dye

Dyes	Sample	K/S	L	a	b	ΔE
Dye 1	wool	16.89	59	30.45	34.10	53.84
	nylon	12.39	64.83	23.85	36.68	42.92
Dye 2	wool	16.29	68.10	13.63	19.81	31.73
	nylon	12.06	71.62	10.94	25.99	25.35

TABLE 3. The fastness properties of the dyed nylon and wool fabrics at optimum pH and 2% dye concentration

Dye	**Dyed sample**	**K/S**	**Fastness to rubbing**		**Wash fastness**			**Fastness to perspiration**						**light**
								Alkaline			**Acidic**			
			Dry	**Wet**	**Alt.**	**SC**	**SW**	**Alt**	**SC**	**SW**	**Alt**	**SC**	**SW**	
1	**W**	16.89	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	6
	N	12.39	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	5-6
2	**W**	16.29	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	5
	N	12.06	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	5

N, nylon, W wool, Alt alteration, SC staining to cotton, SW staining to wool

TABLE 4. The antimicrobial activity screening of the prepared compounds

Sample	**Inhibition zone diameter (mm / mg Sample) Concentration 2mg/disc**			
	G^-		**G^+**	
	Escherichia coli	***Pseudomonas aeruginosa***	***Bacillus subtilis***	***Staphylococcus aureus***
Ampicillin	25	26	26	21
Dye1	15	14	15	13
Dye2	14	13	11	12
Dye1 W/N	19/15	18/12	19/14	18/15
Dye 2 W/N	18/13	18/11	19/11	19/12

to the fabric with excellent fastness properties. The antibacterial affinity of the synthesized dyes as well as the dyed fabric against gram (+) and gram (-) bacteria showed good affinity.

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