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Material applications of novel heterocyclic disperse and mordent dyes based on 2-butyl-3-(4-hydroxybenzoyl)benzofuran

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ABSTRACT

The novel mordent and disperse heterocyclic dyes were prepared by coupling of various diazo solution of aromatic amines with 2-Butyl-3-(4-hydroxybenzoyl)benzofuran. The resultant mordent and disperse heterocyclic dyes were characterized by elemental analyses, IR and NMR spectral studies. The UV-visible spectral data have also been discussed in terms of structural property relationship. The dyeing assessment of all the mordent and disperse heterocyclic dyes was evaluated on wool and polyester textile fibers. The results of antibacterial studies of chrome pretreated fabrics revealed that the toxicity of mordented dyes against Escherichia coli, Staphylococcus aureus, Salmonella typhi, Bacillus subtilis bacteria was fairly good.

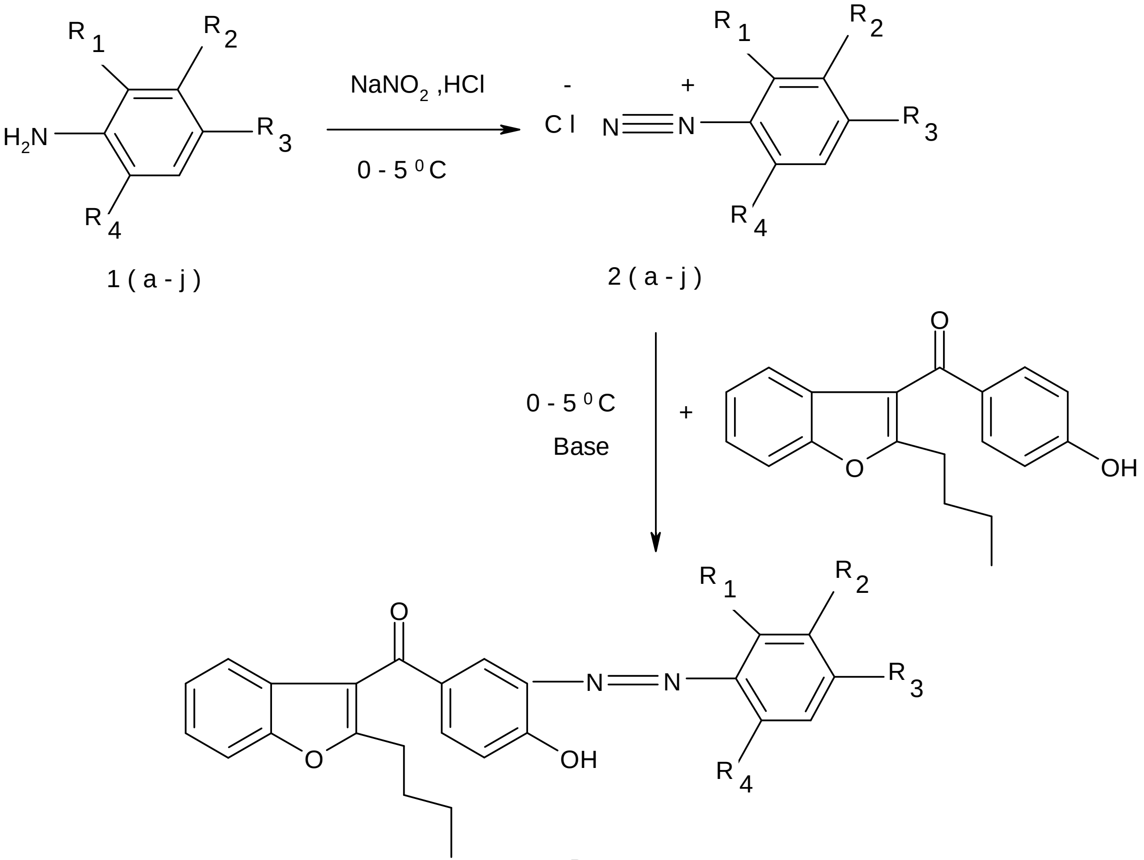
Keywords: heterocyclic dyes, UV absorber, disperse and mordent dyeing, fastness property, microbial activity.

INTRODUCTION

In the field of dyes, the heterocyclic compounds play the important role for the most of commercial dyes [1-6]. Most of the heterocyclic dyes [7] are marketed in the form of azo disperse, azo-vat, azo-acid dyes etc. All these have part of phenols and naphthols having hydroxyl group as auxochrome group. One of the hydroxy compound i.e. 2-Butyl-3-(4-hydroxybenzoyl)benzofuran having hydroxy (as auxochrome) groups and a keto (chromophore) group. This compound has wide applications as an excellent UV absorber [8- 9] which prevents the photo degradability of most of vinyl polymers [10]. The area in which the azo dye formation based on this compound has not been developed except of few patents [11-12]. The formation of dyes based on this heterocyclic compounds may yield with good hue properties. Hence, in continuation of earlier work [13], it was thought interesting to explore the field of azo dyes based on 2-Butyl-3-(4-hydroxybenzoyl)benzofuran. The present communication comprises the studies on novel mordent and disperse heterocyclic dyes based on 2-Butyl-3-(4-hydroxybenzoyl)benzofuran.

MATERIALS AND METHODS

All the chemicals used were of commercial grade and were further purified by crystallization. Melting points were determined by open capillary method and are uncorrected. The visible absorption spectra were measured on a Carl Zeiss UV/VIS Specord spectrometer, elemental analysis were carried out on Perkin elmer CHNS/O Analyzer 2400 Series II, Infrared spectra were recorded in KBr pellets on a Perkin-elmer Spectrum GX FT-IR model, NMR were recorded on Hitachi R-1500. TLC (Thin layer chromatograthy) was run on a glass plate using methanol-water-acetic acid (12:3:7) solvent system. A spot color was visualized by U.V. chamber. The equipment used for dying purpose was done by using HTHP dyeing machine (model – LL).



Dyes $_{1-7}$

Dyes No	R_1	R_2	R_3	R_4	Amines 1(a-j)
D_1	H	H	H	H	Aniline
D_2	OH	H	H	H	2- hydroxyaniline
D_3	H	Br	H	H	3 -bromoaniline
D_4	H	H	Br	H	4-bromoaniline
D_5	Br	H	H	Br	2,6-Dibromoaniline
D_6	Br	H	NO_2	Br	2,6-Dibromo-4-nitroaniline
D_7	NO_2	H	NO_2	H	2,4 -Dinitroaniline

Scheme1. Synthesis of 2-Butyl-3-(4-hydroxybenzoyl)benzofuran based dyes (D_{1-7})

Preparation of azo disperse dyes

Diazotization

Diazotization of various aromatic amines 1(a-j) was carried out by the method reported in literature [14].

Coupling procedure

The coupling of above mentioned diazotized aromatic amines was carried out in the similar manner. The general procedure adopted is given below :

2-Butyl-3-(4-hydroxybenzoyl)benzofuran 2.64 x 10^{-3} Kg (0.02 M) was dissolved in 1.6 x 10^{-5} m^3 sodium hydroxide (0.02 M) solution. The clear solution was cooled in ice-bath and diazonium solution of amine was added dropwise over 30 minutes with vigorous stirring. The pH was maintained between 7.5 to 8 by simultaneous addition of 10% w/v sodiumcarbonate solution. Stirring was continued for 2 hours, allowing the temperature to rise to ambient. The dye was then filtered off, washed with warm water and with cold water until it becomes acid free, and was dried at 323.15^0K 50^0 C in an oven. The dyes were designated as dye (D_{1-10}) reprecipitated from DMF.

Disperse dyeing method

Dyeing of polyester and wool fiber [15] is convenient at 363.15^0 – 408.15^0 K and at high pressure (165.6 K pa - 207 K pa) in the laboratory. A model glycerin-bath high-temperature beaker and HTHP-LL dyeing machine was used. For this purpose a paste of finely powdered dye (5x10^{-5} Kg) was prepared with dispersing agent dodamol (9 x 10^{-5} Kg) wetting agent Tween-80 (5x10^{-6} Kg) and water (1x10^{-6} m^3) in a ball mill. To this paste, water (9.9x10^{-5} m^3) was added with stirring and the pH was adjusted to 4-5 using acetic acid, the previously mentioned dye suspension ($1x10^{-4}$ m^3) was added to a beaker provided with a lid and a screw cap. Before closing the lid and lightening the metal cap over the beaker a wetted pattern of polyester was rolled in to the beaker, and then placed vertically on the rotatory carrier inside the tank and the clamp plate was firmly tightened. The rotatory carrier was then allowed to rotate in the glycerin-bath and the temperature was raised to 363.15^0 K at the rate of 2^0 C/min. The dyeing was continued for 1 hour under pressure. After cooling for 1 hour, the beaker was removed from the bath and washed with distilled water. The dyed pattern was thoroughly washed with hot water at 323.15^0 K and then with cold water and dried at room temperature.

Mordent dyeing method

The heterocyclic dye pattern of polyester and wool obtained as mentioned above was treated with potassium dichromate solution equal to half of the weight of dye was allowed to rolled in to the beaker and again beaker was then placed vertically on the rotatory carrier in side the tank and the dyeing was continue for 1 hour under the pressure. After cooling for 1 hour, the beaker was removed from the bath and wash with distilled water. The dyed pattern was thoroughly washed with warm water and then with cold water and air dried at room temperature.

Characteristic data of dyes

1-(2-butylbenzofuran-3-yl)-1-[4-hydroxy-3-phenyldiazenyl-phenyl]-methanone ***(D_1)***.
Calculated for $C_{25}H_{22}O_3$ N_2 : m.wt.: 398, % yield 72, m.p. : $242\text{-}243^0$ C, Rf. value: 0.80; C , 75.36%; H, 5.57% ; N, 7.03%. Found: C , 75.32%; H, 5.54% ; N, 7.01%. IR : 3460 cm^{-1}(-OH); 3077 cm^{-1}(=CH. aromatic); 1630 cm^{-1} (C=O, diaryl), 1525 cm^{-1} (N=N); 1484 cm^{-1}(C=C aromatic); 1338 cm^{-1} (C-N), 1123 cm^{-1} (C-O); 737 cm^{-1}, 586 cm^{-1}, 480 cm^{-1}. ^{1}H-NMR : 7.2-8.0 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2-hydroxyphenyl)diazenyl-phenyl]-methanone ***(D_2)***.
Calculated for $C_{25}H_{22}O_4$ N_2: m.wt. : 414, %yield : 75, m.p. : $225\text{-}226^0$ C, Rf. value: 0.73; C, 72.45%; H, 5.35%; N, 6.76%. Found: C, 72.41%; H, 5.33%; N, 6.71%. IR : 3480 cm^{-1}(-OH); 3072 cm^{-1}(=CH aromatic), 1631 cm^{-1} (C=O, diaryl), 1540 cm^{-1} (N=N), 1480 cm^{-1}(C=C aromatic), 1339 cm^{-1}(C-N), 1104 cm^{-1} (C-0), 738 cm^{-1}, 560 cm^{-1}, 470 cm^{-1}. ^{1}H-NMR : 7.15-8.04 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(3-bromophenyl)diazenyl-phenyl]-methanone ***(D_3)***.
Calculated for $C_{25}H_{21}O_3$ N_2Br: m.wt.: 477, %yield : 80, m.p. : $245\text{-}246^0$ C, Rf. value: 0.77; C, 62.90%; H, 4.43%; N, 5.87%. Found : C, 62.83%; H, 4.41%; N, 5.82%. IR : 3453 cm^{-1}(-OH); 3087 cm^{-1}(=CH aromatic); 1628 cm^{-1} (C=O, diaryl), 1528 cm^{-1} (N=N); 1490 cm^{-1}(C=C. aromatic); 1348 cm^{-1} (C-N), 1100 cm^{-1} (C-O); 740 cm^{-1}, 568 cm^{-1}, 479 cm^{-1}. ^{1}H-NMR : 7.32-8.31 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(4-bromophenyl)diazenyl-phenyl]-methanone ***(D_4)***.
Calculated for $C_{25}H_{21}O_3$ N_2Br: m.wt.: 477, %yield : 72, m.p. : $235\text{-}236^0$ C, Rf. value: 0.79; C, 62.90%; H, 4.43%; N, 5.87%. Found : C, 62.82%; H, 4.40%; N, 5.85%. IR : 3453 cm^{-1}(-OH); 3087 cm^{-1}(=CH aromatic); 1628 cm^{-1} (C=O, diaryl), 1528 cm^{-1} (N=N); 1490 cm^{-1}(C=C. aromatic); 1348 cm^{-1} (C-N), 1100 cm^{-1} (C-O); 740 cm^{-1}, 568 cm^{-1}, 479 cm^{-1}. ^{1}H-NMR : 7.32-8.31 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2,6-dibromophenyl)diazenyl-phenyl]-methanone ***(D_5)***.
Calculated for $C_{25}H_{20}O_3N_2Br_2$: m.wt. : 556, % yield : 81, m.p. : $215\text{-}216^0$C, Rf. value : 0.74 ; C, 53.98%; H, 3.62%; N, 5.04%. Found: C, 53.94%; H, 3.60%; N, 5.01%. IR : 3573 cm^{-1} (-OH); 3075 cm^{-1}(= CH aromatic); 1620 cm^{-1} (C=O, diaryl), 1534 cm^{-1} (N = N); 1484 cm^{-1} (C=C aromatic); 1463 cm^{-1} (C-N), 1343 cm^{-1} (C-O); 1100 cm^{-1}, 736 cm^{-1}, 570 cm^{-1}, 469 cm^{-1} . ^{1}H-NMR : 7.15-8.01 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2,6-dibromo-4-nitrophenyl)diazenyl-phenyl]-methanone ***(D_6)***.
Calculated for $C_{25}H_{19}O_5N_3Br_2$: m.wt. : 601, % yield: 79, m.p. : $258\text{-}259^0$ C, Rf. value : 0.72; C, 49.94%; H, 3.19%; N, 6.99%. Found: C, 49.92%; H, 3.14%; N, 6.91%. IR : 3636 cm^{-1} (-OH), 3080 cm^{-1}(=CH aromatic); 1658 cm^{-1} (C=O, diaryl), 1531 cm^{-1} (N=N); 1478 cm^{-1} (C=C aromatic); 1343 cm^{-1}(C-N), 1103 cm^{-1} (C-O); 780 cm^{-1}, 741 cm^{-1}, 584 cm^{-1}, 489 cm^{-1} ^{1}H-NMR : 7.32-8.71 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2,4-dinitrophenyl)diazenyl-phenyl]-methanone ***(D$_7$)***.
Calculated for $C_{25}H_{20}O_7N_4$: m.wt. : 488, %yield : 83, m.p.: 118-119^0 C, R.f. value : 0.76, C, 61.47%; H, 4.13%; N, 11.47%. Found : C, 61.44%; H, 4.10%; N, 11.43%. IR : 3591 cm^{-1}(-OH); 3060 cm^{-1}(=CH aromatic); 1629 cm^{-1} (C=O, diaryl), 1532 cm^{-1}. (N=N); 1470 cm^{-1} (C=C aromatic); 1323 cm^{-1} (C-N), 1104 cm^{-1} (C-O); 780 cm^{-1}, 737 cm^{-1}, 581 cm^{-1}, 470 cm^{-1}. ^{1}H-NMR : 7.32-9.12 δ (Ar-H, multiplet), 5.35 δ (Ar-OH, singlet), 1.31 to 2.40 δ ($-CH_2-$, multiplet),0.90 δ ($-CH_3$, triplet).

Fastness property

The fastness to light, sublimation and perspiration of dye pattern was assessed according to British standard: 1006-1978 and the wash fastness test according to Indian standard: IS: 765–1979. The rubbing fastness was tested by using Crock meter (Atlas) AATCC-1961, shown in Table 1 to Table 4.

Table 1. Result of disperse dyeing and various fastness properties of heterocyclic dyes onto polyester.

Dyes No.	Color shades on wool	Light fastness	Wash fastness	Perspiration fastness		Sublimation fastness	Rubbing fastness	
				Acid	Alkaline		Dry	Wet
D_1	Lemon Yellow	4	4	4	4	4	4	4
D_2	Yellowish Orange	5	4	4	4	5	4	4
D_3	Yellowish	5	5	5	4	4	4	4
D_4	Yellowish Orange	4	4	4	5	4	4	3
D_5	Reddish	5	4	4	4	5	4	4
D_6	Brown	4	5	5	5	4	4	3
D_7	chocolate	5	4	4	4	4	4	4

Table 2. Result of Mordent dyeing and various fastness properties of heterocyclic dyes onto polyester.

Dyes No.	Color shades on wool	Light fastness	Wash fastness	Perspiration fastness		Sublimation fastness	Rubbing fastness	
				Acid	Alkaline		Dry	Wet
D_1	Lemon Yellow	4	4	5	5	5	4	4
D_2	Yellowish Orange	4	5	4	4	4	4	4
D_3	Yellowish brown	5	5	5	5	5	5	4
D_4	Orange	5	5	5	5	5	4	4
D_5	Reddish Brown	5	4	4	4	4	5	4
D_6	Dark-Brown	5	5	5	5	5	4	4
D_7	chocolate brown	5	4	5	5	5	5	4

Table 3. Results of disperse dyeing and various fastness properties of heterocyclic dyes onto wool.

Dyes No.	Color shades on wool	Light fastness	Wash fastness	Perspiration fastness		Sublimation fastness	Rubbing fastness	
				Acid	Alkaline		Dry	Wet
D_1	Lemon Yellow	4	4	4	4	4	4	4
D_2	Yellowish Orange	5	4	4	4	5	4	4
D_3	Yellowish	5	5	5	4	4	4	4
D_4	Yellowish Orange	4	4	4	5	4	4	3
D_5	Reddish	5	4	4	4	5	4	4
D_6	Brown	4	5	5	5	4	4	3
D_7	chocolate	5	4	4	4	4	4	4

Table 4. Results of mordent dyeing and various fastness properties of heterocyclic dyes onto wool.

Dyes No.	Color shades on wool	Light fastness	Wash fastness	Perspiration fastness		Sublimation fastness	Rubbing fastness	
				Acid	Alkaline		Dry	Wet
D_1	Lemon Yellow	4	4	5	5	5	4	4
D_2	Yellowish Orange	4	5	4	4	4	4	4
D_3	Yellowish brown	5	5	5	5	5	5	4
D_4	Orange	5	5	5	5	5	4	4
D_5	Reddish Brown	5	4	4	4	4	5	4
D_6	Dark-Brown	5	5	5	5	5	4	4
D_7	chocolate brown	5	4	5	5	5	5	4

Determination of the percentage exhaustion and fixation

The dye bath percentage exhaustion and fixation of the dyed fabric was determined according to the known method [16] shown in Table 5 and Table 6.

Table 5 Absorption maxima, exhaustion (E) and fixation (F) of heterocyclic dyes on polyester.

Dyes No.	Absorption maxima λmax/nm in DMF		Mordent dyeing on polyester		Disperse dyeing on polyester	
		Log ε	%E	%F	%E	%F
D_1	416	4.1	81	90	71	80
D_2	424	4.2	78	89	76	78
D_3	435	4.3	78	85	80	84
D_4	439	4.4	81	92	75	78
D_5	446	4.5	76	92	75	80
D_6	452	4.6	74	91	72	78
D_7	442	4.4	82	93	75	77

Table 6 Absorption maxima, exhaustion (E) and fixation (F) of heterocyclic dyes on wool.

Dyes No.	Absorption maxima λmax/nm in DMF		Mordent dyeing on wool		Disperse dyeing on wool	
		Log ε	%E	%F	%E	%F
D_1	416	4.1	81	90	71	80
D_2	424	4.2	78	89	76	78
D_3	435	4.3	78	85	80	84
D_4	439	4.4	81	92	75	78
D_5	446	4.5	76	92	75	80
D_6	452	4.6	74	91	72	78
D_7	442	4.4	82	93	75	77

Antimicrobial activity

The *in vitro* antimicrobial activities [17-18] of the heterocyclic chrome dyes were tested against *Escherichia coli*, *Staphylococcus aureus*, *Salmonella typhi* and *Bacillus subtilis* bacteria using agar nutrient as the medium. A stock solution of 250 ppm was prepared by dissolving the compounds in 20 % DMSO solution. The antimicrobial activity was performed at a concentration 100 µg/ml, using the agar-cup method in which the well diameter was 4 mm [19]. Benzyl penicillin was used as a standard drug for antibacterial screening and solvent DMSO was used as a control. Antimicrobial activities of the heterocyclic chrome dyes are shown in Table 7.

Table 7. Antibacterial activity of heterocyclic crome dyes (100 µg/mL) of (D_{11}-D_{20})

Dye	Zone of inhibition (mm)			
No.	*E. coli*	*S. aureus*	*S. typhi*	*B. subtilis*
D_1	18	17	16	17
D_2	18	20	20	19
D_3	20	21	20	21
D_4	19	17	19	19
D_5	21	20	22	22
D_6	20	22	21	22
D_7	19	20	21	22
Benzyl penicillin	28	26	27	29

RESULTS AND DISCUSSION

Physical properties of dyes

All the dyes are obtained as amorphous powder ranging from yellow to reddish brown in colour. The purity of the dyes were checked by TLC using methanol-water-acetic acid (12:3:7) solvent system. The TLC results show that only single spot observed for each dye. The purified dyes have melting points measured by open capillary tube. The melting points were uncorrected.

The results of elemental analysis content of each dye are consistent with the predicted structure as shown in Scheme 1.

The number of azo group is almost one for each dye. The nitrogen content and number of azo group for each dye are co-related with each other. The IR spectrum of each dye comprises the important features (shown above) of aromatic, azo, hydroxyl and keto groups. The NMR spectra also giving important signals at their respective positions. All diazo compounds based on the well known structure of 2-Butyl-3-(4-hydroxybenzoyl)benzofuran and reactive site for azo coupling, the structure of azo dyes shown in Scheme 1 are confirmed.

The visible absorption spectroscopic properties of dye were recorded in DMF solution. The absorption maxima (λ_{max}) of all dyes fall into the range of 416-452 nm in DMF, as shown in Table- 5 to 6. The value of the logarithm of molar extinction coefficient (log ε) of all the dyes were in the range of 4.1 – 4.6 consistent with their high intensity of absorption.

More ever the presence of electron donating or electron attracting groups did not bring about any marked increase or decreased in λ_{max} in the visible region and that log ε remained nearly constant. However electron attracting substituent's like –Br and -NO_2 in the substituent group of the coupler increase polarizability, and will result in bathochromic shifts. This leads to decrease in energy between the highest occupied molecular orbital and lowest unoccupied molecular orbital and thus $\pi \rightarrow \pi^*$ electron transition takes place at lower frequency photon resulting in the bathochromic shift of the visible absorption band.

Dyeing properties of dyes

The mordent and dispersed heterocyclic dyes were applied at 2% depth on wool and polyester fabric respectively. Their dyeing properties are given in Table 1 to Table 4. These heterocyclic dyes gave a wide range of colour varying from yellowish brown to reddish brown shades with good levelness, brightness and depth on the fabric. The variation in the shades of the heterocyclic dyes fabric results from both the nature and position of the substituent present on the diazotized compound. The light fastness values of the heterocyclic dyes are more consistent (as shown in Table 7). The dyeing showed an excellent fastness to light, with very good to excellent fastness to washing, perspiration and sublimation and it shows poor rubbing fastness.

A remarkable degree of levelness after washing is observed. This may be attributed to good penetration and affinity of the heterocyclic dyes in to fiber structure. The most prominent features of these heterocyclic dyes is that, the dye pattern treated with Cr(III) salt solution afford excellent shining shade of dyes. This might be the chrome complex formation on fiber. The bacterial activity of chrome complexes of heterocyclic dyes was monitored against the plant pathogens. The results (Table 7) show that these heterocyclic dyes are inhibiting the bacteria about 70%. The dye pattern of chrome treated heterocyclic dyes may be affordable for human body.

CONCLUSION

Produced heterocyclic dyes have good fastness to light, sublimation and perspiration but show poor rubbing fastness properties. The nature of the substituent in the coupling components has a little influence on the visible absorption and shade of the dyeing. Comparison of above two heterocyclic dyes reveals that mordent heterocyclic dyes have good shades and fastness properties rather than heterocyclic disperse dyes due to chrome complex.

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