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### **Synthesis, characterization and application of some novel mordent and heterocyclic disperse 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.

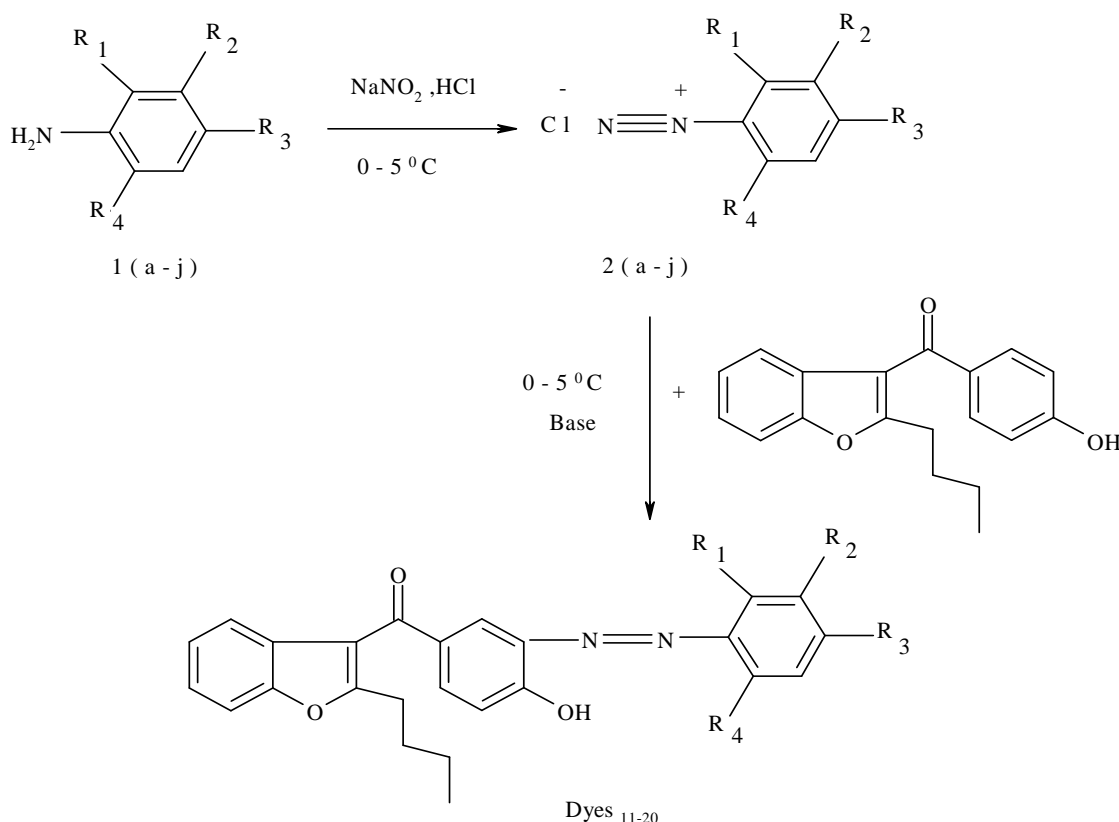
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#### **INTRODUCTION**

In the field of dyes, the heterocyclic compounds play the important role for the most of commercial dyes<sup>1, 6</sup>. Most of the heterocyclic dyes<sup>7</sup> 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<sup>8, 9</sup> which prevents the photo degradability of most of vinyl polymers<sup>10</sup>. The area in which the azo dye formation based on this compound has not been developed except of few patents<sup>11, 12</sup>. The formation of dyes based on this heterocyclic compounds may yield with good hue properties. Hence, 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 chromatography) 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 dyeing purpose was done by using HTHP dyeing machine (model – LL).



Dyes No	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	R <sub>4</sub>	Amines 1(a-j)
D <sub>11</sub>	NO <sub>2</sub>	H	H	H	2- Nitroaniline
D <sub>12</sub>	H	H	NO <sub>2</sub>	H	4- Nitroaniline
D <sub>13</sub>	H	NO <sub>2</sub>	H	H	3 -Nitroaniline
D <sub>14</sub>	Cl	H	NO <sub>2</sub>	Cl	2,6 -Dichloro-4-nitroaniline
D <sub>15</sub>	CH <sub>3</sub>	H	CH <sub>3</sub>	H	2,4-Dimethylaniline
D <sub>16</sub>	H	Cl	H	H	3-Chloroaniline
D <sub>17</sub>	H	H	Cl	H	4-Chloroaniline
D <sub>18</sub>	H	H	OH	H	4-hydroxyaniline
D <sub>19</sub>	H	H	CH <sub>3</sub>	H	4-Methylaniline
D <sub>20</sub>	CH <sub>3</sub>	H	H	H	2-Methylaniline

Scheme 1. Synthesis of 2-Butyl-3-(4-hydroxybenzoyl)benzofuran based dyes (D<sub>11-20</sub>)

## Preparation of azo disperse dyes

## Diazotization

Diazotization of various aromatic amines 1(a-j) was carried out by the method reported in literature<sup>13</sup>.

### 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  $2.64 \times 10^{-3}$  Kg (0.02 M) was dissolved in  $1.6 \times 10^{-5}$  m<sup>3</sup> 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 sodium carbonate 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  $323.15^{\circ}\text{K}$   $50^{\circ}\text{C}$  in an oven. The dyes were designated as dye (D<sub>1-10</sub>) reprecipitated from DMF.

### Disperse dyeing method

Dyeing of polyester fiber<sup>14</sup> is convenient at  $363.15^{\circ}\text{K}$  –  $408.15^{\circ}\text{K}$  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 ( $5 \times 10^{-5}$  Kg) was prepared with dispersing agent dodamol ( $9 \times 10^{-5}$  Kg) wetting agent Tween-80 ( $5 \times 10^{-6}$  Kg) and water ( $1 \times 10^{-6}$  m<sup>3</sup>) in a ball mill. To this paste, water ( $9.9 \times 10^{-5}$  m<sup>3</sup>) was added with stirring and the pH was adjusted to 4.5 - 5 using acetic acid, the previously mentioned dye suspension ( $1 \times 10^{-4}$  m<sup>3</sup>) 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  $363.15^{\circ}\text{K}$  at the rate of  $2^{\circ}\text{C}/\text{min}$ . 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  $323.15^{\circ}\text{K}$  and then with cold water and dried at room temperature.

### Mordent dyeing method

The heterocyclic dye pattern of 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-(2-nitrophenyl)diazenyl-phenyl]-methanone (D<sub>11</sub>)*.

Calculated for C<sub>25</sub>H<sub>21</sub>N<sub>3</sub>O<sub>5</sub> : m.wt.: 443, % yield 78, m.p. :  $232-233^{\circ}\text{C}$ , Rf. value: 0.84; C , 67.71%; H, 4.77% ; N, 9.48%. Found: C, 67.65%; H, 4.73%; N, 9.39%. IR :  $3460\text{ cm}^{-1}$ (-OH);  $3077\text{ cm}^{-1}$ (=CH. aromatic);  $1630\text{ cm}^{-1}$  (C=O, diaryl),  $1525\text{ cm}^{-1}$  (N=N);  $1484\text{ cm}^{-1}$ (C=C aromatic);  $1338\text{ cm}^{-1}$  (C-N),  $1123\text{ cm}^{-1}$  (C-O);  $737\text{ cm}^{-1}$ ,  $586\text{ cm}^{-1}$ ,  $480\text{ cm}^{-1}$ . <sup>1</sup>H-NMR : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(4-nitrophenyl)diazenyl-phenyl]-methanone (D<sub>12</sub>)*.

Calculated for C<sub>25</sub>H<sub>21</sub>N<sub>3</sub>O<sub>5</sub>: m.wt.: 443, % yield : 79, m.p. :  $215-216^{\circ}\text{C}$ , Rf. value: 0.87; C, 67.71%; H, 4.77%; N, 9.48%. Found : C, 67.65%; H, 4.72%; N, 9.41%. IR :  $3453\text{ cm}^{-1}$ (-OH);  $3087\text{ cm}^{-1}$ (=CH aromatic);  $1628\text{ cm}^{-1}$  (C=O, diaryl),  $1528\text{ cm}^{-1}$  (N=N);  $1490\text{ cm}^{-1}$ (C=C.

aromatic); 1348  $\text{cm}^{-1}$  (C-N), 1100  $\text{cm}^{-1}$  (C-O); 740  $\text{cm}^{-1}$ , 568  $\text{cm}^{-1}$ , 479  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(3-nitrophenyl)diazanyl-phenyl]-methanone (D<sub>13</sub>).*

Calculated for C<sub>25</sub>H<sub>21</sub>N<sub>3</sub>O<sub>5</sub> : m.wt. : 443, % yield : 79, m.p. : 215-216<sup>0</sup> C, Rf. value: 0.79; C, 67.71%; H, 4.77%; N, 9.48%. Found: C, 67.69%; H, 4.75%; N, 9.43%. IR : 3480  $\text{cm}^{-1}$  (-OH); 3072  $\text{cm}^{-1}$  (=CH aromatic), 1631  $\text{cm}^{-1}$  (C=O, diaryl), 1540  $\text{cm}^{-1}$  (N=N), 1480  $\text{cm}^{-1}$  (C=C aromatic), 1339  $\text{cm}^{-1}$  (C-N), 1104  $\text{cm}^{-1}$  (C-O), 738  $\text{cm}^{-1}$ , 560  $\text{cm}^{-1}$ , 470  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2,6-dichloro-4-nitrophenyl)diazanyl-phenyl]-methanone (D<sub>14</sub>).*

Calculated for C<sub>25</sub>H<sub>19</sub>O<sub>5</sub>N<sub>3</sub>Cl<sub>2</sub> : m.wt. : 512, % yield: 77, m.p. : 258-299<sup>0</sup> C, Rf. value : 0.82; C, 58.61%; H, 3.74%; N, 8.20%. Found: C, 58.58%; H, 3.71%; N, 8.18%. IR : 3636  $\text{cm}^{-1}$  (-OH), 3080  $\text{cm}^{-1}$  (=CH aromatic); 1658  $\text{cm}^{-1}$  (C=O, diaryl), 1531  $\text{cm}^{-1}$  (N=N); 1478  $\text{cm}^{-1}$  (C=C aromatic); 1343  $\text{cm}^{-1}$  (C-N), 1103  $\text{cm}^{-1}$  (C-O); 780  $\text{cm}^{-1}$ , 741  $\text{cm}^{-1}$ , 584  $\text{cm}^{-1}$ , 489  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2,4-dimethylphenyl)diazanyl-phenyl]-methanone (D<sub>15</sub>).*

Calculated for C<sub>27</sub>H<sub>26</sub>O<sub>3</sub>N<sub>2</sub> : m.wt. : 427, % yield : 83, m.p. : 205-206<sup>0</sup> C, Rf. value : 0.88 ; C, 76.03%; H, 6.14%; N, 6.57%. Found: C, 76.0%; H, 6.10%; N, 6.52%. IR : 3573  $\text{cm}^{-1}$  (-OH); 3075  $\text{cm}^{-1}$  (= CH aromatic); 1620  $\text{cm}^{-1}$  (C=O, diaryl), 1534  $\text{cm}^{-1}$  (N = N); 1484  $\text{cm}^{-1}$  (C=C aromatic); 1463  $\text{cm}^{-1}$  (C-N), 1343  $\text{cm}^{-1}$  (C-O); 1100  $\text{cm}^{-1}$ , 736  $\text{cm}^{-1}$ , 570  $\text{cm}^{-1}$ , 469  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet). 2.34  $\delta$  ( -CH<sub>3</sub>, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet). 2.34  $\delta$  (-CH<sub>3</sub>, singlet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(3-chlorophenyl)diazanyl-phenyl]-methanone (D<sub>16</sub>).*

Calculated for C<sub>25</sub>H<sub>21</sub>O<sub>3</sub>N<sub>2</sub>Cl : m.wt. : 433, % yield : 87, m.p. : 165-166<sup>0</sup> C, Rf. value : 0.92, C, 69.36%; H, 4.89%; N, 6.47%. Found C, 69.34%; H, 4.83%; N, 6.41%. IR : 3430  $\text{cm}^{-1}$  (-OH); 3066  $\text{cm}^{-1}$  (=CH aromatic); 1630  $\text{cm}^{-1}$  (C=O, diaryl), 1584  $\text{cm}^{-1}$  (N=N); 1482  $\text{cm}^{-1}$  (C=C aromatic); 1350  $\text{cm}^{-1}$  (C-N), 1104  $\text{cm}^{-1}$  (C-O); 786  $\text{cm}^{-1}$ , 740  $\text{cm}^{-1}$ , 586  $\text{cm}^{-1}$ , 489  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(4-chlorophenyl)diazanyl-phenyl]-methanone (D<sub>17</sub>).*

Calculated for C<sub>25</sub>H<sub>21</sub>O<sub>3</sub>N<sub>2</sub>Cl : m.wt. : 433, % yield : 89, m.p.: 115-116<sup>0</sup> C, Rf. value : 0.86, C, 69.36%; H, 4.89%; N, 6.47%. Found : C, 69.30%; H, 4.80%; N, 6.43%. IR : 3591  $\text{cm}^{-1}$  (-OH); 3060  $\text{cm}^{-1}$  (=CH aromatic); 1629  $\text{cm}^{-1}$  (C=O, diaryl), 1532  $\text{cm}^{-1}$ . (N=N); 1470  $\text{cm}^{-1}$  (C=C aromatic); 1323  $\text{cm}^{-1}$  (C-N), 1104  $\text{cm}^{-1}$  (C-O); 780  $\text{cm}^{-1}$ , 737  $\text{cm}^{-1}$ , 581  $\text{cm}^{-1}$ , 470  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  ( -CH<sub>2</sub>-, multiplet), 0.90  $\delta$  ( -CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(4-hydroxyphenyl)diazanyl-phenyl]-methanone (D<sub>18</sub>).*

Calculated for C<sub>25</sub>H<sub>22</sub>O<sub>4</sub>N<sub>2</sub> : m.wt. : 414, % yield : 84, m.p.: 137-138<sup>0</sup> C, Rf. value : 0.78; C, 72.45%; H, 5.35%; N, 6.76%. Found: C, 72.42%; H, 5.31%; N, 6.74%. IR : 3580  $\text{cm}^{-1}$  (-OH); 3056  $\text{cm}^{-1}$  (=CH aromatic); 1635  $\text{cm}^{-1}$  (C=O, diaryl), 1523  $\text{cm}^{-1}$  (N=N); 1475  $\text{cm}^{-1}$  (C=C aromatic); 1327  $\text{cm}^{-1}$  (C-N), 1101  $\text{cm}^{-1}$  (C-O); 742  $\text{cm}^{-1}$ , 578  $\text{cm}^{-1}$ , 470  $\text{cm}^{-1}$ .  $^1\text{H-NMR}$  : 7.3-8.4  $\delta$

(Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet), 1.31 to 2.40  $\delta$  (-CH<sub>2</sub>-, multiplet), 0.90  $\delta$  (-CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(4-methylphenyl)diazenyl-phenyl]-methanone (D<sub>19</sub>).*

Calculated for C<sub>26</sub>H<sub>24</sub>O<sub>3</sub>N<sub>2</sub> : m.wt.: 412, % yield : 80, m.p. : 104-105<sup>0</sup> C , Rf. value : 0.87 ; C, 75.71 %; H, 5.86 %; N, 6.79%. Found: C, 75.68 %; H, 5.82 %; N, 6.74%. IR : 3630 cm<sup>-1</sup>(-OH); 3024 cm<sup>-1</sup> (=CH aromatic); 1626 cm<sup>-1</sup> (C = O, diaryl), 1527 cm<sup>-1</sup> (N=N); 1468 cm<sup>-1</sup> (C=C aromatic); 1446 cm<sup>-1</sup> (CH<sub>3</sub>), 1351 cm<sup>-1</sup> (C-N); 1111 cm<sup>-1</sup> (C-O); 744 cm<sup>-1</sup>, 580 cm<sup>-1</sup>, 473 cm<sup>-1</sup>. <sup>1</sup>H-NMR : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet). 2.34  $\delta$  (-CH<sub>3</sub>, singlet), 1.31 to 2.40  $\delta$  (-CH<sub>2</sub>-, multiplet) 0.90  $\delta$  (-CH<sub>3</sub>, triplet).

*1-[2-Butyl-benzofuran-3-yl]-1-[4-hydroxy-3-(2-methylphenyl)diazenyl-phenyl]-methanone (D<sub>20</sub>).*

Calculated for C<sub>26</sub>H<sub>24</sub>O<sub>3</sub>N<sub>2</sub> : m.wt.: 412, % yield : 79, m.p. : 116-117<sup>0</sup> C , Rf. value : 0.81; C, 75.71 %; H, 5.86 %; N, 6.79%. Found: C, 75.66 %; H, 5.81 %; N, 6.73%. IR : 3503 cm<sup>-1</sup> (-OH); 3053 cm<sup>-1</sup> (=CH aromatic); 1630 cm<sup>-1</sup> (C=O, diaryl), 1535 cm<sup>-1</sup> (N=N); 1478 cm<sup>-1</sup> (C=C aromatic); 1450 cm<sup>-1</sup> (CH<sub>3</sub>), 1320 cm<sup>-1</sup> (C-N); 1109 cm<sup>-1</sup> (C-O); 747 cm<sup>-1</sup>, 579 cm<sup>-1</sup>, 468 cm<sup>-1</sup>. <sup>1</sup>H-NMR : 7.3-8.4  $\delta$  (Ar-H, multiplet), 5.35  $\delta$  (Ar-OH, singlet). 2.34  $\delta$  (-CH<sub>3</sub>, singlet), 1.31 to 2.40  $\delta$  (-CH<sub>2</sub>-, multiplet) 0.90  $\delta$  (-CH<sub>3</sub>, triplet).

**Table 1. Result of 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 <sub>11</sub>	Dark-brown	5	5	5	5	5	4	4
D <sub>12</sub>	Yellowish brown	5	4-5	4-5	5	5	4	4
D <sub>13</sub>	Reddish Brown	5	5	5	5	5	5	4
D <sub>14</sub>	Dark-Brown	4-5	5	4-5	5	4-5	4	3-4
D <sub>15</sub>	Green	5	4-5	5	5	5	5	4
D <sub>16</sub>	Yellowish Orange	5	5	5	5	5	4-5	3-4
D <sub>17</sub>	Yellowish Orange	5	4	5	5	5	5	4
D <sub>18</sub>	chocolate brown	5	4-5	4-5	4	4-5	4-5	4
D <sub>19</sub>	Lemon Yellow	5	5	5	5	5	5	3-4
D <sub>20</sub>	Yellowish Orange	5	5	4-5	5	5	5	4

**Table 2. Results of mordent heterocyclic 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 <sub>11</sub>	Dark-brown	4-5	4-5	4	4-5	5	4-5	3-4
D <sub>12</sub>	Yellowish brown	4-5	4	4-5	4-5	4-5	4	3-4
D <sub>13</sub>	Reddish Brown	4-5	4-5	4-5	5	5	4-5	4
D <sub>14</sub>	Dark-Brown	4-5	4	4-5	5	4-5	4	3-4
D <sub>15</sub>	Green	4-5	4-5	4	4-5	4	4-5	3-4
D <sub>16</sub>	Yellowish Orange	4-5	4-5	4-5	4-5	5	4-5	3-4
D <sub>17</sub>	Yellowish Orange	4	4-5	4-5	4-5	5	5	3-4
D <sub>18</sub>	chocolate brown	4	4-5	4-5	4-5	4-5	3-4	3
D <sub>19</sub>	Lemon Yellow	4-5	4-5	4-5	5	5	4-5	3-4
D <sub>20</sub>	Yellowish Orange	4-5	4-5	4-5	4-5	5	4-5	3-4

**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 and Table 2.

**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<sup>15</sup> shown in Table 3.

**Table 3 Absorption maxima, exhaustion (E) and fixation (F) of heterocyclic dyes on polyester / wool.**

Dyes No.	Absorption maxima $\lambda_{\max}/\text{nm}$ in DMF	Log $\epsilon$	Mordent dyeing on wool		Disperse dyeing on polyester	
			%E	%F	%E	%F
D <sub>11</sub>	430	4.3	80	93	71	90
D <sub>12</sub>	427	4.2	74	91	76	87
D <sub>13</sub>	431	4.3	78	93	80	92
D <sub>14</sub>	455	4.5	81	96	75	84
D <sub>15</sub>	436	4.4	78	92	75	89
D <sub>16</sub>	440	4.4	74	91	72	91
D <sub>17</sub>	442	4.3	86	92	75	90
D <sub>18</sub>	410	4.1	80	92	71	83
D <sub>19</sub>	432	4.3	77	91	72	90
D <sub>20</sub>	424	4.3	73	90	78	91

**Antimicrobial activity**

The *in vitro* antimicrobial activities<sup>16, 17</sup> 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  $\mu\text{g}/\text{ml}$ , using the agar-cup method in which the well diameter was 4 mm.<sup>18</sup> 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 4.

**Table 4. Antibacterial activity of heterocyclic chrome dyes (100  $\mu\text{g}/\text{mL}$ ) of (D<sub>11</sub>-D<sub>20</sub>)**

Dye No.	Zone of inhibition ( mm)			
	<i>E. coli</i>	<i>S. aureus</i>	<i>S. typhi</i>	<i>B. subtilis</i>
D <sub>11</sub>	20	19	18	21
D <sub>12</sub>	18	18	16	20
D <sub>13</sub>	23	21	20	24
D <sub>14</sub>	24	23	22	26
D <sub>15</sub>	14	14	13	16
D <sub>16</sub>	19	17	20	21
D <sub>17</sub>	21	20	19	22
D <sub>18</sub>	14	17	19	19
D <sub>19</sub>	12	15	17	14
D <sub>20</sub>	13	17	18	16
<b>Benzyl penicillin</b>	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 point's 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 comprise 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 ( $\lambda_{\max}$ ) of all dyes fall into the range of 410-455 nm in DMF, as shown in Table- I. The value of the logarithm of molar extinction coefficient ( $\log \epsilon$ ) of all the dyes were in the range of 4.1 – 4.5 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  $\lambda_{\max}$  in the visible region and that  $\log \epsilon$  remained nearly constant. However electron attracting substituents like -Cl and -NO<sub>2</sub> 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 mordant and dispersed heterocyclic dyes were applied at 2% depth on wool and polyester fabric respectively. Their dyeing properties are given in Table II and Table III. 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 result 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 III). 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 IV) 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 than heterocyclic disperse dyes.

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