Research Article

Disperse Dyes Based on Thiazole, Their Dyeing Application on Polyester Fiber and Their Antimicrobial Activity

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Various diazotized aryl amines were coupled with *N*-(4-nitrophenyl)-2-[(4-phenyl-1,3-thiazol-2-yl)amino]acetamide to give the corresponding various azo disperse dyes (D_1 - D_{13}). These dyes were applied to polyester fiber by HTHP method and their fastness properties were evaluated. Dyes were characterized by IR, elemental analysis, and NMR spectral studies. These dyes showed very good antibacterial and antifungal activities.

1. Introduction

Disperse dyes are organic colorants with less water solubility and are applied in colloidal dispersions to hydrophobic textile fibers to produce desired color. With increasing use of polyester fibers and their blends, there has been significant increase in the development of disperse dyes because over 90% of disperse dyes' usage is for the dyeing of polyester and its blends and with significant increase in the world production of polyester fibers compared to other fibers. Many diazo components have been used in the production of disperse dyes in recent time [1–6]. Derivatives of 2-aminothiazole [7–9] have been used as heterocyclic components since long for different disperse dyes. It was our main objective to synthesize [10–13] the disperse dyes consisting thiazole with azo substituent which further utilized to dye some hydrophobic fibers, to characterize, to evaluate their fastness properties. The dyes were screened for their antimicrobial properties because a large number of natural products and drugs comprises of this heterocyclic moiety [14–16].

2. Experimental

2.1. Materials and Methods. Melting points were determined in open capillary tubes and are uncorrected. The purity of dyes was determined by thin-layer chromatography (TLC) using silica gel-G-coated Al-plates. The visible absorption

spectra were measured using Shimadzu UV-160 PC Spectrophotometer. Infrared spectra were recorded on a Shimadzu FT-IR 8400S model using KBr pellets. ^1H NMR spectra were recorded on a Varian 400 MHz Spectrophotometer using DMSO solvent and TMS as internal reference (chemical shifts in δ , ppm). Elemental analysis was carried out on Perkin Elmer (USA) 2400 Series instrument. The fastness to light, wash, and sublimation was assessed in accordance with ISO 105. A convenient laboratory method was used for dyeing polyester to employ high temperature (130°C) and high pressure (24–30 psi.). The dye bath exhaustion (%E) of the dyed fiber was determined according to the method. The synthesized dyes were screened for their antimicrobial activity using the Kirby-Bauer method. All the compounds were screened for their in vitro antimicrobial activity against bacterial strains such as *Escherichia coli*, *Pseudomonas aeruginosa*, *Staphylococcus aureus*, and fungi *Candida albicans* at 40 $\mu\text{g/mL}$ concentration.

2.2. Preparation of *N*-(4-Nitrophenyl)-2-[(4-Phenyl-1, 3-Thiazol-2-yl)Amino]Acetamide (2). In 250 mL R.B.F., 4-nitroaniline (2.76 gm, 0.02 mole) in dry benzene (60 mL) was cooled to $0-5^\circ\text{C}$ and 2-3 drops of TEA were added. Chloroacetyl chloride (2.26 mL, 0.02 mole) in dry benzene (20 mL) was slowly added to RBF with vigorous stirring then the reaction mixture was refluxed for 3 hours. Excess of solvent was removed in vacuum and the residue stirred with water

TABLE 1: Yield, λ_{max} , melting points, and nitrogen analysis of disperse dyes.

Dye number	R	Yield (%)	λ_{max} (nm)	M.P. °C	Nitrogen (%)	
					Found	Calcd.
D ₁	H	81	456	148	18.35	18.33
D ₂	4-NO ₂	72	427	153	19.55	19.47
D ₃	3-NO ₂	78	472	142	19.51	19.47
D ₄	2-CN,4-NO ₂	58	412	157	21.0	21.2
D ₅	2-OH	70	429	137	17.50	17.71
D ₆	3-OH	69	432	144	17.66	17.71
D ₇	4-OH	73	414	131	17.64	17.71
D ₈	4-CH ₃	77	419	141	17.86	17.79
D ₉	3-CH ₃	72	458	137	17.77	17.79
D ₁₀	4-Cl	68	438	146	16.85	17.05
D ₁₁	2,4-di NO ₂	60	425	164	20.05	20.43
D ₁₂	2,4-di NO ₂ , 6-Cl	62	484	178	19.28	19.22
D ₁₃	2,6-di Br,4-NO ₂	65	421	183	14.72	14.83

FIGURE 1: Reaction scheme.

(50 mL) and washed with 5% NaHCO₃ and subsequently with water. The crude product was dried and crystallized from ethanol; it yielded pale yellow solid 2-chloro-N-(4-nitrophenyl)acetamide (1). Yield: 72%, M.P.: 116°C. IR (KBr, cm⁻¹): 3339 (N-H str.), 3012 (C-H str.), 1681 (C=O str.), 1514 (NO₂ str.), 741 (C-Cl); ¹H NMR (399.76 MHz, DMSO) δ , ppm: 3.30 (s, 2H, -COCH₂), 4.43 (s, 1H, -NH), 6.81–7.65 (m, 4H, Ar-H).

2-chloro-N-(4-nitrophenyl)acetamide (1) (4.28 gm 0.02 mole) and 2-amino-4-phenylthiazole (3.52 gm 0.02 mole) in 20 mL dry benzene were refluxed for 4 hours. Benzene was removed in vacuo and crude product n-(4-nitrophenyl)-2-[(4-phenyl-1,3-thiazol-2-yl)amino] acetamide (2) was dried

and recrystallized from ethanol. Yield: 68%, M.P.: 119°C. C-a: IR (KBr, cm⁻¹) 3328 (N-H str.), 2993 (C-H str. aromatic), 1685 (C=O str.), 1512 (C-S-C str.), 776 (C-S str. in thiazole); ¹H NMR (399.76 MHz, DMSO), δ , ppm: 3.32 (s, 2H, -COCH₂), 4.41 (s, 1H, -NH attached with thiazole ring), 6.78 (s, 1H, -NH attached with aromatic ring), 6.13 (s, 1H, thiazole ring), 7.00–8.10 (m, 9H, Ar-H).

2.3. Diazotization and Coupling Reaction. The different aryl amines (0.01 mole) dissolved in HCl (6 mL, 50%) were cooled to 0–5°C. A solution of sodium nitrite (0.01 mole, 0.69 gm) in water (4 mL) previously cooled to 0°C was added dropwise

TABLE 2: Shade, fastness properties, K/S value, R value, and % exhaustion of the dyes.

Dye number	Shade on polyester	Fastness to		Sublimation at 180°C		K/S value	R value	% exhaustion
		Light	Washing	Staining of cotton	Staining of polyester			
D ₁	Yellowish brown	4	5	4	4	5.07	8.29	70.42
D ₂	Maroon	5	5	5	4-5	6.85	6.4	76.30
D ₃	Orange	3-4	4	4-5	4-5	11.53	4.0	79.90
D ₄	Light yellow	4-5	5	4	4	2.97	12.78	74.32
D ₅	Light brown	4-5	4	5	5	2.17	16.19	81.75
D ₆	Reddish brown	5	5	5	4-5	3.96	10.19	71.60
D ₇	Dull yellow	5	5	5	4	3.36	11.61	77.00
D ₈	Reddish yellow	4	5	4	4	8.84	5.10	71.72
D ₉	Yellowish orange	4	3-4	5	5	11.38	4.05	73.00
D ₁₀	Yellow	4-5	5	3-4	3	8.36	5.36	80.00
D ₁₁	Light yellow	5	5	5	4	3.64	10.9	66.95
D ₁₂	Reddish orange	5	4	5	5	6.48	6.71	72.5
D ₁₃	Cream	4-5	5	4-5	4	5.40	7.86	70.42

TABLE 3: Antimicrobial screening results of disperse dyes.

Dye compound	R	Zone of inhibition (mm)			
		Antibacterial activity			Antifungal activity
		<i>E. coli</i>	<i>P. aeruginosa</i>	<i>S. aureus</i>	<i>C. albicans</i>
D ₁	H	10	11	17	12
D ₂	4-NO ₂	11	13	16	13
D ₃	3-NO ₂	10	14	18	15
D ₄	2-CN,4-NO ₂	10	12	18	14
D ₅	2-OH	12	10	17	17
D ₆	3-OH	12	10	20	13
D ₇	4-OH	11	15	19	14
D ₈	4-CH ₃	10	10	15	11
D ₉	3-CH ₃	10	10	14	10
D ₁₀	4-Cl	13	12	19	13
D ₁₁	2,4-di NO ₂	11	13	20	12
D ₁₂	2,4-di NO ₂ , 6-Cl	12	11	21	11
D ₁₃	2,6-di Br,4-NO ₂	10	11	18	14
Metronidazole		26	23	29	—
Fluconazole		—	—	—	19

maintaining the temperature at 0–5°C; stirring was continued for an hour, with positive test of nitrous acid on starch iodide paper. Excess of nitrous acid was destroyed by adding required amount of sulphamic acid. The resulting solution was used for coupling reaction (Figure 1).

N-(4-nitrophenyl)-2-[(4-phenyl-1, 3-thiazol-2-yl) amino]acetamide (2) (0.01 mole) was dissolved in glacial acetic acid (30 mL) and cooled below 5°C. To this well stirred solution, above mentioned diazonium chloride solution was added drop wise maintaining the pH 7.5 to 8.0 by addition of aqueous sodium acetate (10% w/v). The stirring was continued for 3 hours at 0–5°C. Then reaction mixture was poured into ice to obtain dyes D₁ to D₁₃, these dyes were filtered and dried at 70°C and were recrystallized from acetone, the properties are presented in Table 1.

3. Results and Discussion

All the dyes showed good performance with polyester fiber. Table 2 shows moderate to fairly good light fastness. Compounds D₂, D₆, D₇, D₁₁, and D₁₂ showed better light fastness. The wash fastness of all the compounds was also of very good order. Introduction of terminal amino group for better dispersibility observed no notable change in the percentage exhaustion. Overall, synthesised dyes gave good dyeing on polyester fibers. All the samples showed moderate activities against *E. coli* and *P. aeruginosa*. Dyes D₃, D₇, D₁₀, and D₁₁ showed good antibacterial activity against *S. aureus*, while D₃ and D₅ showed good antifungal activity against *C. albicans*. Standards used were Metranidazole and Flucanazole for the comparison purpose as described in Table 3. The structures

TABLE 4: IR and NMR data of D₁–D₁₃ dyes.

Dye number	IR (KBr) cm ⁻¹	¹ H NMR (DMSO) chemical shift in δ ppm
D ₁	3341 (N–H str.), 3011 (C–H), 1652 (C=O), 1578 (N=N), 1520 (NO ₂), 1442 (C–S–C), 776 (C–S)	3.36 (s, 2H, –COCH ₂), 4.30 (s, 1H, –NH), 6.92 (s, 1H, –NH), 7.37–8.48 (m, 14H, Ar–H),
D ₂	3329 (N–H), 3008 (C–H), 1660 (C=O), 1581 (N=N), 1514 (NO ₂), 1434 (C–S–C), 781 (C–S)	3.34 (s, 2H, –COCH ₂), 4.32 (s, 1H, –NH), 6.86 (s, 1H, –NH), 7.20–8.31 (m, 14H, Ar–H),
D ₃	3337 (N–H), 3007 (C–H), 1655 (C=O), 1578 (N=N), 1517 (NO ₂), 1419 (C–S–C), 779 (C–S)	3.35 (s, 2H, –COCH ₂), 4.35 (s, 1H, –NH), 6.87 (s, 1H, –NH), 7.15–8.21 (m, 12H, Ar–H), 8.59 (s, 1H)
D ₄	3319 (N–H), 3010 (C–H), 2224 (C $\equiv$ N), 1664 (C=O), 1581 (N=N), 1510 (NO ₂), 1415 (C–S–C), 778 (C–S)	3.32 (s, 2H, –COCH ₂), 4.31 (s, 1H, –NH), 6.84 (s, 1H, –NH), 7.12–8.22 (m, 11H, Ar–H), 8.78 (s, 1H)
D ₅	3322 (N–H), 3202 (O–H), 2998 (C–H), 1661 (C=O), 1584 (N=N), 1511 (NO ₂), 1417 (C–S–C), 1181 (O–C), 774 (C–S)	3.34 (s, 2H, –COCH ₂), 4.28 (s, 1H, –NH), 5.64 (s, 1H, –OH), 6.82 (s, 1H, –NH), 7.10–8.27 (m, 13H, Ar–H)
D ₆	3317 (N–H), 3209 (O–H), 3007 (C–H), 1669 (C=O), 1580 (N=N), 1514 (NO ₂), 1416 (C–S–C), 1176 (O–C), 776 (C–S)	3.31 (s, 2H, –COCH ₂), 4.32 (s, 1H, –NH), 5.67 (s, 1H, –OH), 6.79 (s, 1H, –NH), 7.15–8.22 (m, 13H, Ar–H)
D ₇	3324 (N–H), 3205 (O–H), 3002 (C–H), 1667 (C=O), 1584 (N=N), 1510 (NO ₂), 1414 (C–S–C), 1177 (O–C), 768 (C–S)	3.35 (s, 2H, –COCH ₂), 4.29 (s, 1H, –NH), 5.59 (s, 1H, –OH), 6.80 (s, 1H, –NH), 7.10–8.21 (m, 13H, Ar–H)
D ₈	3334 (N–H), 3208 (CH ₃), 3012 (C–H), 1669 (C=O), 1579 (N=N), 1509 (NO ₂), 1411 (C–S–C), 775 (C–S)	2.59 (s, 3H, –CH ₃), 3.39 (s, 2H, –COCH ₂), 4.26 (s, 1H, –NH), 6.81 (s, 1H, –NH), 7.06–8.28 (m, 13H, Ar–H)
D ₉	3327 (N–H), 3017 (CH ₃), 2912 (C–H), 1658 (C=O), 1580 (N=N), 1512 (NO ₂), 1418 (C–S–C), 769 (C–S)	2.62 (s, 3H, –CH ₃), 3.37 (s, 2H, –COCH ₂), 4.31 (s, 1H, –NH), 6.70 (s, 1H, –NH), 7.11–8.18 (m, 13H, Ar–H)
D ₁₀	3318 (N–H), 2998 (C–H), 1665 (C=O), 1578 (N=N), 1517 (NO ₂), 1413 (C–S–C), 763 (C–S), 729 (C–Cl)	3.30 (s, 2H, –COCH ₂), 4.19 (s, 1H, –NH), 6.74 (s, 1H, –NH), 7.05–8.20 (m, 13H, Ar–H)
D ₁₁	3305 (N–H), 3009 (C–H), 1663 (C=O), 1583 (N=N), 1512 (NO ₂), 1416 (C–S–C), 766 (C–S)	3.32 (s, 2H, –COCH ₂), 4.18 (s, 1H, –NH), 6.67 (s, 1H, –NH), 7.12–8.19 (m, 11H, Ar–H), 8.51 (s, 1H),
D ₁₂	3315 (N–H), 3004 (C–H), 1659 (C=O), 1582 (N=N), 1516 (NO ₂), 1418 (C–S–C), 772 (C–S), 724 (C–Cl)	3.34 (s, 2H, –COCH ₂), 4.23 (s, 1H, –NH), 6.73 (s, 1H, –NH), 7.10–8.12 (m, 9H, Ar–H), 8.44 (s, 1H, between –NO ₂ and Cl), 8.79 (s, 1H, between two –NO ₂ group).
D ₁₃	3312 (N–H), 3007 (C–H), 1661 (C=O), 1581 (N=N), 1521 (NO ₂), 1420 (C–S–C), 767 (C–S), 694 (C–Br)	3.31 (s, 2H, –COCH ₂), 4.17 (s, 1H, –NH), 6.79 (s, 1H, –NH), 7.00–8.05 (m, 9H, Ar–H), 8.53 (s, 2H).

of synthesized dyes were confirmed by spectral analysis as mentioned in Table 4.

4. Dyeing Procedure

The dyeing of the polyester fabric samples was carried out by HTHP dyeing method [17, 18].

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