

Synthesis of 4-Oxoquinazoline Derivatives and Their Application on Polyester Fibre

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Thirteen azo disperse dyes of the type 2-phenyl-3-[H]-substituted-4'''-(N,N-disubstituted amino phenyl-4''-aryl azo-4'-benzanilid-1'-yl)-4-oxoquinazoline have been synthesised by the condensation of the diamino benzanilide with 2-phenyl 4-oxo 3,1-benzoxazine, then after diazotisation, subsequent coupling with various substituted amino benzene derivatives was carried out. Their spectral characteristics are evaluated. Their dyeing properties like washing, light and rubbing fastness percentage % exhaustion were assessed.

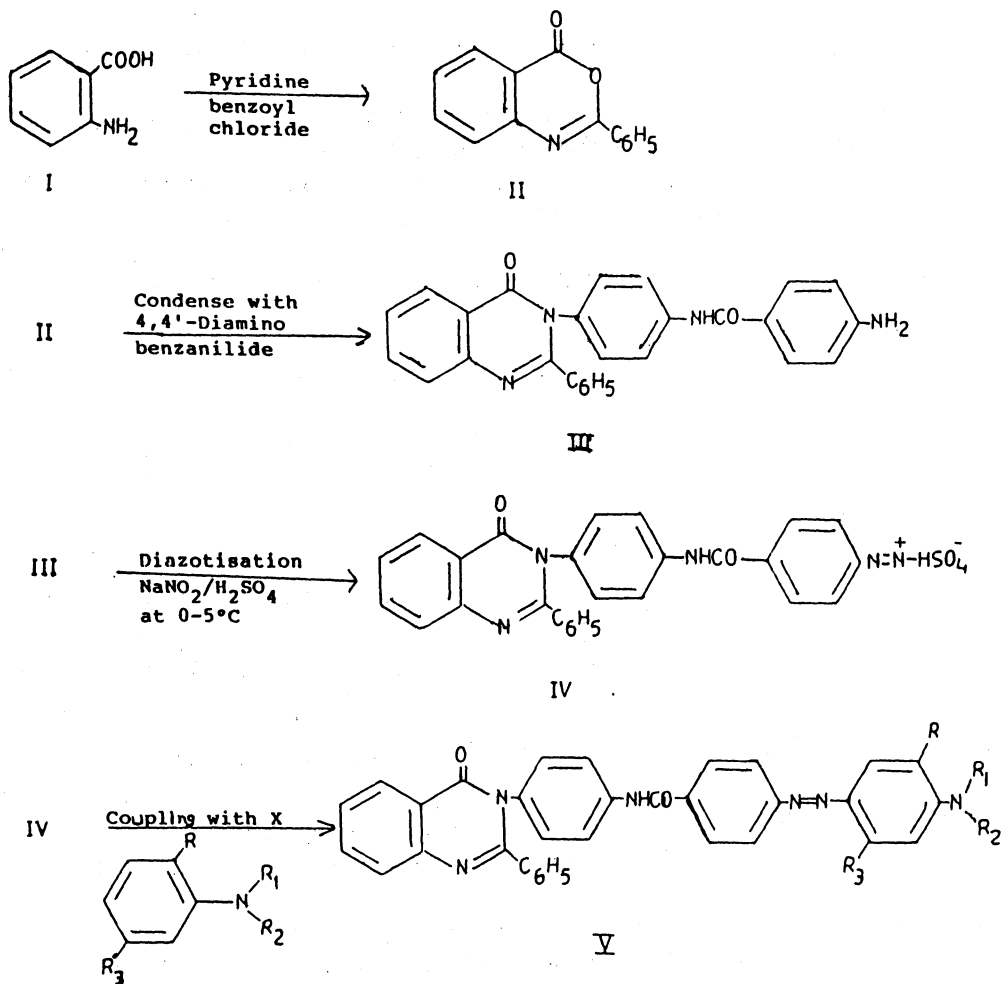
INTRODUCTION

The utility of quinazoline derivatives for the production of some commercial dyes and pigments, useful both to natural and man-made fibres, has been known.^{1,2} It has been reported that 4-oxoquinazoline functions as an azo component and coupled with various coupling components, it shows good tinctorial strength. Due to the high penetration power of azo quinazoline derivatives into the fibres^{4,5} as azo disperse dyes, it shows good colour value and high washing, light and sublimation fastness.⁶⁻⁸ A number of azo disperse dyes from 3-(*m*- or *p*-amino phenyl)-2-methyl-4-(3H)-quinazoline have been reported^{9,10} showing good colour over various polyester and nylon blends.

Because of the encouraging reports on the industrial application of some of the dyes based on azo aromatic systems, it enchants us to prepare the dyes from 2-phenyl-4-oxo-quinazoline system and to study their dyeing performance. The azo disperse dyes of the following structure were prepared (D₁-D₁₃) (Scheme-1).

EXPERIMENTAL

All the melting points were determined in open capillaries and are uncorrected. IR spectra (in KBr disc) were recorded on a Perkin-Elmer 377 spectrophotometer; elemental analysis was done on Carlo Erba 1108 analyser; λ_{\max} was determined in DMF at 28°C using Bausch and Lomb Spectronic-20 at 2×10^{-3} M dye conc. R_f values are determined on TLC using chloroform : methanol (95:5) solvent system. Fastness tests were assessed by the standard method of testing (BS : 1006-1978 and IS : 765-1979).



where, R₁, R₂, R₃ various substituents table-I

Scheme I

2-Phenyl-3-(4''-amino-4'-benzanilid-1'-yl)-4-oxoquinazoline [I]

This is synthesised by the cyclisation and subsequent condensation reaction of anthranilic acid with benzoyl chloride in presence of pyridine, and condensed with 4,4'-diamino benzanilide and finally diazotised and coupled with various substituted benzene derivatives.

2-Phenyl-3,1-benzoxazin-4(H)-one [II]

To a solution of anthranilic acid (0.1 mole, 13.7 g) in pyridine (6.0 mL) benzoyl chloride (14.0 g, 0.1 mole) was added dropwise with constant stirring at 0-8°C over 30 min. The solid mass obtained was washed with sodium bicarbonate

solution (20%) to remove unreacted anthranilic acid. The product was filtered, washed with cold water, dried and recrystallised from ethanol 99%. m.p. = 110°C, yield = 78% (Found, N: 6.27%; Calcd., N: 6.35%).

Preparation of 2-phenyl-3-(4''-amino-4'-benzanilid-1'-yl)-4-oxoquinazoline [III]

2-Phenyl-3,1-benzoxazin-4(H)-one (22.3 g, 0.1 mole) and 4,4'-diamino benzanilide (22.7 g, 0.1 mole) were intimately mixed and heated on a free flame for 5 minutes with vigorous shaking. To the hot reaction, 20 mL of ethanol was added and the contents of the flask were allowed to cool. Scratching the side with glass rod, reaction mixture was poured in ice-cold water with continuous stirring. A crystallised solid was obtained which was then further recrystalline from ethanol. Yield = 70%, m.p. = 250°C (Found, N: 13.02%; Calcd., N: 12.96%).

Diazotisation of 2-phenyl-3-(4''-amino-4'-benzanilid-1'-yl)-4-oxoquinazoline [IV]

2-Phenyl-3-(4''-amino-4'-benzanilid-1'-yl)-4-oxoquinazoline (4.32 g, 0.01 mole) was diazotised in the usual manner. The resulting diazo solution was used for subsequent coupling reaction.

Preparation of 2-phenyl-3-[H/substituted-4'''-(N,N-disubstituted)-amino phenyl-4''-aryl azo-4'-benzanilid-1'-yl]-4-oxo quinazoline [D₁ to D₁₃] [V]

m-Chloro-N,N-bis- β -hydroxy ethyl aniline X (2.15 g, 0.1 mole) was dissolved in hydrochloric acid (1.8 mL, 0.05 mole). The above solution was cooled below 5°C in an ice-bath. To this well stirred solution, the above mentioned diazo solution was added maintaining the pH at 6-7 by the addition of sodium carbonate solution (10% w/v). The stirring was continued for 3 h at the same temperature. The precipitated solid was then filtered, washed with water, dried and recrystallised from DMF-chloroform to give dye D₁. Yield = 62%, m.p. = 275°C (Found, N: 11.27%; Calcd., N: 11.29%).

The same procedure was used to prepare dye D₂ to D₁₃. The coupling components X are commercially available.

Dyeing of polyester in 2% shade

The dyeing of the unwhitened (100%) polyester fabric was done under pressure in a high temperature dyeing laboratory equipment. Pressure maintained = 30 psi; temperature of dye-bath = 130°C; dyeing duration = 45 min; dye-bath material : liquor ratio = 1 : 50.

RESULTS AND DISCUSSION

Melting points are uncorrected. Data on yield, elemental analysis and the dyeing assessments are shown in Table-1 and Table-2, respectively. λ_{max} , R_f, % E, shade on polyester and various fastness properties are shown in Table-2.

(a) *Light-fastness*: All the compounds show good light-fastness. The com-

TABLE-1
YIELD, MELTING POINT AND ELEMENTAL ANALYSIS OF OXOQUINAZOLINE DYES

Dye No.	Coupling component (X)			Yield (%)	m.p. (°C)	Nitrogen (%)		
	R	R ₁	R ₂			R ₃	Found	Calcd.
D ₁	H	CH ₂ CH ₂ OH	CH ₂ CH ₂ OH	Cl	68	275	12.80	12.75
D ₂	H	CH ₂ CH ₂ OCOCH ₃	CH ₂ CH ₂ OCOCH ₃	Cl	67	279	11.27	11.29
D ₃	H	CH ₂ CH ₂ OH	CH ₂ CH ₂ OH	CH ₃	68	272	13.18	13.16
D ₄	H	CH ₂ CH ₂ OCOCH ₃	CH ₂ CH ₂ OCOCH ₃	CH ₃	76	298	11.67	11.63
D ₅	H	CH ₂ CH ₂ OH	CH ₂ CH ₂ OH	NHCOCH ₃	72	285	14.37	14.39
D ₆	H	CH ₂ CH ₂ OH	CH ₂ CH ₂ OH	H	70	282	13.48	13.46
D ₇	OCH ₃	CH ₂ CH ₂ OCOCH ₃	CH ₂ CH ₂ OCOCH ₃	NHCOCH ₃	78	>300	12.28	12.32
D ₈	OCH ₃	CH ₂ CH ₂ CN	CH ₂ CH ₂ OH	NHCOCH ₃	72	295	15.50	15.55
D ₉	H	CH ₂ CH ₂ CN	CH ₂ CH ₂ OCOCH ₃	H	76	279	14.57	14.51
D ₁₀	H	CH ₂ CH ₂ CN	CH ₂ CH ₃	H	82	282	15.92	15.88
D ₁₁	H	CH ₂ CH ₃	CH ₂ CH ₃	NHCOCH ₃	78	285	15.15	15.10
D ₁₂	H	CH ₂ CH ₃	CH ₂ CH ₃	NHCOCH ₂ CH ₃	80	>300	14.73	14.78
D ₁₃	OCH ₃	CH ₂ CH ₃	CH ₂ CH ₃	NHCOCH ₃	82	>300	14.40	14.43

pounds D₈–D₁₀ show somewhat better light-fastness. This may be due to the presence of an additional acceptor substituent such as cyano group, which results into increasing electron mobility in these compounds and thus results into good light-fastness. However, in case of compound D₂, the presence of strong electron acceptor substituents results into somewhat low light-fastness compare to that of compound D₁. Further, hydrogen bonding involving azo and acetylamino substituents could be considered as having stabilizing aspect. In other words, too strong electron acceptor substituents in the azo dyes can dramatically increase light-fastness because of their consequent tendency to stabilize the azo disperse dyes.

TABLE-2
λ_{max}, PERCENTAGE EXHAUSTION, SHADE AND FASTNESS OF OXOQUINAZOLINE

Dye No.	Shade on polyester	λ _{max}	R _f value	% Exhaustion	Fastness to:			
					Light	Washing	Rubbling	
							Dry	Wet
D ₁	Orange yellow	485	0.75	57	5	4–5	5	5
D ₂	Golden yellow	480	0.82	52	4	5	4	4
D ₃	Bright orange	495	0.80	59	4	4	4	4
D ₄	Orange yellow	475	0.72	45	4	4–5	5	5
D ₅	Brown	510	0.85	50	4	4	5	5
D ₆	Khakhi	457	0.87	55	4–5	4	4	4
D ₇	Brown pink	490	0.72	42	4	5	4	4
D ₈	Light brown	465	0.75	47	5	4	4	4
D ₉	Khakhi yellow	435	0.83	56	4–5	4	5	5
D ₁₀	Yellow brown	492	0.89	59	5	3	4	4–5
D ₁₁	Pink	510	0.80	52	4	4	5	5
D ₁₂	Scarlet-red	485	0.85	50	4	4	5	5
D ₁₃	Bright orange	495	0.82	53	4	5	4	4

(b) *Wash-fastness*: Compared to that of compound D₁₀ all the compounds show good to excellent wash-fastness. This is due to the higher molecular size of the compounds, the larger substitution capability of hydrogen bond formation which results into lowering the magnitude of the dye removal. The presence of the electron-attracting substituents, with respect to the hydroxyl group in case of dye D₁ results into somewhat good wash-fastness. Overall, the wash fastness of compounds D₁ to D₁₃ is good.

(c) *Rubbing Fastness*: The rubbing fastness of the compounds D₁ to D₁₃ appears to be good to excellent.

IR Spectra

All the compounds show two strong intense bands with N—H stretching at 3400 cm^{-1} , N—H bending (medium) at 1575 cm^{-1} and carbonyl stretching at 1680 cm^{-1} due to the presence of the anilide group (—NHCO) in their ring structure. Compounds D₁ to D₁₃ also show aromatic stretching at 1635 cm^{-1} due to the presence of phenyl ring at second position in 4-oxo-quinazoline nucleus. The characteristic of —N=N— stretching at 1475 cm^{-1} and aromatic carbonyl stretching at $1660\text{--}1620\text{ cm}^{-1}$ is also found. Respective stretching and bending of various substituents in all compounds is also found.

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