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RESEARCH ARTICLE

SYNTHESIS AND APPLICATIONS OF SOME REACTIVE DYES HAVING QUINAZOLIN-4(3H)-ONE MOIETY

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Abstract

Some reactive dyes (6a-6l) having Quinazolin-4(3H)-one reactive system have been synthesized by coupling of diazotized 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl) phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid with cyanurated coupling components such as H-acid, J-acid, N-methyl J-acid, Gamma acid, Peri acid, Sulpho Tobias acid, Koch acid, Bronner's acid, Chicago acid, K-acid, N-Phenyl J-acid and M-acid. The synthesized dyes were characterized by nitrogen elemental analysis, IR, and ¹H-NMR spectra. They were applied on wool, silk, and cotton fibres. Their dyeing properties, fastness properties, and colorimetric data (L*, a*, b*, C*, H*, and K/S) have also been investigated.

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Introduction:-

Reactive dyes are colored compounds which contain one or two groups capable of forming covalent bonds between a carbon or phosphorus atom of the dye ion or molecule and an oxygen, nitrogen or sulphur atom of a hydroxy, an amino or mercapto group, respectively, of the substrate. Such covalent bonds are formed with the amino, hydroxyl groups of cellulosic fibres, with the amino, hydroxyl and mercapto groups of protein fibres and with the amino groups of polyamides [1]. Reactive dyes are known for their bright colors and very good to excellent light and wash fastness; however, their resistance to chlorine bleach is poor [2]. Reactive dyes are extensively used in the textile industry not only for cotton, but also for wool and silk because of their wide variety of color shades, high wet fastness, ease of application and brilliant colors. In addition, investigations into the development of reactive dyes for polyester and polypropylene fibres have been demonstrated to the level of technical possibility but such dyes are not yet of commercial interest [3, 4]. A steady increase in the reactive dye usage has been observed as a result of the increased cotton use worldwide [5].

The first reactive dyes for cellulose, Procion M dyes, were introduced in 1956 by ICI [6]. These dyes contain a highly reactive dichlorotriazinyl group, which reacts with cellulose in the presence of alkali at room temperature. In subsequent years, ICI and Ciba [7] marketed dyes containing the lower reactivity (hot dyeing) monochlorotriazinyl (MCT) reactive group, whilst Hoechst [8] introduced Remazol (warm dyeing) dyes bearing the sulfatoethylsulfonyl 'vinyl sulfonyl' (VS) reactive group.

Quinazolinones are a very important class of heterocyclic carrier systems of reactive dyes due to their high chromophoric potential. As a part of our continuous interest in the reactivity of Quinazolinone derivatives we have synthesized some reactive dyes having quinazolin-4(3H)-one moiety and investigated their applications as reactive

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dyes to silk, wool and cotton fiber [9-11]. Advantages claimed for reactive dyes having Quinazolin-4(3H)-one moiety include high exhaustion, good levelness, high fastness, and good wash-off properties [12, 13]. Patel et al. [14-18] have synthesized cold brand reactive dyes and hot brand reactive dyes for silk, wool, and cotton fibres.

Experimental

Materials and Methods:-

Commercially available silk, wool and cotton fibres were used for dyeing. The chemicals and reagents were obtained from Sigma-Aldrich and all the coupling components were received from Atul Ltd., Valsad, Gujarat, India and used without further purification. All melting points taken by open capillary method are uncorrected and are expressed in °C. TLC analysis was carried out on silica gel G F254-precoated aluminium sheets [19]. IR spectra were recorded on a Shimadzu Model 8400S system using the potassium bromide wafer technique. ¹H-NMR (400 MHz) and spectra were determined on Bruker Avance II in DMSO solvent using TMS as internal standard, UV-vis absorption spectra were recorded on a Thermo Scientific Evolution 300 Spectrophotometer at the wavelength of maximum absorption (λ_{max}) using water as solvent. Elemental analysis (C, H, and N) were carried out using a CHN analyser, Carlo Erba, Italy. The dyeing was done by using a Laboratory Rota Dyer instrument. Colorimetric data (L^* , a^* , b^* , C^* , H^* and K/S) were recorded on a Reflectance Spectrophotometer Gretag Macbeth CE: 7000.

Synthesis of Dyes

Synthesis of 4-oxo-2-phenyl-4H-benzo[d][1,3]oxazine-6-sulphonic acid (1) [20]

To a stirred solution of 2-amino-5-sulphobenzoic acid (2.17 g, 0.01 mol) in pyridine (60 ml), benzoyl chloride (1.16 ml, 0.01 mol) was added dropwise, maintaining the temperature at 0-5 °C for 1 hour. The reaction mixture was stirred for another 2 hours at room temperature until a solid product was formed. The reaction mixture was neutralized with saturated Na₂CO₃ solution. A yellow separated solid was filtered, washed with water, and recrystallized from ethanol.

Yield: 78 %, m. p. 344-346 °C. (Scheme 1)

Synthesis of 4,4'-(1,3,4-oxadiazole-2,5-diyl)dianiline (2) [21]

4-Amino benzoic acid (1.37 g, 0.01 mol) and 50 ml methanol were refluxed on water bath for 3 hours with a few drops of concentrated H₂SO₄ as a catalyst. After completion of the reaction, reaction mixture was poured onto ice cold water to give methyl 4-aminobenzoate. It was washed with NaHCO₃ solution (5%), dried and recrystallized from methanol. Methyl 4-aminobenzoate (1.51 g, 0.01 mol) in 30 ml ethanol was added to hydrazine hydrate (0.015 mol) and refluxed on a water bath for 4 hours. After completion of reaction, excess ethanol was distilled off and the reaction mass was poured onto ice cold water to give 4-Aminobenzohydrazide. It was filtered, washed and recrystallized from methanol. 4-Aminobenzohydrazide (1.51 g, 0.01 mol) and 4-amino benzoic acid (1.37 g, 0.01 mol) were refluxed in 5 ml POCl₃ for 8 hours. After completion of the reaction, reaction mixture was poured onto ice cold water to give 4,4'-(1,3,4-Oxadiazole-2,5-diyl)dianiline. It was washed with NaHCO₃ solution (5%), dried and recrystallized from methanol.

Yield: 75 %, m. p. 428-430 °C. (Scheme 1)

Synthesis of 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid (3) [24]

4-Oxo-2-phenyl-4H-benzo[d][1,3]oxazine-6-sulphonic acid (3.03 g, 0.01 mol) and 4,4'-(1,3,4-oxadiazole-2,5-diyl)dianiline (2.52 g, 0.01 mol) were dissolved in pyridine (40 ml) and refluxed for 6 hours under anhydrous reaction conditions and then allowed to cool at room temperature. The reaction mixture was then treated with ice cooled dilute HCl solution and stirred. A solid separated out which was filtered off and washed with water to remove any adhered pyridine. 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid thus obtained was dried under vacuum and recrystallized from ethanol.

Yield: 84 %, m. p. above 450 °C. (Scheme 2)

Diazotization of 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid (4) [25]

Sodium nitrite (0.69 g, 0.01 mol) was added to a cooled concentrated sulphuric acid (5 ml) and then the mixture was warmed gradually on water bath at 65-70 °C. The solution was cooled to 0-5 °C and then a mixture of propionic acid

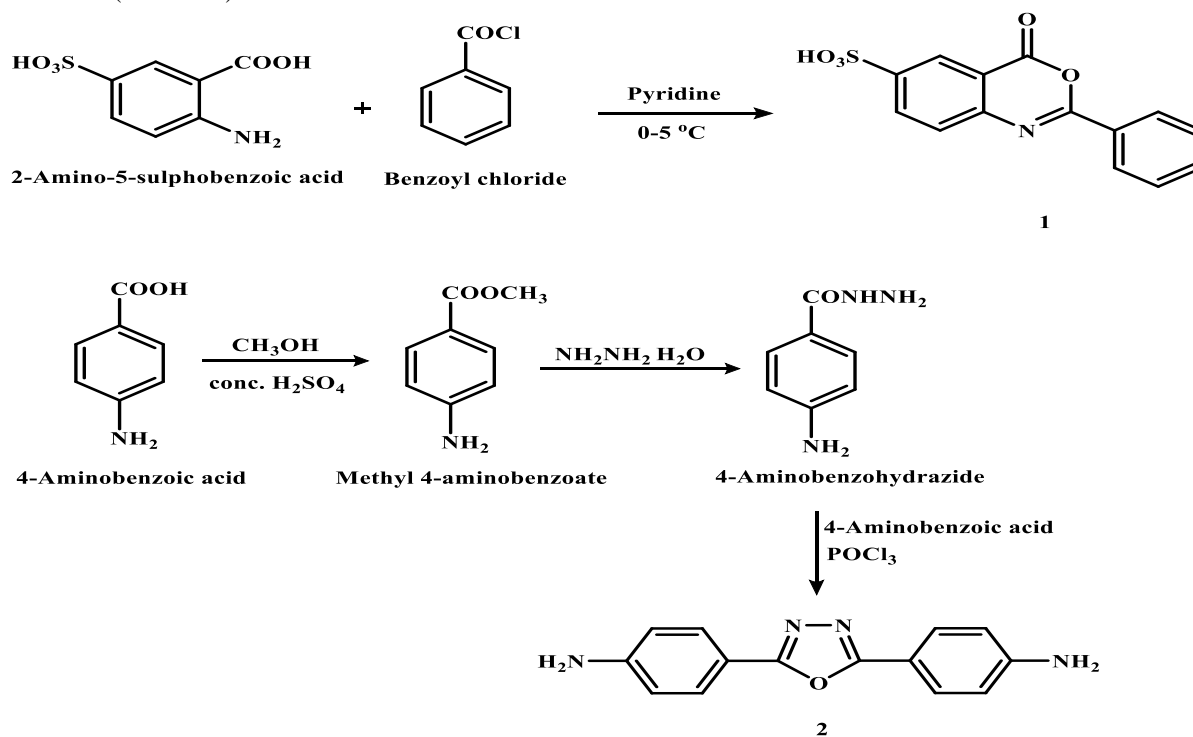
(2 ml) and acetic acid (3 ml) was added to it. 3-(4-(5-(4-Aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid (5.37 g, 0.01 mol) was added gradually to a nitrosylsulphuric acid paste at 0 °C, and then stirred for 30 minutes, maintaining the temperature at 0-5 °C. After completion of reaction, excess of nitrous acid was destroyed using sulphamic acid. The diazotized solution at 0-5 °C was obtained and used for subsequent coupling reaction. (Scheme 3)

Cyanuration of H-acid (5)

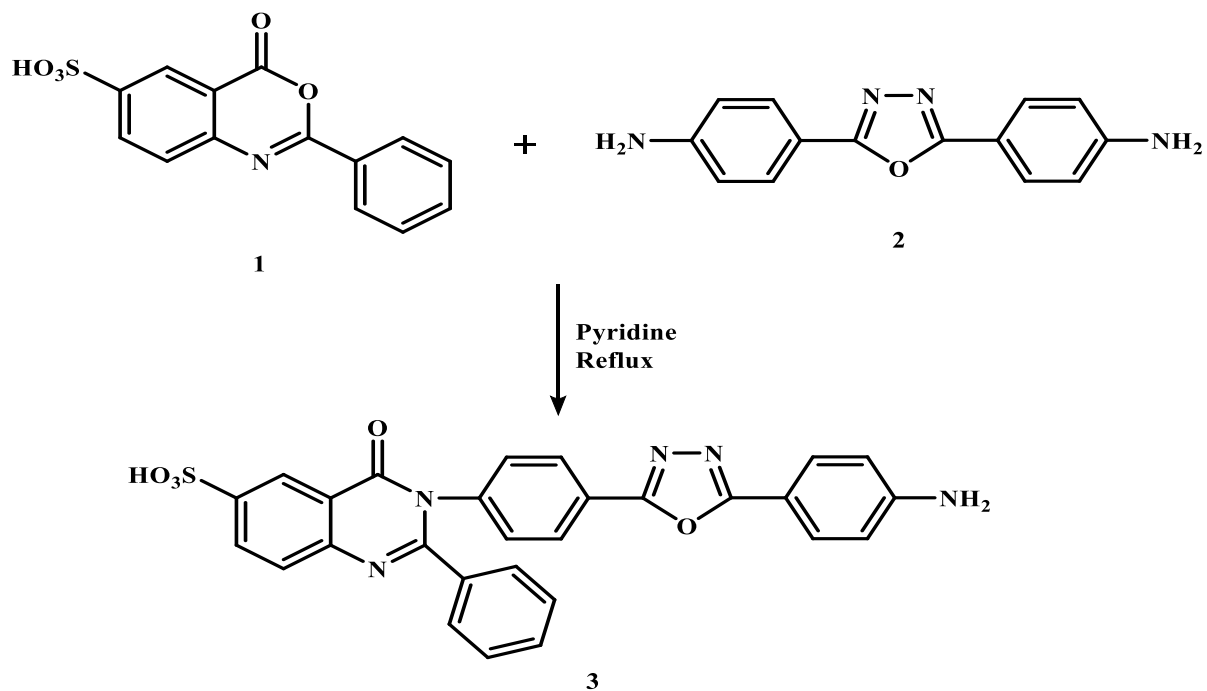
Cyanuric chloride (1.85 g, 0.01 mol) was stirred in acetone (25 ml) at a temperature below 5 °C for a period of an hour. A neutral solution of H-acid (3.19 g, 0.01 mol) in aqueous sodium carbonate solution (10% w/v) was then added in small lots in about an hour. The pH was maintained neutral by simultaneous addition of sodium carbonate solution (1% w/v). The reaction mass was stirred at 0-5 °C for further 4 hours. The cyanurated H-acid solution was used for subsequent coupling reaction. (Scheme 4)

Coupling of diazotized 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid with cyanurated H-acid: Synthesis of dyes (6ato 6l)

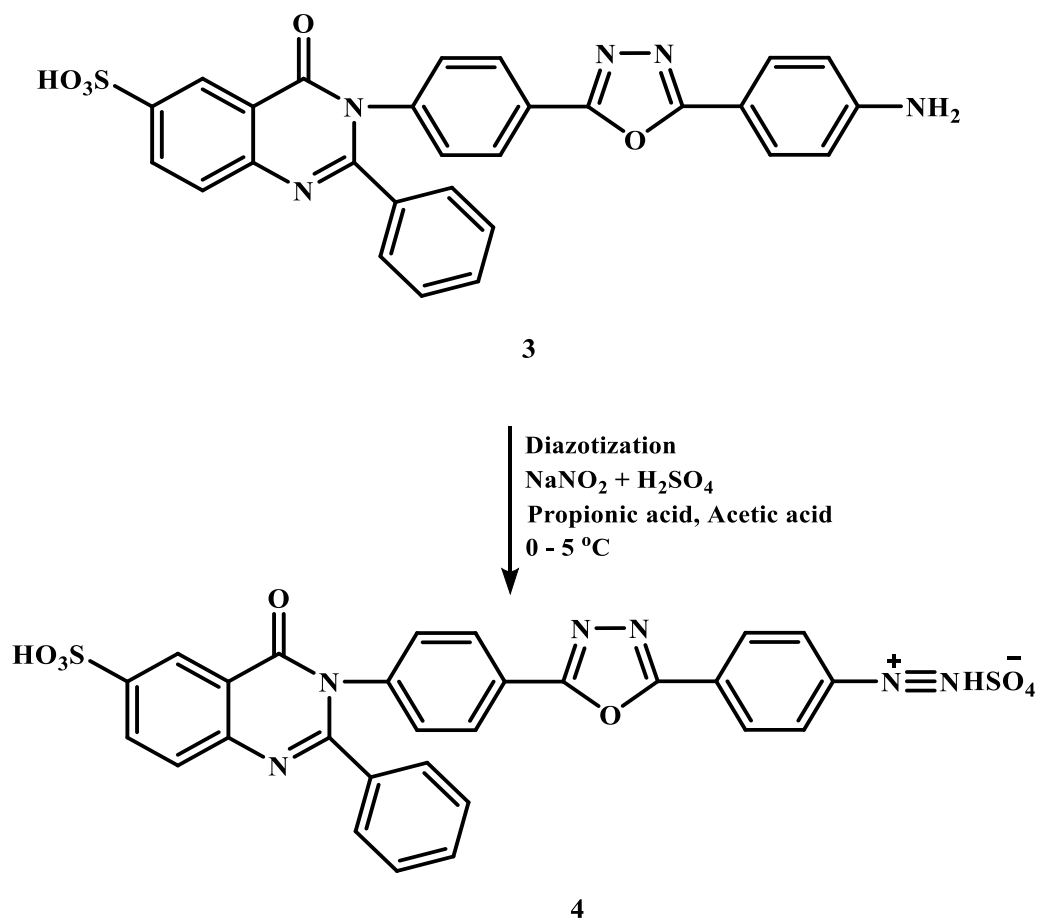
The above mentioned freshly prepared diazotized 3-(4-(5-(4-aminophenyl)-1,3,4-oxadiazol-2-yl)phenyl)-4-oxo-2-phenyl-3,4-dihydroquinazoline-6-sulphonic acid was added to an ice cooled and well stirred solution of cyanurated H-acid over a period of 10-15 minutes. The pH was maintained at 8.0 to 8.5 by simultaneous addition of sodium carbonate solution (10% w/v) and stirring was continued for 4 hours. Sodium chloride (15 g) was then added and the mixture was stirred for an hour. The solid dye separated out was filtered, washed with minimum amount of acetone and dried. (Scheme 5)



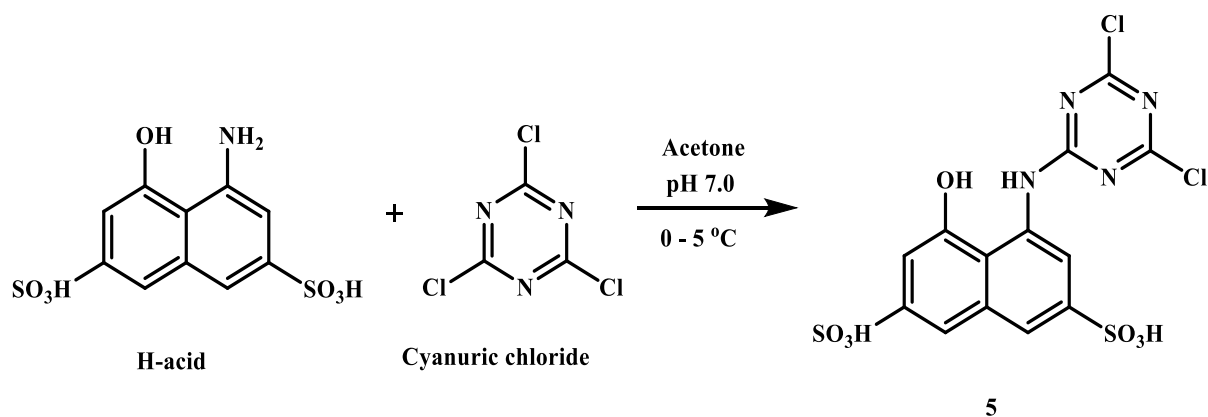
Scheme 1: Synthesis of 1 and 2



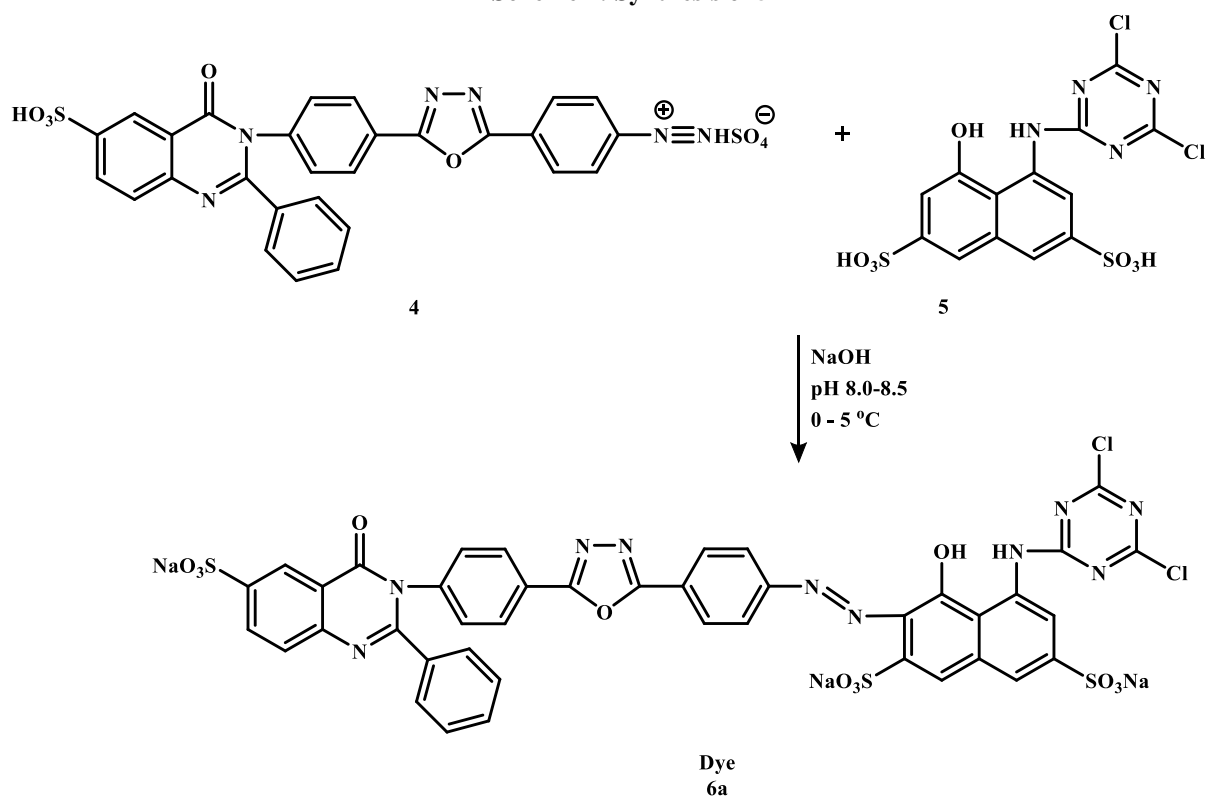
Scheme 2: Synthesis of 3



Scheme 3: Synthesis of 4



Scheme 4: Synthesis of 5



Scheme 5:- Synthesis of Dye 6a.

Following the above procedure, other reactive dyes 6b to 6l were synthesized by using various cyanurated coupling components such as J-acid, N-Methyl J-acid, Gamma acid, Peri acid, Sulpho Tobias acid, Koch acid, Bronner's acid, Chicago acid, K-acid, N-Phenyl J-acid and M-acid. (Table 1). All the synthesized dyes were recorded in Table 2. IR and $^1\text{H-NMR}$ characterization results of all dyes are given in Table 3.

Table 1:- Various coupling components (a-l), arrow indicates coupling position.

Dye No.	Coupling component	Structural formula	Dye No.	Coupling component	Structural formula
6a	H-acid		6g	Koch acid	
6b	J-acid		6h	Bronner's acid	
6c	N-Methyl J-acid		6i	Chicago acid	
6d	Gamma acid		6j	K-acid	
6e	Peri acid		6k	N-Phenyl J-acid	
6f	Sulpho Tobias acid		6l	M-acid	

Table 2:- Characterization of dyes (6a-6l).

Dye No.	Coupling component (R)	Molecular Formula	Mol. Weight g/mol	Yield (%)	m. p. °C	Nitrogen %		Rf Value
						Found	Req.	
6a	H-acid	C ₄₁ H ₂₁ Cl ₂ N ₁₀ Na ₃ O ₁₂ S ₃	1081.73	82	>300	12.88	12.95	0.60
6b	J-acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₉ S ₂	979.69	76	>300	14.22	14.30	0.56
6c	N-Methyl J-acid	C ₄₂ H ₂₄ Cl ₂ N ₁₀ Na ₂ O ₉ S ₂	993.71	78	>300	14.05	14.10	0.64
6d	Gamma acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₉ S ₂	979.69	74	>300	14.24	14.30	0.61
6e	Peri acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₈ S ₂	963.69	84	>300	14.44	14.53	0.57
6f	Sulpho Tobias acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₈ S ₂	963.69	86	>300	14.45	14.53	0.54
6g	Koch acid	C ₄₁ H ₂₀ Cl ₂ N ₁₀ Na ₄ O ₁₄ S ₄	1167.77	82	>300	11.92	11.99	0.57
6h	Bronner's acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₈ S ₂	963.69	79	>300	14.47	14.53	0.63
6i	Chicago acid	C ₄₁ H ₂₁ Cl ₂ N ₁₀ Na ₃ O ₁₂ S ₃	1081.73	86	>300	12.88	12.95	0.54
6j	K-acid	C ₄₁ H ₂₁ Cl ₂ N ₁₀ Na ₃ O ₁₂ S ₃	1081.73	78	>300	12.87	12.95	0.56
6k	N-Phenyl J-acid	C ₄₇ H ₂₆ Cl ₂ N ₁₀ Na ₂ O ₉ S ₂	1055.79	75	>300	13.22	13.27	0.59
6l	M-acid	C ₄₁ H ₂₂ Cl ₂ N ₁₀ Na ₂ O ₉ S ₂	979.69	83	>300	14.22	14.30	0.62

Dyeing

All the dyes were applied on silk, wool, and cotton fibres by using the reported procedure [22]. The variation in the hues of the dyed fabric results from both the nature and position of the substituent present on the coupler ring. The remarkable degree of levelness after washing indicates good penetration and affinity of these dyes to the fabric. The percentage dye bath exhaustion (%E) and percentage dye bath fixation (%F) of the dyed fibres were determined by using reported method [23]. The light fastness was assessed in accordance with BS: 1006-1978 and the wash fastness was carried out in accordance with IS: 765-1979. The rubbing fastness test was carried out by using Crock meter (Atlas) in accordance with the AATCC-1961.

Dyeing of Silk

Dye (0.2 g) was dissolved by pasting up in cold water and then added hot water. Anhydrous Glauber's salt solution (1.0 ml, 20 % w/v) was added to it. The pH of the dye bath was adjusted to 3.0 by adding acetic acid solution and the total volume was adjusted to 80 ml by adding required amount of distilled water. The dye bath solution was transferred to 250 ml conical flask fitted with an air condenser. The temperature of the dye bath was adjusted to 50 °C and the silk fibre was introduced into the dye liquor with stirring. The temperature of the dye bath was gradually increased to 85 °C over a period of 10 min and it was maintained for another 30 min. The dyed fibre was washed with cold water, soaping, and dried.

Dyeing of Wool

Dye (0.2 g) was pasted with a few drops of cold water, then about 80 ml cold water was added, and the mixture was stirred and made up to 100 ml with dye solution (20 ml), acetic acid (1.5 ml of 10 % v/v), Glauber's salt solution (4 ml of 10 % w/v), and water (14.4 ml). Then wool fabric (2 g) was introduced into the dye bath at 30 °C and the temperature was raised up to 80 °C for 20 min. Sulfuric acid (0.4 ml of 10 % v/v) was added and the dyeing was continued for 40 min more at the same temperature. The dyed fibre was then removed, rinsed with cold water, soaping, and dried.

Dyeing of Cotton

Dye (0.2 g) was pasted with a few drops of cold water, then about 80 ml cold water was added, and the mixture was stirred and made up to 100 ml with dye solution (20 ml), Glauber's salt solution (4 ml of 10 % w/v), and water (14.4 ml). Then cotton fabric (2 g) was introduced into the dye bath at 30 °C and the temperature was raised up to 80 °C for 20 min. Soda ash (Na₂CO₃) solution (0.4 ml of 10 % v/v) was added to bring about fixation and the

dyeing was continued for 40 min more at the same temperature. The dyed fibre was then removed, rinsed with cold water, soaping, and dried.

Result and Discussion:-

Visible Absorption Spectra

The visible absorption spectra of all synthesized dyes have been recorded in water and observed in the range of 550-410 nm (Table 4). The introduction of electron donating or electron attracting groups at the suitable position in the coupler ring affected the absorption characteristic of dyes. Some substances appear colored due to the presence of such functional groups like C=C, C=O, N=N, etc. which are capable of absorbing visible and/or ultraviolet radiation (800-200 nm). Such functional groups, which confer color on substances, are known as chromophores. Auxochrome is a functional group which does not absorb radiation longer than 200 nm, but when attached to a given chromophore, it causes a shift in absorption to longer wavelength and also increases the depth of absorption. Dyes 6a-6l consist of many chromophoric groups like C=O, N=N, etc. These chromophores are capable of absorbing visible and/or UV radiation (800-200 nm) to exhibit a color. With comparison of dyes 6a-6l, dyes 6a-6d and 6i-6l have λ_{\max} greater than 6e-6h because 6a-6d and 6i-6l have hydroxyl group (auxochrome) which extends the conjugation due to the lone pair of electrons on the oxygen atom. Dye 6a has highest absorption maxima due to the presence of -OH and -NH₂ groups and more places for oscillation of electrons. This effect is slightly lower than other dyes because of fewer electrons. Moreover, the chromophoric group -N=N- is attached ortho to -OH group, causing resonance effects, which in turn increases λ_{\max} . The magnitude of bathochromic shift depends on the nature of substituents at the terminal amino group. The introduction of auxo-chromic group, like sulfonic acid group which causes bathochromic shift by increasing polarizability in the dye molecule.

IR Spectra

The IR spectra of dyes (Table 3) showed characteristic broad absorption band at 3503-3562 cm⁻¹ corresponding to the O-H stretching vibration of -OH group, N-H stretching vibration of secondary amine at 3445-3451 cm⁻¹, C-H stretching vibration of -CH₂- group at 2982 cm⁻¹, C=C stretching vibration of aromatic ring at 1638-1652 cm⁻¹ and 1408-1414 cm⁻¹, N=N stretching vibration of azo group at 1455 cm⁻¹, C-N stretching vibration of tertiary amine group at 1439-1440 cm⁻¹, S=O stretching vibration of -SO₃Na group at 1154-1191, and 1048-1095 cm⁻¹ and C-Cl stretching vibration of chloride group at 879-880 cm⁻¹.

¹H-NMR Spectra

¹H-NMR spectra of dyes (Table 3) singlet at 3.90-3.93 ppm of -NH- protons, singlet at 4.70-4.85 ppm of -OH protons, singlet at 3.44 ppm of -CH₃ protons and multiplet at 6.78-8.10 ppm of Ar-H protons.

Table 3:- IR and ¹H-NMR spectral data of dyes (6a, 6c and 6d).

Dye no.	IR (KBr): ν_{\max} (cm ⁻¹)	¹ H-NMR (D ₂ O) chemical shift in δ ppm
6a	3503 (O-H), 3445 (N-H), 1662 (C=O), 1638, 1408 (C=C), 1454 (N=N), 1372 (O-H), 1439 (C-N), 1193 (C-O-C), 1154, 1053 (S=O), 880 (C-Cl)	4.73 (s, 1H, -OH), 3.90 (s, 1H, -NH-), 7.26-7.84 (m, 19H, Ar-H)
6c	3562 (O-H), 3446 (N-H), 2982 (C-H), 1700 (C=O), 1642, 1412 (C=C), 1455 (N=N), 1394 (O-H), 1439 (C-N), 1192 (C-O-C), 1050, 1095 (S=O), 879 (C-Cl)	3.44 (s, 3H, -CH ₃), 4.85 (s, 1H, -OH), 6.78-8.10 (m, 20H, Ar-H)
6d	3560 (O-H), 3451 (N-H), 1699 (C=O), 1652, 1414 (C=C), 1455 (N=N), 1378 (O-H), 1440 (C-N), 1261 (C-O-C), 1191, 1048 (S=O), 880 (C-Cl)	4.70 (s, 1H, -OH), 3.93 (s, 1H, -NH-), 6.82-7.98 (m, 20H, Ar-H)

Exhaustion and Fixation Study

Dye bath exhaustion was evaluated with the percentage of dye absorbed on fibre from the dye bath at the end of dyeing process. After dyeing process, the dye liquor was decanted into a 250 ml volumetric flask. The dyed fibre was then washed several times with cold water (100 ml). The washing was transferred to the volumetric flask. The combined solution of the dye liquor and washing was then diluted to 250 ml with water. 5 ml of this solution was further diluted to 25 ml with water and the absorbance of this solution was measured. The percentage dye bath exhaustion was calculated by measuring the absorbance of the above solution and reading the corresponding

concentration on the calibration curve (concentration versus absorbance). The results of the exhaustion study of each dye are presented in Table 4.

Fixation is the percentage of dye chemically bonded to the fabric. After dyeing, the fabric was rinsed with water. Dyed fibre (0.1 g) was placed in a coning tube and sulfuric acid (10 ml) was poured over it. The fabric was stirred in sulfuric acid at room temperature for about 10-15 min. The solution was diluted to 50 ml with sulphuric acid. The percentage fixation of the dye was then calculated by measuring the absorbance of this solution and reading the corresponding concentration on the calibration curve (concentration versus absorbance). A solution of the same amount of undyed fabric in sulphuric acid (50 ml) was used as reference solution. The results of the fixation study of each dye are presented in Table 4.

The percentage exhaustion of 2 % dyeing on silk fibre ranges from 60.95 to 70.85 %, for wool ranges from 58.89 to 67.23 %, and for cotton ranges from 65.44 to 73.60 %. The percentage fixation of 2 % dyeing on silk fibre ranges from 88.43 to 92.67 %, for wool ranges from 89.03 to 91.34 %, and for cotton ranges from 82.95 to 87.88 % (Table 4). The exhaustion shows good attachment of the dye to the fibre. The presence of s-triazine group improves the exhaustion and fixation value.

Table 4:- Dyeing performance of dyes (6a-6l).

Dye no.	λ_{max} (nm)	% Exhaustion			% Fixation		
		Silk	Wool	Cotton	Silk	Wool	Cotton
6a	550	69.48	64.06	65.92	91.69	89.72	84.74
6b	530	70.85	67.23	67.36	92.67	89.03	85.11
6c	510	68.38	64.06	65.92	92.45	91.20	83.25
6d	515	69.75	61.48	66.64	91.33	89.63	83.82
6e	425	67.83	61.76	65.68	91.03	90.75	83.56
6f	420	65.08	58.89	66.16	90.36	91.15	84.43
6g	425	64.53	60.33	73.60	90.37	91.34	87.88
6h	410	62.05	60.61	68.56	92.39	90.91	85.05
6i	485	62.88	60.33	66.88	91.18	92.13	84.26
6j	460	64.53	61.76	66.16	91.13	90.75	82.95
6k	470	60.95	60.33	66.88	88.43	90.55	84.99
6l	440	63.98	62.34	65.44	91.91	90.68	83.11

Fastness Properties

All the dyes (Table 5) showed generally fair to good light fastness properties. The washing and rubbing fastness properties ranges from very good to excellent fastness on silk, wool, and cotton. The higher values of rubbing fastness are associated with higher molecular weights of the dyes.

Table 5:- Fastness properties of dyes (6a-6l).

Dye No.	Light fastness			Wash fastness			Rubbing fastness					
							Dry			Wet		
	S	W	C	S	W	C	S	W	C	S	W	C
6a	5-6	5-6	5	4-5	4-5	4	4-5	4	4-5	4-5	4-5	4
6b	5	5	5	4-5	4	4	4	5	4-5	4	3	4
6c	5	5	5	4	5	4-5	4-5	4	4	4	3	3-4
6d	5-6	5-6	4-5	4-5	4	4	4	4	4	4-5	4	4
6e	5-6	5	4-5	4-5	3	4	4	4-5	4	3-4	4	4
6f	5	5	4	4	4	3-4	3-4	4	3-4	4	4-5	3-4
6g	4-5	4-5	3-4	3-4	4	3	4	3-4	3-4	3-4	3	4
6h	4	5	3-4	3	4-5	3-4	4	4-5	4	3-4	4-5	4
6i	4	5	4	3	4	4	3-4	4	4	3	4	4
6j	5	4-5	4	4	4	3-4	4	5	3-4	3-4	5	3-4
6k	4-5	4-5	3-4	3-4	4-5	4	3-4	3-4	4	4	4	4
6l	4	4-5	4	4	4	3	4	4	4	4	3-4	4

Computer Color Matching Data (CCM)

The colors on silk, wool, and cotton fibres are expressed in terms of CIELab values and the coordinates measured are lightness (L^*), chroma (C^*), hue angle from 0° to 360° (H), a^* value representing the degree of redness (positive) and greenness (negative), and b^* representing the degree of yellowness (positive) and blueness (negative). K/S values given by the reflectance spectrophotometer were calculated at λ_{max} and are directly correlated with the dye concentration on the substrate according to the Kubelka-Munk equation [26]. The colorimetric data of silk, wool, and cotton fibres for all dyes are summarized in Tables 6, 7, and 8. The K/S value graph of all the dyes is shown in Figure 1 and graphs of b^* versus a^* for silk, wool, and cotton fibres are shown in Figures 2, 3, and 4, respectively.

For silk fibre, the data summarized in Table 6 showed that dye 6l is the lightest and 6k is the darkest. Dye 6k is the reddest and dye 6e is the least red, dye 6j is the most yellow and dye 6e is the least yellow, dye 6a is the bluest. Dye 6k has the highest K/S value and dye 6e has the lowest K/S value.

For wool fibre, the data showed in Table 7 that dye 6l is the lightest and 6k is the darkest. Dye 6i is the reddest and dye 6a is the least red, dye 6l is the most yellow and dye 6k is the least yellow, and dye 6a is the bluest. Dye 6k has the highest K/S value and dye 6d has the lowest K/S value.

For cotton fibre, in Table 8 the data showed that dye 6e is the lightest and dye 6k is the darkest. Dye 6k is the reddest and dye 6a is the least red, dye 6j is the most yellow and dye 6e is the least yellow. Dye 6a is the bluest. Dye 6k has the highest K/S value and dye 6e has the lowest K/S value.

Table 6:- Color measurement (CIELab) data of dyes (6a-6l) on silk fibre.

Dye No.	L^*	a^*	b^*	C^*	H^*	K/S
6a	36.86	13.02	-2.58	13.28	348.79	5.42
6b	47.89	16.33	12.53	20.59	37.50	3.41
6c	54.33	10.64	15.52	18.82	55.57	3.15
6d	60.01	11.11	16.05	19.52	55.31	2.04
6e	59.38	9.54	10.81	14.42	48.56	1.68
6f	52.34	13.50	18.09	22.57	53.26	3.77
6g	57.34	12.73	15.56	20.10	50.70	2.10
6h	65.95	13.06	22.57	26.07	59.93	1.72
6i	48.56	18.33	15.44	23.97	40.10	3.93
6j	61.72	14.21	30.68	33.81	65.16	2.91
6k	27.92	20.31	-0.58	20.31	358.35	13.26
6l	66.05	13.16	23.81	27.21	61.07	1.82

Table 7:- Color measurement (CIELab) data of dyes (6a-6l) on wool fibre.

Dye No.	L^*	a^*	b^*	C^*	H^*	K/S
6a	40.60	10.16	-1.20	10.23	353.28	3.98
6b	49.38	17.97	16.31	24.27	42.23	3.92
6c	57.20	11.25	15.45	19.11	53.94	2.78
6d	62.30	11.05	17.73	20.89	58.07	2.19
6e	55.63	14.29	18.41	23.31	52.20	2.99
6f	52.47	13.72	18.62	23.13	53.61	4.08
6g	38.02	12.73	10.67	16.61	39.97	6.43
6h	62.70	14.19	26.65	30.19	61.97	2.91
6i	46.14	21.40	16.10	26.78	36.96	4.80
6j	57.89	15.15	28.42	32.21	61.94	3.98
6k	26.49	20.23	0.81	20.25	2.28	15.41
6l	63.48	14.48	29.28	32.66	63.69	3.20

Table 8:- Color measurement (CIELab) data of dyes (6a-6l) on cotton fibre.

Dye No.	L*	a*	b*	C*	H*	K/S
6a	40.48	-1.41	-14.42	14.49	264.40	4.76
6b	51.80	12.70	2.78	13.00	12.35	2.02
6c	67.95	7.31	11.07	13.27	56.55	0.95
6d	55.15	10.75	12.47	16.46	49.25	2.16
6e	78.22	5.25	6.99	8.74	53.06	0.30
6f	55.83	16.24	19.70	25.53	50.49	2.70
6g	73.56	10.81	15.54	18.93	55.19	0.68
6h	72.77	14.18	21.35	25.63	56.41	0.93
6i	56.97	12.71	13.21	18.33	46.12	2.03
6j	62.38	14.25	27.71	31.16	62.79	2.44
6k	28.99	22.72	-6.87	23.73	343.18	11.61
6l	72.86	14.35	20.77	25.25	55.36	0.90

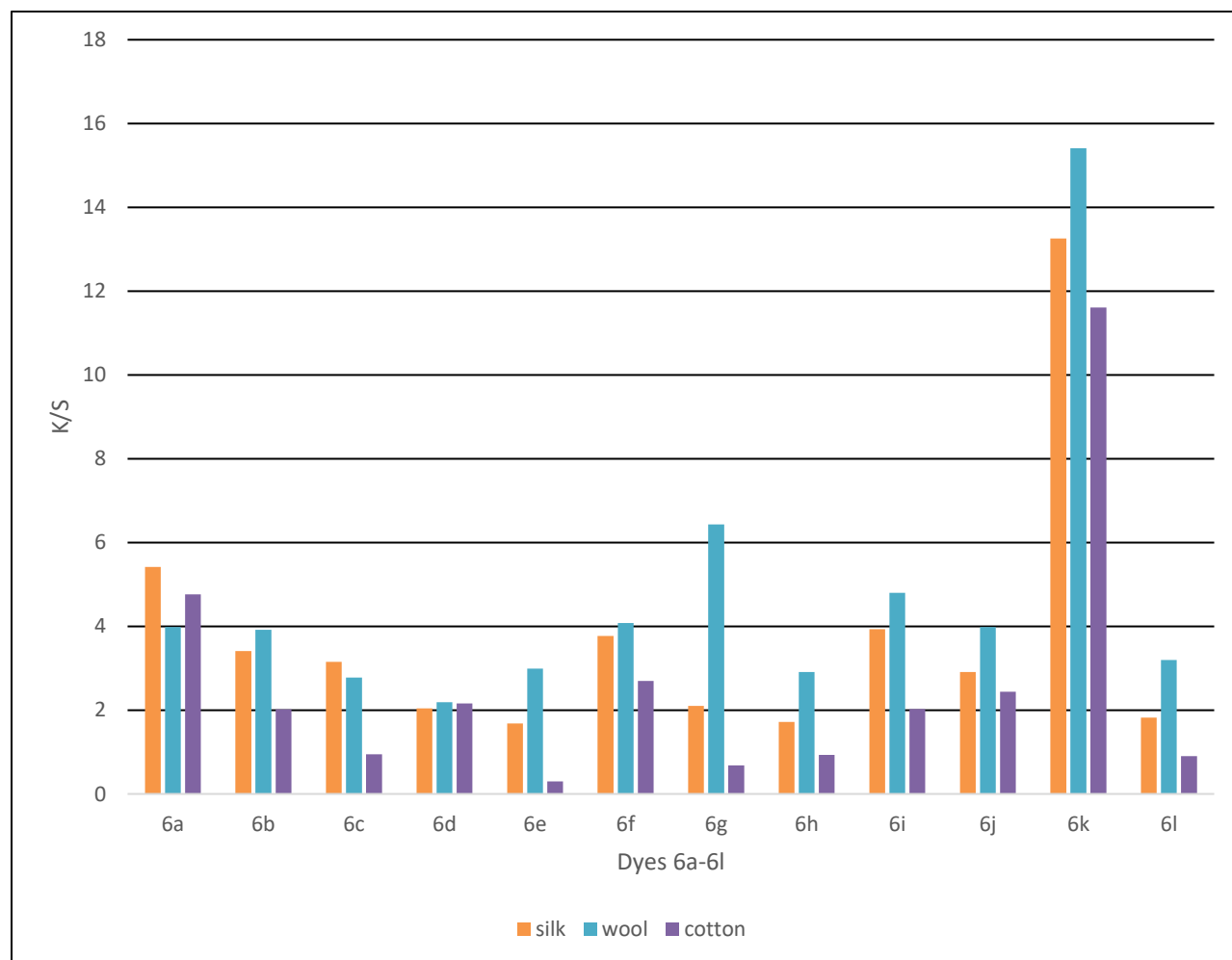
Figure 1:- Graph of K/S values of dyes (6a-6l).

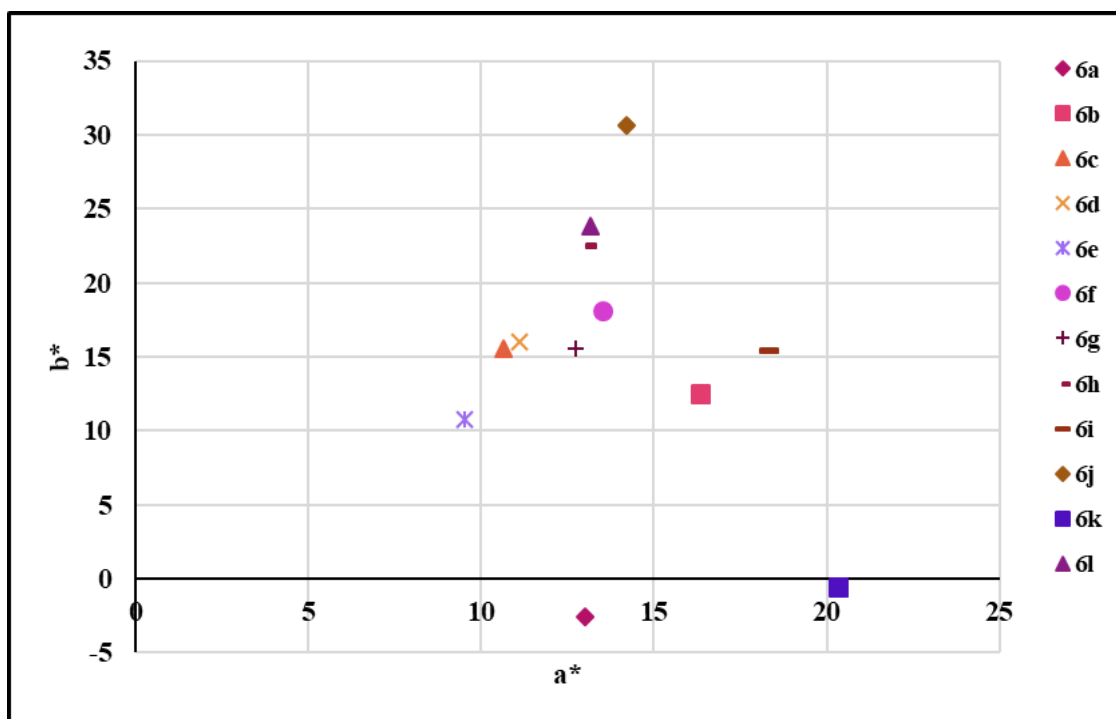
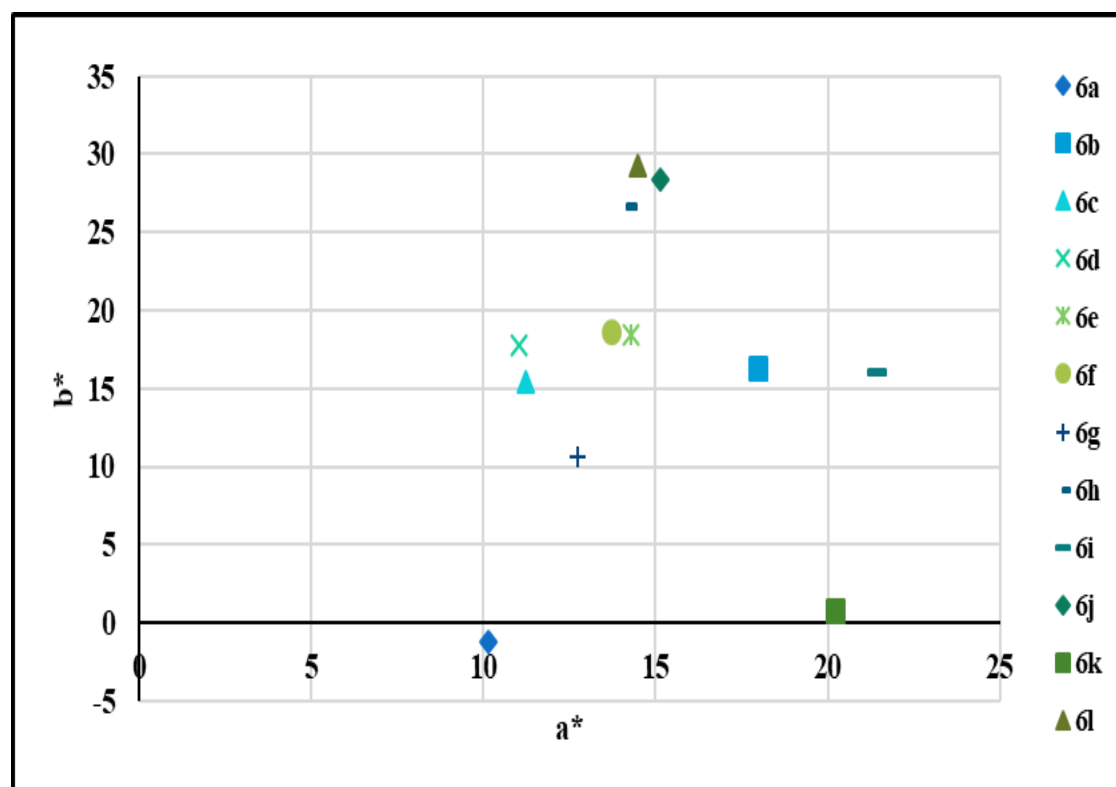
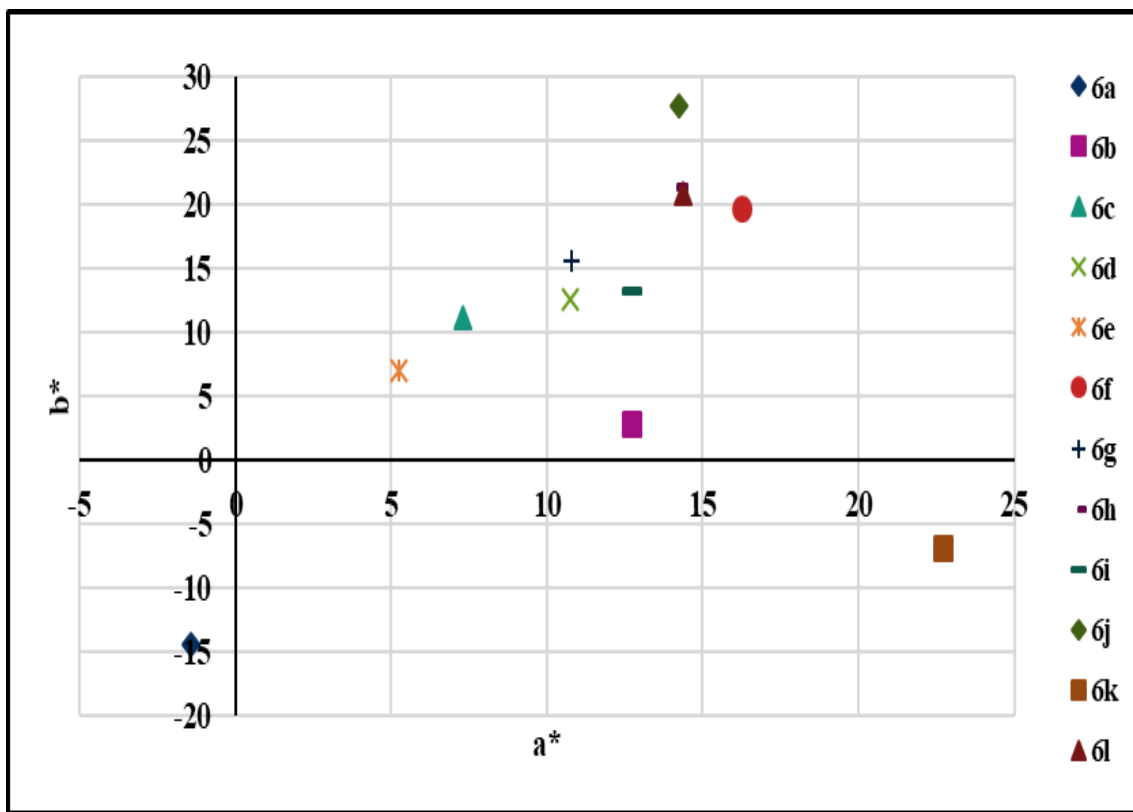
Figure 2:- Graph of b^* vs. a^* for silk fibre.**Figure 3:-** Graph of b^* vs. a^* for wool fibre.

Figure 4:- Graph of b^* vs. a^* for cotton fibre.**Conclusion:-**

A series of new water soluble reactive dyes derived from Quinazolin-4(3H)-one moiety were synthesized in good yield. The synthesized dyes gave brown to blue-violet shades on silk, wool and cotton fibres, showed good to very good with respect to washing, rubbing, and light fastness. The presence of azo group have major influence on color coordinates (L^* , a^* , b^* and K/S). Dye 6k has the highest K/S value for silk, wool and cotton fibres. Thus, dye 6k has more dye concentration on silk, wool and cotton fabric.

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References:-

1. H. Zollinger, Color Chemistry, 2nd Ed., Weinheim: VCH 167 (1991).
2. M. R. Fox and H. H. Sumner, "Dyeing with Reactive Dyes" (C. Preston Ed.), pp.142-195, The Dyers' Company Publication Trust, England, 1986.
3. R. M. Christie, "Color Chemistry, pp.135-147, The Royal Society of Chemistry, UK, 2001.
4. I. D. Rattee, Rev. Prog. Coloration, 14, 50 (1984).
5. W. J. Epolito, Y. H. Lee, L. A. Bottomley, S. G. Pavlostathis, Dyes Pigm., 67 (2005) 35.
6. W. E. Stephen and I. D. Rattee, J. Soc. Dyers Color., 76, 6 (1960).
7. E. Siegel, "The Chemistry of Synthetic Dyes" (K. Venkataraman Ed.), Vol. VI, p.131 Academic Press, New York and London, 1972.
8. FH, DAS 1153029 (1960).
9. D. R. Patel, and K. C. Patel, Fiber. Polym., 11, 537-544 (2010).
10. D. R. Patel, and K. C. Patel, Arab. J. Chem., 4, 279-285 (2010).
11. D. R. Patel, J. T. Bilimoriya and K. C. Patel, V. N. S. G. U., J. Sci. Technol., 1, 128 (2009).

12. J. P. Luttringer, Text. Chem. Colorist, 25, 25 (1993).
13. S. Abeta, T. Yoshida, and K. Imada, Am. Dyestuff Reporter, 73, 26 (1984).
14. D. R. Patel A. L. Patel, and K. C. Patel, Arab. J. Chem., 9, S161 (2016).
15. D. R. Patel, B. M. Patel, N. B. Patel, and K. C. Patel, J. Saudi Chem. Soc., 18, 245 (2014).
16. A. B. Patel, P. I. Vashi, V. D. Patel, D. H. Patel, K. C. Patel, and P. S. Patel, J. Appl. Chem., 8, 1091 (2019).
17. S. M. Patel, E. A. Gamit, S. S. Savant, S. V. Patel, H. U. Hajiyani, R. B. Tailor, P. S. Patel and K. C. Patel, Int. J. Rec. Sci. Res., 8, 19793 (2017).
18. P. S. Patel, D. H. Patel and K. C. Patel, Fiber. Polym., DOI 10.1007/s12221-021-0413-3 (2020).
19. B. Fried, and J. Sherma,, "Thin Layer Chromatography Techniques and Application", Marcel-ekker Inc., New York and Basel, 1982.
20. D. R. Patel, A. L. Patel, K. C. Patel and L. A. Patel, Colorage, 57, 72 (2010).
21. N. B. Patel, J. C. Patel, Sci. Pharm., 78, 171-193 (2010).
22. X. Gao, X. Cai, K. Yan, B. Song, L. Gao and Z. Chen, Molecules, 12, 2621 (2007).
23. M. H. Mistry, S. J. Parmar, G. C. Desai, J. Chem. Pharm. Res., 3, 831-837 (2011).
24. V. A. Shenai, "Technology of Textile Processing", Vol. VI, Technology of Dyeing, p.260, Sevak Publisher, Bombay, 1994.
25. H. R. Maradiya, J. Serb. Chem. Soc., 67, 709 (2002).
26. F. W. Billmeyer and M. Saltzman, "Principles of Color Technology", 2nd ed., Wiley, 1981.