King Saud University

Journal of Saudi Chemical Society

www.ksu.edu.sa
www.sciencedirect.com

ORIGINAL ARTICLE

Disperse dyes based on 2-aminothiazole derivatives for cellulose triacetate fabric

Hari Raghav Maradiya *

V.P. and R.P.T.P. Science College, Vallabh Vidyanagar, Gujarat 388 120, India

Abstract   A range of azo disperse dyes was prepared by coupling diazotized 2-amino-4-(*p*-nitrophenyl)-5-nitrothiazole with various substituted arylamines. Spectral properties in the infrared and visible range of the dyes obtained were investigated. All the dyes, when applied on cellulose triacetate fabric as 2% shade, showed fairly good to very good light fastness and very good to excellent fastness to washing, perspiration, rubbing and sublimation. All the dyes gave a wide range of reddish brown to indigo shades with very good depth and levelness on fabric. The purity of dyes was checked by thin layer chromatography. The percentage dyebath exhaustion and fixation on fabric was reasonably good and acceptable.

1. Introduction

Monoazo disperse dyes with thiazole diazo components have been intensively investigated to produce bright and strong colour shades ranging from red to greenish blue on synthetic fiber. These results led to commercial products to replace the conventional azobenzene disperse dyes (Colour Index, 1971). Most heterocyclic dyes of technical interest for application to textiles are derived from diazo components consisting of five-membered rings containing one sulphur heteroatom and to which a diazotizable amino group is directly attached, the ring may also possess one or more nitrogen heteroatoms. The discovery of the fusion reaction between *p*-toluidine and sulphur to give dehydrothio-*p*-toluidine in 1887 constitutes the beginning of thiazole dyestuff technology. Colour Index described various basic, direct, vat and disperse dyes wherein thiazole nucleus occurs (Annen et al., 1987).

Before 1950, almost all the disperse blue dyes used were prepared from the anthraquinone class and had the limitations of poor dischargeability and sensitivity to oxide of nitrogen. Derivative of 2-aminothiazole has a long history of use as heterocyclic diazo components for disperse dyes (Shuttleworth and Weaver, 1990). Blue dischargeable monoazo derivatives based on 2-amino-5-nitrothiazole caught the attention of dye chemists in the 1950. With colourants (Hu et al., 1987) such as C.I. Disperse Blue 339 having been produced commercially since then, 2-aminothiazole containing nitro groups are prized for their bathochromism and high tinctorial strength relative

to their carbocyclic analogues (Schwander, 1982). These properties, which have aided the rise to prominence of such dyes as replacement for blue anthraquinone colourants (Egli, 1991) were first reported by Dickey nearly 40 years ago (Dickey et al., 1959). 2-Amino-5-nitrothiazole could be used to produce bright blue dyes with adequate light fastness combined with excellent dischargeability, gas fastness and dyeability on hydrophobic fabric. The commercial success of dyes for cellulose triacetate, which were derived from 2-amino-5-nitrothiazole, resulted in considerable interest in this class of dyes. Early dyes showed moderate light fastness on cellulose triacetate and polyesters, and light fastness on polyamides was very poor. Dyes with increased light fastness and/or sublimation fastness were prepared from aniline type coupling components containing one or more N-alkyl groups substituted with cyano (Gosei Senryo Gijutsu Kenkyu Kumiai, 1983a), amylcarbonyloxy (Gosei Senryo Gijutsu Kenkyu Kumiai, 1983b), alkoxy (Mitsubishi Chemical Industries Co. Ltd., 1979a,b), sulphato (Sumitomo Chemical Co. Ltd., 1981), diphenylamine (Erwin et al., 1991), pyridinium (Parton, 1978), dicarboximide (Weaver and Straley, 1970), l-naphthylamine (Modena and Risaliti, 1977) and N-alkyl-1,2,3,4-tetrahydroquinolines (Gosei Senryo Gijutsu Kenkyu Kumiai, 1984) were used as coupling components. These coupler types have the advantages of providing dyes, which have a neutral or green cast when exposed to artificial light, whereas the corresponding dyes from N-alkylaniline couplers have an extremely red cast. The improved fastness to light and sublimation provided by 3-acylaminoaniline type coupling components was not overlooked and dyes were patented (Nippon Kayaku Co. Ltd., 1979; Peter, 1979; Mitsubishi Chemical Industries Co. Ltd., 1979a,b; Gosei Senryo Gijutsu Kenkyu Kumiai, 1982). Greenish blue dyes can be prepared by choosing a coupler containing electron donating groups such as 5-acetamido-2-methoxyaniline derivatives. Additional early work on the 2-thiazolylazo dyes resulted in violet dyes from 2-amino-4-trifluoromethyl-5-carbalkoxythiazole (Towne and Hill, 1956), 2-amino-5-alkylsulphonylthiazole (Merian, 1961), 4-halo-5-formylthiazole (Egli, 1979) and even more bathochromic dyes from 2-amino-4-alkylsulphonyl-5-nitrothiazole (Towne et al., 1958).

During the last 15 years, the level of interest as indicated by the patent literature has grown in the field of thiazolylazo disperse dyes (Tsien et al., 1995; Sens and Etzbach, 1994; Beckmann et al., 1994). There has been a considerable growth in the volume of data reporting outside of patent literature (Sekar, 1994; Peters and Gbadamosi, 1995; Bello et al., 1995; Desai et al., 1995; Bello, 1995; Griffihs and Riepl, 1998; Chikhalia and Desai, 1998; Rangnekar et al., 1998; Hallas and Choi, 1999; Towns, 1999; Bajpai et al., 2000; Rangnekar and Malankar, 2000; Elkholy et al., 2001; Helal, 2001, 2004; Yao, 2001; Sonawane and Rangnekar, 2002; Georgiadou and Tsatsaroni, 2002; El-Mossalomy and Ibrahim, 2002; Kaparir et al., 2004; Saylam et al., 2008; Helal et al., 2008; Abdet-Latif et al., 2009). Earlier we had studied the disperse dyes based on 2-aminothiazole derivatives (Maradiya and Patel, 2000, 2001a,b,c,d,e, 2002a,b, 2003; Maradiya, 2002, 2004a,b, 2009, 2010).

The encouraging results prompted to extend the study on thiazole moiety. Hence, a series of monoazo dyes of general formula shown in Fig. 1 were synthesized and tested as disperse dyes for cellulose triacetate fabric. In addition to the characterization of the dyes, an evaluation of their technical properties and a colour assessment was performed.
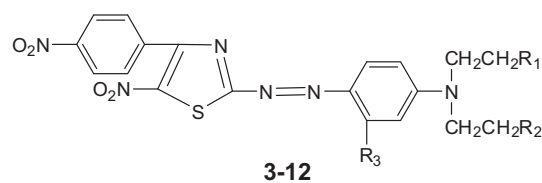

Figure 1 General structure of dyes **3–12**; **3** $R_1 = R_2 = OH$, $R_3 = H$; **4** $R_1 = R_2 = OAc$, $R_3 = H$; **5** $R_1 = R_2 = OH$, $R_3 = H$; **6** $R_1 = R_2 = OH$, $R_3 = Cl$; **7** $R_1 = R_2 = OH$, $R_3 = NHAc$; **8** $R_1 = R_2 = OAc$, $R_3 = CH_3$; **9** $R_1 = R_2 = Cl$, $R_3 = H$; **10** $R_1 = R_2 = CN$, $R_3 = H$; **11** $R_1 = R_2 = CN$, $R_3 = CH_3$; **12** $R_1 = R_2 = CN$, $R_3 = Cl$.

2. Experimental

2.1. Materials and methods

All of the chemicals used were of commercial grade and were further purified by crystallization and distillation. All solvents used were either of analytical grade or redistilled commercial grade.

2.2. Preparation of 2-amino-4-phenylthiazole (1)

The title compound was synthesized by the following method.

Bromine (160 g, 2.0 mol) was added dropwise with stirring to acetophenone (120 g, 1.0 mol) and thiourea (152 g, 2.0 mol). The mixture was heated overnight on the steam bath, diluted with 2.5 l hot water (85 °C), stirred, and filtered hot. The filtrate was cooled, made slightly basic with concentrated ammonium hydroxide, and filtered. The solid residue recrystallized from ethanol gave 126 g (71%) yield of 2-amino-4-phenylthiazole, m.p. 143–144 °C, literature m.p. 144–145 °C (see Scheme 1).
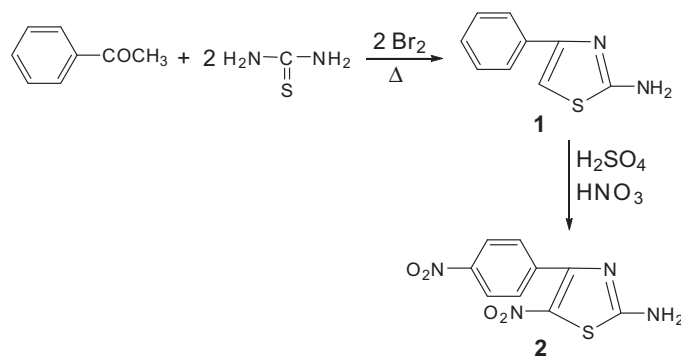
2.3. Preparation of 2-amino-4-(p-nitrophenyl)-5-nitrothiazole (2)

The title compound was synthesized by the following method.

To 47 ml (0.88 mol) of concentrated sulphuric acid in a 100 ml three-necked flask, 8.8 g (0.05 mol) of 2-amino-4-phenylthiazole was added at 10–14 °C with stirring. The stirred solution was treated at 3–5 °C over a period of 30 min with a mixture of 7.0 g (0.11 mol) of fuming nitric acid (sp. gr. 1.49–1.50) and 6 ml of concentrated sulphuric acid, stirring was continued for 15 min at this temperature. After the stirred reaction mixture warmed to room temperature, it was allowed to stand overnight, protected from moisture and the reaction mixture was poured into 500 ml of an ice–water mixture, yielding a reddish solid, which was collected on a filter. The product was washed with cold water, then with saturated sodium acetate solution until it was neutral to Congo Red paper and then the product was washed with water. The crude product has m.p. 221–226 °C, weight 10.5 g (78%). A sample recrystallized from a benzene–ethyl alcohol mixture melted with decomposition at 249–251 °C, literature m.p. 250–251 °C (see Scheme 1).

2.4. Preparation of 2-[4′-[N,N-bis(2-hydroxyethyl)amino]phenylazo]-4-(4″-p-nitro phenyl)-5-nitrothiazole (3)

Synthesis of **3** involves two steps.

Scheme 1 Synthetic route of compounds **1** and **2**.

2.4.1. Diazotization of 2-amino-4-(p-nitrophenyl)-5-nitrothiazole (2)

Dry sodium nitrite (1.38 g, 0.02 mol) was slowly added with stirring to concentrated H_2SO_4 (1.2 ml) on a water bath, allowing the temperature to rise to 65 °C but no higher. The solution was then cooled to 5 °C and a mixture of 20 ml acetic acid and propionic acid (17:3) was added dropwise with stirring, allowing the temperature to rise to 15 °C but no higher. The reaction mixture was then cooled to 0–5 °C, and 2-amino-4-(p-nitrophenyl)-5-nitrothiazole (**2**) (5.32 g, 0.02 mol) was added portion wise and stirring was continued at this temperature for 2 h. The excess nitrous acid (given positive test on starch-iodide paper) was decomposed with urea. The clear diazonium salt solution thus obtained was used immediately in the coupling reaction as exemplified below (see Scheme 2).

2.4.2. Coupling with N,N-bis(2-hydroxyethyl)aniline

Aryl bis(2-hydroxyethyl)aniline (3.62 g, 0.02 mol) was dissolved in 10 ml acetic acid and 5 ml water together 5 g with sodium acetate. This solution was cooled in an ice-bath and the diazonium solution previously prepared was added dropwise over 30 min with vigorous stirring. The pH was maintained between 4 and 5 by simultaneous addition of 10% sodium acetate solution. Stirring was continued for 2 h, allowing the temperature to rise to ambient. The dye was then filtered off, washed with warm and cold water until acid-free, dried at 50 °C in an oven to give azo dye **3**. Recrystallization from ethanol provided the pure dye. The characterization data are given in Table 1.

The same procedure was repeated for the preparation of the dyes **4–12**. The synthetic scheme is shown in Scheme 2.

2.5. Dyeing method

Dyeing of cellulose triacetate fabric was carried out according to the following procedure.

A convenient laboratory method for dyeing polyester is to employ high temperature (130–135 °C) and high pressure (24–30 psi), using a laboratory dyeing machine with a glycerin-bath. A paste of finely powdered dye (40 mg) was prepared with dispersing agent Dodamol (80 mg), wetting agent Tween-80 (5 mg) and water (1 ml) in a ball mill. To this paste, water (99 ml) was added with stirring and the pH was adjusted to 4.5–5 using acetic acid. The previously mentioned dye suspension (100 ml) was added to a beaker provided with a lid and a screw cap. Before closing the lid and tightening the metal cap over the beaker, a wetted pattern of polyester fabric was rolled into the beaker. The beaker was 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 raised to 130 °C at the rate of $2\,°C\,min^{-1}$. The dyeing was continued for 1 h, under pressure. After cooling for 1 h, the beaker was removed from the bath and washed with water. The dyed pattern was thoroughly washed with hot water at 50 °C and then with cold water and dried at room temperature.

2.6. Determination of the percentage exhaustion and fixation

The dyebath exhaustion percentage (% E) and fixation percentage (% F) of dyed fabric were determined according to the known method (Maradiya and Patel, 2001c).
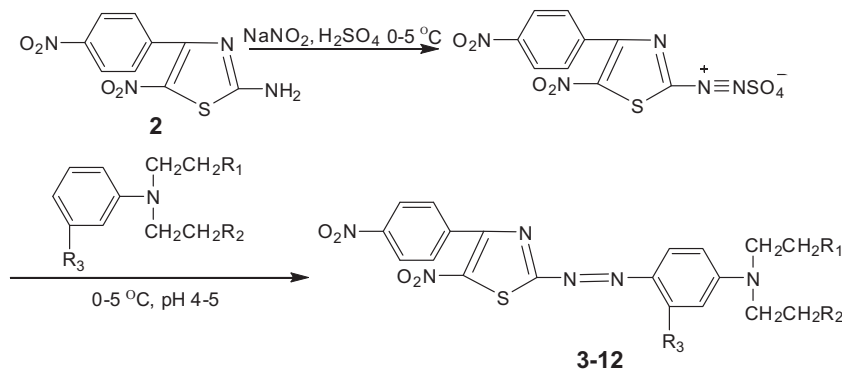
Scheme 2 Synthetic route of dyes **3–12**.

Table 1 Characterization data, intensities, exhaustion and fixation of dyes.

Dye no.	Melting point (°C) (C_2H_5OH)	Yield (%)	R_f value	Absorption max. (λ_{max}/nm)		log ε (DMF)	Cellulose triacetate	
				DMF	Conc. H_2SO_4		% F	% E
3	125–127	81	0.81	525	470	4.47	76	88
4	95–96	86	0.79	540	465	4.69	75	92
5	124–125	78	0.89	540	475	4.60	84	89
6	118–120	80	0.90	520	463	4.23	78	91
7	113–115	79	0.78	545	460	4.80	84	90
8	98–99	82	0.83	550	470	4.82	77	91
9	125–126	81	0.96	510	460	4.75	85	89
10	140–141	85	0.78	505	480	4.41	84	87
11	160–161	82	0.77	510	483	4.23	83	89
12	127–128	76	0.92	500	460	4.55	85	92

2.7. Colour fastness test

The fastness to light, sublimation and perspiration were assessed in accordance with BS: 1006-1978. The wash fastness test in accordance with IS: 765-1979. The details of the various fastness tests are reported in literature (Maradiya and Patel, 2001c).

2.8. General

Melting points were determined by open capillary method. The visible absorption spectra were measured in a Carl Zeiss UV/VIS Specord spectrometer. Elemental analysis was carried out on a Carlo Erba Elemental Analyser 1108. Infrared spectra were recorded in KBr pellets, on a Perkin-Elmer model 983 Spectrophotometer.

3. Results and discussion

3.1. Preparation of diazo component 2

Acetophenone was reacted overnight on a steam bath with thiourea in equimolar ratio in the presence of 1 mol bromine (Dickey and Towne, 1956). The nitration of compound 1 with 2 mol of nitric acid gave compound 2 (Dickey et al., 1959). In each case, the thiazole derivative obtained from the reaction mixture agreed in IR and physical properties with those reported in the literature (Dickey et al., 1959). The compound 2 obtained was recrystallized in ethanol. The nitro group reduces the basicity of the compound 2 and thus efficient diazotization can only be achieved using nitrosylsulphuric acid obtained from dry solid sodium nitrite and concentrated H_2SO_4. The resulting diazonium salt solution was coupled to N,N-dialkylaniline derivatives to obtain dyes. In order to determine the end point of diazotization, it was found useful to check the presence of unreacted diazo component on thin layer chromatography (TLC) by sampling the diazotization mixture and extracting with ethyl acetate. Thus, when unreacted diazo component no longer persisted on TLC, the diazotization was ended. The diazonium salt solution was used immediately since this decomposed on standing, even when cold. Subsequent coupling reactions took place readily on adding the diazonium salt continuously to the solution of the coupling component in acetic acid. Some evidence of decomposition usually accompanied coupling; however by careful addition of the diazonium salt solution at 0–5 °C to a solution

of the coupling component in acetic acid, a 76–86% yield of the dye was usually obtained. To complete the coupling particularly for reaction using nitrosylsulphuric acid in the previous diazotization, the pH of the reaction mixture was eventually adjusted to approximately 4–5. Thus, an appropriate amount of 10% sodium acetate solution was slowly added below 5 °C.

3.2. Physical properties of dyes

All the solid dyes are dark brown in colour. The purity of the dyes was checked by TLC using methanol–water–acetic acid (12:3:7, v/v) as the solvent system. When adsorbed onto silica chromatography plates, each dye produced a reddish-violet colour sport. All the dyes recrystallized from ethyl alcohol and exhibited well-defined melting points characteristic of pure compounds. It would be premature to attempt to explain in detail their relative values because of the complex dependence of the melting point on a number of factors (e.g., polarity, geometry, size, and molecular interaction). The purified dyes were all found to have satisfactory elemental analysis (Table 2).

3.3. Visible absorption spectroscopic properties of dyes

The visible absorption spectroscopic properties of dye 3–12 were recorded in DMF and concentrated H_2SO_4 solution, and are presented in Table 1, The value of the logarithm of molar extinction coefficient (log ε) of the dyes 3–12 was in the range of 4.23–4.82, consistent with their high intensity of absorption. One cause of the increased intensity might be attributed to the greater planarity of the thiazole dyes, because of the lower steric interaction of a five-membered ring. The colour of the dyes 3–12 is affected by substituents in the coupler constituent. The bathochromism of the dyes is an inherent property of the thiazole system itself; the bathochromism does not derive from the substituent groups. However, the greater polarizability of the thiazole system (relative to the benzene system) affecting a general bathochromism of the dyes. As can be seen from the data in Table 1, additional bathochromic shifts can also be obtained by enhancing the electron donor properties of the coupler ring, e.g. by introducing an acetamido group *ortho* to the azo group, as in dye 7. This dye, however, gave shift of 20 nm relative to dye 3. The acetamido group is a weak electron donor, but more importantly, it can take part in hydrogen bonding with the azo group. This enhances the planarity of the dye containing the acetamido group. When replacing the OH group in dye 3 and 5 by the

Table 2 Elemental analysis of dye.

Dye no.	Molecular formula	Molecular weight	C%		H%		N%	
			Require	Found	Require	Found	Require	Found
3	$C_{19}H_{18}N_6O_6S$	458.49	49.76	49.60	3.96	3.75	18.33	18.20
4	$C_{23}H_{22}N_6O_8S$	542.57	50.91	50.76	4.09	3.86	15.49	15.28
5	$C_{20}H_{20}N_6O_6S$	472.52	50.83	50.62	4.27	4.08	17.78	17.56
6	$C_{19}H_{17}N_6O_6SCl$	492.91	46.29	46.10	3.48	3.25	17.05	16.82
7	$C_{21}H_{21}N_7O_7S$	515.55	48.92	48.49	4.11	3.89	19.02	18.88
8	$C_{24}H_{24}N_6O_8S$	556.60	51.78	51.60	4.35	4.30	15.10	14.89
9	$C_{19}H_{16}N_6O_4SCl_2$	495.33	46.06	45.83	3.26	3.10	16.97	16.69
10	$C_{21}H_{16}N_8O_4S$	476.51	52.92	52.79	3.39	3.18	23.52	22.70
11	$C_{22}H_{18}N_8O_4S$	490.54	53.86	53.58	3.70	3.50	22.84	22.61
12	$C_{21}H_{15}N_8O_4SCl$	510.93	49.36	49.12	2.96	2.78	21.93	21.68

Table 3 Dyeing properties of dye.

Dye no.	Shade on cellulose triacetate	Fastness properties				
		Light	Wash	Perspiration fastness		Sublimation
				Acid	Alkaline	
3	Reddish violet	5–4	5–4	5–4	5	5
4	Indigo	4–5	5–4	5–4	5–4	5
5	Indigo	5–4	4–5	5–4	5–4	5
6	Reddish blue	4–5	5–4	4–5	5–4	5
7	Indigo	7–8	5	5	5	5
8	Indigo	5–4	5–4	5–4	5–4	5
9	Reddish violet	5–4	5–4	5-4	5–4	5
10	Reddish brown	5–4	5–4	5–4	5–4	5
11	Reddish violet	6–5	5	5	5	5
12	Maroon	5–6	5	5–4	5–4	5

Cl and CN groups, the resulting dyes **9** and **11** are absorbed at 510 nm. The hypsochromic shift is due to a decrease in the donor strength of the coupler ring.

The results summarized in Table 1, clearly shows that the incorporation of electron withdrawing group in this position leads to red shift. Similarly, when the electron donating ability of the group on the coupler ring enhanced the depth of the colour in the resulting dyes. The shift in λ_{max} to longer wavelengths is due to increase in polarisability of the dye molecule resulting from enhanced density movement from the donor groups (coupler) to the acceptor group (diazo ring).

3.4. Infrared spectra of dyes

The C–H stretching vibrations of aromatic ring appear at 700–820 cm^{-1} and 1580–1595 cm^{-1}. The strong absorption band in the range 1500–1510 cm^{-1} is corresponding to C–N stretching vibration. The N–H stretching vibration confirmed at 3400–3450 cm^{-1}. The azo and O–H group stretching vibration band appears at 1500–1525 cm^{-1} and 3500–3600 cm^{-1}, respectively. The C–H stretching vibration of alkyl group appears at 2895–2900 cm^{-1}.

3.5. Dyeing properties of dyes

The disperse dyes **3**–**12** were applied at 2% depth on cellulose triacetate fabric. Their dyeing properties are given in Table 3. These dyes gave a wide range of colour varying from reddish brown to indigo shades with good levelness, brightness and depth on the fabric. The variation in the shades of the dyed fabric result from both the nature and position of the substituent present on the coupler ring. For example, introduction of an acetamido group *ortho* to the azo linkage in dye **7** results in a significant improvement in the light fastness. This may be due to the intramolecular hydrogen bond formed between the acetamido group and azo group. The dyeings showed fairly good to very good fastness to light with very good to excellent fastness to washing, rubbing, perspiration and sublimation, A remarkable degree of levelness after washing indicated good penetration and affinity of these dyes to the fabric.

3.6. Exhaustion and fixation properties of dyes

Table 1 shows that the fixation and exhaustion values of dyes **3**–**12** are in the range of 87–92% and 75–85%, respectively. The exhaustion values depends on the structure of fabric, diffusion rate and dye molecules. The fixation values related to affinity of the dyes to the fabric. The substantivity of the dye, which determines its tendency to partition in favour of the fabric, depends on factors such as molecular size, geometry and polarity of the dye molecule.

4. Conclusion

A series of thiazolylazo disperse dyes containing various N, N-dialkylaniline residues have been synthesized and their

colour properties examined both in solution and on application to cellulose triacetate fabric. The colour vary from reddish brown to indigo shades on fabric and offer excellent affinity and intensity of colour. Their synthesis is generally convenient and economical. The small size of thiazolylazo dyes provided better dyeability. The intrinsic conjugation in the thiazolylazo structure results in excellent colour strength. The outstanding property of the thiazolylazo dyes is their high sublimation fastness. The compactness of structure improves sublimation fastness. The nature of the substituent in the coupling components has a little influence on the visible absorption and shade of the dyeing. Furthermore these dyes show level dyeing and display excellent dischargeability which is an additional dyeing property.

Acknowledgement

References

Abdet-Latif, E., Amer, F.A., Metwally, M.A., Khalifa, M.E., 2009. Pigm. Resin Technol. 38, 105–110.

Annen, O., Egli, R., Hansler, R., Henzi, B., Jakob, H., Matzinger, P., 1987. Rev. Prog. Colour. 17, 72–85.

Bajpai, R., Tiwari, S., Tiwari, K.S., Tiwari, B.M.L., 2000. Asian J. Chem. 12, 1199–1208.

Beckmann, S., Etzbach, K.-H., Sens, R. 1994. PTC Int. Appl. WO 9,529,958.

Bello, K.A., 1995. Dyes Pigm. 27, 45–54.

Bello, K.A., Martins, C.M.O.A., Adamu, I.K., 1995. Colourage 42, 29–32.

Chikhalia, K.H., Desai, K.R., 1998. J. Inst. Chem. (India) 70, 142–143.

Colour Index, 1971. Society of Dyers and Colourist, third ed. America Association of Textiles Chemist and Colourist, Bradford. p. 3.

Desai, K.R., Modi, B.R., Mistry, B.D., 1995. J. Indian Chem. Soc. 72, 141–142.

Dickey, J.B., Towne, E.B., 1956. US Patent 2,730,523.

Dickey, J.B., Towne, E.B., Bloom, M.S., Moore, W.H., Hill, H.M., Heynemann, H., Hedberg, D.G., Sievers, D.C., Ottis, M.V., 1959. J. Org. Chem. 24, 187–196.

Egli, R., 1979. US Patent 4,395,544.

Egli, R., 1991. The design and synthesis of organic dyes and pigments. In: Peters, A.T., Freeman, H.S. (Eds.), Colour Index. Elsevier, London, pp. l–27.

Elkholy, Y.M., Erian, A., Helal, M.H., 2001. Pigm. Resin Technol. 30, 168–171.

El-Mossalomy, E.H., Ibrahim, A.A., 2002. Pigm. Resin Technol. 31, 375–380.

Erwin, H., Matthias, W., Helmut, R., Sabine, G., 1991. Ger. Offen DE 4120362.

Georgiadou, K.L., Tsatsaroni, E.G., 2002. Dyes Pigm. 53, 73–78.

Gosei Senryo Gijutsu Kenkyu Kumiai, 1982. JP 5,991,152.

Gosei Senryo Gijutsu Kenkyu Kumiai, 1983a. Jpn. Kokai Tokkyo Koho JP 59,213,765.

Gosei Senryo Gijutsu Kenkyu Kumiai, 1983b. Jpn. Kokai Tokkyo Koho JP 59,170,142.

Gosei Senryo Gijutsu Kenkyu Kumiai, 1984. Jpn. Kokai Tokkyo Koho 5,920,356.

Griffihs, J., Riepl, C.J., 1998. Chem. Commun. (Cambridge) 13, 1349–1350.

Hallas, G., Choi, J.-H., 1999. Dyes Pigm. 42, 249–265.

Helal, M.H., 2001. Pigm. Resin Technol. 30, 296–300.

Helal, M.H., 2004. Pigm. Resin Technol. 33, 165–171.

Helal, M.H., Elgemele, G.H., Masoud, D.M., 2008. Pigm. Resin Technol. 37, 402–409.

Hu, J.-Z., Skrabal, P., Zollinger, H., 1987. Dyes Pigm. 8, 189–195.

Kaparir, M., Cansiz, A., Ahmedzade, M., 2004. Russ. J. Org. Chem. 40, 1813–1818.

Maradiya, H.R., 2002. J. Saudi Chem. Soc. 6, 299–310.

Maradiya, H.R., 2004a. Adv. Colour. Sci. Technol. 7, 29–35.

Maradiya, H.R., 2004b. J. Saudi Chem. Soc. 8, 495–503.

Maradiya, H.R., 2009. Chem. Heterocycl. Compd. 10, 1558–1563.

Maradiya, H.R., 2010. J. Saudi Chem. Soc. 14, 77–81.

Maradiya, H.R., Patel, V.S., 2000. J. Serb. Chem. Soc. 65, 773–780.

Maradiya, H.R., Patel, V.S., 2001a. Adv. Colour. Sci. Technol. 4, 54–58.

Maradiya, H.R., Patel, V.S., 2001b. Fibers Polym. 2, 153–158.

Maradiya, H.R., Patel, V.S., 2001c. Bull. Chem. Technol. Macedonia 20, 51–60.

Maradiya, H.R., Patel, V.S., 2001d. J. Braz. Chem. Soc. 12, 710–714.

Maradiya, H.R., Patel, V.S., 2001e. Man-Made Textile (India) 44, 468–473.

Maradiya, H.R., Patel, V.S., 2002a. Bull. Chem. Technol. Macedonia 21, 57–64.

Maradiya, H.R., Patel, V.S., 2002b. Fibers Polym. 3, 43–48.

Maradiya, H.R., Patel, V.S., 2003. Chem. Heterocycl. Compd. 3, 404–409.

Merian, E., 1961. US Patent 3,007,915.

Mitsubishi Chemical Industries Co. Ltd., 1979a. JP 80,112,266.

Mitsubishi Chemical Industries Co. Ltd., 1979b. Jpn. Kokai Tokkyo Koho 80,133,458.

Modena, Risaliti, A., 1977. Bull. Sci. Fac. Chem. Ind. Borogna 11, 65–69.

Nippon Kayaku Co. Ltd., 1979. JP 8,060,558.

Parton, B., 1978. British UK Patent 2,028,856.

Peter, M., 1979. Ger. Offen 3001945.

Peters, A.T., Gbadamosi, A., 1995. J. Chem. Tech. Biotechnol. 53, 301–308.

Rangnekar, D.W., Malankar, G.J., 2000. Phosphorus Sulfur Silicon Relat. Elem. 164, 199–205.

Rangnekar, D.W., Kanetkar, V.R., Shankarling, G.S., Malanker, J.V., 1998. J. Indian Counc. Chem. 15, 28–30.

Saylam, A., Seferoglu, Z., Ertan, N., 2008. Russ. J. Org. Chem. 44, 587–594.

Schwander, H.R., 1982. Dyes Pigm. 3, 133–160.

Sekar, N., 1994. Colourage 41, 38–41.

Sens, R., Etzbach, K.-H., 1994. Ger Offen DE 4426023.

Shuttleworth, L., Weaver, M.A., 1990. In: Waring, D.R., Hallas, G. (Eds.), The Chemistry and Application of Dyes. Plenum Press, New York, pp. 107–162.

Sonawane, N.D., Rangnekar, D.W., 2002. J. Heterocycl. Chem. 39, 303–308.

Sumitomo Chemical Co. Ltd., 1981. Jpn. Kokai Tokkyo Koho 57,161,653.

Towne, E.B., Hill, H.M., 1956. US Patent 2,746,953.

Towne, E.B., Dickey, J.B., Bloom, M.S., 1958. US Patent 2,852,504.

Towns, A.D., 1999. Dyes Pigm. 42, 3–28.

Tsien, H., Chu, K.-P., Chang, H., 1995. US Patent 5,652,344.

Weaver, M.A., Straley, J.M., 1970. US Patent 3,526,733.

Yao, W.H., 2001. Jiemian Kexue Huizhi. 23, 106–115 (Chem. Abstr. 137 (2001) 34443j).