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Synthesis and solvatochromic properties of some disazo dyes derived from pyrazolo[1,5-*a*]pyrimidine derivatives

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Abstract

4-Arylazo-3,5-diaminopyrazole compounds have been synthesized by reaction of arylazomalononitrile with hydrazine hydrate. The symmetrical and asymmetrical 3,6-diarylazo-2,5,7-triaminopyrazolo[1,5-*a*]pyrimidine heterocyclic disazo dyes have been prepared by the cyclization of 4-arylazo-3,5-diaminopyrazoles with different arylazomalononitriles. The solvatochromic behaviour of these disazo dyes in various solvents was evaluated.

Keywords: Arylazopyrazole; Diarylazopyrazolo[1,5-*a*]pyrimidine; Disazo dyes; Solvents effect

1. Introduction

It is well known that nitriles are widely used as intermediates for a large number of heterocyclic compounds. The aminopyrazole compounds have been easily obtained by the reaction of nitrile derivatives with hydrazine hydrate [1–6], and are very useful as precursors for the synthesis of fused heterocyclic ring systems. The fused heterocyclic compounds play an important role in biological and pharmacological activities [7–9], and they can also be used as intermediates in the dyestuff industry [10,11]. There are few papers dealing with the preparation of fused heterocyclic compounds as dye intermediates. In the present study, we prepare some symmetrical and asymmetrical 3,6-diarylazo-2,5,7-triaminopyrazolo[1,5-*a*]pyrimidine heterocyclic disazo dyes and evaluate their absorption properties in various solvents in order to study their relationship between colour and constitution.

2. Results and discussion

2.1. Preparation of symmetrical and asymmetrical heterocyclic disazo dyes

As shown in Scheme 1, aniline derivatives **1** were diazotized using sodium nitrite in hydrochloric acid; the temperature was maintained below 5 °C in an ice bath. The diazotized products were then coupled with malononitrile **2** to produce yellow solids of 2-(*p*-substituted-phenylazo)-malononitrile derivatives **3a–3c** [11–16], followed by the cyclization with hydrazine hydrate **4** in ethanol (95%) under reflux for 3–4 h, to give the 3,5-diamino-4-(*p*-substituted-phenylazo)-1H-pyrazole derivatives **5a–5c** as yellow to red-brown solids in low to good yield. Symmetrical 3,6-bis-(4-substituted-phenylazo)-2,5,7-triaminopyrazolo[1,5-*a*]pyrimidine heterocyclic disazo dyes **6a–6c** were synthesized by the cyclization,

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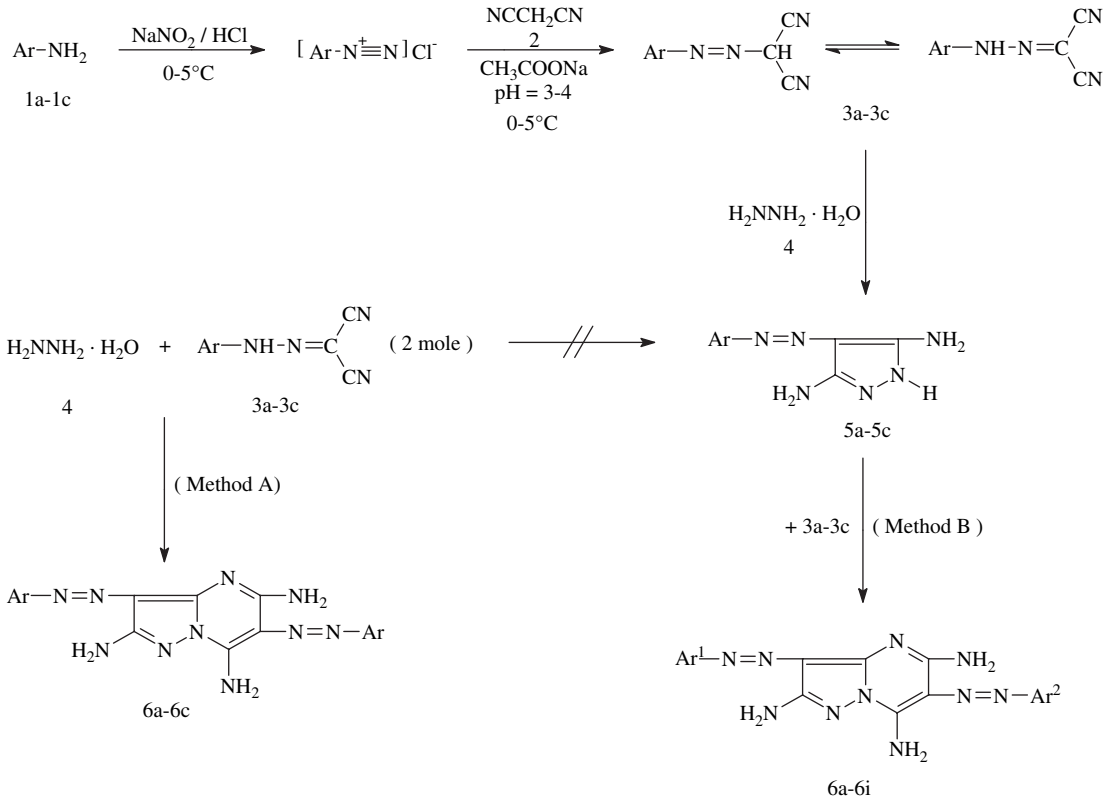


1, 3, 5	Ar
a	Ph
b	p-ClC_6H_4
c	p-$NO_2C_6H_4$

6	Ar1	Ar2
a	Ph	Ph
b	p-ClC_6H_4	p-ClC_6H_4
c	p-$NO_2C_6H_4$	p-$NO_2C_6H_4$
d	Ph	p-ClC_6H_4
e	Ph	p-$NO_2C_6H_4$
f	p-ClC_6H_4	Ph
g	p-ClC_6H_4	p-$NO_2C_6H_4$
h	p-$NO_2C_6H_4$	Ph
i	p-$NO_2C_6H_4$	p-ClC_6H_4

Scheme 1.

involving the reaction of **3a–3c** with **4** in the molar ratio 2:1 in ethanol (95%) under reflux for 3–4 h (Method A), or by the cyclization, involving the reaction of **5a–5c** derivatives with equimolar **3a–3c** under the same procedure (Method B). Asymmetrical heterocyclic disazo dyes **6d–6i** were synthesized by the cyclization of compounds **5a–5c** with different **3a–3c** derivatives in ethanol (95%) under reflux for 3–4 h. The compounds **6a–6i** were orange to red-brown solids. The physical data are summarized in Section 3 and spectral characterization is listed in Table 1.

2.2. Electronic absorption properties of heterocyclic disazo dyes

The absorption spectra of some heterocyclic disazo dyes **6a–6i** in various solvents at a concentration range of 10^{-6}–10^{-7} M are given in Table 2. It was found that the electronic absorption properties of these heterocyclic disazo dyes are strongly solvent dependent and vary with solvent polarity. The influence of solvents for these dyes increases in this order: DMSO > DMF > CH_3CO-CH_3 > $CH_3COOC_2H_5$ > $CHCl_3$. The spectral shifts of dye **6a** in various solvents are shown in Fig. 1. The dye **6a** showed absorption maxima at 490 nm in DMSO, 477 nm in DMF, 465 nm in acetone, 456 nm in ethyl acetate, and 454 nm in chloroform, in other words, the dye **6a** showed significantly larger bathochromic shifts in stronger polar solvent (DMSO) than the weaker polar solvents, such as DMF, acetone, ethyl acetate and chloroform. And the same trends of absorption shifts in various solvents were observed for the entire series of dyes **6a–6i**, as shown in Table 2. The spectral shifts of dyes **5a–5c** and **6a–6c** in acetone at a concentration

Table 1
Spectral data of dyes **5a–5c** and **6a–6i**

Dye	MS (m/z M^+)	IR (KBr) ν (cm^{-1})	^{1}H-NMR[a] (DMSO-d_6) δ (ppm)
5a	202.1	3330 (N–H), 1618 (C=N)	5.96 (2H, s, NH_2), 6.25 (2H, s, NH_2), 7.28 (2H, d, 3,5-PhH), 7.42 (1H, s, 4-PhH), 7.76 (2H, d, 2,6-PhH) and 10.75 (1H, s, NH)
5b	236.0	3305 (N–H), 1620 (C=N)	5.97 (2H, s, NH_2), 6.26 (2H, s, NH_2), 7.49 (2H, d, 3,5-PhH), 8.02 (2H, d, 2,6-PhH) and 10.76 (1H, s, NH)
5c	247.2	3371 (N–H), 1620 (C=N), 1561,1331 (NO_2)	6.31 (2H, s, NH_2), 6.52 (2H, s, NH_2), 7.83 (2H, d, 3,5-PhH), 8.21 (2H, d, 2,6-PhH) and 10.95 (1H, s, NH)
6a	372.1	3371 (N–H), 1654 (C=N)	6.97 (2H, s, NH_2), 7.45–8.02 (10H, m, ArH)
6b	440.1	3276 (N–H), 1615 (C=N)	6.95 (2H, s, NH_2), 7.49–8.05 (8H, m, ArH–Cl)
6c	461.8	3214 (N–H), 1615 (C=N), 1561, 1331 (NO_2)	7.16 (2H, s, NH_2), 7.85–8.35 (8H, m, ArH–NO_2)
6d	406.1	3261 (N–H), 1605 (C=N)	6.96 (2H, s, NH_2), 7.49–8.07 (5H, m, ArH), 7.69 (2H, d, 3,5-ArH–Cl), 8.02 (2H, d, 2,6-ArH–Cl)
6e	417.1	3275 (N–H), 1602 (C=N), 1514, 1325 (NO_2)	6.98 (2H, s, NH_2), 7.47–8.03 (5H, m, ArH), 8.26 (2H, d, 2,6-ArH–NO_2), 8.32 (2H, d, 3,5-ArH–NO_2)
6f	406.1	3261 (N–H), 1605 (C=N)	6.96 (2H, s, NH_2), 7.38–7.71 (5H, m, ArH), 7.48 (2H, d, 3, 5-ArH–Cl), 8.01 (2H, d, 2,6-ArH–Cl)
6g	451.1	3325 (N–H), 1601 (C=N), 1513, 1327 (NO_2)	6.98 (2H, s, NH_2), 7.52 (2H, d, 3,5-ArH–Cl), 7.72 (2H, d, 2,6-ArH–Cl), 8.25 (2H, d, 2,6-ArH–NO_2), 8.29 (2H, d, 3,5-ArH–NO_2)
6h	417.1	3258 (N–H), 1629 (C=N), 1517, 1324 (NO_2)	7.15 (2H, s, NH_2), 7.38–7.84 (5H, m, ArH), 8.02(2H, d, 3,5-ArH–NO_2), 8.32 (2H, d, 2,6-ArH–NO_2)
6i	451.1	3253 (N–H), 1611 (C=N), 1516, 1324 (NO_2)	7.15 (2H, s, NH_2), 7.53 (2H, d, 3,5-ArH–Cl), 7.84 (2H, d, 2,6-ArH–Cl), 8.11 (2H, d, 3,5-ArH–NO_2), 8.32 (2H, d, 2,6-ArH–NO_2)

[a] Abbreviations: s, singlet; d, doublet; m, multiplet.

Table 2
Absorption spectra of dyes **6a–6i** in various solvents

Compounds	DMSO	DMF	Acetone	Ethyl acetate	Chloroform	log ε (acetone)
6a	490	477	465	456	454	4.19
6b	508	492	483	468	464	4.28
6c	564	539	508	504	500	4.14
6d	507	486	476	466	462	4.29
6e	554	523	488	486	481	4.27
6f	494	483	467	464	455	4.08
6g	547	523	489	488	482	4.26
6h	546	518	491	483	475	4.41
6i	549	525	495	490	478	4.34

The ε_r value of solvents: DMSO = 48.9; DMF = 36.7; acetone = 20.7; ethyl acetate = 6.02; chloroform = 4.9.

range of 10^{-6}–10^{-7} M are given in Table 3. The absorption maxima of dyes **6a–6c** ranged from 465 to 508 nm and are shown in Fig. 2. It was found that dye **6c** contains an electron donor group (methoxy) in the *p*-position of phenylsubstituted on the 3-, 6-position of the pyrazolo[1,5-*a*]pyrimidine ring, so that λ_{max} of the dye **6c** showed bathochromic shift of +43 nm relative to dye **6a** in acetone; λ_{max} of the dye **6b** is +18 nm longer than that of dye **6a** in acetone, due to weaker electron acceptor of the chloro group in the *p*-position of the phenylsubstituted on the 3-, 6-position of the pyrazolo[1,5-*a*]pyrimidine ring, as shown in Table 3. The spectral shifts of dyes **6a–6i** in acetone at a concentration range of 10^{-6}–10^{-7} M are given in Table 4. The absorption maxima of dyes **6a**, **6d** and **6e** ranged from 465 to 488 nm and are shown in Fig. 3. It was found that dye **6e** contains an electron donor group (methoxy) in the *p*-position of phenylsubstituted on the 6-position of the pyrazolo[1,5-*a*]pyrimidine ring, so that the λ_{max} of dye **6e** showed bathochromic shift of +23 nm relative to dye **6a** in acetone; λ_{max} of dye **6d**

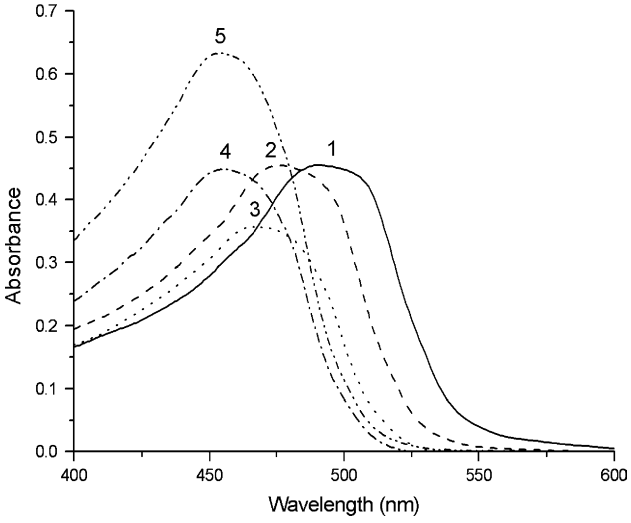
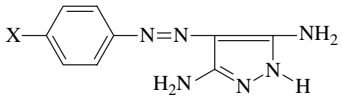
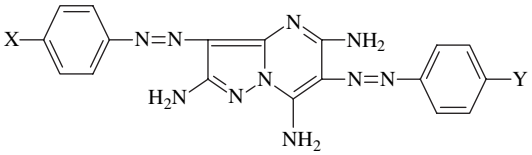


Fig. 1. Absorption spectra of dye **6a**: 1. DMSO; 2. DMF; 3. Acetone; 4. Ethyl acetate; 5. Chloroform (conc.: 10^{-6}–10^{-7} mol/l).

Table 3
Substituent effect of dyes **5a–5c**, **6a–6c** in acetone





Compounds	λ_{max}	X	$\Delta\lambda$[a]
5a	362	H	–
5b	369	Cl	+7
5c	445	NO_2	+83

Compounds	λ_{max}	X	Y	$\Delta\lambda$[b]
6a	465	H	H	–
6b	483	Cl	Cl	+18
6c	508	NO_2	NO_2	+43

[a] Relative to (**5a**:X = H).
[b] Relative to (**6a**:X = Y = H).

is +11 nm longer than that of dye **6a** in acetone, due to weaker electron accepter of chloro group in the *p*-position of the phenylsubstituted on the 6-position of the pyrazolo[1,5-*a*]pyrimidine ring. Similar effects of the substituent were also observed in the absorption maxima of dyes **6a–6i**, as shown in Table 4.

3. Experimental

All melting points are uncorrected and are in °C. The IR spectra were recorded on a JASCO FTIR-3 spectrometer (KBr). The ^{1}H-NMR spectra were obtained on a Joel-EX-400 MHz NMR spectrometer, and the chemical shifts are expressed in δ ppm using TMS as an internal standard. The mass spectra were obtained from a Finnigan TSQ-700 GC/LC/MS spectrometer. Microanalyses for C, H and N were performed on a Perkin–Elmer 2400(II) elemental analyzer. Absorption spectra were recorded on a Heliosa UV1 in various solvents.

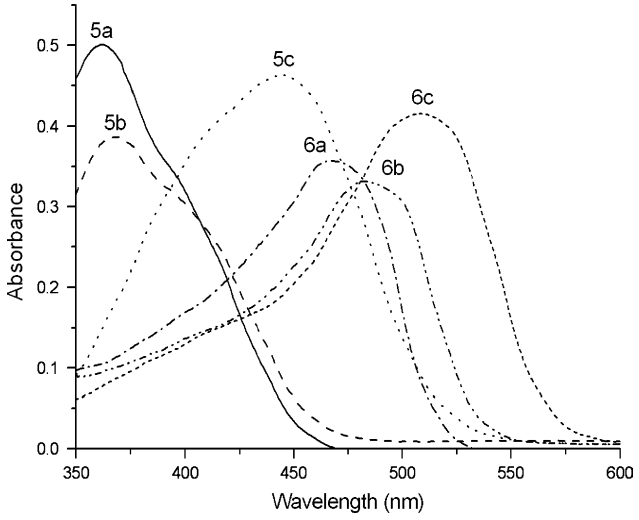


Fig. 2. Absorption spectra of dyes **5a–5c**, **6a–6c** in acetone (conc.: 10^{-6}–10^{-7} mol/l).

*3.1. Preparation of 2-arylazo-malononitrile derivatives **3a–3c** [11–15]*

The same procedures were used for the syntheses of compounds **3a–3c**, as represented by the preparation of compound **3a** below.

*3.1.1. 2-Phenylazo-malononitrile (**3a**)*

A hydrochloric acid solution (6 ml) of aniline **1a** (0.93 g, 0.01 mol) and an aqueous solution (3 ml) of sodium nitrite (0.72 g, 0.0105 mol) were mixed and stirred at 0 °C for 1 h, followed by the addition of an aqueous solution (10 ml) of the coupling component malononitrile **2** (0.66 g, 0.01 mol) and continued stirring at 0 °C for 2 h. The resulting product was filtered and the pre-cake washed with water, dried, and recrystallized from ethanol to give 2-phenylazo-malononitrile (**3a**) as yellow crystals (1.26 g, 74%), m.p. 134–136 °C; *m*/*z* 170.0 (M^+); FT-IR (KBr, cm^{-1}): 2233 ν (C ≡ N).

*3.1.2. 2-(4-Chloro-phenylazo)-malononitrile (**3b**)*

This compound was obtained from **1b** and malononitrile as yellow crystals (1.65 g, 81%), m.p. 183–185 °C; *m*/*z* 204.0 (M^+); FT-IR (KBr, cm^{-1}): 2228 ν (C ≡ N).

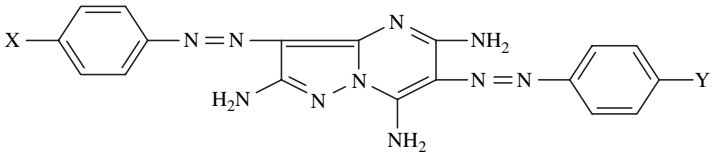
*3.1.3. 2-(4-Nitro-phenylazo)-malononitrile (**3c**)*

This compound was obtained from **1c** and malononitrile as yellow crystals (1.99 g, 92%); m.p. 140–142 °C; *m*/*z* 215.1 (M^+); FT-IR (KBr, cm^{-1}): 2230 ν (C ≡ N), 1513 and 1344 ν (NO_2).

*3.2. Preparation of heterocyclic monoazo dyes **5a–5c***

The syntheses of azo dyes **5a–5c** were carried out by the same procedures, as described below for the preparation of dye **5a**.

Table 4
Substituent effect of dyes **6a–6i** in acetone



Compounds	λ_{max}	X	Y	$\Delta\lambda$
6a	465	H	H	–
6d	476	H	Cl	+11[a]
6e	488	H	NO_2	+23[a]
6b	483	Cl	Cl	–
6f	467	Cl	H	−16[b]
6g	489	Cl	NO_2	+6[b]
6c	508	NO_2	NO_2	–
6h	491	NO_2	H	−17[c]
6i	495	NO_2	Cl	−13[c]

[a] Relative to (**6a**:X = Y = H).
[b] Relative to (**6b**:X = Y = Cl).
[c] Relative to (**6c**:X = Y = NO_2).

*3.2.1. 3,5-Diamino-4-phenylazo-1H-pyrazole (**5a**)*

Hydrazine hydrate **2** (0.59 g, 0.01 mol) was added to a solution of **3a** (1.7 g, 0.01 mol) and pyridine 0.5 ml in 30 ml ethanol. The reaction mixture was heated under reflux for 3–4 h, then cooled to room temperature. The separated solid was filtered, washed with water, dried and recrystallized from ethanol to give a pale yellow solid 3,5-diamine-4-phenylazo-1H-pyrazole (**5a**) (1.1 g, 54%), m.p. 260–262 °C.

$C_9H_{10}N_6$ Calcd: C, 53.46; H, 4.95; N, 41.58; Found: C, 53.41; H, 4.92; N, 41.63.

*3.2.2. 3,5-Diamino-4-(4-chloro-phenylazo)-1H-pyrazole (**5b**) [5]*

This compound was obtained from **3b** and hydrazine hydrate as yellow crystals (1.35 g, 57%), m.p. 270–272 °C (lit. m.p. 267–270 °C).

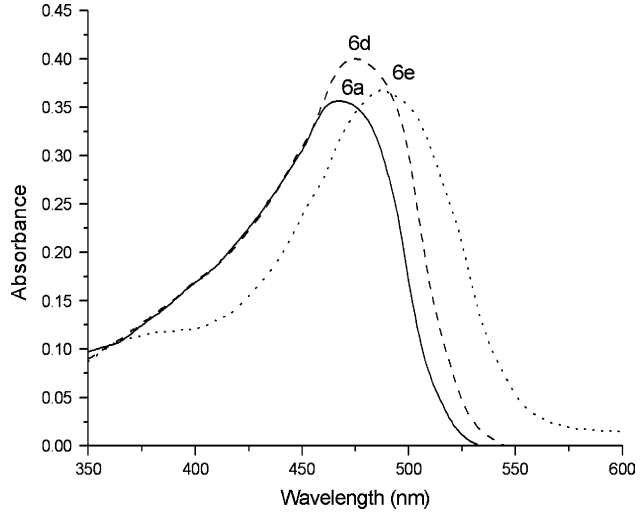


Fig. 3. Absorption spectra of dyes **6a**, **6d** and **6e** in acetone (conc.: 10^{-6}–10^{-7} mol/l).

$C_9H_9N_6Cl$ Calcd: C, 45.76; H, 3.81; N, 35.59; Found: C, 45.78; H, 3.83; N, 35.63.

*3.2.3. 3,5-Diamino-4-(4-nitro-phenylazo)-1H-pyrazole (**5c**)*

This compound was obtained from **3c** and hydrazine hydrate as red-brown crystals (1.48 g, 60%); m.p. 255–257 °C.

$C_9H_9N_7O_2$ Calcd: C, 43.72; H, 3.64; N, 39.68; Found: C, 43.71; H, 3.65; N, 39.72.

*3.3. Preparation of symmetrical and asymmetric heterocyclic disazo dyes **6a–6i***

*3.3.1. Preparation of symmetrical heterocyclic disazo dyes **6a–6c***

The syntheses of disazo dyes **6a–6c** followed the same procedures as described below for the preparation of dye **6a**.

*3.3.1.1. 3,6-Bis-phenylazo-2,5,7-triaminopyrazolo[1,5-a] pyrimidine (**6a**) [6].* (Method A) To a solution of **3a** (3.4 g, 0.02 mol) and pyridine 1 ml in 30 ml ethanol was added hydrazine hydrate **2** (0.59 g, 0.01 mol). The reaction mixture was heated under reflux for 3–4 h, then cooled to room temperature. The separated solid was filtered, washed with hot ethanol, dried and recrystallized from DMF to give a pale orange solid 3,6-bis-phenylazo-2,5,7-triaminopyrazolo[1,5-α]pyrimidine (**6a**) (1.48 g, 39%). (Method B) To a solution of **5a** (2.02 g, 0.01 mol) and pyridine 0.5 ml in 30 ml ethanol was added compound **3a** (1.7 g, 0.01 mol). The reaction mixture was heated under reflux for 3–4 h, then cooled to room temperature. The separated solid was filtered, washed with hot ethanol, dried and recrystallized from DMF to give a pale orange solid 3,6-bis-phenylazo-2,5,7-triaminopyrazolo

[1,5-*a*]pyrimidine (**6a**) (1.63 g, 44%), m.p. >300 °C (lit. m.p. >300 °C)

$C_{18}H_{16}N_{10}$ Calcd: C, 58.06; H, 4.30; N, 37.63; Found: C, 58.02; H, 4.28; N, 37.65.

*3.3.1.2. 3,6-Bis-(4-chloro-phenylazo)-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6b**) [6].* This compound was obtained from **3b** and hydrazine hydrate, or obtained from **5b** and **3b** as orange crystals (2.07 g, 47%) m.p. >300 °C (lit. m.p. >300 °C).

$C_{18}H_{14}N_{10}Cl_2$ Calcd: C, 49.09; H, 3.18; N, 31.82; Found: C, 49.11; H, 3.15; N, 31.79.

*3.3.1.3. 3,6-Bis-(4-nitro-phenylazo)-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6c**).* This compound was obtained from **3c** and hydrazine hydrate, or obtained from **5c** and **3c** as red-brown crystals (1.43 g, 31%), m.p. >300 °C.

$C_{18}H_{14}N_{12}O_4$ Calcd: C, 47.75; H, 3.03; N, 36.36; Found: C, 47.71; H, 3.06; N, 36.31.

*3.3.2. Preparation of asymmetric heterocyclic disazo dyes **6d–6i***

The syntheses of disazo dye **6d–6i** followed the same procedures, as represented below by the preparation of dye **6d**.

*3.3.2.1. 6-(4-Chloro-phenylazo)-3-phenylazo-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6d**).* To a solution of **5a** (2.02 g, 0.01 mol) and pyridine 0.5 ml in 30 ml ethanol was added compound **3b** (2.04 g, 0.01 mol). The reaction mixture was heated under reflux for 3–4 h, then cooled to room temperature. The separated solid was filtered, washed with hot ethanol, dried and recrystallized from DMF to give a pale reddish-orange solid 6-(4-chloro-phenylazo)-3-phenylazo-2,5,7-triaminopyrazolo [1,5-*α*]pyrimidine **(6d)** (2.35 g, 57%), m.p. >300 °C.

$C_{18}H_{15}N_{10}Cl$ Calcd: C, 53.20; H, 3.69; N, 34.48; Found: C, 53.17; H, 3.72; N, 34.46.

*3.3.2.2. 6-(4-Nitro-phenylazo)-3-phenylazo-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6e**).* This compound was obtained from **5a** and **3c** as red-brown crystals (3.13 g, 75%), m.p. >300 °C.

$C_{18}H_{15}N_{11}O_2$ Calcd: C, 51.80; H, 3.60; N, 36.93; Found: C, 51.86; H, 3.58; N, 36.88.

*3.3.2.3. 3-(4-Chloro-phenylazo)-6-phenylazo-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6f**) [6].* This compound was obtained from **5b** and **3a** as orange crystals (2.11 g, 52%), m.p. >300 °C (lit. m.p. >300 °C).

$C_{18}H_{15}N_{11}O_2$ Calcd: C, 51.80; H, 3.60; N, 36.93; Found: C, 51.88; H, 3.55; N, 36.89.

*3.3.2.4. 3-(4-Chloro-phenylazo)-6-(4-nitro-phenylazo)-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6g**).* This compound was obtained from **5b** and **3c** as red-brown crystals (2.07 g, 45%), m.p. >300 °C.

$C_{18}H_{14}N_{11}Cl\ O_2$ Calcd: C, 47.89; H, 3.10; N, 34.15; Found: C, 47.83; H, 3.12; N, 34.12.

*3.3.2.5. 3-(4-Nitro-phenylazo)-6-phenylazo-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6h**).* This compound was obtained from **5c** and **3a** as red-brown crystals (1.34 g, 32%), m.p. >300 °C.

$C_{18}H_{15}N_{11}O_2$ Calcd: C, 51.80; H, 3.60; N, 36.93; Found: C, 51.75; H, 3.63; N, 36.86.

*3.3.2.6. 3-(4-Nitro-phenylazo)-6-(4-chloro-phenylazo)-2,5,7-triaminopyrazolo[1,5-a]pyrimidine (**6i**).* This compound was obtained from **5c** and **3b** as red-brown crystals (1.85 g, 41%), m.p. >300 °C.

$C_{18}H_{14}N_{11}Cl\ O_2$ Calcd: C, 47.89; H, 3.10; N, 34.15; Found: C, 47.83; H, 3.12; N, 34.09.

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