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Synthesis of some novel pyrazolo[5,1-*c*][1,2,4]triazine derivatives and investigation of their absorption spectra

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Abstract

In this study, 5-amino-4-hetarylazo-3-methyl-1*H*-pyrazoles were diazotised and coupled with malononitrile and ethyl cyanoacetate to give pyrazolylazo malononitriles and ethyl pyrazolylazo cyanoacetate, respectively. Ten novel pyrazolo[5,1-*c*][1,2,4]triazine dyes were synthesized by heating of pyrazolylazo malononitriles and ethyl pyrazolylazo cyanoacetate in glacial acetic acid. The dyes were characterized by elemental analysis and spectral methods. The solvatochromic behaviour of these diazo dyes in various solvents was evaluated. Substituent, acid and base effects on the visible absorption maxima of the dyes are also reported.

Keywords: Active methylene compounds; Pyrazole; Intramolecular cyclization; Diazo-coupling reaction; Tautomerism; Solvatochromism

1. Introduction

Polyfunctionally substituted heteroaromatics are biologically interesting molecules and their synthesis has recently received considerable attention [1–3]. 5-Amino pyrazoles are versatile reagents and have been extensively utilized as synthetic starting components for preparation of several polysubstituted fused pyrazoles [4–6]. Also, fused pyrazoles are important compounds that have many derivatives with a wide range of interesting properties, such as anti-hyperglycemic, analgesic, anti-inflammatory, anti-pyretic, anti-bacterial, hypoglycaemic and sedative–hypnotic activities. Recently, some pyrazoles were reported to have non-nucleoside HIV-1 reverse transcriptase inhibitory activity [7–13]. Some azopyrazole derivatives also find application in dyes, biological and pharmacological studies and complexes [14–20].

The cyclization reactions of hydrazones take up a very important place among the synthesis of various heterocyclic compounds. These reactions are concerned with many unfused five- and six-membered heterocyclic compounds. Among these reactions, cyclization reactions that result in 7-aminopyrazolo[5,1-*c*][1,2,4]triazines, of pyrazolylazoketonitriles found by Partridge and Stevens [21] can be given as an example. The above stated cyclization was applied to the whole series of variously substituted pyrazolylhydrazones [22–30]. All the above mentioned cyclization reactions are concerned with pyrazolylhydrazones unsubstituted at nitrogen atoms, which due to the NH group in the pyrazole ring readily succumb to the ring closure to pyrazolo[5,1-*c*][1,2,4]triazine system.

Azo dyes based on heterocyclic amines have been developed, and the resultant dyes give higher tinctorial strength and brighter dyeing than those derived from aniline-based diazo components. For instance, amino-substituted thiazole, isothiazole, and thiophene compounds afford very electronegative diazo components and consequently, provide a pronounced bathochromic effect compared to the corresponding benzenoid compounds [31–39]. Although many patents and papers describe the synthesis and dyeing properties of

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Table 1
Spectral data for dyes **4a–e**

Dye no.	FT-IR (cm^{-1}, in KBr)				^{1}H NMR (δ, ppm)		
	ν_{NH_2}	$\nu_{Ar\text{-}H}$	$\nu_{Al\text{-}H}$	ν_{CN}	Aro.-H	Alip.-H	NH_2
4a	3434, 3379	3078	2982	2229	8.05 (1H, d), 7.83 (1H, d)	2.70 (3H, s, pyrazole CH_3)	9.72 (2H, br)
4b	3430, 3382	3085	2987	2228	7.77 (1H, s)	2.82 (3H, s, thiazole CH_3), 2.72 (3H, s, pyrazole CH_3)	9.66 (2H, br)
4c	3436, 3385	3077	2978	2235	8.12–7.50 (4H, m)	2.78 (3H, s, pyrazole CH_3)	9.82 (2H, br)
4d	3433, 3382	3081	2988	2231	7.99–7.14 (3H, m)	4.14 (2H, q, ethoxy CH_2), 2.78 (3H, s, pyrazole CH_3), 1.40 (3H, t, ethoxy CH_3)	9.77 (2H, br)
4e	3428, 3379	3075	2977	2228	6.56 (1H, s)	2.79 (3H, s, isoxazole CH_3), 2.73 (3H, s, pyrazole CH_3)	9.69 (2H, br)

monoazo dyes based on heterocyclic coupling component [40–43], an investigation involving monoazo dyes based on pyrazolo[5,1-*c*][1,2,4]triazine ring has not been found. In continuation of our work, we report here the synthesis of a series of new monoazo dyes based on pyrazolo[5,1-*c*][1,2,4]triazine ring. The visible absorption spectra in various solvents of these dyes are also discussed.

2. Experimental

2.1. General

The chemicals used for the synthesis of the compounds were obtained from Sigma–Aldrich Chemical Company without further purification. The solvents used were of spectroscopic grade.

IR spectra were determined using a Mattson 1000 Fourier Transform-infrared (FT-IR) spectrophotometer on a KBr disc. Nuclear magnetic resonance (^{1}H NMR) spectra were recorded on a Bruker-Spectrospin Avance DPX 400 Ultra-Shield in deuterated dimethylsulphoxide (DMSO-d_6) using tetramethylsilane (TMS) as the internal reference; chemical shifts were (δ) given in ppm. Ultraviolet–visible (UV–vis) absorption spectra were recorded on an ATI Unicam UV-100 spectrophotometer at the wavelength of maximum absorption (λ_{max}) in a range of solvents, i.e. DMSO, dimethylformamide (DMF), acetonitrile, methanol, acetic acid and chloroform, at various concentrations (1×10^{-6}–10^{-8}). Change of λ_{max} was also investigated when 0.1 ml base (potassium hydroxide, 0.1 M) and 0.1 ml acid (hydrochloric acid, 0.1 M) were added to dye solutions in methanol (1 ml). The methods used for the syntheses of 5-amino-3-methyl-4-hetarylazo-1*H*-pyrazoles (**1a–e**) were described in the previous part of our study [44].

2.2. Synthesis of pyrazolo[5,1-c][1,2,4]triazines (*4a–e, 5a–e*)

Diazotisation of **1a–e** was effected with nitrosylsulphuric acid. A typical procedure is that described below for 5-amino-3-methyl-4-thiazolylazo-1*H*-pyrazole (**1a**); all other

Table 2
Spectral data for dyes **5a–e**

Dye no.	FT-IR (cm^{-1}, in KBr)					^{1}H NMR (δ, ppm)		
	ν_{NH_2}	$\nu_{Ar\text{-}H}$	$\nu_{Al\text{-}H}$	$\nu_{C=O}$	ν_{C-O}	Aro.-H	Alip.-H	X–H
5a	3436, 3389	3090	2985	1690	1071	8.09 (1H, d), 7.87 (1H, d)	4.51 (2H, q, 6-ethoxycarbonyl CH_2), 2.80 (3H, s, pyrazole CH_3), 1.46 (3H, t, 6-ethoxycarbonyl CH_3)	9.70 (br, NH_2), 8.95 (br, tautomeric NH)
5b	3435, 3377	3082	2980	1689	1078	7.82 (1H, s)	4.51 (2H, q, 6-ethoxycarbonyl CH_2), 2.85 (3H, s, thiazole CH_3), 2.79 (3H, s, pyrazole CH_3), 1.45 (3H, t, 6-ethoxycarbonyl CH_3)	9.71 (br, NH_2), 8.87 (br, tautomeric NH)
5c	3429, 3382	3087	2982	1687	1067	8.13–7.50 (4H, m)	4.47 (2H, q, 6-ethoxycarbonyl CH_2), 2.79 (3H, s, pyrazole CH_3), 1.41 (3H, t, 6-ethoxycarbonyl CH_3)	9.80 (br, NH_2), 8.90 (br, tautomeric NH)
5d	3437, 3378	3083	2982	1689	1056	8.00–7.09 (3H, m)	4.47 (2H, q, 6-ethoxycarbonyl CH_2), 4.15 (2H, q, 6-ethoxybenzothiazole CH_2), 2.78 (3H, s, pyrazole CH_3), 1.39 (6H, m, ethoxy $2CH_3$)	9.72 (br, NH_2), 8.86 (br, tautomeric NH)
5e	3424, 3368	3076	2982	1687	1078	6.54 (1H, s)	4.46 (2H, q, 6-ethoxycarbonyl CH_2), 2.80 (3H, s, isoxazole CH_3), 2.73 (3H, s, pyrazole CH_3), 1.40 (3H, t, 6-ethoxycarbonyl CH_3)	9.50 (br, NH_2), 8.88 (br, tautomeric NH)

Table 3
Element analysis of dyes **4a–e** and **5a–e**

Dye no.	Molecular formula	Molecular mass	Yield (%)	Colour	%C		%H		%N		%S		Melting point[a] (°C)
					Calcd.	Found	Calcd.	Found	Calcd.	Found	Calcd.	Found	
4a	$C_{10}H_7N_9S$	285.29	87	Orange	42.10	42.28	2.47	2.53	44.19	43.91	11.24	11.02	dec. >330
4b	$C_{11}H_9N_9S$	299.32	79	Red	44.14	44.25	3.03	3.10	42.12	41.89	10.71	10.59	dec. >330
4c	$C_{14}H_9N_9S$	335.35	84	Red	50.14	49.93	2.71	2.82	37.59	37.38	9.56	9.45	dec. >300
4d	$C_{16}H_{13}N_9OS$	379.40	88	Red	50.65	50.78	3.45	3.54	33.23	33.08	8.45	8.22	dec. >240
4e	$C_{11}H_9N_9O$	283.25	67	Yellow	46.64	46.52	3.20	3.09	44.51	44.37	–	–	dec. >315
5a	$C_{12}H_{12}N_8O_2S$	332.34	78	Red	43.37	43.16	3.64	3.56	33.72	33.59	9.65	9.52	262–263
5b	$C_{13}H_{14}N_8O_2S$	346.37	73	Orange	45.08	44.87	4.07	4.14	32.35	32.23	9.26	9.04	245–246
5c	$C_{16}H_{14}N_8O_2S$	382.40	72	Red	50.25	50.39	3.69	3.75	29.30	29.06	8.39	8.17	268–269
5d	$C_{18}H_{18}N_8O_3S$	426.45	75	Black	50.70	50.58	4.25	4.16	26.28	25.96	7.52	7.39	269–270
5e	$C_{13}H_{14}N_8O_3$	330.30	70	Yellow	47.27	47.38	4.27	4.33	33.92	33.76	–	–	242–243

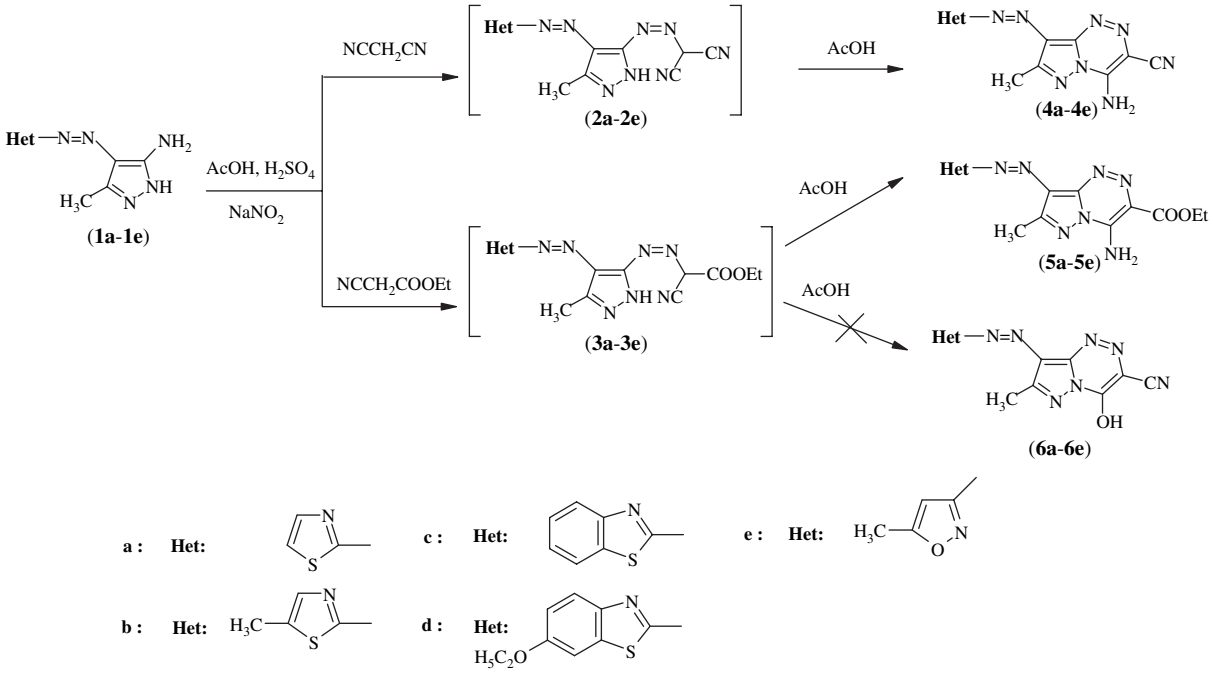
[a] Recrystallization from DMF–H_2O.

dyes were prepared in a similar manner. Characterization data are shown in Tables 1–3.

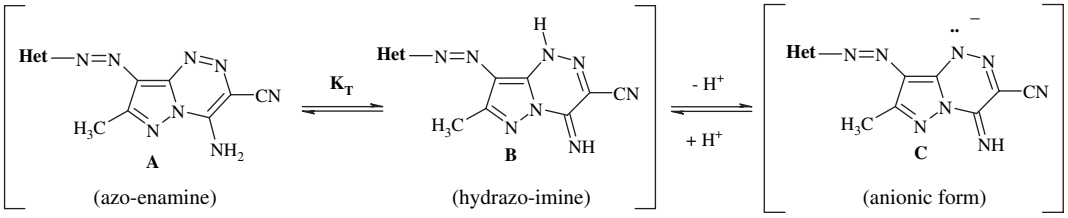
2.2.1. 7-Amino-6-cyano-2-methyl-3-thiazolylazopyrazolo [5,1-c][1,2,4]triazine (*4a*)

Nitrosylsulphuric acid was prepared by dissolving sodium nitrite (1 g) in concentrated sulphuric acid (7 ml) at 0 °C. 5-Amino-3-methyl-4-thiazolylazo-1*H*-pyrazole (**1a**) (2.0×10^{-3} mol) was dissolved in hot glacial acetic acid (2.5 ml) and rapidly cooled in an ice/salt bath to −5 °C. The solution was then added in portions over 30 min to nitrosylsulphuric acid at 0–5 °C and the mixture stirred for a further 1 h at this temperature. Then the resulting diazonium solution was added in portions over 30 min to a vigorously stirred solution of malononitrile (2.0×10^{-3} mol) in pyridine (10 ml) at 0–5 °C, maintaining the pH at 7–8 by simultaneous addition of solid sodium acetate at 0–5 °C. The mixture was then stirred for 2 h at 0–5 °C. The progress of the reaction was followed by thin layer chromatography (TLC) using a DMF–H_2O mixture (5:2 by volume) as a developing solvent and silica gel TLC plates as the stationary phase. The resulting solid (**2a**) was filtered, washed with cold water (3×50 ml) and dried in air. A solution of the precipitated solid (**2a**) in glacial acetic acid (30 ml) was refluxed for 4 h. The solvent was then evaporated in vacuo and the remaining product was collected by filtration and dried in air. Recrystallization from DMF–H_2O (3:1) gave orange crystals of the product dye **4a**.

This procedure was also used to synthesize dyes **4b–e** and **5a–e** (Scheme 1).



Scheme 1.



Scheme 2.

3. Result and discussion

3.1. Azo–hydrazo tautomerism

Azo–hydrazo tautomerism is not only of importance to the dyestuff manufacturer but also to other areas of chemistry. Azo–hydrazo tautomers not only have different colours, but also have different tinctorial strengths (and hence economics) and different properties, e.g. light fastness. Azo dyes **4a–e** and **5a–e** can exist in two possible tautomeric forms, namely the azo-enamine forms **A**, **D** and the hydrazo-imine forms **B**, **E** as shown in Schemes 2 and 3. The FT-IR spectra of dyes **4a–e** and **5a–e** showed intense amino (NH_2) bands at 3437–3424 and 3389–3368 cm^{-1}. This suggests that these dyes are predominantly in azo-enamine form, as opposed to the hydrazo-imine form, in the solid state. FT-IR spectra of dyes **4a–e** showed a band at 2235–2228 cm^{-1}, assigned to cyano group (CN). On the other hand, FT-IR spectra of dyes **5a–e** showed a band at 1691–1672 cm^{-1}(C=O), a band at 1078–1056 cm^{-1} (C–O) and did not show a band at 2200–2300 cm^{-1} (C≡N). These results suggest that these dyes are predominantly in **5a–e** form, as opposed to **6a–e** form (Scheme 1). The other ν_{max} values of 3090–3075 cm^{-1} (aromatic C–H) and 2988–2977 cm^{-1} (aliphatic C–H) were recorded.

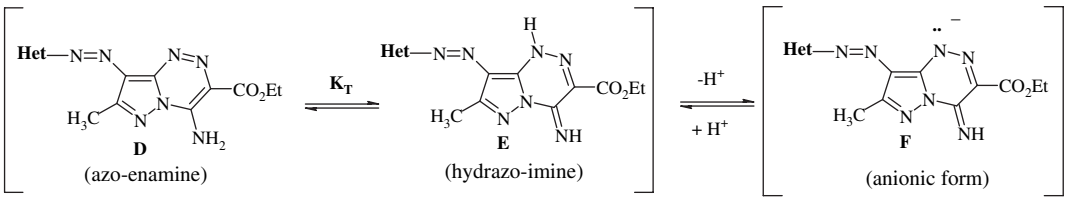
The ^1H NMR spectra of dyes **4a–e** showed a broad NH_2 peak at 9.82–9.66 ppm. This result suggests that dyes **4a–e** are present as a single tautomeric form in DMSO, namely the azo-enamine form **A** as shown in Scheme 2. The ^1H NMR spectra of dyes **5a–e** showed a broad NH_2 peak at 9.80–9.50 ppm and a broad NH peak at 8.95–8.86 ppm. These results suggest that dyes **5a–e** are present as a mixture of two tautomeric forms in DMSO, namely the azo-enamine form **D** and the hydrazo-imine form **E** as shown in Scheme 3. Also, ^1H NMR spectra of dyes **5a–e** showed a quartet methylene ($COOCH_2CH_3$) peak at 4.51–4.46 ppm and a triplet methyl ($COOCH_2CH_3$) peak at 1.46–1.39 ppm. These results suggest that these dyes are predominantly in **5a–e** form, as opposed to **6a–e** form (Scheme 1).

3.2. Effect of solvent

The UV–vis absorption spectra of dyes **4a–e** and **5a–e** were recorded over the range of λ between 300 and 700 nm, using a variety of solvents in concentrations 10^{-6}–10^{-8} M and the results are summarised in Table 4. The visible absorption spectra of the dyes did not correlate with the polarity of the solvent.

Dyes **4a–e** gave a single dominant absorption peak without a shoulder in all used solvents. It was also observed that λ_{max} values of dyes **4a–e** were the largest in DMF when compared with those in the other five solvents. Also, λ_{max} values of dyes **4a–e** in DMSO are larger than the λ_{max} in acetonitrile, methanol, acetic acid and chloroform (Fig. 1). These results suggest that dyes **4a–e** are present as an anionic form **C** in DMF, as a tautomeric form (azo-enamine form **A**) in DMSO and as a different tautomeric form (hydrazo-imine form **B**) in acetonitrile, methanol, acetic acid and chloroform (Scheme 2). Dyes **5a–e** gave a single dominant absorption peak without a shoulder in acetonitrile, methanol, acetic acid and chloroform and a maximum absorption peak with a shoulder in DMSO and DMF. Shoulders of dyes **5a–e** in DMSO and DMF are similar to absorption maxima in acetonitrile, methanol, acetic acid and chloroform (Fig. 2). The reason for this is probably that dyes **5a–e** are present predominantly in a single tautomeric form in acetonitrile, methanol, acetic acid and chloroform and in a mixture of azo-enamine form **D** and hydrazo-imine form **E** in DMSO and DMF (Scheme 3).

It was observed that although λ_{max} values of dyes **4a–e** in DMSO and DMF were shifted bathochromically with respect to the λ_{max} in chloroform, λ_{max} values of dyes **4a–e** in acetonitrile, methanol and acetic acid were shifted a little



Scheme 3.

Table 4
Influence of solvent on λ_{max} (nm) of dyes **4a–e** and **5a–e**

Dye no.	DMSO	DMF	Acetonitrile	Methanol	Acetic acid	Chloroform
4a	464	498	444	444	440	446
4b	470	499	462	465	460	467
4c	484	520	475	474	475	480
4d	486	528	478	480	477	482
4e	410	460	396	399	395	400
5a	465, 442 s	440, 460 s	429	431	429	427
5b	467, 452 s	463, 447 s	435	437	435	431
5c	479, 461 s	473, 462 s	444	450	444	439
5d	493, 476 s	489, 475 s	472	470	476	468
5e	405, 426 s	401, 427 s	395	398	395	390

s: Shoulder.

hypsochromically with respect to the λ_{max} in chloroform; for example for **4c**, λ_{max} was at 484 nm in DMSO and 520 nm in DMF, but at 475 nm in acetonitrile and 474 nm in methanol (480 nm in chloroform) (Fig. 1). It was also observed that λ_{max} values of dyes **5a–e** in DMSO, DMF, acetonitrile, methanol and acetic acid were shifted bathochromically with respect to the λ_{max} in chloroform. But, bathochromic shifts of λ_{max} of dyes **5a–e** in DMSO and DMF were larger than the bathochromic shifts of λ_{max} in acetonitrile, methanol and acetic acid with respect to the λ_{max} in chloroform; for example for **5d**, λ_{max} was at 493 nm in DMSO and 489 nm in DMF, but at 472 nm in acetonitrile and 470 nm in methanol (468 nm in chloroform) (Fig. 2).

3.3. Effect of acid and base

The effects of acid and base on the absorption of dye solutions were investigated and the results are shown in Table 5. The absorption spectra of the dyes in methanol were quite sensitive to the addition of base (potassium hydroxide, 0.1 M), with λ_{max} of dyes **4a–e** and **5a–e** showing a large bathochromic shift (Fig. 3). The absorption curves of dyes **4a–e** resembled those in DMF when base (potassium hydroxide, 0.1 M) was added to a solution in methanol. Also, the absorption curves of dyes **5a–e** were different from those in DMSO and DMF when base (potassium hydroxide, 0.1 M) was added to a solution in methanol. These results indicate that dyes **4a–e** and **5a–e** are present in anionic forms (**C**, **F**) in basic solution.

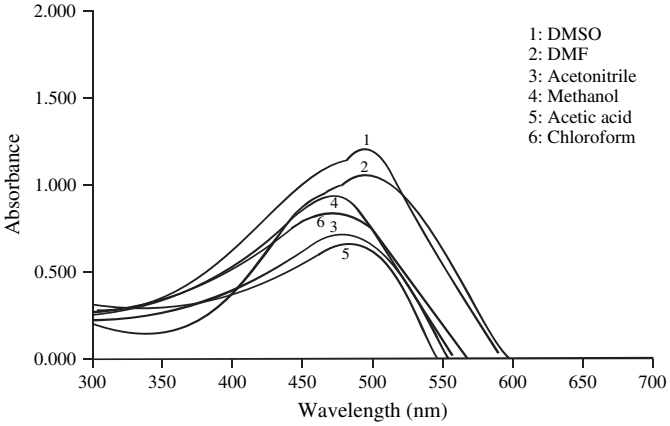


Fig. 2. Absorption spectra of dye **5d** in various solvents.

Also, λ_{max} of dyes **4a–e** and **5a–e** showed large bathochromic shifts when a small amount of piperidine was added to each of the dye solutions in chloroform (Table 5) and absorption curves of the dyes resembled those in methanol + KOH (Fig. 3). This result indicates that dyes **4a–e** and **5a–e** are present in anionic forms (**C**, **F**) in chloroform + piperidine.

When hydrochloric acid (0.1 M) was added to dye solutions in methanol, λ_{max} did not change significantly, with the exception of dyes **4a** and **4b** and the absorption curves of the dyes resembled those in acetic acid. When hydrochloric acid (0.1 M) was added to dye solutions of **4a** and **4b** in methanol, λ_{max} showed bathochromic shifts (Fig. 4). The reason for this is probably that dyes **4a**, **4b** are present in a cationic form (**G**) in strong acidic solutions (Scheme 4).

3.4. Effect of substituent

As seen in Table 4, the introduction of electron-donating methyl group into the thiazole ring and the introduction of electron-donating ethoxy group into the benzothiazole ring

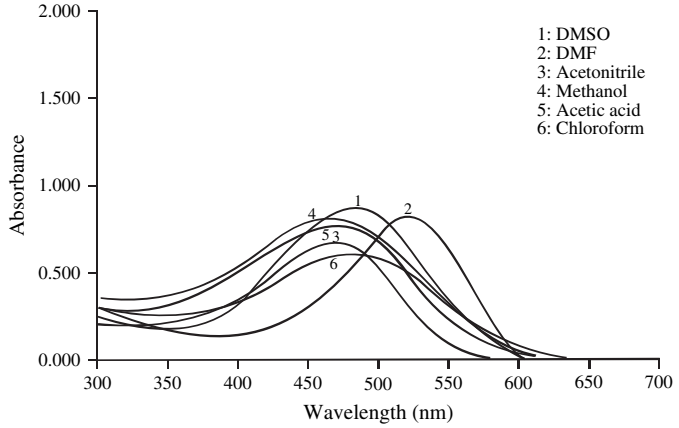


Fig. 1. Absorption spectra of dye **4c** in various solvents.

Table 5
Absorption maxima of dyes **4a–e** and **5a–e** in acidic and basic solutions

Dye no.	λ_{max} (nm)					
	Methanol	Methanol + KOH	Methanol + HCl	Chloroform	Chloroform + piperidine	Acetic acid
4a	444	493	487	446	508	440
4b	465	496	503	467	509	460
4c	474	508	460	480	522	475
4d	480	514	488	482	528	477
4e	399	450	397	400	478	395
5a	431	499	439	427	514	429
5b	437	501	440	431	514	435
5c	450	514	444	439	528	444
5d	470	507	474	468	529	476
5e	398	460	388	390	465	395

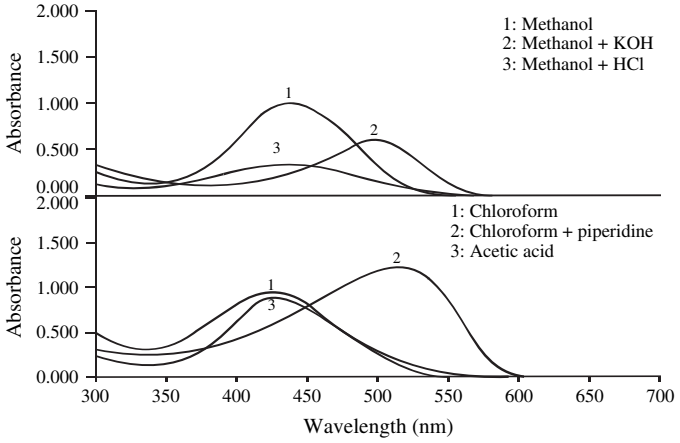


Fig. 3. Absorption spectra of dye **5a** in acidic and basic solutions.

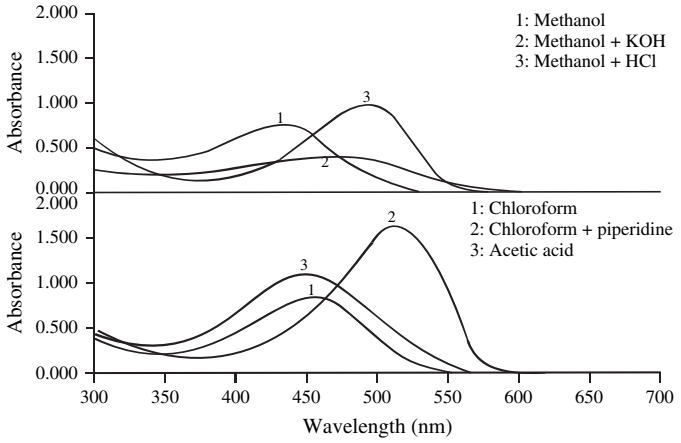


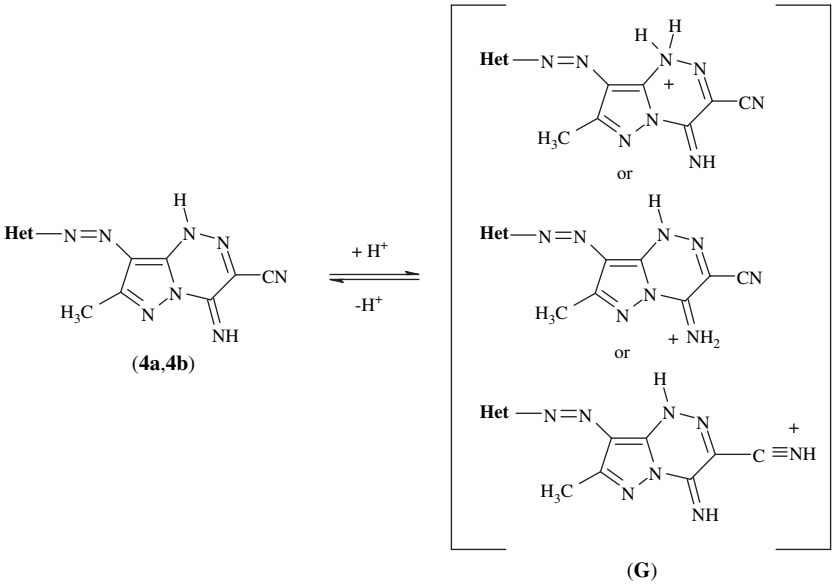
Fig. 4. Absorption spectra of dye **4a** in acidic and basic solutions.

result in bathochromic shifts in all solvents (for dye **4b** $\Delta\lambda = 21$ nm relative to dye **4a** for spectra in methanol, for dye **5b** $\Delta\lambda = 6$ nm relative to dye **5a** for spectra in methanol, for dye **4d** $\Delta\lambda = 6$ nm relative to dye **4c** for spectra in methanol and for dye **5d** $\Delta\lambda = 20$ nm relative to dye **5c** for spectra in methanol). The observed bathochromism upon introduction of electron donors, methyl and ethoxy groups, is in agreement with theory. The densities of electrons that are joined to delocalization are increased due to electron donors, methyl and ethoxy groups, that is why bathochromic shift has occurred. Also, λ_{max} values of benzothiazole substituted dyes **4c**, **d** and **5c**, **d** are the largest and λ_{max} values of isoxazole substituted dyes **e** and **e** are the smallest in all used solvents.

4. Conclusions

A series of 10 novel pyrazolo[5,1-*c*][1,2,4]triazine dyes were synthesized by heating of pyrazolylazo malononitriles and ethyl pyrazolylazo cyanoacetate in glacial acetic acid. Solvent and acid–base influences on the wavelength of maximum absorption have been studied. It was observed that although dyes **4a–e** were present in hydrazo-imine form (**B**) in acetonitrile, methanol, acetic acid and chloroform, these dyes were present in azo-enamine form (**A**) in DMSO. Also, dyes **4a–e** were present in an anionic form (**C**) in DMF. Similarly, although dyes **5a–e** were present in a single tautomeric form (**D** or **E**) in acetonitrile, methanol, acetic acid and chloroform, they are present as a mixture of azo-enamine (**D**) and hydrazo-imine (**E**) forms in DMSO and DMF. Moreover, dyes **4a–e** and **5a–e** are present in anionic forms (**C**, **F**) in basic solution. Finally, dyes **4a**, **b** are present in a cationic form (**G**) in strong acidic solutions.

Dyes **4a–e** and **5a–e** may be applied to polyester and/or polyamide fibers as disperse dyes. These dyes may also be used in biological–medical studies.



Scheme 4.

Acknowledgements

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