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Synthesis of some novel hetarylazo disperse dyes derived from 4-hydroxy-2H-1-benzopyran-2-one (4-hydroxycoumarin) as coupling component and investigation of their absorption spectra

Fikret Karcı[a,*], Nermin Ertan[b]

[a] *Department of Chemistry, Pamukkale University, Kınıklı 20017, Denizli, Turkey*
[b] *Department of Chemistry, Gazi University, Teknikokullar 06500, Ankara, Turkey*

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Abstract

Synthesis of some novel hetarylazocoumarin dyes was achieved by diazotisation of nine heterocyclic amines using nitrosyl sulphuric acid, coupling with 4-hydroxy-2*H*-1-benzopyran-2-one (4-hydroxycoumarin). These dyes were characterized by UV–vis, FT-IR and ^{1}H NMR spectroscopic techniques and elemental analysis. Both the effect of varying pH and solvent upon the absorption ability of azocoumarins are discussed using visible absorption spectra of the dyes. The color of the dyes is discussed with respect to the nature of the heterocyclic ring and to the substituent therein. The effects of temperature, concentration as well as acid and base on the visible absorption maxima of the dyes are also reported.

Keywords: Azocoumarin dyes; Heterocyclic disperse dyes; Thiazolylazo dyes; Benzothiazolylazo dyes; Solvent effect; Substituent effect

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1. Introduction

It has been known for many years that the azo compounds are the most widely used class of dyes due to their versatile application in various fields such as the dyeing of textile fiber, the coloring of different materials, colored plastics, biological–medical studies and advanced applications in organic synthesis. Many patents and papers describe the synthesis and dyeing properties of azo compounds [1–6]. Especially, azo dyes based on heterocyclic amines have been developed, and the resultant dyes have higher tinctorial strength and give brighter dyeing than those derived from aniline-based diazo components. For instance, amino-substituted thiazole, isothiazole, thiophene compounds afford very electronegative diazo components and consequently, provide a pronounced bathochromic effect compared to the corresponding benzenoid compounds [7–13].

However, very few comparable investigations have been made using coumarin-based dyes. Examples of coumarin-based dyes are disperse dyes which contain in the 7-position of the coumarin ring an electron-releasing group such as *N*,*N*-diethylamino group and in the 3-position, a heterocyclic electron-acceptor residue [14,15]. These types of coumarin-based dyes have not found the diazo group. Shawali et al. investigated the tautomerism in diazonium coupling products of 4-hydroxycoumarin [16]. They used some aniline derivatives as diazo

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* Corresponding author. Tel.: +90 258 2134030/1162; fax: +90 258 2125546.

E-mail address: fkarci@pamukkale.edu.tr (F. Karcı).

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component. According to this literature, 3-arylazo-4-hydroxycoumarins may exist in four possible tautomeric forms, but the tautomeric equilibrium is in favor of the hydrazone form in solid state and ethanol, acetic acid and pyridine. In this study, the heterocyclic based synthesized dyes contain both the diazo group and coumarin ring.

In a continuation of our work, we report here the synthesis of some hetarylazocoumarin dyes **1–9** resulting from the use of 4-hydroxycoumarin as a coupling component and an evaluation of their visible absorption spectra with respect to the influences of solvent. The color of the dyes is discussed with respect to the nature of the heterocyclic ring and substituents therein. The effects of temperature, concentration as well as acid and base on the visible absorption maxima of the dyes are also reported. The dye structures are shown in Scheme 1.

2. Experimental

2.1. General

The chemicals used in the synthesis of all dyes were obtained from Aldrich Chemical Company or Sigma Chemical Company and were used without further purification. The solvents used were of spectroscopic grade.

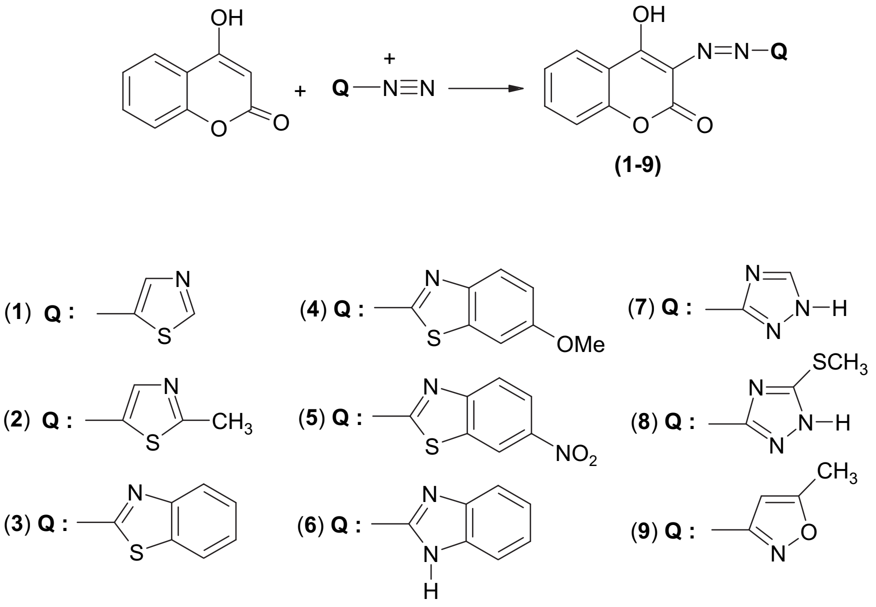
IR spectra were recorded on a Mattson 1000 FT-IR spectrophotometer in KBr. ^{1}H NMR spectra were recorded on a Bruker-Spectrospin Avance DTX 400 Ultra-Shield in DMSO with TMS as internal reference. Absorption spectra were recorded on an ATI-Unicam UV-100 spectrophotometer in various solvents. All melting points were uncorrected.

2.2. Preparation of hetarylazocoumarin dyes (**1–9**)

Diazotisation of the various heterocyclic amines was effected with nitrosyl sulphuric acid. A typical procedure is that described below used for 2-aminothiazole; all other dyes were prepared in a similar manner. The yields of the dyes are in the range of 50–79%. Characterization data are shown in Tables 1 and 2.

2.2.1. Preparation of 4-hydroxy-3-(2′-thiazolylazo)-2H-1-benzopyran-2-one (**1**)

2-Aminothiazole (2.0×10^{-3} mol) was dissolved in hot glacial acetic acid (2.5 ml) and was rapidly cooled in an ice–salt bath to −5 °C. The liquor was then added in portions during 30 min to a cold solution of nitrosyl sulphuric acid (prepared from sodium nitrite (1 g) and concentrated sulphuric acid (7 ml at 70 °C)). The mixture was stirred for an additional 1 h at 0 °C. After diazotisation was complete the azo liquor was slowly added to a vigorously stirred solution of 4-hydroxy-2*H*-1-benzopyran-2-one (2.0×10^{-3} mol) in sodium carbonate (2.0×10^{-3} mol) and water (2 ml). The pH of the reaction mixture was maintained at 7–8 by simultaneous addition of solid sodium carbonate in portions. The mixture was then stirred for 1 h at 0–5 °C. The progress of the reaction was followed by TLC using a DMF–water mixture (5/2 by volume) as the developing solvent and silica gel TLC plates as the stationary phase. The resulting solid was filtered, washed with cold water and dried. Recrystallization



Scheme 1.

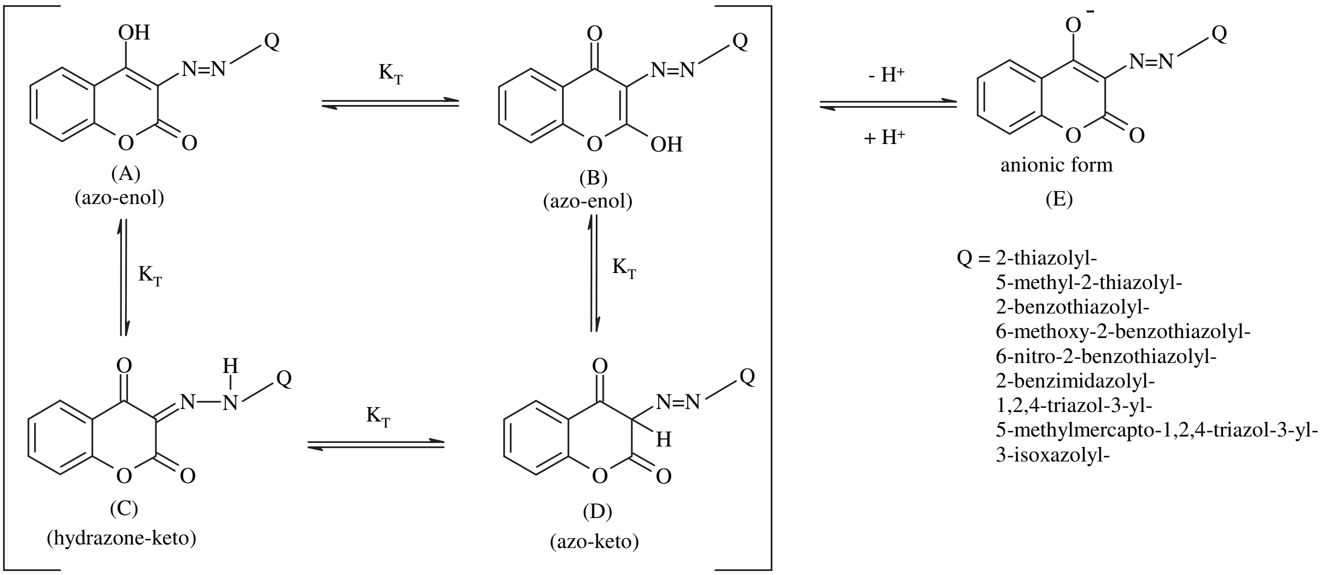
Table 1
Spectral data for dyes **1–9**

Dye no	FT-IR (cm^{-1}, in KBr)					^{1}H NMR (δ, ppm)			
	ν_{O-H}	$\nu_{Aro\text{-}H}$	$\nu_{Al\text{-}H}$	$\nu_{C=O}$	ν_{C-O}	Aro.-H	Alip.-H	X–H	Solvent
1	3536	3128, 3026	–	1766	1092	7.35(2H,m), 7.60(1H,d), 7.70(1H,d), 7.80(1H,m), 8.00(1H,m)	–	10.80(OH, b)	DMSO-d_6
2	3530	3051	2967, 2928	1754	1086	7.20(3H,m), 7.60(1H,s), 7.82(1H,m)	2.25(3H,s)	10.60(OH, b), 13.60(NH, b)	DMSO-d_6
3	3500	3179, 3153	–	1747	1119	7.10–7.61(5H,m), 7.82(1H,m), 7.90(1H,m), 7.98(1H,m)	–	13.60(OH, b), 14.70(NH, b)	DMSO-d_6
4	3500	3230, 3153	2935, 2826	1747	1080	7.15(1H,m), 7.30(2H,m), 7.70(2H,m), 8.05(2H,m)	3.80(3H,s)	13.70(OH, b), 14.80(NH, b)	DMSO-d_6
5	3518	3125, 3071	–	1678	1124	7.65(1H,m), 8.30(1H,m), 8.50(4H,m), 8.90(1H,m)	–	12.70(OH, b)	DMSO-d_6
6	3510	3080, 3054	–	1714	1107	7.35(2H,m), 7.50(1H,m), 7.75(2H,m), 7.92(1H,m), 8.17(2H,m)	–	12.50(OH, b), 14.80(NH, b), 15.30(NH, b)	DMSO-d_6
7	3500	3128	–	1740	1099	7.25(1H,m), 7.65(1H,m), 7.95(1H,m), 8.50(1H,s), 9.35(1H,m)	–	13.70(OH, b), 15.00(NH, b)	DMSO-d_6
8	3500	3144, 3070	2955	1754	1099	7.70(2H,m), 8.05(1H,m), 9.45(1H,m)	2.90(3H,s)	13.60(OH, b), 14.50(NH, b), 15.10(NH, b)	DMSO-d_6
9	3515	3243, 3166	2950	1740	1092	6.45(1H,s), 7.20(2H,m), 7.60(1H,m), 7.82(1H,m)	2.30(3H,s)	14.70(NH, b)	DMSO-d_6

X = –N,O; s, singlet; d, doublet; t, triplet; m, multiplet; b, broad.

Table 2
Element analysis of dyes **1–9**

Dye no	Molecular formula	Molecular mass	Yield (%)	C%		H%		N%		S%		Melting point (°C)
				Calcd.	Found	Calcd.	Found	Calcd.	Found	Calcd.	Found	
1	$C_{12}H_7N_3O_3S$	273	50	52.74	52.78	2.58	2.60	15.38	15.05	11.73	10.72	219–220
2	$C_{13}H_9N_3O_3S$	287	57	53.35	53.81	3.16	3.01	14.64	14.49	11.16	11.13	222–223
3	$C_{16}H_9N_3O_3S$	323	58	59.43	60.17	2.81	3.07	13.00	12.23	9.92	10.08	269–270
4	$C_{17}H_{11}N_3O_4S$	353	63	57.78	58.04	3.14	3.10	11.89	11.27	9.07	9.84	248–249
5	$C_{16}H_8N_4O_5S$	368	72	52.17	52.49	2.17	2.21	15.22	14.87	8.70	8.54	252–253
6	$C_{16}H_{10}N_4O_3$	306	79	62.75	63.17	3.27	3.19	18.30	17.81	–	–	300–301
7	$C_{11}H_7N_5O_3$	257	62	51.37	51.82	2.74	2.61	27.23	27.45	–	–	275–276
8	$C_{12}H_9N_5O_3S$	291	64	47.52	48.12	3.00	2.86	23.10	23.95	10.57	10.80	299–300
9	$C_{13}H_9N_3O_4$	271	69	57.57	57.97	3.34	3.13	15.49	15.36	–	–	220–221



Q = 2-thiazolyl-
5-methyl-2-thiazolyl-
2-benzothiazolyl-
6-methoxy-2-benzothiazolyl-
6-nitro-2-benzothiazolyl-
2-benzimidazolyl-
1,2,4-triazol-3-yl-
5-methylmercapto-1,2,4-triazol-3-yl-
3-isoxazolyl-

Scheme 2.

from DMF/H_2O mixture gave dark red (yield: 50 %, m.p: 219–220 °C).

3. Result and discussion

The hetarylazocoumarin dyes **1–9** were prepared by coupling 4-hydroxy-2*H*-1-benzopyran-2-one with diazotized heterocyclic amines in nitrosyl sulphuric acid (Scheme 1). The dyes may exist in four possible tautomeric forms, namely two azo-enol forms A and B, the hydrazone-keto form C and the azo-keto form D, as shown in Scheme 2. The deprotonation of the four tautomers leads to a common anion.

The infrared spectra of all the compounds (in KBr) showed intense carbonyl bands at 1766–1678 cm^{-1} and showed broad hydroxy bands at 3536–3500 cm^{-1}. It can be suggested that these compounds do not exist as the hydrazone-keto form and the azo-keto form in solid state. The FT-IR spectra also show a weak band or shoulder at 3243–3026 cm^{-1}, which was assigned to aromatic C–H. The other ν_{max} values of 2967 and 2928 cm^{-1} (aliphatic C–H, dye **2**), 2935 and 2826 cm^{-1} (aliphatic C–H, dye **4**), 2955 cm^{-1} (aliphatic C–H, dye **8**), 2950 cm^{-1} (aliphatic C–H, dye **9**), and 1124–1080 cm^{-1} (C–O) were recorded.

The ^1H NMR spectra measured in DMSO-d_6 at 25 °C showed a singlet at 2.25 ppm ($-CH_3$), a singlet at 3.80 ppm ($-OCH_3$), a singlet at 2.90 ppm ($-SCH_3$), a singlet at 2.30 ppm ($-CH_3$), a multiplet from 6.45 to 9.45 ppm for aromatic protons (Aro.-H), a broad peak from 10.60 to 13.70 ppm for hydroxy proton (–OH), and a broad peak from 13.60 to 15.30 ppm (–NH–) (Table 1). ^1H NMR spectra of dye **1** and dye **7** exhibit one broad peak at 10.80 and 13.70 ppm, respectively, which were attributed to –OH protons. ^1H NMR spectrum of dye **9** exhibits one broad peak at 14.70 ppm, which was attributed to –NH– proton. These results show that the dyes may exist as a mixture of several tautomeric forms in DMSO except for dye **1**, dye **7** and dye **9**. Dye **1**, dye **7** and dye **9** are in favor of the predominantly single tautomeric form in DMSO.

Table 3
Influence of solvent on λ_{max} (nm) of dyes **1–9**

Dye no	DMSO	DMF	Acetonitrile	Methanol	Acetic acid	Chloroform
1	424	416	425	410	424, 400 s	435, 405 s
2	419, 488 s	418, 483 s	435	436	440	447
3	359, 346 s, 415 s	356, 370 s, 417 s	369, 353 s, 427 s	366, 352 s, 421 s	367, 354 s, 437 s	370, 355 s, 437 s
4	366, 386 s, 431 s	368, 385 s, 427 s	366, 379 s, 443 s	365, 380 s, 460 s	367, 383 s, 457 s	367, 382 s, 470 s
5	376	370	352	351	335	343
6	472, 446 s	465, 439 s	427	426, 446 s	410, 456 s	414, 446 s
7	378	380	371, 360 s, 412 s	373, 354 s, 410 s	371	377, 370 s, 417 s
8	359, 376 s	358, 376 s	356, 378 s	359, 373 s	359, 373 s	362, 380 s
9	371	371	365	364	365	369

s, shoulder.

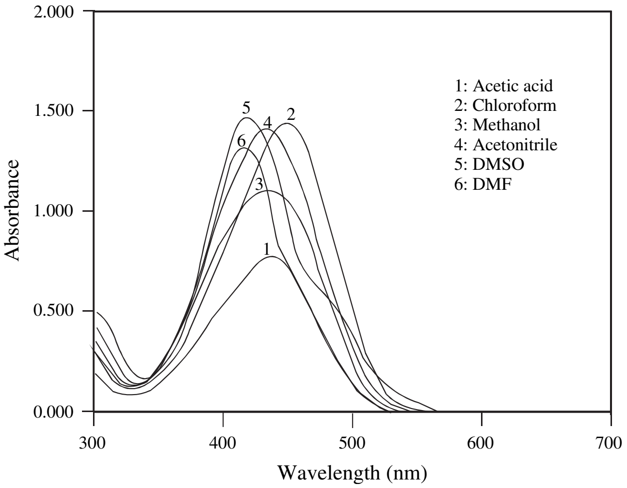


Fig. 1. Absorption spectra of dye **2** in various solvents.

3.1. Solvent effect

UV–vis absorption spectra were recorded using an ATI-Unicam UV-100 spectrophotometer in the wavelength range 300–700 nm. Absorption spectra of hetarylazocoumarin dyes **1–9** were recorded in various solvents at a concentration of $\sim 10^{-6}$–10^{-8} M and these are all run at different concentrations. The results were summarized in Table 3. The pH value of all solutions used was in the range between acidic and basic. The visible absorption spectra of the dyes did not show regular variation with the polarity of solvents.

The dyes showed two or three absorbance in various solvents except for dye **5** and dye **9**. Dye **5** and dye **9** showed single absorbance in all used solvents. It can be suggested that dye **5** and dye **9** are predominantly in the single tautomeric form in all used solvents. But the other dyes may exist as a mixture of tautomeric forms in various solvents.

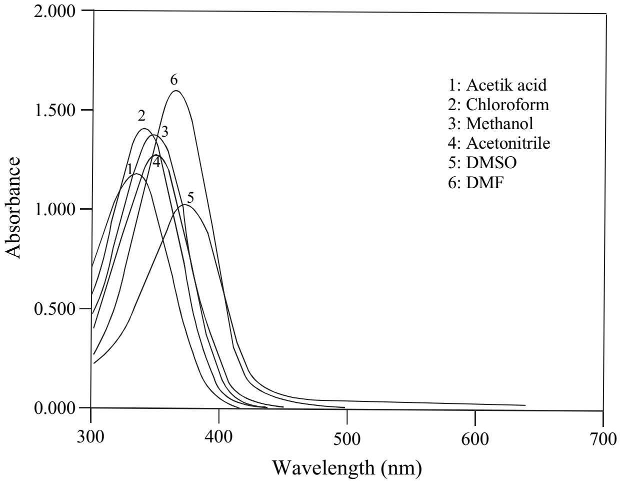


Fig. 2. Absorption spectra of dye **5** in various solvents.

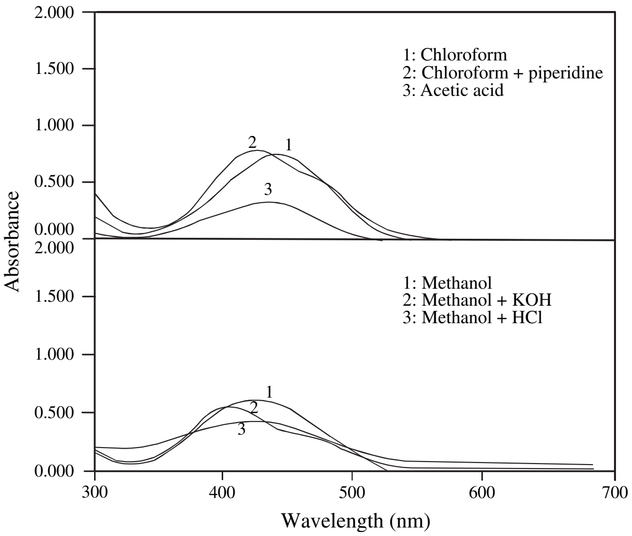


Fig. 3. Absorption spectra of dye **2** in acidic and basic solutions.

It was observed that the absorption spectra of dye **1**, dye **2** and dye **3** in all solvents hypsochromically shifted with respect to the absorption spectra in chloroform (e.g. for dye **2** λ_{max} is 447 nm in $CHCl_3$, 419 nm in DMSO, 418 nm in DMF) (Fig. 1). But, the λ_{max} of dye **5** and dye **6** showed bathochromic shift in DMSO, DMF, acetonitirile and methanol with respect to the λ_{max} in chloroform (e.g. for dye **5** λ_{max} is 343 nm in $CHCl_3$, 376 nm in DMSO, 370 nm in DMF) (Fig. 2). The absorption spectra of dye **4**, dye **7**, dye **8** and dye **9** in various solvents did not significantly change.

Heterocyclic based azo disperse dyes tend to show larger solvatochromic effects than azo-benzene based dyes because of the increased polarity of the dye system, especially in the excited state. Similar effects for the dyes containing benzothiazolyl, thiazolyl and thienyl moities have been reported for some derivatives of *N*-β-cyanoethyl-*N*-β-hydroxyethyl aniline [17–21]. On the other hand, in hetarylazocoumarin dyes, their tautomeric equilibria need to be considered.

Strong evidence for the existence of dyes **1–6** existing in an equilibrium is provided by the single or twin isosbestic points in the visible spectra of, for example, dye **2** and dye **5** in different solvents (Figs. 1 and 2). This equilibrium may exist between the tautomeric forms or between the tautomeric forms and the anionic form. The equilibrium depends on the acidity of the solvents used.

It was also observed that the absorption curves of the dyes were very sensitive to the base with the exception of dyes **3** and **5**. The shoulder of the dyes generally showed large bathochromic shifts when a small amount at piperidine was added to each of the dye solutions in DMSO, DMF and chloroform with the exception of dye **4** (Table 4). A typical example is shown in Fig. 3. The λ_{max} and shoulders of dye **4** showed hypsochromic shifts when a small amount of piperidine was added to dye **4**

Table 4
Absorption maxima of dyes **1–9** in acidic and basic solutions

Dye no	λ_{max} (nm)								
	DMSO	DMSO + piperidine	DMF	DMF + piperidine	Methanol	Methanol + KOH	Methanol + HCl	Chloroform	Chloroform + piperidine
1	423	415, 478 s	414	414, 488 s	410	405, 471 s	419	435, 405 s	423, 478 s
2	422, 488 s	417, 488 s	417, 483 s	416, 483 s	428	410, 470 s	428	446	431, 476 s
3	359, 341 s, 414 s	359, 329 s, 415 s	358, 371 s, 414 s	359, 346 s, 414 s	367, 352 s, 419 s	351, 336 s, 405 s	367, 353 s, 424 s	370, 355 s, 437 s	361, 341 s, 434 s
4	366, 386 s, 431 s	348, 368 s, 429 s	368, 385 s, 428 s	338, 361 s, 400 s	365, 380 s, 461 s	324, 351 s, 424 s	365, 376 s, 449 s	367, 383 s, 452 s	372, 385 s, 447 s
5	376	376	371	371	351	352	341	343	352
6	471, 443 s	485, 439 s	464, 437 s	467, 439 s	426, 446 s	440, 461 s	412, 449 s	414, 446 s	472, 444 s
7	379	397, 427 s, 448 s	380	397, 427 s, 456 s	374, 361 s, 420 s	370, 426 s	358	377, 370 s, 417 s	384, 412 s
8	359, 376 s	388, 328 s, 540 s	358, 373 s	336, 488 s	359, 373 s	324, 501 s	360, 375 s	362, 380 s	358, 380 s
9	370	370, 439 s	371	370, 439 s	365	364, 434 s	367	370	381, 434 s

s, shoulder.

Table 5
Influence of temperature and sample concentration on absorption maxima of dyes **1–9**

Dye no	λ_{max} (nm)													
	DMSO conc. 25 °C	DMSO dil. 25 °C	DMSO 70 °C	DMF conc. 25 °C	DMF dil. 25 °C	DMF 70 °C	A.nitrile conc. 25 °C	A.nitrile dil. 25 °C	Meth. conc. 25 °C	Meth. dil. 25 °C	A. acid conc. 25 °C	A. acid dil. 25 °C	Chl. conc. 25 °C	Chl. dil. 25 °C
1	424	423	417	416	414	413	425	419	410	410	424, 400 s	426, 400 s	435, 405 s	435, 405 s
2	419, 488 s	422, 488 s	420, 487 s	418, 483 s	417, 483 s	418, 483 s	435	432	436	428	440	439	447	446
3	359, 346 s, 415 s	359, 341 s, 414 s	357, 340 s, 413 s	356, 370 s, 417 s	358, 371 s, 414 s	359, 371 s, 414 s	369, 353 s, 427 s	369, 353 s, 427 s	366, 352 s, 421 s	367, 352 s, 419 s	367, 354 s, 437 s	366, 353 s, 436 s	367, 354 s, 437 s	370, 355 s, 437 s
4	366, 386 s, 431 s	366, 386 s, 431 s	367, 386 s, 431 s	368, 385 s, 427 s	368, 385 s, 428 s	368, 388 s, 430 s	366, 379 s, 443 s	351, 380 s, 432 s	365, 380 s, 460 s	365, 380 s, 461 s	367, 383 s, 457 s	367, 381 s, 459 s	367, 382 s, 470 s	367, 383 s, 452 s
5	376	376	372	370	371	368	352	352	351	351	335	335	343	343
6	472, 446 s	471, 443 s	469, 440 s	465, 439 s	464, 437 s	462, 432 s	427	426	414, 468 s	416, 446 s	410, 456 s	412, 458 s	414, 446 s	413, 446 s
7	378	379	378	380	380	380	371, 360 s, 412 s	373, 361 s, 413 s	373, 354 s, 410 s	374, 361 s, 420 s	371	371	377, 370 s, 417 s	377, 370 s, 417 s
8	359, 376 s	359, 376 s	360, 373 s	358, 376 s	358, 373 s	358, 373 s	356, 378 s	357, 372 s	359, 373 s	359, 373 s	359, 373 s	358, 372 s	362, 380 s	362, 380 s
9	371	370	370	371	371	370	365	366	364	365	365	364	369	370

Abbreviations: s, shoulder; A.acid, acetic acid; Chl., chloroform; Meth., methanol; A.nitrile, acetonitrile; conc., concentrated; dil., diluted.

solutions in DMSO and DMF. The λ_{max} of the dyes in methanol also showed bathochromic shift when 0.1 M KOH was added with the exception of dye **4**. The λ_{max} and shoulders of dye **4** showed hypsochromic shifts when 0.1 M KOH was added. The absorption spectra of the dyes in methanol did not significantly change when 0.1 M HCl was added with the exception of dye **5**. The λ_{max} of dye **5** in methanol showed hypsochromic shift when 0.1 M HCl was added. This indicates that the hetarylazocoumarins **1**, **2**, **6**, **7**, **8** and **9** exist in a dissociated state in methanol, DMSO and DMF. The results are in agreement with those obtained previously for both hetarylazopyridones [22] and hetarylazopyrazolones [11,23].

The effects of the dye concentration and temperature on absorption maxima were examined (Table 5). Although the λ_{max} values of dyes **1–9** did not change with dye concentration in chloroform, acetic acid, DMSO and DMF, the λ_{max} values of some dyes in methanol and acetonitrile (e.g. dyes **1**, **2** and **6**) showed a blue shift with decreasing concentration. This also indicates that some hetarylazocoumarins exist in tautomeric form in chloroform and acetic acid, in the common anion form in DMSO and DMF and in a partly dissociated stated in both methanol and acetonitrile.

When solutions of the dyes in DMSO and DMF were examined over the temperature range 25–70 °C (Table 5), the λ_{max} values of dyes **1–9** did not change significantly. These findings support the dissociation equilibrium of hetarylazocoumarins in proton-accepting solvents, which do not involve a change in energy.

3.2. Substituent effects

As is apparent in Table 3, the introduction of electron-donating methyl group into the thiazole ring result in bathochromic shifts in all solvents (for dye **2** $\Delta\lambda = 26$ nm relative to dye **1** for spectra in methanol). It was also observed that the introduction of electron-donating methoxy group into the benzothiazole ring (for dye **4** $\Delta\lambda_{shoulder\ 1} = 27$ nm, $\Delta\lambda_{shoulder\ 2} = 33$ nm relative to dye **3** for spectra in chloroform) resulted in bathochromic shifts at the shoulders in all solvents. The electron-accepting nitro group into the benzothiazole ring result in hypsochromic shifts in acetonitrile, methanol, chloroform and acetic acid (for dye **5** $\Delta\lambda_{max} = 15$ nm relative to dye **3** for the spectra in methanol) while they result in bathochromic shifts in DMSO and DMF (for dye **5** $\Delta\lambda_{max} = 17$ nm relative to dye **3** for spectra in DMSO). The electron-donating methylmercapto group into the 1,2,4-triazole ring result in hypsochromic shifts in all solvents (for dye **8** $\Delta\lambda_{max} = 15$ nm relative to dye **7** for spectra in chloroform).

4. Conclusions

The synthesized 3-hetarylazo-4-hydroxy-2*H*-1-benzopyran-2-one dyes (**1–9**) showed solvatochromic effects because of the increased polarity of the dye system. On the other hand, in hetarylazocoumarin dyes, their tautomeric equilibrium and dissociation need to be considered.

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