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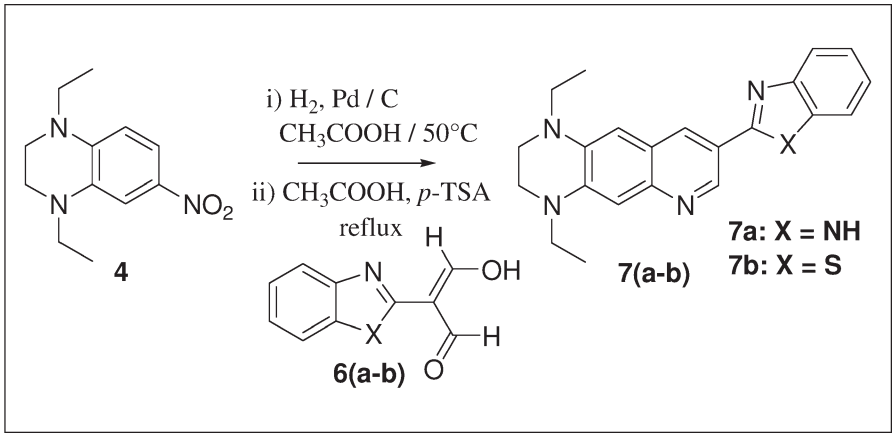
Received October 6, 2009
DOI 10.1002/jhet.422
Published online 13 July 2010 in Wiley Online Library (wileyonlinelibrary.com).



1,4-Diethyl-1,2,3,4-tetrahydro-6-nitroquinoxaline **4** was synthesized by alkylative reduction of 6-nitroquinoxaline. Catalytic reduction of **4** followed by cyclocondensation with heterocyclic malondialdehydes afforded novel 8-(heteroaryl)-1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxalines. The solutions of these novel compounds having 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline framework as an electron releasing system showed absorption in the range of 424–426 nm in the visible region and exhibited brilliant bluish-green fluorescence. The thermogravimetric curve obtained by thermogravimetric analysis displayed that these fluorophores possess excellent thermal stability with one-step thermal decomposition.

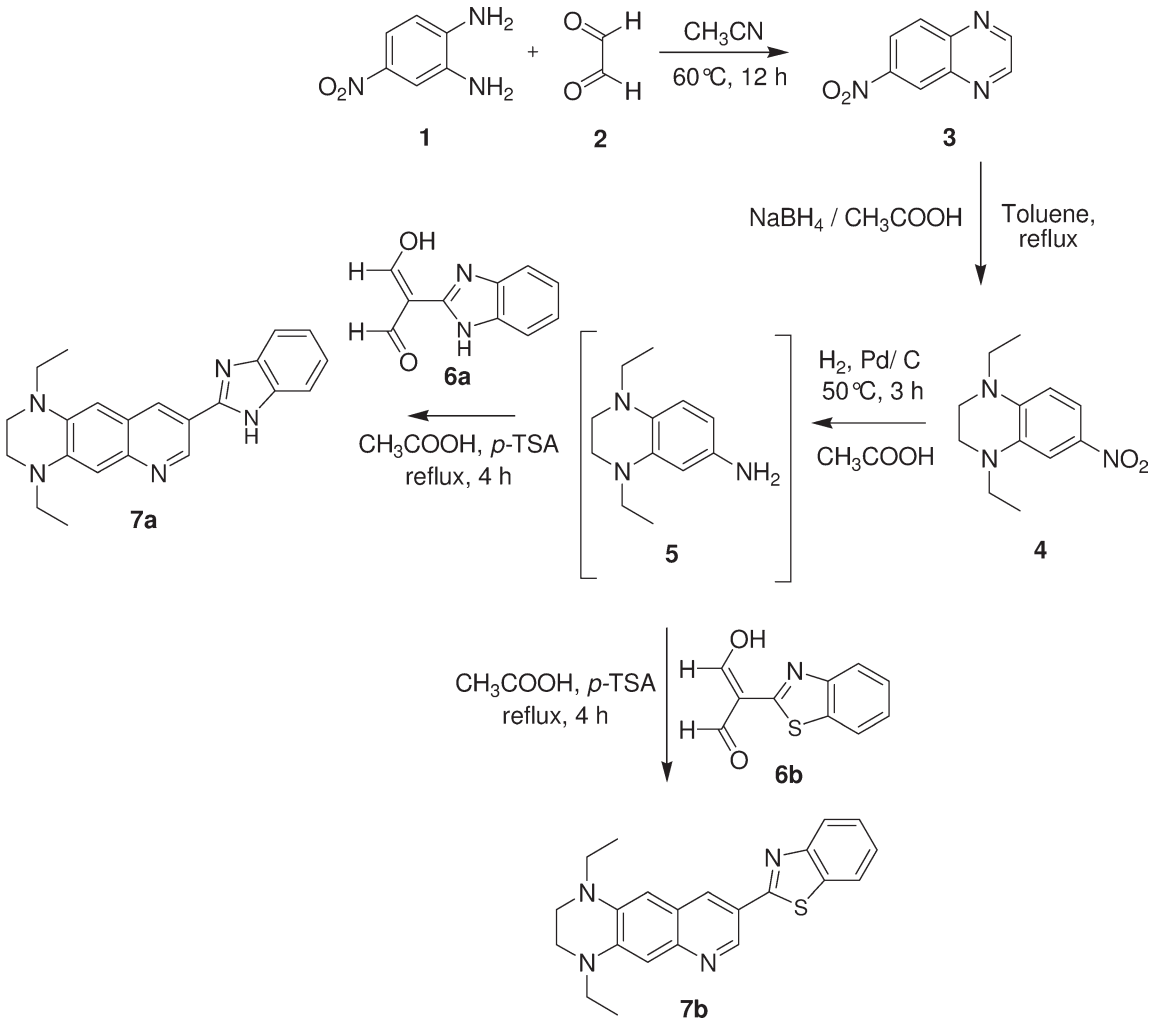
J. Heterocyclic Chem., **47**, 1066 (2010).

INTRODUCTION

The structural diversity, stability, and biological importance of heterocycles have made them attractive synthetic targets over many years, and they have found potential applications in various fields of science and technology. Especially in the field of colorants, heterocycles have gained extreme importance because of their planar and rigid π-conjugation system. Many heterocycles based on rigid ring systems such as coumarins [1], thiazoles [2], benzimidazoles [3], pyrazines [4], naphthalimides [5], and oxadiazoles [6] are well-established fluorescent dye chromophores. Heterocyclic fluorescent compounds have been extensively investigated for various potential applications including tunable dye lasers [7], molecular probes for biochemical research [8], and traditional textile and polymer fields [9].

Quinoxaline is one of the interesting heterocyclic systems. Many quinoxaline scaffolds are found as a core unit in a number of biologically active compounds [10,11]. Its derivatives are used in the development of novel organic dyes and organic semiconductors [12]. Fluorescent styryl dyes based on fused quinoxaline system are reported in the literature. In earlier work from our laboratory, the versatility of quinoxaline has been demonstrated [13–16]. Quinoxalines can be easily reduced to 1,2,3,4-tetrahydroquinoxalines by reducing agents such as lithium aluminium hydride [17] and sodium borohydride [18] in excellent yields. Sequential reduction and alkylation of N-heterocycles such as indole to N-alkylated indoline and quinoline to 1,2,3,4-tetrahydroquinoline by sodium borohydride and trifluoroacetic acid is well known [19–22]. Quinoxalines can also be sequentially reduced and dialkylated using sodium borohydride and carboxylic acids. 6-Nitroquinoxaline has been subjected to similar reductive alkylation using sodium borohydride and glacial acetic acid to obtain 1,4-diethyl-1,2,3,4-tetrahydro-6-nitroquinoxaline [23]. The 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline framework is rigid and highly electron rich. We have

Scheme 1. Synthetic pathway of compounds **7a–7b**.



reported mono- and bis-styryl dyes derived from 1,4-diethyl-1,2,3,4-tetrahydro-6-methoxyquinoxaline [24]. These dyes having orange to violet hue displayed pronounced bathochromicity and good thermal stability. A series of highly fluorescent coumarin derivatives based on 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline framework, synthesized by us, exhibited excellent bathochromicity [25]. These results encouraged us to envisage that, the molecular structures possessing a strong electron donating and rigid 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline unit in conjugation with heterocyclic π-conjugated system should exhibit brilliant fluorescence and display absorption maxima in the yellow region of the electromagnetic spectrum.

In this communication, we report the synthesis and spectroscopic properties of novel pyrido[2,3-g]quinoxaline derivatives having 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline framework as an electron releasing unit in conjugation with electron accepting heterocycles like benzimidazole and benzothiazole. The electronic properties of these highly fluorescent compounds were analyzed by UV-vis absorption spectroscopy and fluorescence emission spectroscopy. The fluorescent compounds **7a** and **7b** were also evaluated for thermal stability by thermogravimetric analysis. The spectroscopic properties of these pyrido[2,3-g]quinoxaline derivatives were compared with the closely related coumarin analogs **8a**–**8b** and styryl derivatives **9a**–**9b**.

RESULTS AND DISCUSSION

Synthesis of compounds 7a–7b. Substituted 1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxalines **7a–7b** were synthesized by cyclocondensation of 6-amino-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline **5** and suitable malondialdehyde derivatives **6a–6b** as depicted in Scheme 1. 4-Nitro-1,2-phenylenediamine **1** was condensed with glyoxal **2** in dry acetonitrile to obtain 6-nitroquinoxaline **3** in excellent yield. Reductive

Table 1
Spectral properties of compounds **7a–7b** in toluene, chloroform, hexane, and methanol.

	Toluene				Chloroform				Hexane				Methanol			
Compd.	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})
7a	418	497	79	34,404	422	502	80	33,290	412	483	71	33,611	424	512	88	29,854
7b	423	519	96	25,956	426	520	93	26,591	418	507	89	26,666	426	533	107	25,469

alkylation of 6-nitroquinoxaline with sodium borohydride and glacial acetic acid in dry toluene yielded 1,4-diethyl-6-nitro-1,2,3,4-tetrahydroquinoxaline **4** as a bright red solid, which was then hydrogenated over 10% palladium charcoal in glacial acetic acid to afford 6-amino-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline **5**. The catalyst was filtered under nitrogen atmosphere and filtrate was immediately used for further reactions as the amino compound **5** was unstable and rapidly oxidized. The reaction of 6-amino-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline **5** with appropriate malondialdehyde derivatives **6a–6b** took place readily in glacial acetic acid in the presence of equivalent amount of *p*-toluenesulphonic acid to yield 8-(heteroaryl)-1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxalines **7a–7b**. This cyclocondensation reaction proceeded in a facile manner owing to the presence of electron releasing 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline framework. The structures of the compounds were confirmed by FT-IR, ^{1}H NMR spectroscopy, mass spectrometry, and elemental analysis. The results are summarized in the experimental section.

Spectral characteristics of compounds 7a–7b. Basic spectral characteristics of the chromophores such as absorption maxima (λ_{max}), emission maxima (λ_{em}), and extinction coefficient (ε) were measured in different solvents and are presented in Tables 1 and 2. The electronic absorption spectra of the compounds **7a–7b** displayed absorption maxima in the visible region from 412 to 427 nm. Compound **7a** showed absorption maxima at 412 nm in hexane, lowest among two derivatives, whereas compound **7b** showed well-pronounced absorption maxima at 427 nm in DMF which is the highest between the two derivatives. To investigate the influence of solvents on the absorption maxima of compounds **7a** and **7b**, their absorption spectra were measured in different solvents such as toluene, chloroform, ethyl acetate, hexane, methanol, DMF, acetonitrile, and ethyl acetate. The solvents differ considerably in polarity and ability to form H-bonds. From the presented values in Tables 1 and 2, it is apparent that practically no solvatochromism was observed. Only in the case of **7a** in hexane, significant hypsochromic shift in the absorption maxima was noticed. Figure 1 displays absorption maxima of the compounds **7a–7b** in methanol.

These 8-(heteroaryl)-1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxalines were expected to be strongly fluorescent in view of the conjugation of rigid and electron rich 1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxaline ring with electron accepting heterocycles. To our delight, both compounds exhibited strong bluish-green fluorescence with large Stokes shift values (Tables 1 and 2). Especially, fluorophore **7b** containing benzothiazole ring showed remarkably high Stokes shift value of 107 in methanol. Figure 2 displays emission maxima of

Table 2
Spectral properties of compounds **7a–7b** in DMF, acetonitrile and ethyl acetate.

	DMF				Acetonitrile				Ethyl acetate			
Compd.	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})	λ_{max} (nm)	λ_{em} (nm)	Stokes shift	ε (l mol^{-1} cm^{-1})
7a	423	505	82	28,054	421	506	85	29,095	418	494	76	30,940
7b	427	528	101	24,995	425	527	102	24,235	421	519	98	24,972

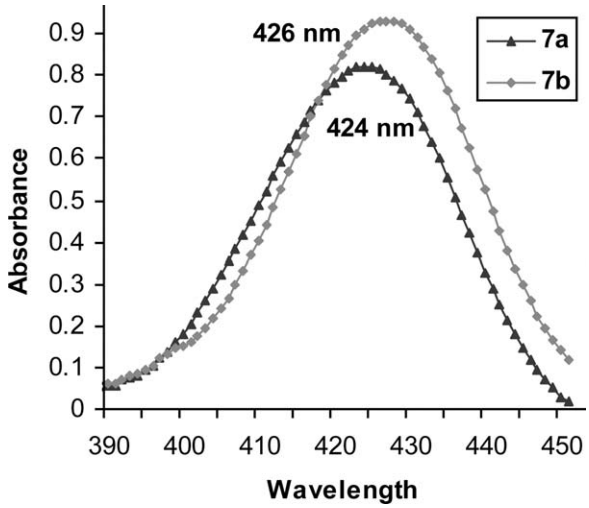


Figure 1. Absorption maxima of compounds **7a–7b** in methanol.

the compounds **7a–7b** in methanol. Figure 3 shows photographs of the fluorophores **7a** and **7b** in UV light (366 nm).

As stated earlier, the fluorophores **7a** and **7b** have rigid and electron rich 1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxaline ring in conjugation with electron accepting heterocycles. The situation is rather similar to the coumarin fluorophores **8a** and **8b** (Table 3) reported by us [25]. Also, the styryl dyes **9a** and **9b** (Table 3) were derived from the same electron donor and acceptors [24]. In short, compounds **7a–7b** are structural analogs of compounds **8a–8b** and **9a–9b**. Hence, the spectral properties of **7a** and **7b** in methanol were also compared with the established dyes **8a–8b** and **9a–9b**. The comparative data are summarized in Table 3. Compounds **7a** and **7b** showed intense yellow hue with absorption maxima at 424 and 426 nm, respectively, whereas compounds **8a** and **8b** showed bright orange

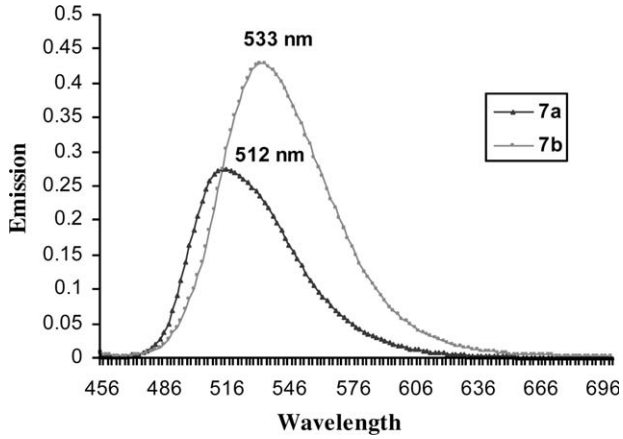


Figure 2. Emission maxima of compounds **7a–7b** in methanol.

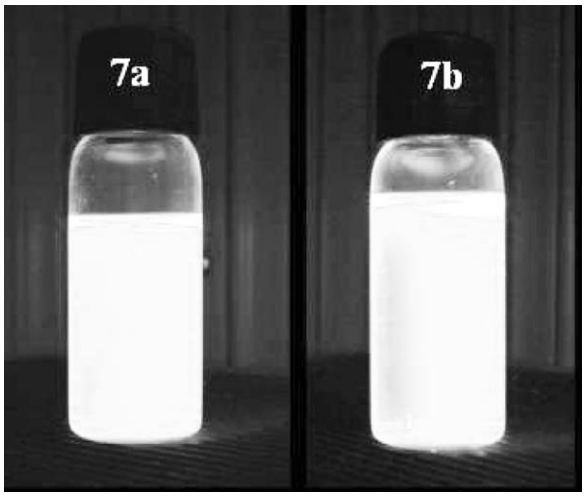
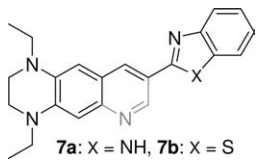
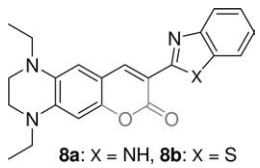
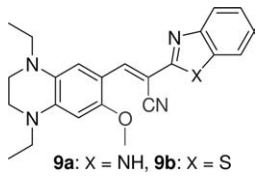
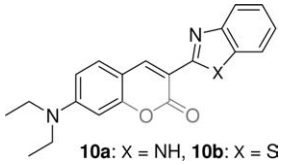


Figure 3. Photographs of fluorophores **7a–7b** in UV light (366 nm).

hue and absorbed at 483 and 501 nm, respectively. The large bathochromic shift in the case of **8a** and **8b** is clearly due to the presence of lactone ring. The compounds **8a** and **8b** were highly fluorescent, as it is usual with coumarin compounds. Stokes shift value of **8a** was almost close to that of **7a**, whereas Stokes shift value of **8b** was lower than that of **7b**. It must be noted that the styryl dyes **9a** and **9b**, having same electron donating 1,4-diethyl-1,2,3,4-tetrahydroquinoxaline skeleton in conjugation with electron accepting benzimidazole and benzothiazole rings, respectively, were nonfluorescent. The absence of fluorescence was probably due to lack of rigidity provided by pyrido ring as in the case of compounds **7a–7b** and lactone ring as in the case of compounds **8a–8b**. However, the styryl dyes **9a** and **9b** showed remarkably higher bathochromic shift with absorption maxima at 501 and 528 nm, respectively, owing to the presence of an additional electron

Table 3

Spectral properties of compounds **7a**–7**b**, **8a**–**8b**, **9a**–**9b**, and **10a**–**10b** in methanol.

Structure	Compd.	λ_{max} (nm)	ε (l mol^{-1} cm^{-1})	λ_{em} (nm)	Stokes shift	Quantum yield[a] (Φ)
 **7a**: X = NH, **7b**: X = S	**7a**	424	24,054	512	88	0.1
	7b	426	25,469	533	107	0.14
 **8a**: X = NH, **8b**: X = S	**8a**	483	24,600	574	91	0.28
	8b	501	33,900	598	97	0.34
 **9a**: X = NH, **9b**: X = S	**9a**	501	29,218	–	–	–
	9b	528	25,514	–	–	–
 **10a**: X = NH, **10b**: X = S	**10a**	435	52,200	480	45	0.62
	10b	465	54,000	491	26	0.7

[a] Quantum yields were measured in methanol using Rhodamine-6G (Φ = 0.94) as standard [26,27].

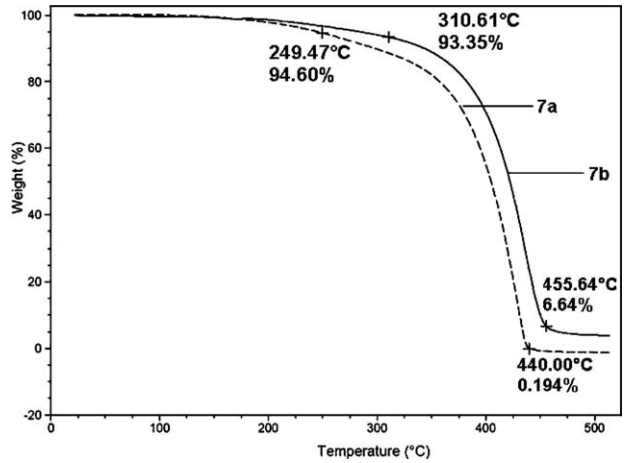


Figure 4. Thermogravimetric curves of fluorophores **7a–7b**.

accepting cyano group. In all the three classes, the compounds **7b**, **8b**, and **9b** having benzothiazole ring underwent bathochromic shift relative to their respective analogs having benzimidazole ring, owing to high electronegativity of sulfur atom in the ring.

To complement this study, spectral properties of fluorescent compounds **7a–7b** and **8a–8b** were compared with that of commonly encountered commercial fluorescent coumarin dyes **10a** (Coumarin 535) and **10b** (Coumarin 540) (Table 3) having same electron accepting groups. As expected, the rigid coumarins **8a** and **8b** showed significant bathochromic shift in absorption maxima compared with the nonrigid coumarins **10a** and **10b**. The compounds **10a** and **10b**, however, have remarkably high molar extinction coefficient values and absorb at longer wavelength compared with quinoxaline derivatives **7a** and **7b**. The Stokes shift values of compounds **10a** and **10b** are lower than that of compounds **7a–7b** and **8a–8b**. The fluorescence quantum yields (Φ) of the compounds were measured in methanol using Rhodamine-6G ($\Phi = 0.94$) [26,27] as standard. Compound **7b** showed Φ value of 0.14 which is marginally higher than that of **7a** ($\Phi = 0.1$). The fluorescence quantum yields of compounds **7a** and **7b** were found to be lower than that of coumarin derivatives. Although, the quinoxaline derivatives **7a–7b** possess similar electron donor and acceptor groups as coumarins **8a–8b** and **10a–10b**, they exhibit significant hypsochromic shift in absorption maxima and lower fluorescence quantum yields. The lower fluorescence in quinoxaline derivatives is due to the absence of lactone framework in coumarins which impart rigidity, high electronegativity, and excellent planarity to the molecule.

Thermal properties of fluorophores 7a and 7b. The fluorophores were subjected to the thermogravimetric analysis to investigate their thermal stability. The change in weight of the compound was measured as a function of temperature. Figure 4 displays thermograph of the fluorophores **7a** and **7b**. The thermogravimetric curves for the compounds show a clear plateau followed by a sharp and smooth decomposition curve. The loss in weight of the compound **7a** was rapid when heated above 250°C. This fact indicates that the compound is stable up to 250°C after which it decomposes rapidly and decomposition completes at 440°C. Among the two compounds, compound **7b** in particular showed excellent thermal stability up to 310°C. Rapid decomposition of **7b** occurred when it was heated above 310°C. The decomposition completed at about 455°C. Both the fluorophores underwent one-step thermal decomposition. Coumarin chromophores **8a–8b** and styryl dyes **9a–9b** also showed thermal stability above 250°C with smooth, one step thermal decomposition curve [24,25].

CONCLUSION

In conclusion, novel 8-(heteroaryl)-1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxalines are valuable as new fluorescent chromophores having absorption maxima at 412–427 nm and emission maxima at 502–533 nm in different solvents. These compounds did not show any appreciable solvatochromism and have lower fluorescence quantum yields than coumarins having same electron donating and accepting groups. The compounds displayed good thermal stability.

EXPERIMENTAL

All melting points were uncorrected and are in °C. IR spectra were recorded on a Bomem Hartmann and Braun MB-Series FT-IR spectrometer (KBr). ^{1}H NMR spectra were recorded on Varian 300 MHz mercury plus spectrometer, and chemical shifts are expressed in δ ppm using TMS as an internal standard. Mass spectra were recorded on Micromass: Q-T of micro (YA-105) mass spectrometer. Microanalysis for C, H, N, and S were performed on Thermofignnin elemental analyzer. Electronic spectra were recorded on Spectronic spectrophotometer. The fluorescence maxima of the compounds were recorded on Jasco FP-1520 fluorimeter. Thermogravimetric analysis was carried out on SDT Q600 v8.2 Build 100 model of TA instruments.

Synthesis of 6-amino-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline (5). 1,4-Diethyl-1,2,3,4-tetrahydro-6-nitroquinoxaline **4** [23] (5.0 g, 0.021 moles) and catalytic amount of palladium on charcoal (10%) in glacial acetic acid (100 mL) were stirred in Parr hydrogenator at 50°C under an atmosphere of hydrogen until think layer chromatography (TLC) (eluent: 10% ethyl acetate in *n*-hexane) of the reaction mixture showed no red colored spot of reactant. The reaction mixture was then filtered under nitrogen atmosphere to separate the catalyst. 6-Amino-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline **5** thus obtained was not isolated and subsequently used for further reaction immediately after filtration as it was found to be unstable [23]. (The

pale yellow reaction mixture was filtered under nitrogen blanket as it turns black on exposure to air).

General method for the synthesis of compounds (7a–7b). Above reaction mixture, appropriate malondialdehyde derivative **6a** or **6b** [28] (0.021 moles) and *p*-toluenesulphonic acid (*P*-TSA) (3.6 g, 0.021 moles) were heated to reflux under nitrogen atmosphere for 4 h. The reaction mixture was then cooled to room temperature, neutralized with dilute sodium hydroxide solution (10%) maintaining the temperature below 15°C. Dark brown solid obtained was filtered, washed with water and dried. The crude compound was purified by column chromatography on activated neutral aluminium oxide using toluene–ethyl acetate (7:3) system.

8-(Benzimidazol-2-yl)-1,4-diethyl-1,2,3,4-tetrahydropyrido[2,3-g]quinoxaline (7a). (5.17 g, 69%); mp 160–162°C; ir (KBr) ν_{max} cm^{-1}: 3090–3015, 2900–2850, 1688, 1612, 1279; ^1H NMR: δ 1.20 (t, $J = 7.1$ Hz, 3H, CH_3), δ 1.26 (t, $J = 7.1$ Hz, 3H, CH_3), δ 3.15–3.21 (m, 2H), δ 3.29 (q, $J = 7.1$ Hz, 2H, CH_2), δ 3.39 (q, $J = 7.1$ Hz, 2H, CH_2), δ 3.47–3.53 (m, 2H), δ 6.69 (s, 1H, phenyl proton), δ 7.03 (s, 1H, phenyl proton), δ 7.31–7.37 (m, 2H, protons on benzimidazole ring), δ 7.53–7.58 (m, 1H, proton on benzimidazole ring), δ 7.72–7.77 (m, 1H, proton on benzimidazole ring), δ 8.55 (d, $J = 1.95$ Hz, 1H, proton *para* to N of pyrido ring), δ 9.26 (d, $J = 1.95$ Hz, 1H, proton *ortho* to N of pyrido ring), Anal. Calcd for $C_{22}H_{23}N_5$: C, 73.92; H, 6.49; N, 19.59. Found: C, 73.95; H, 6.48; N, 19.55; ms: *m*/*z* 358 (M^++H).

8-(Benzthiazol-2-yl)-1,4-diethyl-1,2,3,4-tetrahydroquinoxaline (7b). (5.74 g, 73%); mp 174–176°C; ir (KBr) ν_{max} cm^{-1}: 3100–3010, 2911–2850, 1682, 1608, 1280; ^1H NMR: δ 1.21 (t, $J = 6.9$ Hz, 3H, CH_3), δ 1.28 (t, $J = 6.9$ Hz, 3H, CH_3), δ 3.13–3.18 (m, 2H), δ 3.31 (q, $J = 6.9$ Hz, 2H, CH_2), δ 3.41 (q, $J = 6.9$ Hz, 2H, CH_2), δ 3.49–3.54 (m, 2H), δ 6.71 (s, 1H, phenyl proton), δ 7.04 (s, 1H, phenyl proton), δ 7.31–7.51 (m, 2H, phenyl protons on benzthiazole ring), δ 7.87–7.91 (m 1H, phenyl proton on benzthiazole ring), δ 8.03–8.07 (m, 1H, phenyl proton on benzthiazole ring), δ 8.42 (d, $J = 2.42$ Hz, 1H, proton *para* to N of pyrido ring), δ 9.11 (d, $J = 2.42$ Hz, 1H, proton *ortho* to N of pyrido ring); Anal. Calcd for $C_{22}H_{22}N_4S$: C, 70.56; H, 5.92; N, 14.96; S, 8.56. Found: C, 70.59; H, 5.88; N, 14.97; S, 8.57; ms: *m*/*z* 375 (M^++H).

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