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Synthesis, properties, and solvatochromism of 1,3-dimethyl-5-{(thien-2-yl)-[4-(1-piperidyl) phenyl]methylidene}-(1H,3H)-pyrimidine-2,4,6-trione

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ABSTRACT: A new merocyanine dye, 1,3-Dimethyl-5-{(thien-2-yl)-[4-(1-piperidyl)phenyl]methylidene}-(1H, 3H)-pyrimidine-2,4,6-trione **3**, has been synthesized by condensation of 2-[4-(piperidyl)benzoyl]thiophene **1** with N,N'-dimethyl barbituric acid **2**. The solvatochromic response of **3** dissolved in 26 solvents of different polarity has been measured. The solvent-dependent long-wavelength UV/Vis spectroscopic absorption maxima, v_{max}, are analyzed using the empirical Kamlet–Taft solvent parameters π^* (dipolarity/polarizability), α (hydrogen-bond donating capacity), and β (hydrogen-bond accepting ability) in terms of the well-established linear solvation energy relationship (LSER):

$$\tilde{v}_{max} = (\tilde{v}_{max})_0 + s\pi^* + a\alpha + b\beta \tag{1}$$

The solvent independent coefficients **s**, **a**, and **b** and $(v_{max})_0$ have been determined. The McRae equation and the empirical solvent polarity index, $E_T(30)$ have been also used to study the solvatochromism of **3**. Copyright © 2007 John Wiley & Sons, Ltd.

KEYWORDS: LSER correlations; solvent polarity; solvatochromism; barbituric acid derivatives; push-pull system; merocyanine dyes

INTRODUCTION

The design of new solvatochromic dyes requires a proper understanding of their behavior in solution, and how specific structural features affect their spectral properties in different media. These dyes should be fulfill all criteria for establishing solvent parameters: namely, good spectral sensitivity, absorption in the Vis range in which all solvents are transparent, high intensity of the charge-transfer (CT) transition not masked by other electronic transition, and good solubility in all organic solvents.[1]

The incorporation of distinct electron-donor and -acceptor moieties at both ends of conjugated π-systems has long been known to induce long-wavelength UV/Vis absorptions and emissions, both sensitive to the solvent environment. Compounds based on this principle have received considerable attention in the literature.[1,2] They are attractive from several points of view, such as their application as dyestuffs and laser dyes, their non-linear optical properties, and their use as fluorescent sensors.[3] Therefore, it is of importance to establish correlations between the structural parameters of organic solids and their optical properties as function of structure variations.[4–19] Some of the results in this area were obtained by our group using dipolar aromatic aminoketones of furan and thiophene moieties which are functionalized in the periphery of the molecule.[5,6]

Solvatochromic indicators displaying positive solvatochromism show smaller specific interaction and are therefore generally better suited for the description of solvent polarity parameters in terms of unspecific electrostatic interactions.[15b]

Merocyanines seem especially promising candidates as solvatochromic dyes owing to their electronic structure.[1] The ground-state electronic structure of merocyanine dyes having a push-pull system can be described in terms of a two-state valence-bond model. This model assumes

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resonance between neutral and zwitterionic molecular structures and provides a useful qualitative interpretation of their solvatochromism and bond-length alternation phenomena.[1,4]

Quantum yield modulation for formation of the lowest-energy, excited triplet state by insertion of different heteroatoms in the terminal aromatic and barbiturate subunits of merocyanine dyes were reported.[20] This strategy not only affects the rate of intersystem crossing from singlet to triplet levels but also modifies the lipophilicity of the dye and, thereby, the rate of uptake by intact biological cells.

Therefore, we intended to prepare 1,3-Dimethyl-5-{(thien-2-yl)-[4-(1-piperidyl)phenyl] methylidene}-(1H, 3H)-pyrimidine-2,4,6-trione 3 as a particular model compound of merocyanine dyes, in order to establish the strong positive solvatochromic effect for this special type of merocyanines containing specific electron-donor thienyl and piperidyl substituents (Scheme 1).

The objective is to investigate the solvent influence on the UV/Vis spectral changes of 3 to check its suitability as a probe for optical chemical sensor, laser dye and optoelectronic applications.

The well-established Kamlet–Taft LSE relationship,[21–30] the McRae equation[31,32] and the empirical solvent polarity index, E_T (30)[1] have been used to study its solvatochromism.

RESULTS AND DISCUSSION

The pronounced positive solvatochromism of the longest-wavelength UV/Vis absorption bands of 3 is noteworthy.

Compound 3 shows four bands in the wavelength regions $\lambda = 288–301$, $344–384$, $406–505$, and $500–587$ nm called λ_{max} I, λ_{max} II, λ_{max} III, and λ_{max} IV, respectively (Fig. 1 and Table 1).

The longest-wavelength Vis band appearing in the wavelength range of 500–587 nm (λ_{max} IV) is the most sensitive band towards the nature of the organic solvent. The corresponding ν_{max} values were used in the correlation analysis. Solvents are used with wide-ranging properties for which α, β, and π^* are known.[24,27]

On going from cyclohexane to formamide as solvent, a bathochromic shift of $\Delta\lambda = 87$ nm ($\Delta\nu = 2960$ cm^{-1}) is observed (Fig. 1 and Table 1). The large positive solvatochromism of the long-wavelength Vis band of 3 is in agreement with a distinct intramolecular CT character of its $S_0 \rightarrow S_1$ electronic transition, going from a less dipolar ground to a highly dipolar excited state ($\mu_E > \mu_G$).

The solvatochromic behavior of 3 is caused by the intramolecular CT from the electron-donor piperidino and thienyl groups to the electron-accepting carbonyl groups. The strong electron-pushing piperidino group can raise the HOMO energy level of 3. Therefore, the
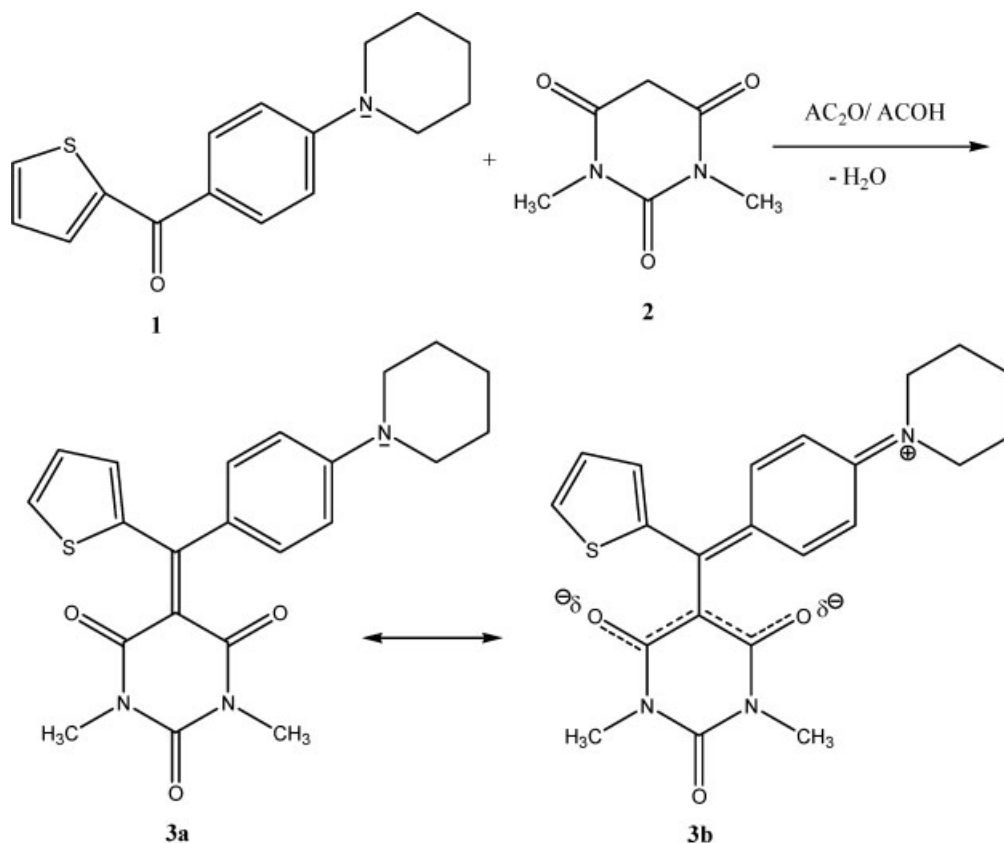
Scheme 1. Synthesis of **3**
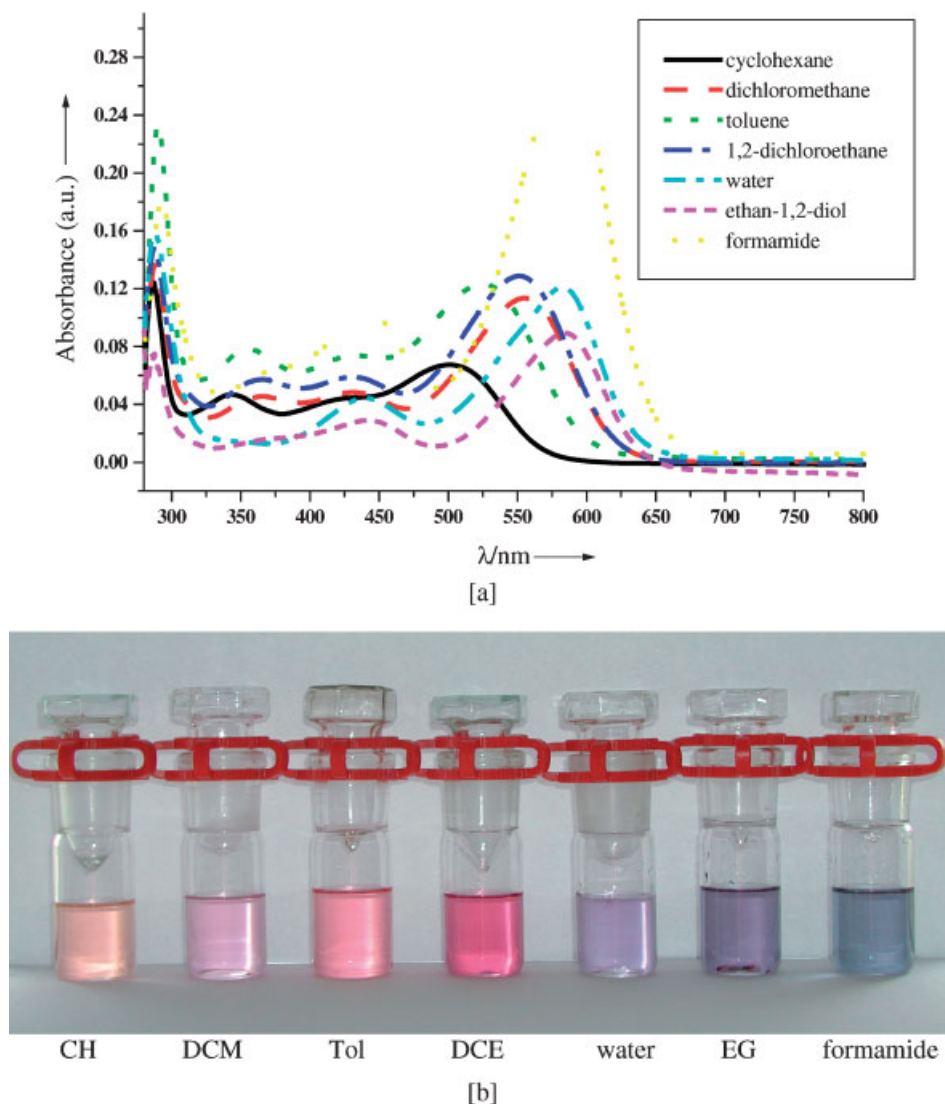
Figure 1. [a] UV/Vis absorption spectra of **3** measured in seven different solvents, i.e. cyclohexane (CH), dichloromethane (DCM), toluene (Tol), 1,2-dichloroethane (DCE), water, ethane-1,2-diol (EG), and formamide at 25°C. [b] Photograph of the color changes of **3** dissolved in the same seven solvents

intramolecular charge transfer band of **3** shifts to the longer wavelength.

To determine the respective contributions of the various solvent properties on v_{max}, a simplified form of the Kamlet–Taft LSER was used. The simplified Kamlet–Taft equation applied to single solvatochromic shifts, $XYZ = 1/\lambda_{max} = v_{max}$ (probe),[1,24–27] is given in Eqn. (1).

$$XYZ = (XYZ)_0 + a\alpha + b\beta + s(\pi^* + d\delta) \quad (2)$$

$(XYZ)_0$ is the property of the solute under study measured in a reference medium, for example, in a non-polar solvent or in the gas phase, α describes the hydrogen-bond donating (HBD) ability or acidity, β the hydrogen-bond accepting (HBA) ability or basicity, and π^* the dipolarity/polarizability of the respective solvent. δ is a polarizability correction term, which is 1.0 for aromatics, 0.5 for polyhalogenated, and zero for aliphatic

solvents; **a**, **b**, **s**, and **d** are solvent-independent correlation coefficients.[1,24]

The calculated LSER shows a high quality in particular as indicated by correlation coefficients larger than r = 0.90 of $\tilde{v}_{max}$ with α, β, and π^*, respectively (See Fig. 2a).

The multiple square analyses of the wave number of the longest-wavelength Vis absorption bands of **3** has been made from the data of Table 1 and are given by Equations (3) and (4); v_{max} is expressed in cm^{-1}; r: correlation coefficient, SD: standard deviation, n: number of solvents, F: significance.

$$v_{max} * 10^{-3}[3] = 20.031 - 0.827\alpha - 0.531\beta - 1.935\pi^*$$
$$r = 0.933 \ n = 25 \ SD = 0.293 \ F < 0.0001$$
$$(3)$$

Table 1. UV/Vis absorption maxima, $\tilde{v}_{max}$, of **3** measured in 26 solvents[1] of different polarity and hydrogen-bond ability

Solvent	$\tilde{v}_{max}$ I (10^{-3} cm^{-1})	$\tilde{v}_{max}$ II (10^{-3} cm^{-1})	$\tilde{v}_{max}$ III (10^{-3} cm^{-1})	$\tilde{v}_{max}$ IV (10^{-3} cm^{-1})	α	β	π^*
Cyclohexane	34.72	29.04	—	20.00	0.00	0.00	0.00
p-Xylene	—	28.49	22.27	19.34	0.00	0.12	0.43
Ethyl acetate	—	28.49	—	19.12	0.00	0.45	0.55
Toluene	34.36	28.09	22.99	19.05	0.00	0.11	0.54
Benzene	—	28.17	—	18.94	0.00	0.10	0.59
Tetrahydrofuran	—	28.33	—	18.80	0.00	0.55	0.58
Methoxybenzene	—	27.78	—	18.55	0.00	0.32	0.73
Acetonitrile	—	26.81	—	18.32	0.19	0.40	0.75
Tetramethyl urea	—	26.25	20.12	18.28	0.00	0.80	0.83
1,2-Dichloroethane	34.60	27.32	23.20	18.12	0.00	0.10	0.81
N,N-Dimethylformamide	34.36	27.40	20.00	18.12	0.00	0.69	0.88
Pyridine	33.22	27.47	19.80	18.12	0.00	0.64	0.87
4-Butyrolactone[2]	—	27.32	20.08	18.12	0.00	0.49	0.87
Nitromethane	34.48	26.18	—	18.08	0.22	0.06	0.85
Dichloromethane	34.60	27.32	23.20	18.02	0.13	0.10	0.82
Dimethylsulfoxide	34.13	27.10	19.84	17.76	0.00	0.76	1.00
1,1,2,2-Tetrachloroethane	—	27.17	—	17.70	0.00	0.00	0.95
1,1,1,3,3,3-Hexafluoropropan-2-ol	34.60	30.40	24.63	17.64	1.96	0.00	0.65
Propan-2-ol	—	27.40	—	17.57	0.76	0.84	0.48
Ethanol	—	27.17	—	17.42	0.86	0.75	0.54
Acetic acid	—	26.95	—	17.42	1.12	0.45	0.64
Mercaptoacetic acid	33.44	30.40	—	17.42	—	—	—
2,2,2-Trifluoroethanol	34.13	26.04	—	17.27	1.51	0.00	0.73
Water	34.60	—	22.68	17.15	1.17	0.47	1.09
Ethane-1,2-diol	34.72	—	22.62	17.09	0.90	0.52	0.92
Formamide	34.13	—	22.57	17.04	0.71	0.48	0.97

[1] α, β and π^* for all solvents were taken from reference 27.
[2] Solvatochromic parameters for this solvent were taken from reference 24.

$$v_{max} * 10^{-3}[3] = 19.949 - 0.804\alpha - 2.104\pi*$$
$$r = 0.913 \quad n = 25 \quad SD = 0.325 \quad (4)$$
$$F < 0.0001$$

It is clear from these Equations that the changes of v_{max} induced by HBD and non-HBD solvents show an excellent linear dependence on the π^* and α parameters.

The influence of the solvents dipolarity/polarizability overcomes the solute-solvent hydrogen- bond interactions. However, the contribution of the α term to the Vis band shift $\tilde{v}_{max}$ [3] is not so strong, because coefficient **a** is significantly smaller than coefficient **s**. The β term can be neglected as shown by Eqn. (4) compared to Eqn. (3).

$$v_{max} * 10^{-3}[3] = 20.260 + 0.168\beta - 2.542\pi^*$$
$$r = 0.980 \quad n = 14 \quad SD = 0.143 \quad F < 0.0001$$
$$(5)$$

$$v_{max} * 10^{-3}[3] = 20.253 - 2.443\pi^*$$
$$r = 0.978 \quad n = 14 \quad SD = 0.144 \quad F < 0.0001$$
$$(6)$$

In non-HBD solvents ($\alpha = 0$), the correlation coefficient r is improved according to Eqn. (5). Neglecting again the less important β-term leads to Eqn. (6) and a good linear correlation is even obtained with the single parameter π^* only.

Also, the McRae equation was used to express the solvatochromic shift of **3** in this work:[31,32]

$$v - v_0 = c(\tilde{v} - \tilde{v}_0) = A\left[\frac{n^2 - 1}{2n^2 + 1}\right] + B\left[\frac{\varepsilon_r - 1}{\varepsilon_r + 2} - \frac{n^2 - 1}{n^2 + 2}\right]$$
$$(7)$$

where v and v_0 is the frequency of the transition of the dye in a given solvent and in a vacuum, respectively, ε_r is the relative permittivity of the solvent, n is the refractive index of the solvent, and c is the velocity of light.

The constant v_0 is obtained, together with A and B, by linear regression of the v_{max} of **3** measured in 15 non-HBD solvents. The calculated values of v_0, A, and B were 23289, -17865 and -2471 cm^{-1}, respectively, with a correlation coefficient of 0.933 and standard deviation 240 cm^{-1} for the regression analysis. Figure 2b compares the experimental wavenumber values for these solvents with those calculated with the regression values of v_0, A and B.

(a)

(b)
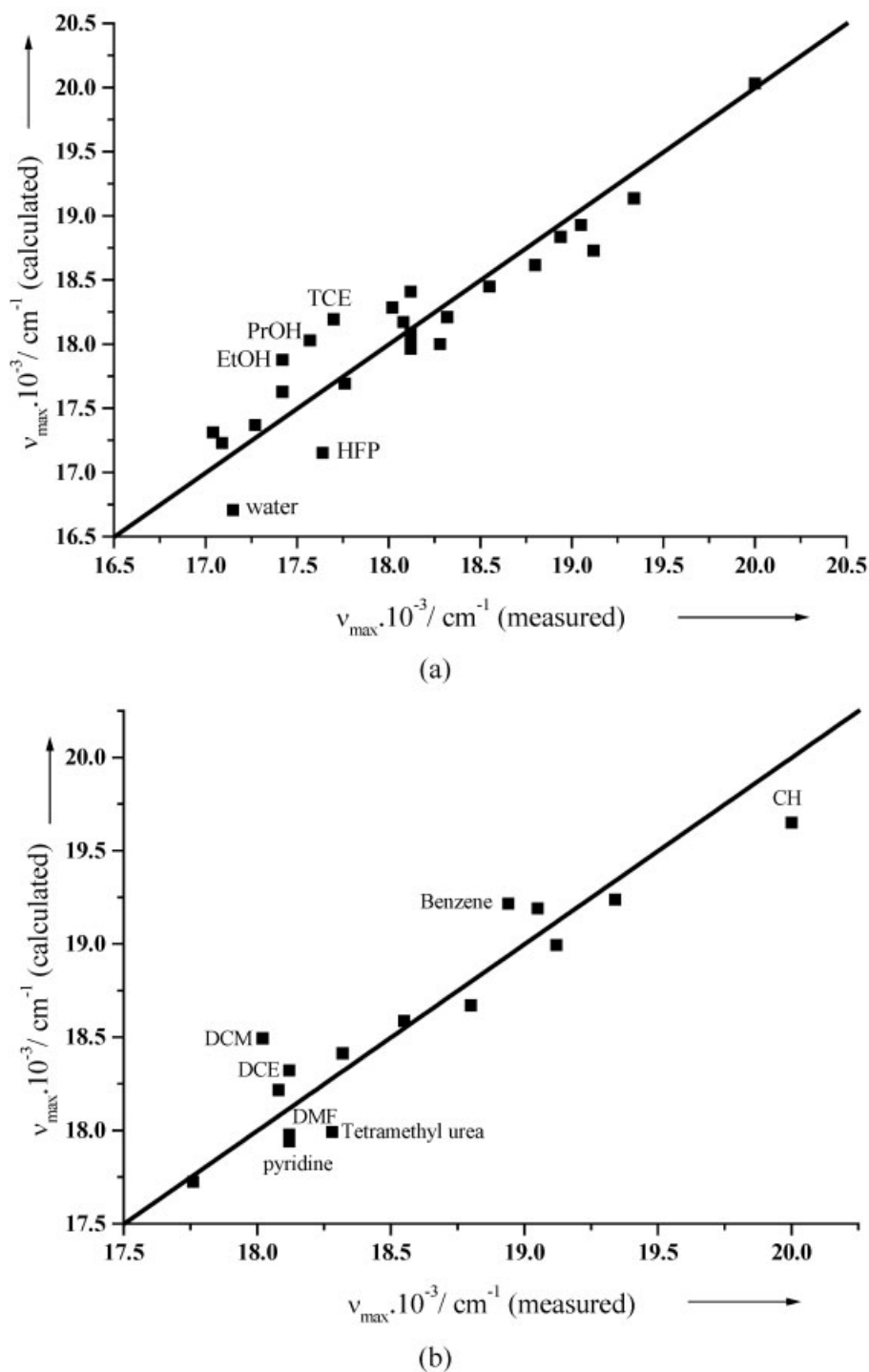
Figure 2. Relationship between calculated and measured v_{max} values for **3** dissolved in: [a] 25 solvents of different polarity and hydrogen bond ability, according to Eqn (3) and [b] 15 non-HBD solvents, obtained from data fitted to Eqn (7)

$E_T (30)$ scale develop by Reichardt[1] was also used to estimate the solvent-solute interactions on electronic transitions for **3**.

$$v_{max} * 10^{-3}[3] = 21.343 - 0.072 \, E_T(30)$$
$$\text{r} = 0.867 \quad \text{n} = 25 \qquad (8)$$
$$\text{SD} = 0.389 \, \text{F} < 0.0001$$

As expected, Fig. 3 and Eqn. 8 show a non-perfect linear correlation in a plot of the CT transition energies of **3** *vs.* $E_T(30)$ values, when the full range of employed solvents is considered. This is because the solvatochromic shifts of Reichardt's betaine, which is the molecular probe used for $E_T(30)$ scale results from a blend of dipolarity/polarizability and acidity contributions, whose proportions are, in a way, unique for each compound.
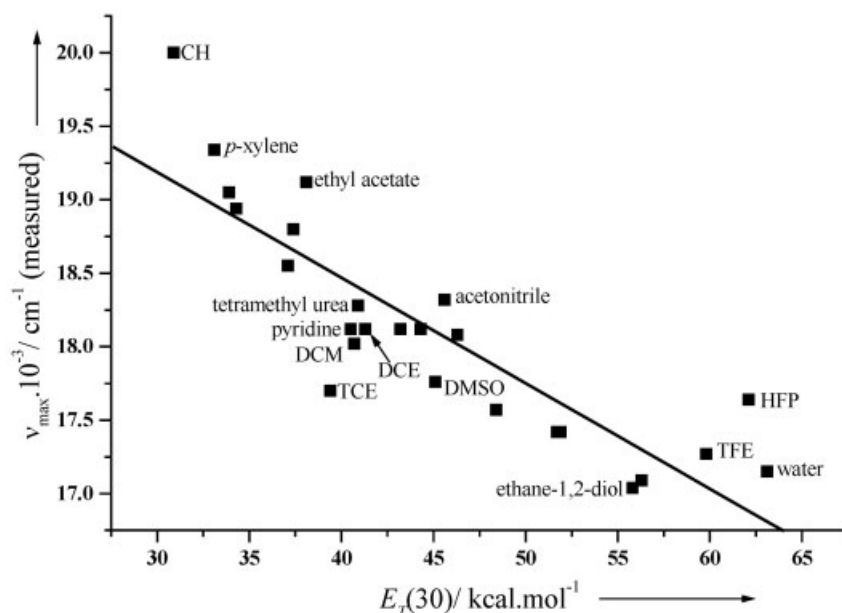
Figure 3. Solvatochromic shifts in wavenumber values v_{max} of **3** dissolved in 25 solvents of different polarity and hydrogen bond ability as a function of E_T (30) values of the same solvents

The negative slope observed for **3** indicate that it follows the opposite pattern as the Reichardt's betaine, which is the molecular probe used for $E_T(30)$ scale.

In non-HBD solvents ($\alpha = 0$), the correlation coefficient r is slightly improved according to Eqn. (9).

$$v_{max} * 10^{-3}[3] = 23.718 - 0.134\ E_T(30)$$
$$r = 0.881 \quad n = 14 \quad SD = 0.325 \quad (9)$$
$$F < 0.0001$$

CONCLUSION

The synthesis and solvatochromism of **3** were reported. This compound reflects solvent influence by manifold shifts of its absorption band in the UV/Vis spectrum. The LSE analyses show that dipolarity/polarizability interactions preferably contribute to the solvent induced color change. The internal dipolarity of **3** arises because of the presence of zwitterionic resonance forms generated upon electron donation from the piperidinophenyl group to the keto groups of the barbiturate acceptor. The introduction of thienyl and piperidyl functionalities at the periphery of pyrimidinetrione system of barbituric acid are expected to change solid-state structures in relation to UV/Vis absorption properties, which makes this kind of compounds promising for investigating chromophores in terms of environmental effects relating to optical properties for application.

EXPERIMENTAL SECTION

Materials

Solvents from Merck, Fluka, Lancaster, Uvasol, and Aldrich were redistilled over appropriate drying agents prior to use.[1a,33] The N,N′-dimethyl barbituric acid, purity >99%, was used as received.

Spectral measurement

The UV/Vis absorption spectra were recorded at 25 °C by means of the UV/Vis MCS 400 diode-array spectrophotometer from Carl Zeiss, Jena, connected with an immersion cell (TSM 5) via glass-fiber optics. NMR measurements were recorded at 20°C on a VARIAN GEMINI 300 FT NMR spectrometer, operating at 300 MHz for ¹H and 75 MHz for ¹³C. The signals of the solvent (CDCl₃) were used as internal standards. C, H, N quantitative analysis was performed with a Vario–EL from the company Elementaranalysen GmbH, Hanau. Electrospray ionization mass spectra were obtained with a Finnigan MATSSQ 710 spectrometer.

Correlation analysis

Multiple regression analysis was performed with the Origin 5.0 statistic programs.

1,3-DIMETHYL-5-{(THIEN-2-YL)-[4-(1-PIPERIDYL)PHENYL]METHYLIDENE}-(1*H*,3*H*)- PYRIMIDINE-2,4,6-TRIONE 3

The synthesis of 2-[4-(piperidyl)benzoyl]thiophene (**1**) has been reported elsewhere.[6,34]

To a solution of **1** (2.72 g, 10 mmol) and *N*,*N*′-dimethyl barbituric acid **2** (1.56 g, 10 mmol) in acetic acid (10 mL) was added acetic anhydride (5 mL). The mixture was refluxed overnight followed by removal of the solvent in a rotary evaporator. The residue was dissolved in dichloromethane (100 mL) and washed three times with 100 mL of 10% aqueous Na_2CO_3 solution and finally with water. The organic phase was dried with anhydrous Na_2SO_4 and then evaporated. The crude product was purified by chromatography with silica gel column and 1,2-dichloroethane as eluent, to give 2.20 g (54%) of **3** as red plates with m.p. 225 °C.

$C_{22}H_{23}N_3O_3S$ (409.50) (calcd. C 64.53%; H 5.66%; N 10.26%; S 7.83% found: C 64.16%; H 5.52%; N 10.18%; S 7.96%); MS (ESI), $m/z = 410.0$ [$M^+ + 1$]; ^{1}H-NMR (CDCl$_3$): $\delta = 7.87$ (d, J = 8.79 Hz, 2H, ArH-2,6), 7.61 (dd, J = 4.94, 6.04 Hz, 2H, ThH-3′,4′), 7.11 (t, J = 4.94 Hz, 1H, ThH-5′), 6.65 (d, J = 8.79 Hz, 2H, ArH-3,5), 3.39 (t, J = 7.69 Hz, 4H, NCH$_2$), 3.01 (s, 6H, NCH$_3$), 1.62 (dd, J = 7.69, 14.28 Hz, 4H, NCH$_2$*CH$_2$*); 1.37 (m, 2H, NCH$_2$CH$_2$*CH$_2$*); ^{13}C-NMR (CDCl$_3$): $\delta = 169.8$ (2 C=O), 161.5 (1 C=O), 155.5 (ArC-4), 150.4 (benzal C), 144.1 (ThC-2′), 137.9 (ArC-2,6), 135.0 (ThC-3′), 131.7 (ThC-5′), 127.5 (ArC-1), 128.4 (ThC-4′), 112.3 (ArC-3,5), 96.2 (barbituric C-5), 48.4 (NCH$_2$), 28.9 (NCH$_3$), 25.0 (NCH$_2$*CH$_2$*), 24.6 (NCH$_2$CH$_2$*CH$_2$*).

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