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SPECTRAL-LUMINESCENCE PROPERTIES AND ACID-BASE PROPERTIES OF LUMINOPHORES OBTAINED FROM 3-IODO-7-DIALKYLAMINOCOUMARINS

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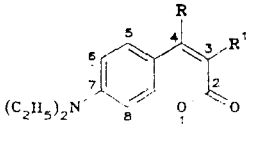
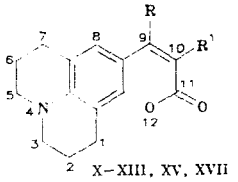
The absorption and fluorescence spectra of the conjugate acids of 7-dialkylaminocoumarins were studied, and their pK_a^I, pK_a^{II}, and pK_a^* values were determined. It was established with the aid of PMR data that the primary protonation generally involves the nitrogen-containing substituent in the 3 or 4 position, while the secondary protonation involves the nitrogen atom in the 7 position.

The spectral-luminescence and acid-base characteristics of a series of 7-aminocoumarins with a mono- or dialkylamino group in the 4 position were described in [1]. We have recently [2] reported the synthesis of new 7-aminocoumarin dyes I-XIII as a result of reactions of 3-iodo-7-dialkylaminocoumarins with secondary amines. In the present publication we discuss the absorption and fluorescence spectra and the pK_a values of the conjugate acids of I-XIII to ascertain the effect on these properties of a heteroaromatic substituent in the 4 position (in the 9 position for julolidine derivatives X-XIII, XV, and XVII) (coumarins I, II, X), of a dialkylamino group in the 3(10) position (coumarins VIII, IX, and XIII), and of the same substituents but bonded to the $C_{(4)}$ [$C_{(9)}$] atom by a methylene link (III-VII, XI, XII). As models for comparison we used the known coumarins XIV-XVIII [3, 4].



I–IX, XIV, XVI, XVIII



X–XIII, XV, XVII

I, X R = **N-imidazolyl** II R = **N-benzimidazolyl** III R = $CH_2N(C_2H_5)_2$; IV R = $=CH_2N(CH_2)_5$; V, XI R = $CH_2N(CH_2CH_2)_2O$; VI, XII R = CH_2-**N-imidazolyl**; VII R = CH_2-**N-benzimidazolyl**; VIII, IX, XIII, XVI–XVIII R = CH_3; XIV, XV R = H; I–VII, X–XII, XIV–XVII R^1 = H; VIII $R^1 = N(CH_2)_5$; IX, XIII $R^1 = N(CH_2CH_2)_2O$; XVIII $R^1 = NH_2$

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TABLE 1. Spectral-Luminescence Properties of Coumarins I-XIII

Compound	Solvent	λ_{max}^{ab}, nm (log ε)	Fluorescence	
			λ_{max}^{fl}, nm	φ_f*
I	C_2H_5OH	256 (4,34); 305 (3,41); 319 (3,41); 394 (4,43)	480	<0,10
	CH_3CN	254 (4,39); 304 (3,47); 319 (3,51); 387 (4,51)	460	<0,10
II	C_2H_5OH	256 (4,48); 282 (4,05); 300 (3,94); 320 (3,70); 399 (4,54)	480	<0,10
	CH_3CN	255 (4,37); 264 (3,95); 300 (3,87); 320 (3,67); 392 (4,44)	466	<0,10
III	C_2H_5OH	251 (4,28); 306 (3,64); 316 (3,67); 379 (4,45)	468	<0,10
	CH_3CN	251 (4,25); 256 (4,23); 310 (3,51); 325 (3,56); 371 (4,44)	451	<0,10
IV	C_2H_5OH	254 (4,20); 284 (3,49); 308 (3,60); 318 (3,63); 382 (4,38)	465	<0,10
	CH_3CN	254 (4,07); 284 (3,25); 308 (3,49); 318 (3,62); 375 (4,40)	450	<0,10
V	C_2H_5OH	256 (4,18); 308 (3,56); 318 (3,60); 384 (4,40)	470	0,35
	CH_3CN	252 (4,21); 308 (3,66); 320 (3,75); 376 (4,47)	456	0,38
VI	C_2H_5OH	253 (4,20); 306 (3,52); 317 (3,56); 378 (4,38)	480	0,28
	CH_3CN	250 (4,06); 306 (3,54); 319 (3,64); 380 (4,41)	478	0,38
VII	C_2H_5OH	255 (4,34); 276 (3,88); 284 (3,84); 308 (3,51); 319 (350); 385 (4,35)	480	0,22
	CH_3CN	254 (4,33); 276 (3,90); 285 (3,88); 308 (3,66); 318 (3,73); 379 (4,38)	466	0,38
VIII	C_2H_5OH	252 (4,18); 310 (3,74); 318 (3,77); 378 (4,38)	490	<0,10
	CH_3CN	252 (4,12); 310 (3,75); 320 (3,82); 374 (4,39)	476	<0,10
IX	C_2H_5OH	256 (4,10); 280 (3,31); 307 (3,51); 317 (3,58); 377 (4,37)	480	<0,10
	CH_3CN	253 (4,18); 305 (3,68); 316 (3,79); 370 (4,49)	470	<0,10
X	C_2H_5OH	266 (4,24); 294 (3,85); 324 (3,59); 415 (4,41)	495	0,63
	CH_3CN	264 (4,17); 292 (3,69); 326 (3,37); 407 (4,42)	480	0,57
XI	C_2H_5OH	252 (4,06); 290 (3,65); 400 (4,35)	490	1,00
	CH_3CN	250 (4,05); 290 (3,62); 393 (4,34)	480	0,55
XII	C_2H_5OH	254 (4,03); 290 (3,75); 316 (3,63); 400 (4,27)	500	0,83
	CH_3CN	252 (4,05); 290 (3,70); 394 (4,34)	480	0,65
XIII	C_2H_5OH	252 (4,05); 270 (3,93); 290 (3,64); 398 (4,32)	490	0,95
	CH_3CN	250 (4,06); 270 (3,90); 290 (3,66); 390 (4,34)	480	0,74

*Note that φ_f is the relative quantum yield.

TABLE 2. Acid-Base Properties of Coumarins I-XIII and XVIII in Aqueous Ethanol (1:1) Solutions

Compound	Neutral molecule		Monocation				Dication		
	λ_{max}^{ab}	λ_{max}^{fl}	pK_a^I	$pK^*{}_a^I$	λ_{max}^{ab}	λ_{max}^{fl}	pK_a^{II}	$pK^*{}_a^{II}$	λ_{max}^{ab}
I	403	489	3,76	5,23	415	512	0,60	−15,45	316
II	407	491	2,16	3,02	413	506	0,36	−16,33	312
III	389	478	6,63	9,20	408	505	1,09	−14,75	313
IV	391	479	6,63	9,20	410	508	0,96	−15,09	313
V	391	481	3,99	6,77	411	507	1,04	−12,23	314
VI	392	490	5,60	6,24	397	500	1,18	−13,93	311
VII	392	487	3,20	3,63	395	496	1,20	−15,71	310
VIII	385	506	2,94	5,94	407	514	0,37	−14,82	315
IX	385	496	2,47	−3,31	348	—	−0,70	−6,91	316
X	422	508	3,57	5,07	435	530	−0,78	−15,33	336
XI	407	500	4,18	7,39	433	524	−0,45	−15,22	333
XII	409	511	5,44	6,40	415	520	−0,12	−14,46	325
XIII	397	505	0,98	3,33	415	525	−0,25	−13,09	332
XVIII	383	496	3,81	−5,18	330	—	0,12	−3,95	311

It is known that the excited singlet state of 7-aminocoumarins is a state of charge transfer (an ICT state [5]) from the nitrogen atom to the lactone carbonyl group. The fact of a nonfluorescing TICT state [6] with a rotated dialkylamino group is also considered to be established for some 7-aminocoumarins. It is known that the introduction of electron-acceptor substituents into the pyrone ring of 7-aminocoumarin molecules increases intramolecular charge transfer [7], is accompanied by a bathochromic shift of the long-wave absorption band and a bathofluoric shift of the emission band in the electronic spectra, and usually facilitates conversion of the ICT state to the TICT state [6].

TABLE 3. PMR Spectra of Coumarins VIII and IX and Their Monoprotonated Forms VIIIa and IXa in CD_3OD

Compound	Chemical shift, δ, ppm (SSCC, Hz)								
	5-H, d (J=9,0)	6-H, dd	8-H, d	3-NCH_2	3-NCH_2CH_2	4-CH_3, s	7-NCH_2, q (J=7,0)	7-NCH_2CH_3 (J=7,0)	Dication
VIII	7,48	6,70 (J=9,0; J=2,6)	6,44	2,93 m	1,62 m	2,42	3,43	1,19 t	1,62 (2H, m, CH_2)
VIIIa	7,80	7,10 (J=9,0; J=2,6)	6,92	3,62 m	1,95 m	2,65	3,60	1,20 t	1,72 (2H, m, CH_2)
IX	7,44	6,65 (J=9,0; J=2,5)	6,40	2,95 m	3,71 t (J=4,8)	2,42	3,40	1,13 t	—
IXa	7,87	7,38 (J=9,0; J=2,5)	7,42	3,05 t (J=4,8)	3,75 t (J=4,8)	2,50	3,62	1,09 q	—

In the electronic spectra of solutions of hetaryl-substituted derivatives I, II, and X in ethanol or acetonitrile (Table 1) the long-wave absorption maximum and the fluorescence maximum are shifted to the long-wave region by 30-40 nm as compared with the 4,7-bis(dialkylamino) derivatives [1], which can be ascribed to lengthening of the overall π,π-conjugation chain, as, for example, in the case of 3- or 4-alkylcoumarins [8, 9]. However, considering the fact that 4(9)-unsubstituted aminocoumarins — 7-diethylaminocoumarin (XIV) and 6H-coumarin (XV) — absorb and emit at shorter (by 15-20 nm) wavelengths [10] than I, II, and X, it may be asserted that the effect of a hetaryl substituent on the electron-density distribution in the S_0 and S_1 states of 7-aminocoumarins is small. The reason for this may be steric hindrance [1], leading to a nonplanar orientation of the heterocyclic fragments. In addition, the presence of an N-imidazolyl or N-benzimidazolyl residue in the 4(9) position leads to appreciable quenching of the fluorescence as compared with the unsubstituted analogs (XIV and XV), which is most appreciable for the 7-diethylamino derivatives (see Table 1).

The separation of the heteroaromatic fragments by a methylene link in coumarins VI, VII, and XII is accompanied by a hypsochromic shift in the absorption maximum as compared with I, II, and X ($\Delta \approx$ 10-15 nm) and has virtually no effect on the location of the fluorescence maximum. The latter circumstance indicates, in our opinion, the small role played by electron interactions of the heterocyclic fragments "through the bond," in contrast to interactions "through space" due to the high polarizability of the aromatic systems. As a result, the stabilizing influence of the substituent in the polar CT states of the I and VI, II and VII, or X and XII molecules is similar in magnitude. As compared with 4-methyl-7-diethylaminocoumarin (XVI) and coumarin-102 (XVII), the absorption maximum of VI, VII, and XII experiences a small bathochromic ($\Delta\lambda \approx$ 5-10 nm) shift, while the emission maximum experiences a substantial bathofluoric shift ($\Delta\lambda \approx$ 20 nm), which confirms the higher sensitivity of the fluorescence spectra to the effects of substituents [11]. The transition from I, II, and X to coumarins VI, VII, and XII is also accompanied by intensification of the luminescence properties. The quenching of the fluorescence in the case of coumarins VI and VII as compared with coumarin XVI is possibly a consequence of an increase in the energy transfer in the excited state, in which the imidazole or benzimidazole residue acts as an acceptor.

The absorption and fluorescence spectra of III-V and XI are similar to the spectra of the corresponding methyl derivatives XVI and XVII [10]. As in the case of 4,7-bis(dialkylamino)coumarins [1], the fluorescence quantum yield of morpholino derivative V is higher than for the diethylamino or piperidino derivatives III and IV. This peculiarity is evidently associated with the great "rigidity" of the morpholine ring, which may lead to a decrease in the vibrational losses of energy upon excitation of the molecule.

Intensification of the fluorescence in julolidine derivatives X-XIII as compared with the 7-diethylamino analogs is also a general principle.

It was quite unexpected that the presence of a dialkylamino group in the 3(10) position in the case of coumarins VIII, IX, and XIII is virtually unaccompanied by a shift in the long-wave absorption maximum as compared with unsubstituted coumarins XVI and XVII. This principle could be associated with steric hindrance that interferes with conjugation of the 3-dialkylamino group with the pyrone ring. An alternative explanation, which includes a "balanced" interaction of the 3- and 7-amino groups, was previously examined [2] in a discussion of the ^1H and ^{13}C NMR spectra.

To shed some light on the problem we studied the spectral properties of the known coumarin XVIII [4], for which the steric interactions of the substituents in the 3 and 4 positions were expected to be substantially smaller than for VIII and IX. We observed that the locations of the absorption maxima for coumarins VIII, IX, and XVIII virtually coincide (see Table 1), and, thus, the reasons for the peculiar behavior of these compounds are electronic [2] rather than steric effects.

It is interesting that coumarins VIII, IX, XIII, and XVIII fluoresce in a significantly longer-wave region of the spectrum than model compounds XVI and XVII ($\Delta \approx$ 20-30 nm). This fact does not, in principle, contradict the hypothesis of the possibility of rotation of the 3-amino group about the $C_{(3)}$—N bond, as a consequence of which the latter, in the ICT state, acts mainly as a σ acceptor of electron density. It is characteristic that coumarins VIII and IX, in contrast to coumarin XIII, have weak fluorescence. In our opinion, is the more facile formation of the TICT state in coumarins VIII and IX, in which the $C_{(7)}$—N bond is weakened under the influence of the 3-amino group. Additional confirmation of this was provided by our observed (from NMR spectral data) rotation of the 7-diethylamino group in solutions of these compounds [2].

We have previously investigated the basicities of a series of 4,7-diethylaminocoumarins with mono- or dialkylamino groups in the 4 position and have shown that primary protonation takes place at the 7-N atom and is accompanied by a hypsochromic shift ($\approx$ 100 nm) of the long-wave absorption maximum and quenching of the fluorescence. In this research we also measured the pK_a values of the conjugate acids of I-XIII in aqueous ethanol (1:1) solutions (Table 2). We found that for coumarins I-VII and X-XII primary protonation involves the nitrogen-containing substituent in the pyrone ring. Proof for this is provided by the following: the substantially greater basicities (usually 1.5-4.5 orders of magnitude) of the investigated compounds as compared with other 7-aminocoumarins [1, 12], the long-wave absorptions of the monocations as compared with the neutral forms, as well as the retention of the weak fluorescence of the cations in the longer-wave region (see Table 2).

Coumarins VIII, IX, and XIII constitute a special group. The character of the observed (upon acidification) changes for the enumerated compounds differs: the monocations of coumarins VIII and XIII fluoresce weakly and absorb in a longer-wavelength region, while the monocation of coumarin IX does not fluoresce and absorbs in a shorter-wavelength region as compared with the neutral forms. A hypsochromic shift of the absorption maximum and losses in the luminescence properties were also observed in the case of protonation of model coumarin XVIII (see Table 2). Taking into account the data on the basicities of known 4,7-diaminocoumarins [1], one may assert that the protonation of coumarins IX an XVIII takes place at the $N_{(7)}$ atom, whereas protonation proceeds at the $N_{(3)}$ atom for coumarins VIII and XIII.

An independent confirmation of this concept follows from the PMR spectra of the monoprotonated forms (VIIIa and IXa) of coumarins VIII and IX, obtained in solutions in CD_3OD with acidification by CF_3COOH (Table 3). Upon protonation of coumarin VIII, the signals of the α- and β-methylene protons of the piperidine fragment experience a strong shift to weak field, at the same time that, or coumarin IX, analogous signals of the morpholino substituent undergo an insignificant change. On the other hand, in the case of monoprotonated form IXa, as compared with cation VIIIa, one observes more pronounced changes in the chemical shifts of the 6-H and 8-H protons, which attests to protonation of the nitrogen atom in the 7 position. It is noteworthy that monoprotonation in both cases is accompanied by contraction of the "diffuse" signals of the $N—CH_2$ protons of the substituent in the 3 position as a consequence of a decrease in the conformational lability of the heterocyclic substituent as a result of steric reasons (in the case of cation VIIIa) or shortening of the $C_{(3)}—N$ bond (in the case of cation IXa).

The change in the center of highest basicity in VIII, IX, and XIII becomes understandable if one takes into account that the basicities of piperidine and diethylamine are higher by three orders of magnitude than the basicity of morpholine and that the fastening of the nitrogen atom in the julolidine fragment is accompanied by a sharp decrease in basicity [1].

We then investigated the behavior of coumarins I-XIII, as well as coumarin XVIII, under diprotonation conditions in solution in H_2SO_4 (see Table 2). The transition to more acidic media is accompanied by disappearance of the long-wave absorption maximum at 310-340 nm, as well as the loss of fluorescence. These principles constitute evidence that secondary protonation involves the nitrogen atom in the 7 position. The pK_a^{II} values of X-XIII are lower by 1.0-1.5 orders of magnitude than those of the 7-diethylamino derivatives, as was indeed previously observed for other 7-aminocoumarins [1, 12]. Coumarins IX and XVIII also undergo secondary protonation, in which the 3-amino group is protonated.

Using the Fuerster method for the investigated series of 7-aminocoumarins we estimated the acidities of the conjugate acids (pK_a^{*I}) in the excited state (see Table 2). For most of the investigated compounds the basicity increases by one to three orders of magnitude upon excitation; coumarins IX an XVI, for which one observes a decrease in the basicity upon excitation, constitute exceptions, a state of affairs that is in agreement with protonation of the $N_{(7)}$ atom [1, 12]. The small increase in the basicities for hetaryl derivatives VI. VII, and XII upon excitation ($\Delta pK_a \approx 0.4$-0.6) is explained by the greater delocalization of the positive charge in the $N_{(3)}$-protonated heteroring and, as a consequence, the smaller effect of this substituent on the π system of coumarin.

The pK_a^{**II} values calculated by the Fuerster method for the monocations of all of the investigated compounds are also presented in Table 2. A sharp decrease in the basicity upon excitation of the monocations is also characteristic in most cases for the neutral 7-aminocoumarins, for which the amino group in the 7 position is the most basic center.

EXPERIMENTAL

The spectral-luminescence studies were carried out with Specord M 40 and Hitachi EPS-3T spectrophotometers equipped with a G-3 fluorescence adapter. The fluorescence was excited by irradiation at the long-wave absorption band of the corresponding coumarin. The relative fluorescence quantum yields (φ_f) were determined by the method in [13] using 3-aminophthalimide ($\varphi_f = 0.60$) as the standard.

The PMR spectra were obtained with a Bruker WM 250 spectrometer with hexamethyldisiloxane (HMDS) as the internal standard.

The pK_a^I values were determined by the method in [14] by a spectrophotometric method using solutions in 50% ethanol (H_2SO_4 served as the oxonium-ion donor) with an EV-74 universal pH meter with glass and calomel electrodes. The pK_a^{II} values were determined by the same method using solutions in H_2SO_4 with a known acidity function. The error in the determination of the pK_a^I values was $\pm$ 0.04, while the error in the determination of the pK_a^{II} values was $\pm$ 0.10. Twice-distilled water was used to prepare the solutions.

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PHTHALIMIDOAZIRIDINYLATION OF THE SIMPLEST VINYLACETYLENES

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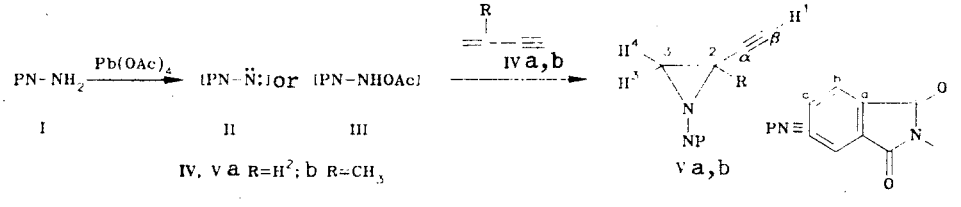
The oxidation of N-aminophthalimide with lead tetraacetate in the presence of butenyne and 2-methylbutenyne leads to the corresponding 1-phthalimido-2-ethynylaziridines in good yields.

In [1] we reported the chemoselective phthalimidoaziridinylation of a number of conjugated phenylbutenynes. The high reactivity of the double bond of these enynes with respect to phthalimidonitrene (II) [or to N-acetoxyaminophthalimide (III)*] may, in principle, be due to both the properties of strictly the enyne fragment and its additional conjugation with the phenyl group, which should lead to an increase in the energy of the highest occupied molecular orbital (HOMO) of the substrate and, consequently, its affinity for electrophilic nitrene II [3] (or N-acetoxy derivative III). Up until now, only two examples of the addition of phthalimidonitrene (II) to enynes that do not contain aromatic substituents have been known: the addition to 2,5,5-trimethyl-1-hexen-3-yne [4] (yield not specified) and to 1-(1-cyclopentenyl)-1-octyne [5], in which the yield of the adduct using a fivefold excess of the substrate was less then 40%. In this connection, we carried out the phthalimidoaziridinylation of the simplest vinylacetylenes, namely butenyne (IVa) and 2-methylbutenyne (IVb):



*The question of the nature of the intermediate in phthalimidoaziridinylation still remains open to debate [1, 2].

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