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Organic Luminophores with Fluorine-Containing Substituents

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SUMMARY

The results of experimental investigations carried out by the authors of this review and their coworkers in the field of fluorine-containing luminescent compounds having a C═N bond either in an open chain or in a heterocyclic ring are discussed. It is shown that the introduction of CF_3, CHF_2SO_2 *or* CF_3SO_2 *groups or of a perfluorophenyl radical significantly affects the spectral and luminescent properties, and in a number of cases the stability of organic luminophores toward UV light is increased.*

INTRODUCTION

The development of the chemistry of organic luminophores and the progress of theoretical investigations in this field have made possible a considerable expansion of their practical applications.[1]

It is well known that the introduction of fluorine-containing substituents into dye molecules frequently improves the lightfastness of the dyes and also, especially importantly, their photostability.[2]

It is feasible therefore to conclude that these substituents would affect the properties of organic luminophores in the same way, despite the fact that in this case the improvement of lightfastness presents a much more complicated problem, since the absorption of UV light, necessary for the activation of luminescence, evokes simultaneously the photodestruction of luminophores.

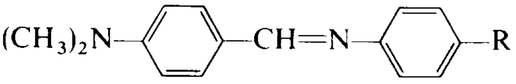
Together with the improvement of photostability, the fluorine-containing substituents, as strong electron acceptors, could markedly affect the spectral and luminescent characteristics of the luminous organic compounds.

To examine these suppositions and with the object of obtaining organic luminophores having the targeted properties fluorine-containing luminous compounds with a C=N bond either in an open chain or in various heterocycles have been synthesized and their properties evaluated.

LUMINOPHORES OF THE AZOMETHINE SERIES

As a rule the flat rigid structure of the molecule essential for the appearance of luminescence[3] is achieved in azomethine derivatives by the introduction of proton-donating substituents into the *ortho*-position of the aldehyde moiety. Such substituents form an intramolecular hydrogen bond with the n-electrons of the nitrogen atom of the C=N bond. These compounds luminesce in the solid state and in frozen solutions, where the most favourable conditions for the existence of a strong hydrogen bond arise.[4]

There is a possibility that intensely luminescing azomethines with a flat rigid structure could be obtained by the synthesis of compounds of formula **I**, in which a dimethylamino group of the 'aldehyde' moiety of benzalaniline is conjugated with a strong electron acceptor substituent R in the 'amine' moiety.



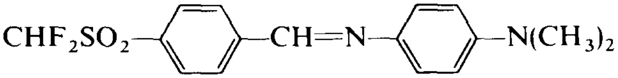
I

It was found, however, that the introduction of a nitro group into such a molecule which was weakly luminous in both the solid state and in frozen solutions resulted in a complete disappearance of the luminescence. Such an effect may be related either to the quenching action of the nitro group or to its effective interaction with n-electrons of the CH=N group. As a result of the latter factor, the coplanarity of the azomethine molecule is disturbed and the conjugation between the 'amine' and the 'aldehyde' parts of the molecule is therefore markedly decreased. Fluorine-containing substituents do not quench luminescence and their introduction into the above instead of the nitro group could therefore give an indication as to which of the above two factors is the main reason for the loss of luminescent properties. For this purpose, azomethines containing CF_3, CHF_2SO_2 and CF_3SO_2 groups have been prepared by condensation of the corresponding *para*-substituted anilines with *p*-dimethylaminobenzaldehyde.[5-7]

Analysis of the spectral, luminescent and polarographic properties of the compounds thus synthesized clearly indicates the substantial contribution of the donor–acceptor interaction between the dimethylamino group and the fluorine-containing substituents to the electronic structure of the molecules both in the ground and excited states. This is especially apparent in the azomethines containing such strong electron acceptors as the CHF_2SO_2 and CF_3SO_2 groups. Their conjugation with the dimethylamino group results in a long wavelength shift of both the absorption and luminescence maxima in comparison with the unsubstituted *p*-dimethylaminobenzaldehyde.

A rather intensive luminescence is observed not only in frozen solutions but also in solutions at ambient temperature. This phenomenon has not been previously observed in other compounds of the azomethine series.

Bathochromic and bathofluoric (i.e. a red shift in fluorescence maxima) effects in the spectra of these compounds could be even more substantial if it were not for the rotation of the *N*-phenyl ring as the result of the interaction of the fluorine-containing substituent with the n-electrons of the nitrogen atom of the C=N group. This is clearly confirmed by the synthesis and investigation of the properties of the isomeric compound (**II**), which contains, in the aldehyde moiety of the azomethine, a CHF_2SO_2 group. This substituent is easily introduced and is a stronger electron acceptor substituent than the nitro group, and thus the above-mentioned interaction is not observed. The absorption maximum of compound **II** in dimethylformamide is shifted bathochromically in comparison with the isomer (**I**, R = CHF_2SO_2) by 45 nm and the luminescence maximum of the frozen solution is shifted by 75 nm; the intensity of absorption is also more than doubled.



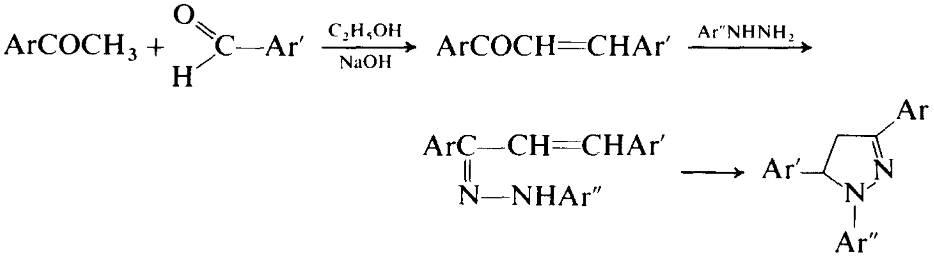
II

The strong donor–acceptor interaction of the substituents is apparent also in the electrochemical reduction of fluorine-containing azomethines. The flatter molecules of compound **II** show a more significant positive shift of the half-wave potential than the isomer with a CHF_2SO_2 group in the amine moiety. The presence of a CF_3 group in compound **I**, whilst not noticeably affecting the spectral and luminescent properties of 4-dimethylaminobenzalaniline, results in behaviour on polarographic reduction, in the case of its conjugation with a dimethylamino group in compound **II**,[8] similar to the CHF_2SO_2 derivative, but notably weaker.

1,3,5-TRIARYL-2-PYRAZOLINES

Fluorine-containing substituents have also been incorporated into azole molecules of various structures and in all cases these substituents markedly affect the spectral and luminescent properties of the compounds. The influence of substituents has been studied in detail with respect to the substituted 1,3,5-triaryl-2-pyrazolines (**III**). These compounds have a predominantly blue luminescence and contain a C=N bond in the heterocycle.

The majority of triarylpyrazolines (TAPs) with fluorine-containing substituents have been prepared by condensation of aromatic aldehydes with methyl aryl ketones in an alkali–alcohol media, followed by reaction of the resultant compounds with arylhydrazines in alkali–alcohol or acetic acid media, and cyclization of the arylhydrazones.[9]

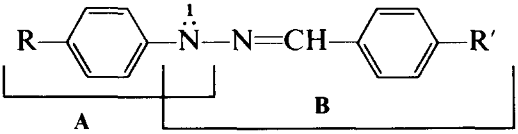


III

Due to low stability of the CHF_2SO_2 group toward alkalis, preparation of the α,β-unsaturated ketones containing this group is carried out in a mixture of acetic and sulphuric acids (10:1).

The structure of the TAP thus synthesized has been confirmed by their IR-spectra, in which an intense absorption band at 1605 cm^{-1}, caused by the CH bond vibrations of the pyrazoline ring, and a number of other bands, which are characteristic for TAP have been observed. This, together with the absence of absorption at 3200–3500 cm^{-1}, characteristic for the NH bond valent vibrations in the hydrazone molecules, confirm the cyclic structure of the compounds synthesized.

It is well known that the spectral and luminescent properties of TAP are determined by the presence of a cross-conjugated chromophoric system in their molecules. The interference of the cross-conjugated fragments **A** and **B** in this system is dependent on the electronic nature of the substituents R and R′ and their effect on the spectral and luminescent properties.



Previous investigations of the spectral and luminescent characteristics of a large number of different 2-pyrazoline derivatives containing electron-acceptor substituents at the *para*-position of the *N*-phenyl ring showed that although the electron transition in these compounds is molecular, it is mainly localized in the fragment **A**. It is connected to intramolecular charge transfer from the nitrogen atom 1 on the substituents R and is particularly apparent with respect to the position of the luminescence maximum.[10]

The results of studies on TAP derivatives with fluorine-containing substituents are summarized in Table 1. A CHF_2SO_2 group in Ar″ (compound **IIIb**), whilst not affecting the position of the long-wave absorption maximum, shifts the luminescence maximum hypsochromically in comparison with the unsubstituted triphenylpyrazoline (**IIIa**). The same substituent at the *para*-position of the 3-phenyl radical (**IIIc**), by increasing the electron density shift toward this radical, exerts bathochromic and bathofluoric shifts of λ_{max} 52 and 55 nm respectively. These effects are enhanced if an electron donor such as the methoxy group is incorporated into the 1-phenyl radical (**IIId**) or if the latter is replaced by biphenyl or 2-naphthyl radicals (**IIIf**, **IIIe**).[11]

In compound **IIIg**, in which the CHF_2SO_2 groups are present both in the 1- and 3-phenyl rings, the interaction between the cross-conjugated fragments is decreased because of the counter-polarizing influence of the electron-acceptor groups. As a result, hypsochromic and hypsofluoric shifts are observed in the spectra of compound **IIIg** in comparison with the compound **IIIc** which does not contain a CHF_2SO_2 group in the 1-phenyl radical.

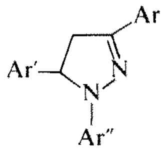
In compounds **IIIh**–**IIIj**, the CF_3 group in the *meta*-position of the 1-phenyl radical is not conjugated with the heterocyclic ring and it can affect the latter only inductively. Nevertheless its effect on the luminescent properties of trifluoromethyl-substituted TAPs is rather strong. Compounds **IIIa**–**IIIj** show blue or yellowish-green luminescence in toluene and in the crystal form, the quantum yields in solutions being in the 0·4–0·5 range.

The perfluorophenyl radical in compounds **IIIk** and **IIIl** exerts bathochromic and bathofluoric shifts, but the quantum yields are lower than those in triphenylpyrazoline.[12,13]

Interesting results have been observed on introduction of a CHF_2SO_2 group into the *para*-position of the 5-phenyl radical (Ar′).[14] Structural

TABLE 1

Spectral and Luminescent Characteristics of TAP with Fluorine-Containing Substituents (in Toluene)



Compound	*Ar*	*Ar′*	*Ar″*	*Absorption*		*Luminescence*	
				λ_{max} (*nm*)	$\varepsilon \times 10^{-4}$	λ_{max} (*nm*)	η
IIIa	C_6H_5	C_6H_5	C_6H_5	360	2·00	440	0·45
IIIb	C_6H_5	C_6H_5	4-$CHF_2SO_2C_6H_4$	365	3·25	400	0·52
				375[a]		410	
IIIc	4-$CHF_2SO_2C_6H_4$	C_6H_5	C_6H_5	412	2·45	495	0·52
IIId	4-$CHF_2SO_2C_6H_4$	C_6H_5	4-$CH_3OC_6H_4$	430	2·36	540	0·27
IIIe	4-$CHF_2SO_2C_6H_4$	C_6H_5	2-$C_{10}H_7$	425	2·76	515	0·45
IIIf	4-$CHF_2SO_2C_6H_4$	C_6H_5	4-$C_6H_5C_6H_4$	424	2·87	520	0·46
IIIg	4-$CHF_2SO_2C_6H_4$	C_6H_5	4-$CHF_2SO_2C_6H_4$	395	2·46	450	0·52
IIIh	C_6H_5	C_6H_5	3-$CF_3C_6H_4$	360	2·08	418	0·46
IIIi	4-$C_6H_5C_6H_4$	C_6H_5	3-$CF_3C_6H_4$	375	3·12	433	0·52
IIIj	4-C_6H_5—CH═CH	C_6H_5	3-$CF_3C_6H_4$	380	4·40	438	0·50
IIIk	C_6F_5	C_6H_5	C_6H_5	370	2·06	462	0·33
IIIl	C_6F_5	C_6H_5	4-$CH_3OC_6H_4$	380	1·81	508	0·12
IIIm	C_6H_5	4-$CHF_2SO_2C_6H_4$	C_6H_5	350	1·88	—	—
IIIn	C_6H_5	4-$CHF_2SO_2C_6H_4$	4-$CHF_2SO_2C_6H_4$	360	3·59	—	—
IIIo	4-$CHF_2SO_2C_6H_4$	4-$CHF_2SO_2C_6H_4$	C_6H_5	408	1·25	485	0·60

[a] Inflexion (shoulder).

changes at the 5-aryl radical do not usually affect the spectral and luminescent properties of TAP.[15] Quite unexpected, therefore, was the complete lack of luminescence both for the compound **IIIm** containing a CHF_2SO_2 group in the 5-phenyl radical and also for compound **IIIn** which additionally contains the same substituent at the *para*-position of the 1-phenyl ring.

This effect is apparently caused by the inductive interaction between the unshared electron pair of the nitrogen atom 1 and the π-electron system of the 5-phenyl radical, substantially reducing the interaction of the aniline fragment with the azomethine grouping, on which is based the luminescent properties of TAPs. In the case of compound **IIIo**, which contains CHF_2SO_2 groups in both the 3- and 5-phenyl radicals, the electron-acceptor substituent at the second radical does not affect the chromophore, and, as in compound **IIIc**, the electron transfers in 1,3-cross-conjugated chromophore systems become decisive for the display of luminescent properties. The position of the absorption and luminescence bands of **IIIo** are not significantly different from those of compound **IIIc**.

The electronic nature of the fluorine-containing substituents is also clearly apparent in the polarographic characteristics. Their presence markedly facilitates the electrochemical reduction of TAP. In compounds containing CF_3 and CHF_2SO_2 groups, not only the pyrazoline ring, but these groups also, are reduced.[16]

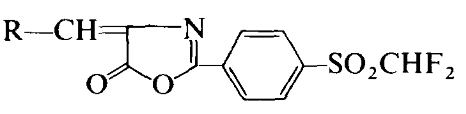
The introduction of the fluorine-containing substituents into the TAP molecule improves the stability toward UV light. The most photostable are luminophores containing a CHF_2SO_2 group in the 1-phenyl radical, viz. where the electron density in the heterocyclic ring, essentially affecting the photostability of TAP, is substantially reduced in comparison with triphenylpyrazoline. A CHF_2SO_2 group in the phenyl rings at the 3- or 5-positions of TAP affects the photostability to a lesser extent.

The intensive luminescence of the majority of fluorine-containing pyrazoline luminophores both in solutions and in polymers, and also their increased stability toward UV irradiation have made possible their application as scintillators[17,18] or daylight fluorescent pigments.[19]

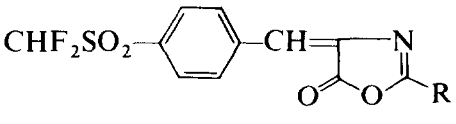
OXAZOL-5-ONE DERIVATIVES

The condensation of aromatic aldehydes with the corresponding hippuric acids leads to the formation of the oxazolone luminophores (**IV**, **V**) containing a CHF_2SO_2 group at the 2-phenyl or arylidene moieties (R is a substituted or unsubstituted aryl radical).[20]

An important feature of the structure of these compounds is the presence



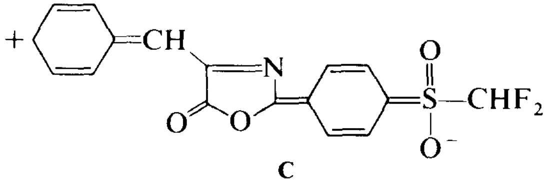
IV



V

of a carbonyl group in the heterocyclic ring. This group is conjugated with the arylidene moiety and the latter is also conjugated with the 2-phenyl radical. The first of these two interactions is increased when an electron donor substituent is introduced into the *para*-positions of the benzylidene moiety, the second by the introduction of an electron acceptor group into the same position.[21]

Both calculations and experimental investigations have shown that the $S_0 \rightarrow S_1^*$, transition in the molecules of compounds **IV** and **V** is connected with the redistribution of the electron density in the arylideneoxazolone moiety, the electron density shift being directed toward the arylidene radical. It might be expected that substitution by a CHF_2SO_2 group in the 2-aryl radical might hinder this effect, thus bringing about a hypsochromic displacement of the absorption band in comparison to compound **V** which contains the same group in the arylidene fragment. The absence of such an effect and in some cases even the red shift of the absorption band has been explained[20] in terms of an increase in the contribution of the quinoid structure (**C**) to the electron mesomeric structure of the substituted oxazolone molecule. The $S_0 \rightarrow S_1^*$ transition is determined in this case by the conjugation between the transformed arylidene and azomethine moieties.



C

Both the above-mentioned intramolecular types of interactions are relevant when investigating compounds incorporating both an electron donor substituent (CH_3O or $(CH_3)_2N$) and a 4'-CHF_2SO_2 group. Electron donor substituents in the arylidene moiety effectively interact with the azomethine grouping, the electron-accepting properties of which are increased by the fluorine-containing substituents in the 2-phenyl radical. Their interaction with the carbonyl group is in this case decreased and a bathochromic displacement of the long-wave absorption band proportional to the electron acceptor force of the substituent is observed. In cases where the electron acceptor substituents are incorporated into both the 2-phenyl and the arylidene moieties ($R = NO_2$), spectral characteristics of the substituted oxazolones **IV** and **V** are very similar.

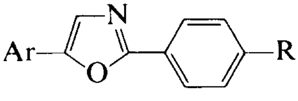
Lengthening of the conjugated chain in the arylidene or 2-aryl fragments leads to a bathochromic shift of the absorption maxima. Comparing compounds having the same length of conjugated chain, the maxima of compounds **V** are however shifted hypsochromically in comparison with compounds **IV**.

In the IR spectra of the compounds **IV** and **V** the electron donor groups diminish, and the electron acceptor groups conversely increase, the value of the absorption frequency of the carbonyl group. The other bands are subject to the effect of the substituents to a lesser extent.

Unlike other known luminophores, the CHF_2SO_2-substituted derivatives of oxazolone luminesce in toluene solution not only in the frozen state but also at ambient temperature. A difluoromethylsulphonyl group increases polarization of the oxazolone molecules in the first excited state and they acquire a more rigid structure. As a result, the possibility of the radiationless energy relaxation of the electron excitation is decreased and the quantum yield of luminescence is correspondingly increased. The intensity of luminescence of compounds containing the CHF_2SO_2 group in the arylidene moiety is higher than that of compounds containing the same group in the 2-aryl radical.

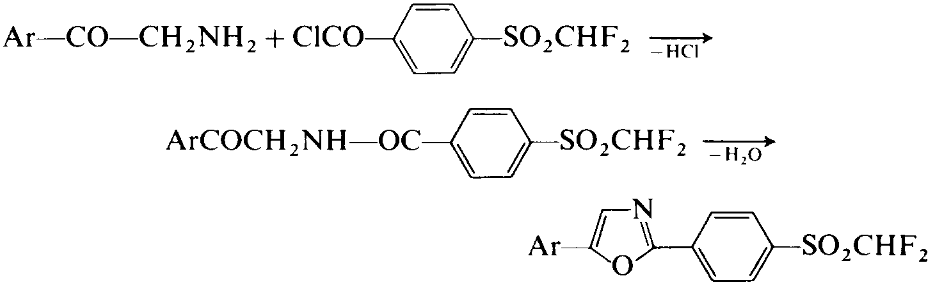
2,5-DIARYLOXAZOLES

An investigation of the effect of the CHF_2SO_2 group on the spectral and luminescent properties of 2,5-diaryloxazoles (**VI**) is also of interest since these compounds are widely used as activators[22,23] of liquid and plastic scintillators.



VI

It has been previously shown that strong electron acceptor substituents introduced into the *para*-position of the 2-phenyl radical exert bathochromic and bathofluoric shifts and also increase the quantum yield of luminescence in hydrocarbon solvents.[24] To understand the influence of fluorine-containing substituents on the spectral and luminescent properties of 2,5-diaryloxazoles a series of CHF_2SO_2-substituted compounds **VI** has been synthesized ($R = CHF_2SO_2$). They have been prepared by the Robinson–Gabriel condensation of 4-difluoromethylsulphonyl benzoyl chloride with ω-aminomethyl aryl ketones, followed by the cyclization of the resultant amides.[25]



Introduction of a CHF_2SO_2 group into 2,5-diphenyloxazole results in a significant long-wave shift of the absorption and luminescence maxima and the intensity of luminescence is also increased. These effects are enhanced by the simultaneous incorporation of donor substituents into the *para*-position of the 5-aryl radical or by lengthening of the conjugated chain in this radical. A substantial Stokes shift of the derivative containing a *p*-dimethylamino group in the 5-aryl residue is also of interest. This compound exhibits a positive solvatofluorochromism and its luminescence maximum is within the 480–655 nm[26] region depending on the polarity of the solvent.

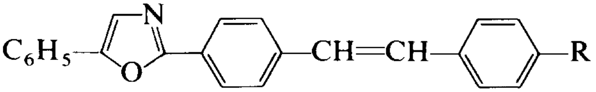
Because of the influence of the CHF_2SO_2 group, apparently as the result of the delocalization of the unshared electrons of the nitrogen atom of the oxazole cycle, the stability of 2,5-diaryloxazoles toward UV light is notably improved.

Under similar conditions the luminescence intensity of the compounds studied ($Ar = Ph$, $4\text{-}CH_3OC_6H_4$, $4\text{-}C_6H_5\text{-}C_6H_4$) under UV irradiation in toluene solution (the PRK-2 UV lamp, distance of sample 12 cm, temperature 36–40°C, irradiation time 20 h) falls up to 61–88% of the original value, whilst in the unsubstituted 2,5-diphenyloxazoles it is decreased up to 25%. This property of 2,5-diaryloxazoles containing the CHF_2SO_2 group can be utilized in liquid lasers with changing frequency of irradiation.[27,28]

2,5-Diaryloxazoles without fluorine-containing substituents show generation in the near-UV and blue regions,[29,30] but the majority of them are sufficiently soluble only in the hydrocarbon solvents, which become unstable under the high energies of pumping. Some of the compounds of this series containing a CHF_2SO_2 group are effective transformers of the irradiation of the second harmonic of the neodymium laser, with the transformation index reaching 20% in some cases. Substituent variations at the *para*-position of the 5-phenyl radical allows the realization of generation in a wide spectral range. The most marked influence both on the wavelength of generation and on the luminescence range is exercised by a dimethylamino group conjugated with a difluoromethylsulphonyl substituent. This compound, due to its solvatofluorochromic properties, allows generation in the 485–705 nm region with the appropriate selection of solvents and one of its advantages is a high solubility in ethanol.[28]

2-(4-Difluoromethylsulphonyl)-5-aryloxazoles are also used as activators of liquid scintillators.[31] The luminescent properties of the structurally simplest compound of this series (λ_{max} 405 nm, η 0·78) allows its application without another luminophore exerting a bathochromic shift of the luminescence maximum.

2-(4-Stilbenyl)-5-phenyloxazole (**VII**, R = H) is regarded as one of the most effective organic luminophores and it is widely used in various fields of science and technology.[32–34]



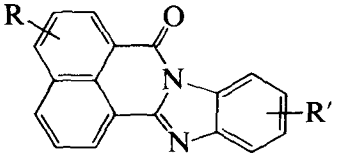
VII

The presence of electron donor substituents such as OCH_3 or NMe_2 at the stilbene moiety of this compound sharply decreases the luminescence intensity. Thus the introduction of the dimethylamino group decreases the quantum yield of luminescence from 0·88 to 0·14 (in toluene).[35] Quantum calculations of the first electron excited state of the unsubstituted compound show that the long-wave band in the absorption spectra, which is responsible for the luminescence, is the band of charge transition from the phenyloxazole moiety to the stilbene moiety. When a dimethylamino group is present, a long-wave absorption band cannot be regarded as being the result of charge transition through the whole electron system, as an oppositely directed electron shift from the dimethylamino-substituted phenyl radical onto the second aromatic ring of the stilbene moiety is observed. It could thus be concluded that the decrease of the quantum yield of luminescence, caused by the introduction of electron donor substituents, is connected with a weaker conjugation in the luminophore molecule and the resultant growth of the radiationless relaxation of the electron excitation energy.[36]

In confirmation of this supposition it was of interest to study the analogous compound containing a strong electron acceptor substituent which did not quench the luminescence, e.g. the nitro group. For this purpose, condensation of Horner reagent, prepared from 2-(4-bromomethylphenyl)-5-phenyloxazole, with 4-difluoromethylsulphonylbenzaldehyde in the presence of sodium methoxide gave 2-(4-difluoromethylsulphonylstilbenyl)-5-phenyloxazole (**VII**, R = CHF_2SO_2). The spectral and luminescent properties of this compound in toluene were investigated (λ_{max} of absorption 370 nm, λ_{max} of luminescence 450 nm, η 0·77).[37] The high quantum yield of luminescence confirms the earlier conclusion, that for the targeted synthesis of luminophores, not only the electron nature of the incorporated substituents but also their influence on the distribution of the electron density in the first excited state must be taken into consideration.[1]

FLUORINE-CONTAINING SUBSTITUTED 1,8-NAPHTHOYLENE-1′,2′-BENZIMIDAZOLES

Luminophores derived from 1,8-naphthoylene-1′,2′-benzimidazole (NBI) and its derivatives (**VIII**) have found broad practical application.[38-40]



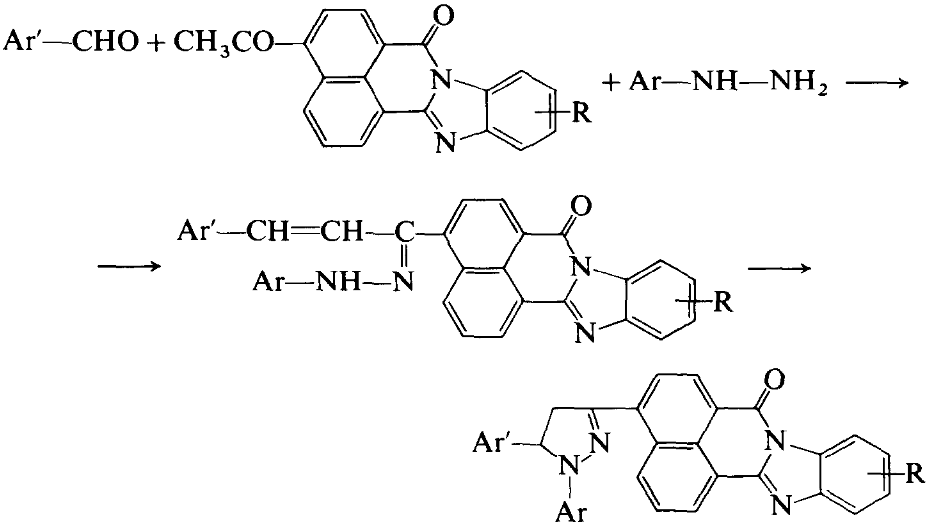
VIII

Unsubstituted NBI, prepared from naphthalic anhydride and *o*-phenylenediamine, luminesces in hydrocarbon solvents in the yellow–green spectral region (λ_{max} in toluene 490 nm, η 0·55).[18]

NBIs with fluorine-containing substituents were prepared by condensation of naphthalic anhydride with CF_3- or CHF_2SO_2-substituted *o*-phenylenediamines or with perfluoro-*o*-phenylenediamine.[41] In accordance with quantum calculations the electron density of the excited state of the NBI molecule is shifted toward the naphthalene ring. The fluorine-containing substituents hamper this shift and exert a noticeable hypsochromic shift (25–30 nm) of the luminescence maximum. The quantum yield of luminescence is increased in this case up to 0·60–0·67.

Incorporation of two fluorophoric groupings, viz. triarylpyrazoline and NBI, into the same molecule, leads to the formation of orange–red and red luminophores, e.g. **IX** (Ar, Ar′ = C_6H_5), which has a luminescence maximum in toluene at 600 nm, and a quantum yield close to 1.[42] Such compounds are usually prepared by the following scheme.

A number of such luminophores with fluorine-containing substituents were synthesized by this method. A CHF_2SO_2 group in the benzene ring of the NBI moiety increases its electron acceptor properties and exerts, as in the case of the triarylpyrazoline series, a bathofluoric shift. When it is present in the 1-phenyl radical (Ar), an alternative effect is observed. In both cases a very high quantum yield is retained. All these luminophores have an increased stability toward UV irradiation and luminesce not only in solution



IX

but also in polystyrene and poly(methyl methacrylate) and impart to them bright and lightfast colours with luminescence in the region of 570–700 nm.[43]

CONCLUSION

The introduction of the fluorine-containing substituents into the molecules of luminophores of different structure has been shown to result in variation of their spectral and luminescent properties and in some cases noticeably increases their stability toward UV and visible light.

REFERENCES

1. B. M. Krasovitskii and B. M. Bolotin, *Organic luminophores*, 2nd edn, (a) p. 336; (b) p. 294. Moscow, Khimia (1984).
2. L. M. Yagupolskii, A. Ya. Ilchenko and L. Z. Gandelsman, *Usp. Khim.*, **52**, 1732 (1983).
3. B. M. Krasovitskii, *Izv. Akad. Nauk SSSR, Ser. Phys.*, **47**, 1301 (1983).
4. Ya. A. Terskoi, B. M. Bolotin, V. G. Brudz and D. A. Drapkina, *Izv. Akad. Nauk SSSR, Ser. Phys.*, **27**, 754 (1963).
5. D. G. Pereyaslova, V. T. Skripkina and L. M. Yagupolskii., *Zh. Prikl. Spektr.*, **17**, 101 (1972).
6. V. T. Skripkina, Yu. M. Vinetskaya, D. G. Pereyaslova and L. M. Yagupolskii, *Zh. Prikl. Spektr.*, **22**, 703 (1975).

7. D. G. Pereyaslova, V. T. Skripkina and L. M. Yagupolskii, *Proc. All-Union Luminescence Meeting, Stavropol, 1971*, p. 79.
8. V. N. Dmitrieva, L. I. Dmitrievskaya, V. T. Skripkina and D. G. Pereyaslova, *Zh. Obshch. Khim.*, **45**, 1371 (1975).
9. D. G. Pereyaslova, V. T. Skripkina, Yu. M. Vinetskaya, L. P. Andreeva and L. I. Bogdanova, *Zh. Prikl. Spektr.*, **25**, 364 (1976).
10. L. A. Kutulya, V. F. Morina, A. E. Shevchenko and Yu. N. Surov, *Zh. Prikl. Spektr.*, **27**, 76 (1977).
11. B. M. Krasovitskii, D. G. Pereyaslova, V. T. Skripkina, V. M. Shershukov, Yu. M. Vinetskaya and L. M. Yagupolskii. *Proc. 13th Ukrainian Organic Chemistry Conference, Donetsk, 1978*, p. 45.
12. D. G. Pereyaslova, V. T. Skripkina, B. M. Krasovitskii and G. G. Yakobson, *Izv. Akad. Nauk SSSR, Ser. Chim.*, 81 (1974).
13. V. I. Natanson, D. G. Pereyaslova and V. T. Skripkina, *Khim. Geterotsikl. Soed.*, 669 (1978).
14. D. G. Pereyaslova, V. T. Skripkina, L. P. Andreeva and L. I. Bogdanova, *Proc. 4th All-Union Conference on Organic Luminophores, Kharkov, 1984*, p. 127.
15. R. N. Nurmuhametov, N. A. Lodigin and G. I. Grishina, *Optika i Spektroskopia*, **25**, 222 (1968).
16. N. P. Shimanskaya, L. A. Kotok, V. T. Skripkina, D. G. Pereyaslova and T. F. Alehina, *Zh. Obshch. Khim.*, **47**, 2054 (1977).
17. V. F. Podujailo, D. G. Pereyaslova, V. T. Skripkina, S. A. Veresubova and L. P. Andreeva, *Zh. Prikl. Spektr.*, **26**, 357 (1977).
18. S. A. Malinovskaya, V. T. Skripkina, S. A. Veresubova and L. E. Pelipyagina, in *Monocrystals and scintillators*, p. 94, Kharkov, All-Union Institute of Monocrystals (1977).
19. D. G. Pereyaslova, V. E. Bondarenko, V. T. Skripkina, Yu. U. Vinetskaya and L. I. Bogdanova, *Ukr. Khim. Zh.*, **45**, 553 (1979).
20. B. M. Krasovitskii, I. V. Lysova, L. Sh. Afanaciadi and Y. N. Surov, *Khim. Geterotsikl. Soedin.*, 1600 (1981).
21. B. M. Krasovitskii, I. V. Lysova and L. Sh. Afanaciadi, *Khim. Geterotsikl. Soedin.*, 158 (1978).
22. V. N. Kerr, F. N. Hayes, D. G. Ott, R. Lieb and H. Hansbury, *J. Org. Chem.*, **24**, 1864 (1959).
23. L. L. Nagornaya, V. D. Bezuglii and N. P. Demchenko, *Optika i Spektroskopia*, **13**, 518 (1962).
24. S. M. Krasovitskii, S. E. Kovalev and V. M. Shershukov, *Khim. Geterotsikl. Soedin.*, 1049 (1974).
25. B. M. Krasovitskii, V. M. Shershukov, D. G. Pereyaslova and V. T. Skripkina, USSR Patent 606859 (1978).
26. B. M. Krasovitskii, V. M. Shershukov and L. M. Yagupolskii, *Khim. Geterotsikl. Soedin.*, 1042 (1982).
27. I. M. Beterev, V. N. Ischenko, B. Ya. Kogan, B. M. Krasovitskii and A. A. Chernenko, *Kwantovaya Elektronika*, **4**, 441 (1977).
28. V. L. Volkov, V. I. Alekseeva, L. Sh. Afanaciadi, B. M. Krasovitskii, E. A. Luk'ianets and V. M. Shershukov, *Zh. Prikl. Spektr.*, **31**, 635 (1979).
29. F. A. Abakumov, A. P. Simonov, V. V. Fadeev, L. A. Kharitonov and R. V. Khohlov, *Pis'ma Zhurnal Eksp. Teor. Phys.*, **9**, 15 (1969).

30. N. A. Borisevitch, V. V. Grusinskii and L. M. Kutsina, *Zh. Prikl. Spektr.*, **12**, 1111 (1970).
31. S. A. Veresuboca and V. M. Shershukov, *Proc. 3rd All-Union Conference on Organic Luminophores, Kharkov, 1980*, p. 22.
32. V. I. Grigor'eva and B. M. Krasovitskii, *Khim. Geterotsikl. Soedin.*, 761 (1967).
33. Yu. V. Naboikin, L. A. Ogurtsova, A. P. Podgornii, F. S. Pokrovskaya, V. I. Grigor'eva, B. M. Krasovitskii, L. M. Kutsyna and V. G. Tischchenko, *Optika i Spektroskopia*, **28**, 974 (1970).
34. V. I. Grigor'eva, V. F. Podujailo and B. M. Krasovitskii, in *Monocrystals, scintillators and organic luminophores*, No. 5, p. 99. Kharkov, Monocrystalreaktiv (1969).
35. V. I. Grigor'eva, R. S. Milner and B. M. Krasovitskii, in *Monocrystals, scintillators and organic luminophores*, No. 4, p. 98. Kharkov, Monocrystalreaktiv (1968).
36. V. I. Grigor'eva, *Investigation in the series of vinyl and arylethylene derivatives of 2,5-diaryloxazoles and 2,5-diaryloxadiazoles-1,3,4.* PhD Thesis, Moscow (1969).
37. B. M. Krasovitskii, V. M. Shershukov, V. L. Volkov and L. Sh. Afanaciadi, *Zh. Org. Khim.*, **19**, 2459 (1983).
38. L. Ya. Malkes, A. F. Zaitsev, V. A. Tiurin, L. I. Sukiasova, A. S. Sajigin and B. M. Sapojnikov, USSR Patent 362038 (1971).
39. B. A. Vainshtok, V. V. Generahova and N. F. Levchenko, in *Radiational dosimetry and spectrometry of ionizing irradiation*, p. 178. Tashkent, Fan (1970).
40. N. F. Levchenko, V. M. Solomonov, E. A. Shevchenko and B. M. Krasovitskii, in *Monocrystals, scintillators and organic luminophores*, No. 4, p. 186. Kharkov, Monocrystalreaktiv (1968).
41. D. G. Pereyaslova, V. T. Skripkina, L. M. Yagupolskii, Yu. M. Vinetskaya and L. P. Andreeva, *Khim. Geterotsikl. Soedin.*, 1245 (1982).
42. B. M. Krasovitskii and E. A. Shevchenko, *Khim. Geterotsikl. Soedin.*, 756 (1968).
43. D. G. Pereyaslova, V. T. Skripkina, E. A. Shevchenko, L. P. Andreeva, S. A. Malinovskaya and L. I. Bogdanova, USSR Patent 907039 (1980).