Reactions of 2,2-disubstituted 1,1-dicyanoethenes with β-dimethylaminoacrolein aminal and 3-dimethylamino-1,1,3-trimethoxypropane

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A reaction of 2,2-disubstituted 1,1-dicyanoethenes with β-dimethylaminoacrolein aminal and 3-dimethylamino-1,1,3-trimethoxypropane leads to substituted 6,6-dicyano-1-dimethylaminohexatrienes and an organic salt containing 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraenide as the anion and (dimethylaminopropenylidene)dimethylammonium as the cation, on the basis of which new cation-anionic polymethine dyes were obtained.

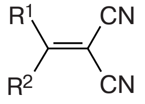
Key words: unsaturated dinitriles, β-dimethylaminoacrolein aminal, 3-dimethylamino-1,1,3-trimethoxypropane, polyenes, cation-anionic dyes.

Earlier, we have developed methods for the synthesis of ω-dimethylaminoaldehyde aminals and 1-dialkylamino-1,3,3-trimethoxypropanes, which, being highly reactive, readily undergo condensation with various CH-acids in the absence of a catalyst (and often of a solvent), leading to various polyfunctional conjugated polyenes, many of which possess unusual spectral properties, whereas some of them are polymethine dyes or intermediate products for their synthesis.[1] The reaction is characteristic, in particular, for the carbonyl compounds, in which the labile proton is located not only near the carbonyl group, but also is separated from it by one or two double bonds or by a heterocycle.[2]

Recently, we have studied interaction of these reagents with CH-acids containing dicyanomethylidene group, including ones with cyclic alkylidenemalononitriles,[3] 3-cyano-2-dicyanomethylidene-4,5,5-trimethyl-2,5-dihydrofuran,[4] 3-(dicyanomethylidene)indan-1-one, and 1,3-bis(dicyanomethylidene)indane.[5] It turned out that the reactions under study take different directions depending on the conditions and result in the formation of analogs of ketocyanine dyes, polycyanopolymethine salts, which are anionic dyes, ω-dimethylaminopolyene dinitriles containing dihydrofuran ring, and other compounds.

The interest to the polyene donor-acceptor chromophores, having nitrile groups in the acceptor fragment, is caused by the fact that some compounds of this type have nonlinear optical properties, as well as other interesting and unusual spectral features.[6—10]

In the present work, a condensation of β-dimethylaminoacrolein aminal **1** (see Ref. 11) and 3-dimethylamino-1,3,3-trimethoxypropane **2** (see Ref. 12) with unsaturated dinitriles **3—6** has been studied.



3—6

Compound	R^1	R^2	Compound	R^1	R^2
3	Me	Ph	**5**	Me	*cyclo*-C_6H_{11}
4	Et	Ph	**6**	Me	*cyclo*-C_3H_5

We assumed that the reaction at the methyl or methylene group in the dinitriles **3—6** will result in the obtaining the earlier unknown substituted 6,6-dicyano-1-dimethylaminohexatrienes, which are merocyanine dyes.

However, it turned out that when aminal **1** and reagent **2** react with dinitrile **3** in benzene, a cyclodimerization of the latter takes place to form (dimethylaminopropenylidene)-dimethylammonium salt **7** in high yield. When the reaction of aminal **1** with dinitrile **3** is carried out solvent-free, salt **7** is obtained in low yield (15%), whereas the earlier undescribed 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraenide salt **8**, which is an anionic dye, becomes the major product (46% yield) (Scheme 1).

A solvent-free interaction of dinitrile **3** with reagent **2** takes predominantly another direction, leading to the formation of a mixture of triene ξ-aminodinitrile **9** and salt **8** in the ratio 3 : 1 (UV spectral data). Dinitrile **9** was iso-

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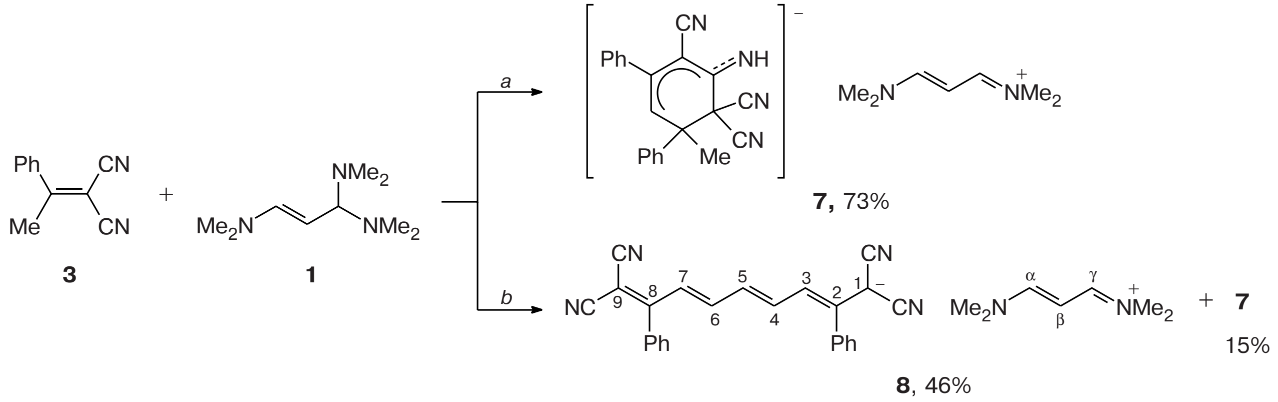
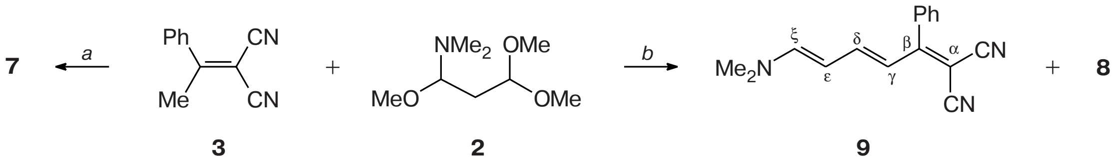
Scheme 1



a. In benzene, 20 °C, 24 h. *b*. Solvent-free, 20 °C, 24 h.

Scheme 2



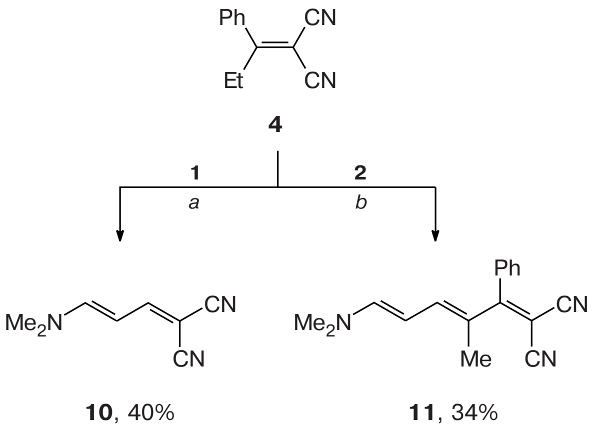
a. In benzene, 50—55 °C, 2.5 h. *b*. Solvent-free, 55—60 °C, 3 h.

lated from this mixture in pure form by chromatography on Al_2O_3 (Scheme 2).

Aminal **1** and reagent **2** react otherwise with dinitrile **4**, containing no Me group at the double bond. The reaction of dinitrile **4** with aminal **1** at 20—25 °C leads only to 4,4-dicyano-1-dimethylaminobutadiene **10** (Scheme 3).

We observed similar transformation in the reaction of aminal **1** with cyclic alkylidenemalononitriles.[3] Earlier, there was no literature reports on the reactions in which alkylidenemalononitriles would have transferred a dicyanomethylene fragment.

Scheme 3

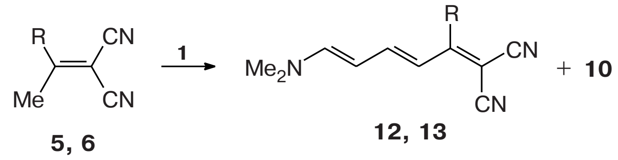


a. Solvent-free 20—25 °C, 24 h. *b*. In benzene or solvent-free, 60 °C, 3 h.

On the reaction of dinitrile **4** with reagent **2**, substituted triene ξ-aminodinitrile **11** was obtained in 34% yield (see Scheme 3).

The interaction of dinitriles **5** and **6** with aminal **1** leads to a mixture of trienes **12** or **13** and diene **10** (Scheme 4).

Scheme 4



R = *cyclo*-C_6H_{11} (**5**, **12**), *cyclo*-C_3H_5 (**6**, **13**)

Trienes **12** and **13**, being the major products in the reaction of dinitriles **5** and **6** with aminal **1** in anhydrous benzene (50—60 °C), were isolated in pure form by chromatography on Al_2O_3.

The structures of the synthesized substituted 6,6-dicyano-1-dimethylaminohexatrienes **9**, **11—13** were established on the basis of the single-dimensional and two-dimensional NMR spectra on the ^1H and ^{13}C nuclei, as well as on the UV and mass spectral data.

Table 1. ^{1}H and ^{13}C NMR spectral data for triene dinitriles **9**, **11—13**

Atom	δ (*J*/Hz)							
	9		**11**		**12**		**13**	
	^{1}H	^{13}C	^{1}H	^{13}C	^{1}H	^{13}C	^{1}H	^{13}C
α	—	59.59	—	59.53	—	68.04	—	70.42
β	—	168.20	—	168.73	—	176.24	—	171.78
γ	6.38 $^3J_{\gamma,\delta}$ = 13.3	112.26	2.16 (Me)	128.55; 15.28 (Me)	6.22 $^3J_{\gamma,\delta}$ = 14.4	114.35	6.25 $^3J_{\gamma,\delta}$ = 14.4	115.47
δ	6.71 $^3J_{\delta,\varepsilon}$ = 12.8	155.13	6.56 $^3J_{\delta,\varepsilon}$ = 12.0	155.73	7.46 $^3J_{\delta,\varepsilon}$ = 11.8	147.54	7.54 $^3J_{\delta,\varepsilon}$ = 12.0	149.02
ε	5.81 $^3J_{\varepsilon,\xi}$ = 14.0	102.67	5.55 $^3J_{\varepsilon,\xi}$ = 12.0	99.26	5.34 $^3J_{\varepsilon,\xi}$ = 12.4	100.50	5.36 $^3J_{\varepsilon,\xi}$ = 12.4	100.34
ξ	7.50	159.81	7.30	159.45	6.90	153.63	6.91	153.99
CN	—	117.70, 117.08	—	118.89, 117.74	—	116.86, 116.75	—	116.29, 116.02
Me_2N	3.11, 2.98	44.87, 37.55	3.11, 3.00	44.87, 37.54	3.01	40.20	3.09	39.80
1*	—	135.35	—	138.32	2.82	43.14	1.74	13.42
2	7.27	128.59	7.23	128.52	1.85; 1.73	30.78	1.10; 0.82	8.15
3	7.46	128.39	7.45	128.23	1.88; 1.35	26.59	—	—
4	7.46	129.38	7.44	129.36	1.79	25.57	—	—

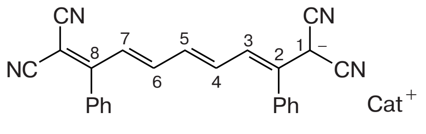
* Positions 1—4 are related to the atoms of substituent in β-position.

Assignment of signals in the ^{1}H and ^{13}C NMR spectra was performed on the basis of two-dimensional spectra COSY, HSQC, and HMBC. Configurations of the double bonds were established on the basis of the NOESY spectra and the values of the vicinal spin-spin coupling constants of the methine protons H_γ—H_δ, H_δ—H_ε, H_ξ—H_ε. Thus in the NOESY spectra of compounds **9** and **11**, a correlation of the *N*-methyl protons with H_ε was recorded, as well as of *ortho*- and *meta*-phenyl protons with H_δ, which, along with the values of the constants $^3J_{\delta-\varepsilon}$ and $^3J_{\varepsilon-\xi}$ being 12—14 Hz (Table. 1), indicates the *E*-configuration of the double bonds and the *s*-*trans*-conformation of the diene fragment. Similarly, in compounds **12** and **13** the values of the vicinal spin-spin coupling constants for protons H_γ—H_δ, H_δ—H_ε, and H_ξ—H_ε (11.8—14.4 Hz) also indicate the *E*-configuration of the double bonds, whereas a correlation between the mentioned protons in the NOESY spectra, the *s*-*trans*-conformation of the diene fragment.

Triene dinitriles **9**, **11—13** have one absorption maximum (λ_{max} = 485—490 nm) in the UV spectrum with high extinction coefficient (ε = 66000—102000). The presence of intensive signals of molecular ions is characteristic of the mass spectra of dinitriles **9**, **11—13**.

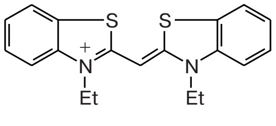
The structures of salts **7** and **8** were confirmed by the ^{1}H NMR and UV spectra and elemental analysis data. In the UV spectrum of salt **7**, there is one absorption maximum (λ_{max} = 312 nm) belonging to the cation, *viz.*, (dimethylaminopropenylidene)dimethylammonium. Tetracyanononamethine salt **8** has two absorption maxima in the UV spectrum: λ_{max} = 312 nm belongs to the cation, whereas λ_{max} = 680 nm (in EtOH) or 720 nm (in $CHCl_3$) to tetracyanononamethine anion.

The iminium cation of salt **8** can be easily exchanged for the cation of a cyanine dye, resulting in the formation of new cation-anionic polymethine dyes **14—16** in 40—45% yield as the dark violet crystals, in the electron spectrum of which, the absorption maxima (λ_{max}) correspond to the λ_{max} of the cationic and anionic components. In dye **16**,

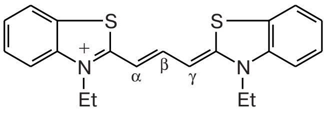


14—16

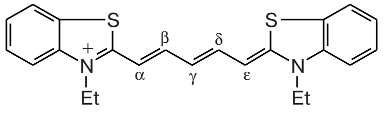
14: Cat^1 =



15: Cat^2 =



16: Cat^3 =



only one absorption band is observed, since the λ_{max} values for its precursors, salts **8** and Cat^{3}OTs, overlap.

Results of photochemical and photophysical study of the synthesized anionic **8**, cation-anionic **14—16**, and merocyanine dyes **9**, **11—13** will be presented in subsequent papers.

Experimental

NMR spectra for compounds **9**, **11—13** were recorded on a Bruker DRX-500 spectrometer (500.13 and 125.76 MHz for ^{1}H and ^{13}C, respectively) in DMSO-d$_6$ (**9**, **11**) and CDCl$_3$ (**12**, **13**) at 30 °C. Chemical shifts of ^{13}C and ^{1}H nuclei were calibrated with respect to the signals of carbon atoms and residual protons of chloroform-d (77.0 and 7.27 ppm) and dimethylsulfoxide-d$_6$ (39.5 and 2.50 ppm). Two-dimensional spectra were recorded following the standard Bruker procedures. ^{1}H NMR spectra of compounds **7**, **8**, **10**, **14—16** were recorded on a Bruker WM-250 spectrometer (250 MHz). Mass spectra (EI) were recorded on a Kratos MS-30 instrument (70 eV). Absorption spectra were recorded on a Specord UV-VIS spectrophotometer. Monitoring of the reaction courses and isolation of compounds by column chromatography on Al$_2$O$_3$ were conducted by UV spectroscopy. Starting dinitriles **3** (see Ref. 13), **4** (see Ref. 13), and **6** (see Ref. 14) were synthesized according to procedures described earlier. Dinitrile **5** (b.p. 147—150 °C (10 mm), n_D^{20} 1.5050) was obtained similarly to **6**. In the UV spectra (EtOH) of starting dinitriles λ_{max}/nm (ε): 230 (8729) and 295 (15482) (**3**), 230 (9341) and 295 (15139) (**4**), 240 (15335) (**5**), 260 (15529) (**6**).

(Dimethylaminopropenylidene)dimethylammonium 1,5,5-tricyano-6-imino-4-methyl-2,4-diphenylcyclohex-2-en-1-ide (7). ***A.*** Aminal **1** (0.64 g, 3.8 mmol) was added dropwise to a solution of dinitrile **3** (0.6 g, 3.6 mmol) in anhydrous benzene (3 mL) under stirring. After 1 day, the mixture was concentrated *in vacuo*, anhydrous acetone (5—7 mL) was added to the residue, and this was cooled down to −10 °C, a precipitate of light gray color was separated and washed with anhydrous acetone to obtain salt **7** (0.6 g, 73%) with m.p. 168—170 °C. Found (%): C, 75.76; H, 6.64; N, 17.92. $C_{29}H_{30}N_6$. Calculated (%): C, 75.32; H, 6.49; N, 18.18. UV (EtOH), λ_{max}/nm (ε): 312 (42280). ^{1}H NMR (DMSO-d$_6$), δ: 1.55 (s, 3 H, Me); 3.05, 3.20 (both s, 6 H each, NMe$_2$); 5.20 (s, 1 H, NH); 5.45 (t, 1 H, H$_\beta$, *J* = 12 Hz); 6.50 (s, 1 H, CH); 7.20—7.45 (m, 10 H, Ph); 7.72 (d, 2 H, H$_\alpha$, H$_\gamma$, *J* = 12 Hz).

B. Reagent **2** (0.094 g, 0.54 mmol) was added dropwise to a solution of dinitrile **3** (0.09 g, 0.54 mmol) in anhydrous benzene (3 mL). The mixture was heated for 2.5 h at 50—55 °C and kept at 20—25 °C for 2 days. A precipitate of light gray color was separated, and washed with anhydrous acetone to obtain salt **7** (0.05 g, 40%) with m.p. 167—169 °C, giving no m.p. depression with salt **7** isolated above. UV (EtOH), λ_{max}/nm (ε): 312 (41100).

(Dimethylaminopropenylidene)dimethylammonium 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraen-1-ide (8). A mixture of dinitrile **3** (0.3 g, 1.8 mmol) and aminal **1** (0.32 g, 1.9 mmol) was kept for 1 day at 20—25 °C and concentrated *in vacuo*. A thick dark red oily residue was washed with anhydrous ether, dissolved in anhydrous acetone, and cooled down to −10 °C. The precipitate was separated and washed with anhydrous acetone to obtain salt **7** (0.06 g, 15%), the m.p., UV spectrum, and ^{1}H NMR spectrum of which agree with those for salt **7** described above. After salt **7** was separated, H$_2$O (10 mL) was added to the solution, which was triturated followed by separation of the precipitate, which in its turn was washed with H$_2$O and dried to obtain salt **8** (0.2 g, 46%) as green crystals with m.p. 65—70 °C. Found (%): C, 77.40; H, 6.35; N, 16.72. $C_{32}H_{30}N_6$. Calculated (%): C, 77.10; H, 6.02; N, 16.86. UV, λ_{max}/nm (ε): (EtOH) 312 (37026), 680 (40272); (CHCl$_3$) 312 (34477), 720 (68953). ^{1}H NMR (CDCl$_3$), δ: 3.04, 3.25 (both s, 6 H each, NMe$_2$); 5.08 (t, 1 H, H$_\beta$, *J* = 12 Hz); 6.35—6.50 (m, 2 H, CH); 7.10—7.45 (m, 13 H, Ph, CH); 7.65 (d, 2 H, H$_\alpha$, H$_\gamma$, *J* = 12 Hz).

(5-Dimethylamino-1-phenylpenta-2,4-dienylidene)malononitrile (9). A mixture of dinitrile **3** (0.9 g, 5.4 mmol) and 3-dimethylamino-1,1,3-trimethoxypropane (**2**) (0.95 g, 5.4 mmol) was stirred for 3 h at 55—60 °C. After the heating was stopped, the UV spectral data showed the formation of a mixture of triene dinitrile **9** (λ_{max} = 480) and salt **8** (λ_{max} = 312 and λ_{max} = 720) in the ratio 3 : 1 in the reaction. Salt **8**, described above, was not isolated, while dinitrile **9** was isolated from the concentrated reaction mixture by chromatography on Al$_2$O$_3$ with elution with CHCl$_3$. A precipitate from chloroform eluate was repeatedly washed with anhydrous ether to obtained triene dinitrile **9** (0.27 g, 20%) as a brown powder with m.p. 152 °C (decomp.). Found (%): C, 76.76; H, 6.24; N, 16.64. $C_{16}H_{15}N_3$. Calculated (%): C, 77.10; H, 6.02; N, 16.86. UV, λ_{max}/nm (ε): (CHCl$_3$) 480 (70550); (EtOH) 490 (79888). MS, *m/z* 249 [M]$^+$.

(3-Dimethylaminoprop-2-enylidene)malononitrile (10). A mixture of dinitrile **4** (0.3 g, 1.6 mmol) and aminal **1** (0.28 g, 1.6 mmol) was kept for 1 day at 20—25 °C and concentrated *in vacuo*. The residue was chromatographed on Al$_2$O$_3$. A precipitate from the chloroform eluate was washed with cold anhydrous ether to obtain nitrile **10** (0.1 g, 40%), identical to the authentic sample in the m.p., UV and NMR-spectra.[15]

(5-Dimethylamino-2-methyl-1-phenylpenta-2,4-dienylidene)-malononitrile (11). A mixture of dinitrile **4** (0.9 g, 5 mmol) and reagent **2** (0.95 g, 5.4 mmol) was heated under stirring for 3 h at 55 °C and concentrated. The residue was chromatographed on Al$_2$O$_3$. After concentration of the chloroform eluate, an oily precipitate of dark brown color was obtained (0.8 g) with λ_{max} = 490 nm in the UV spectrum. After the precipitate was washed with anhydrous ether several times, triene dinitrile **11** was obtained (0.44 g, 34%) as a brown powder with m.p. 138 °C (decomp.). Found (%): C, 77.42; H, 6.45; N, 15.82. $C_{17}H_{17}N_3$. Calculated (%): C, 77.56; H, 6.46; N, 15.97. UV, λ_{max}/nm (ε): (EtOH) 490 (65750)); (CHCl$_3$) 490 (74955). MS, *m/z* 263 [M]$^+$.

(2-Cyclohexyl-5-dimethylaminopenta-2,4-dienylidene)malononitrile (12). Aminal **1** (0.51 g, 3 mmol) was added dropwise to dinitrile **5** (0.52 g, 3 mmol) in anhydrous benzene (15 mL) under stirring, the mixture was kept for 1 h at 20—25 °C and then heated for 3 h at 55—60 °C. After the heating was stopped, the UV spectrum of the reaction mixture showed the presence of absorption maxima λ_{max} = 376 and 485 nm, belonging to dinitriles **10** and **12**, respectively (ratio 1 : 4). The residue left after concentration of the reaction mixture *in vacuo* was chromatographed on Al$_2$O$_3$ with CHCl$_3$ as the eluent.

A precipitate was isolated from the chloroform eluate, which was thoroughly washed with anhydrous ether to obtain dinitrile **12** (0.23 g, 30%) as a dark orange powder with m.p. 140—141 °C. Found (%): C, 74.86; H, 8.05; N, 16.46. $C_{16}H_{21}N_3$. Calculated (%): C, 75.29; H, 8.23; N, 16.47. UV, λ_{max}/nm (ε): (EtOH) 485 (102000); (CHCl$_3$) 470 (94444). MS, *m/z* 255 [M]$^+$.

According to the UV spectral data, a mixture of dinitriles **10** and **12** in the ratio 2 : 1 was obtained from dinitrile **5** and reagent **2** in anhydrous benzene under conditions described above.

(1-Cyclopropyl-5-dimethylaminopenta-2,4-dienylidene)malononitrile (13). Aminal **1** (0.52 g, 3 mmol) was added dropwise to dinitrile **6** (0.4 g, 3 mmol) in anhydrous benzene (15 mL) and this was stirred for 1 h at 20—25 °C followed by heating for 2 h at 55—60 °C. The UV spectrum of the reaction mixture showed the formation of a mixture of dinitriles **13** and **10** in the ratio 2.5 : 1. Dinitrile **13** (0.13 g, 20%) was isolated (as described above for dinitrile **12**) as a brown powder with m.p. 138—139 °C. Found (%): C, 72.88; H, 6.95; N, 19.56. $C_{13}H_{15}N_3$. Calculated (%): C, 73.24; H, 7.04; N, 19.72. UV, λ_{max}/nm (ε): (EtOH) 490 (95343); ($CHCl_3$) 480 (79114). MS, *m/z* 213 $[M]^+$.

Cation-anionic dyes (14—16). Equimolar amounts of salt **8** and cyanine dye tosylate ($[Cat^1]^+OTs^-$, $[Cat^2]^+OTs^-$, $[Cat^3]^+OTs^-$) were separately dissolved in minimum EtOH—CH_2Cl_2 (10 : 1). The solutions after filtration were mixed and kept for 1 h at 20 °C. The crystals of dyes **15** and **16** formed were filtered off and washed with H_2O, EtOH, and ether. In the case of dye **14**, the reaction mixture was concentrated, the residue was suspended in H_2O, the precipitate formed was filtered off, washed with H_2O, EtOH and ether.

3-Ethyl-2-[(3-ethyl-2,3-dihydrobenzothiazol-2-ylidene)methyl]-benzothiazolium 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraen-1-ide (14). M.p. 122—125 °C. UV, λ_{max}/nm (ε): ($CHCl_3$) 430 (74381) 700 (29585); (EtOH) 420 (67620), 680 (26691). ^1H NMR (DMSO-d_6), δ: 1.35 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7.0 Hz); 4.56 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7.0 Hz); 6.65 (s, 1 H, CH); 6.35—6.50 (m, 2 H, CH); 7.10—8.05 (m, 21 H, Ph, CH).

3-Ethyl-2-[(3-ethyl-2,3-dihydrobenzothiazol-2-ylidene)prop-1-en-1-yl]benzothiazolium 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraenide (15), m.p. 132—136 °C. UV, λ_{max}/nm (ε): ($CHCl_3$) 565 (142600) 700 (97750); (EtOH) 560 (149500), 680 (101200). ^1H NMR (DMSO-d_6), δ: 1.30 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7.0 Hz); 4.40 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7.0 Hz); 6.40—6.60 (m, 4 H, H_α, H_γ, CH); 7.10—8.00 (m, 22 H, Ph, H_β, CH).

3-Ethyl-2-[4-(3-ethyl-2,3-dihydrobenzothiazol-2-ylidene)-buta-1,3-dien-1-yl]benzothiazolium 1,1,9,9-tetracyano-2,8-diphenylnona-2,4,6,8-tetraenide (16), m.p. 217—220 °C. UV, λ_{max}/nm (ε): ($CHCl_3$) 670 (255104); (EtOH) 680 (231912). ^1H NMR (DMSO-d_6), δ: 1.30 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7.0 Hz); 4.40 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7.0 Hz); 6.00—6.40 (m, 5 H, H_α, H_γ, H_ε, CH); 7.10—8.05 (m, 23 H, Ph, H_β, H_δ, CH).

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