Reactions of β-dimethylaminoacrolein aminal and 3-dimethylamino-1,1,3-trimethoxypropane with 3-(dicyanomethylidene)indan-1-one and 1,3-bis(dicyanomethylidene)indane

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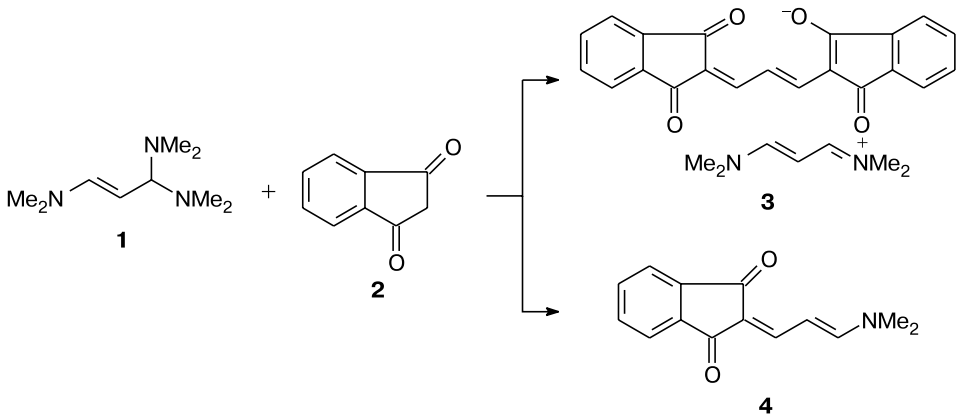
Reactions of 3-(dicyanomethylidene)indan-1-one and 1,3-bis(dicyanomethylidene)indane with β-dimethylaminoacrolein aminal, 3-dimethylamino-1,1,3-trimethoxypropane, DMF acetal, and ethyl orthoformate were studied. The reaction of 3-(dicyanomethylidene)indan-1-one with 3-dimethylamino-1,1,3-trimethoxypropane gave the 1,3-bis(1-dicyanomethylidene-3-oxoindane)trimethineoxanine salt in high yield, which was used to obtain a number of novel cation-anionic polymethine dyes.

Key words: dicyanomethylideneindanes, β-dimethylaminoacrolein aminal, 3-dimethylamino-1,1,3-trimethoxypropane, oxanine salts, cation-anionic dyes.

Earlier,[1] we have demonstrated that condensation of aminals of conjugated ω-dimethylamino aldehydes with various CH acids (including β-dicarbonyl compounds) gives, depending on the reaction conditions, conjugated α,α-difunctionalized ω-amino polyenes or oxanine or tetracyano polymethine salts, which are anionic dyes. The first example of the reaction following the two pathways has been discovered[2] in condensation of β-dimethylaminoacrolein aminal (**1**) with indane-1,3-dione (**2**), yielding oxanine salt **3** and δ-dimethylamino diene diketone **4** (Scheme 1).

Oxanine and tetracyano polymethine salts have been used as anionic components for preparation of novel cation-anionic polymethine dyes possessing two intense bands in the electronic absorption spectrum.[1,3—5] A photochemical study of these dyes has revealed interactions of the polymethine chromophores of the cation and the anion, transfer of excitation energy and electron transfer between them, the formation of fluorescent and nonfluorescent aggregates in weakly polar media, and other interesting and useful properties.[6—11]

Scheme 1



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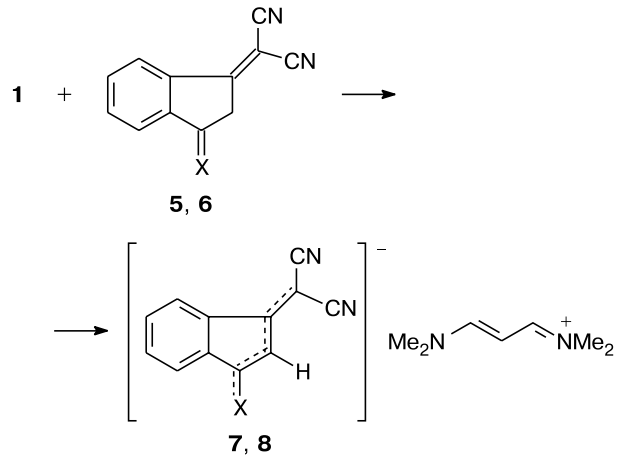
In the last few years, donor-acceptor polyene chromophores containing cyano groups in the acceptor moiety of the molecule are under intensive investigation. It has been found that some conjugated compounds of this type exhibit nonlinear optical properties. This makes them promising for use in optoelectronics, nonlinear optics, data recording and processing, *etc*.[12—16]

The goal of the present work was to obtain novel analogs of oxanine salt **3** and conjugated amino diketone **4** that would contain a dicyanomethylidene group in the indane fragment instead of one or both carbonyl groups.

3-(Dicyanomethylidene)indan-1-one (**5**) and 1,3-bis(dicyanomethylidene)indane (**6**) were employed as starting reagents.

It turned out that aminal **1** reacts with substituted indanes **5** and **6** in a way different from its reaction with indane-1,3-dione (**2**), giving only the corresponding trimethine salts **7** and **8** identified from ^{1}H NMR and UV spectra and elemental analysis data (Scheme 2).

Scheme 2



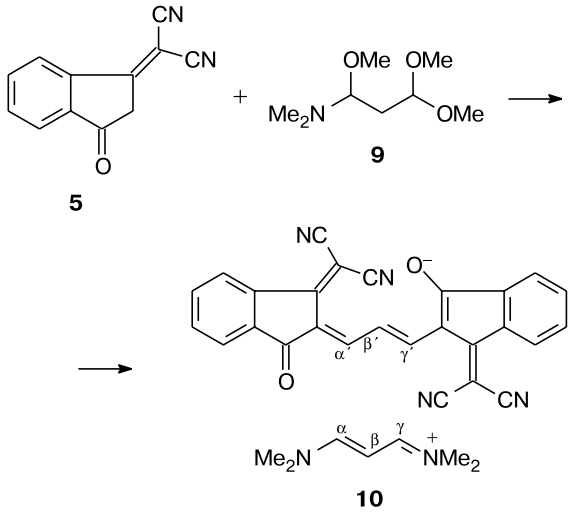
X = O (**5**, **7**), C(CN)$_2$ (**6**, **8**)

Replacement of aminal **1** by 1-dimethylamino-1,3,3-trimethoxypropane (**9**)[17] in reactions with indanes **5** and **6** did not afford the target δ-amino diene dinitriles analogous to diketone **4**. For instance, a reaction of compound **9** with indane **5** gave oxanine salt **10** as the sole product in high yield (80%) (Scheme 3).

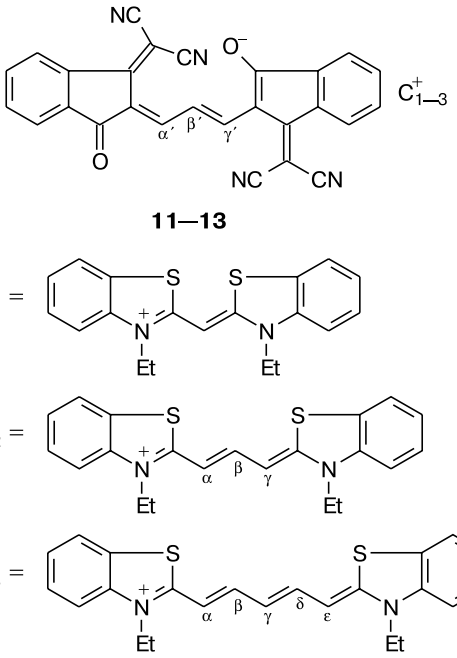
The structure of salt **10** was determined from spectroscopic and elemental analysis data. It should be noted that introduction of the dicyanomethylidene group into salt **3** causes a considerable (125 nm) bathochromic shift of the absorption peak in the electronic absorption spectrum.

The trimethine cation of salt **10** is easily exchanged for the cation of a cyanine dye to give novel cation-anionic dyes **11**—**13** in 45—60% yields. They are dark

Scheme 3



brown crystals (m.p. >260 °C) with the absorption peaks λ_{max} corresponding to those of the cation and the anion.

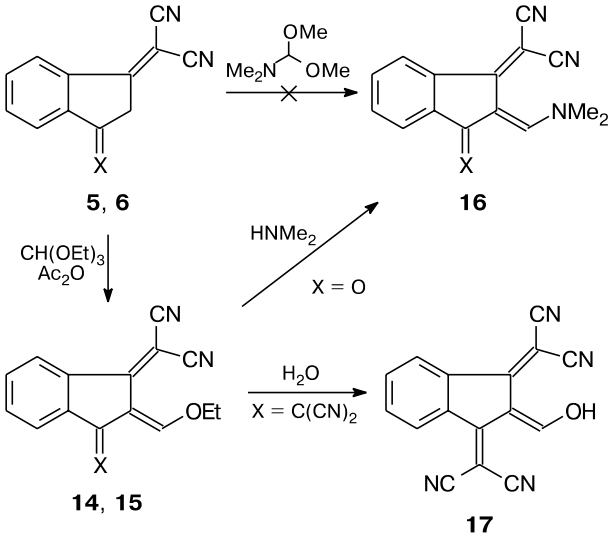


The spectroscopic and luminescence characteristics of dyes **11**—**13** and data on their photophysical and photochemical processes will be published elsewhere.

To synthesize the lower dicyano vinylogs of the type **4**, we carried out condensation reactions of compounds **5** and **6** with triethyl orthoformate (Scheme 4) and obtained ethoxymethylidene derivatives **14** and **15** in high yields.

The ethoxy group in compound **14**, unlike compound **15**, was easily exchanged for the NMe$_2$ group to

Scheme 4



X = O (**5**, **14**, **16**), $C(CN)_2$ (**6**, **15**)

give unsaturated amine **16**. Reflux of compound **15** in 96% EtOH afforded hydroxymethylidene tetranitrile **17** in high yield. Unfortunately, compounds **5** and **6** did not react with DMF acetal.

The structures of novel cyano-containing indanes **14**—**17** were proved by ^{1}H NMR and UV spectroscopy and elemental analysis.

Experimental

UV spectra were recorded on a Specord UV—VIS spectrophotometer. ^{1}H NMR spectra were recorded on a Bruker MW-250 instrument (250 MHz) with reference to Me_4Si. Mass spectra (EI) were recorded on a Kratos MS-30 instrument (70 eV).

3-(Dicyanomethylidene)indan-1-one (5) was prepared in 62% yield according to a known procedure,[18] m.p. 220—224 °C (after triple recrystallization from AcOH). Found (%): C, 73.96; H, 3.21; N, 14.30. $C_{12}H_6N_2O$. Calculated (%): C, 74.22; H, 3.11; N, 14.43. UV (EtOH), λ_{max}/nm (ε): 228 (7632), 305 (6678), 490 (3498). UV ($CHCl_3$), λ_{max}/nm (ε): 245 (10777), 252 (12070), 260 (10777), 310 (16166). ^{1}H NMR (CD_2Cl_2), δ: 3.80 (s, 2 H, CH_2); 7.90—8.10 (m, 3 H, H arom.); 8.70 (m, 1 H, H arom.).

1,3-Bis(dicyanomethylidene)indane (6) was prepared in 79% yield. The known procedure[18] we followed recommends washing of the product until the rinsing water becomes colorless. However, this result was unattainable for us because the water turned blue even in the presence of trace amounts of this compound. UV ($CHCl_3$), λ_{max}/nm (ε): 280 (14519), 285 (29039), 290 (39526), 350 (29039). UV (EtOH), λ_{max}/nm (ε): 245 (36876), 350 (25352), 365 (28810), 385 (23048), 580 (31181), 630 (29962). ^{1}H NMR ($CDCl_3$), δ: 4.30 (s, 2 H, CH_2); 7.90, 8.70 (both m, 2 H each, H arom.).

***N*-(3-Dimethylaminoprop-2-enylidene)-*N,N*-dimethylammonium 3-(dicyanomethylidene)-1-oxoindan-2-ide (7).** A mixture of 3-(dicyanomethylidene)indan-1-one (**5**) (0.2 g, 1 mmol) and aminal **1** (0.17 g, 1 mmol) in dry benzene (2 mL) was stirred at 20—25 °C for 3 h and concentrated. Dry acetone was added, the mixture was cooled, and trimethine salt **7** was separated as a dark violet powder. The yield was 0.2 g (70%), m.p. 80—85 °C. UV ($CHCl_3$), λ_{max}/nm (ε): 312 (60000), 500 (10000). ^{1}H NMR ($CDCl_3$), δ: 2.95, 3.25 (both s, 6 H each, NMe_2); 4.90 (t, 1 H, H_β, J = 12 Hz); 5.20 (s, 1 H, CH); 7.90 (m, 1 H, H arom.); 7.20—7.35 (m, 5 H, H arom., H_α, H_γ).

***N*-(3-Dimethylaminoprop-2-enylidene)-*N,N*-dimethylammonium 1,3-bis(dicyanomethylidene)indan-2-ide (8).** A mixture of indane **6** (0.242 g, 1 mmol) and aminal **1** (0.17 g, 1 mmol) in dry benzene (2 mL) was stirred for 2 h. The precipitate was separated and washed with dry benzene to give salt **8** as a dark brown powder. The yield was 0.25 g (68%), m.p. 181—183 °C. Found (%): C, 71.44; H, 5.22; N, 22.50. $C_{22}H_{20}N_6$. Calculated (%): C, 71.74; H, 5.43; N, 22.82. UV ($CHCl_3$), λ_{max}/nm (ε): 312 (40480), 530 (16560), 595 (22080), 650 (17020). ^{1}H NMR ($CDCl_3$), δ: 3.10, 3.30 (both s, 6 H each, NMe_2); 5.08 (t, 1 H, H_β, J = 12 Hz); 5.85 (s, 1 H, CH); 7.28 (m, 2 H, H arom.); 7.40 (d, 2 H, H_α, H_γ, J = 12 Hz); 8.05 (m, 2 H, H arom.).

***N*-(3-Dimethylaminoprop-2-enylidene)-*N,N*-dimethylammonium 1-dicyanomethylidene-2-[3-(3-dicyanomethylidene-1-oxoindan-2-ylidene)prop-1-enyl]-1*H*-inden-3-olate (10).** 1-Dimethylamino-1,1,3-trimethoxypropane (**9**) (0.36 g, 2 mmol) was added to 3-(dicyanomethylidene)indan-1-one (**5**) (0.4 g, 2 mmol). The reaction mixture was kept at 20—25 °C for 48 h. The voluminous dark brown precipitate that formed was repeatedly washed with dry ether by decantation, filtered off, and washed with anhydrous EtOH and dry ether. The yield of salt **10** was 0.44 g (80%), m.p. 225—228 °C. Found (%): C, 73.72; H, 4.51; N, 14.92. $C_{34}H_{26}N_6O_2$. Calculated (%): C, 74.18; H, 4.72; N, 15.27. UV ($CHCl_3$), λ_{max}/nm (ε): 310 (35351), 510 (11785), 700 (47143). UV (EtOH), λ_{max}/nm (ε): 312 (50400), 510 (13750), 690 (50416). ^{1}H NMR (DMSO-d_6), δ: 3.05, 3.30 (both s, 6 H each, NMe_2); 5.40 (t, 1 H, H_β, J = 12 Hz); 7.60—7.70 (m, 8 H, H arom.); 8.12 (d, 2 H, H_α, H_γ, J = 12 Hz); 8.35 (d, 2 H, $H_{\alpha'}$, $H_{\gamma'}$, J = 12 Hz); 9.15 (t, 1 H, $H_{\beta'}$, J = 12 Hz).

3-Ethyl-2-[(3-ethyl-2,3-dihydrobenzo[1,3]thiazol-2-ylidene)methyl]benzo[1,3]thiazol-3-ium 1-dicyanomethylidene-2-[3-(3-dicyanomethylidene-1-oxoindan-2-ylidene)prop-1-enyl]-1*H*-inden-3-olate (11). A filtered solution of tosylate C_1 (0.1 mmol) in a mixture of CH_2Cl_2 (4 mL) and EtOH (3 mL) was added to a filtered solution of salt **10** (0.1 mmol) in a mixture of dry CH_2Cl_2 (6 mL) and dry DMSO (1 mL). After an hour, the reaction mixture was concentrated *in vacuo* and the residue was triturated with water (5—7 mL) and kept at 5—8 °C. The precipitate was separated and washed with water, EtOH, and ether to give dye **11** (0.045 g, 60%) as dark brown crystals. UV ($CHCl_3$), λ_{max}/nm (ε): 440 (64008), 720 (80010). UV (EtOH), λ_{max}/nm: 425, 680. ^{1}H NMR (DMSO-d_6), δ: 1.35 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7 Hz); 4.60 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7 Hz); 6.65 (s, 1 H, CH); 7.40—8.15 (m, 16 H, H arom.); 8.35 (d, 2 H, $H_{\alpha'}$, $H_{\gamma'}$, J = 12 Hz); 9.05 (t, 1 H, $H_{\beta'}$, J = 12 Hz).

3-Ethyl-2-[3-(3-ethyl-2,3-dihydrobenzo[1,3]thiazol-2-ylidene)prop-1-enyl]benzo[1,3]thiazol-3-ium 1-dicyanomethylidene-2-[3-(3-dicyanomethylidene-1-oxoindan-2-ylidene)prop-1-

enyl]-1*H*-inden-3-olate (12). A filtered solution of tosylate C_2 (0.1 mmol) in a mixture of CH_2Cl_2 (3 mL) and EtOH (2 mL) was added to a filtered solution of salt **10** (0.1 mmol) in a mixture of CH_2Cl_2 (6 mL) and dry DMSO (1 mL). After an hour, the reaction mixture was concentrated *in vacuo* and the residue was triturated with water (5—7 mL) and kept at 5—8 °C. The precipitate was separated and washed with water, EtOH, and ether to give dye **12** (0.036 g, 45%) as dark brown crystals. UV ($CHCl_3$), λ_{max}/nm (ε): 300 (22514), 560 (138087), 720 (69043). UV (EtOH), λ_{max}/nm (ε): 300 (13505), 558 (78799), 680 (45028). ^{1}H NMR (DMSO-d_6), δ: 1.35 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7 Hz); 4.35 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7 Hz); 6.55 (d, 2 H, H_α, H_γ, J = 12 Hz); 7.40—8.20 (m, 17 H, H_β, H arom.); 8.35 (d, 2 H, $H_{\alpha'}$, $H_{\gamma'}$, J = 12 Hz); 9.15 (t, 1 H, $H_{\beta'}$, J = 12 Hz).

3-Ethyl-2-[5-(3-ethyl-2,3-dihydrobenzo[1,3]thiazol-2-ylidene)penta-1,3-dien-1-yl]benzo[1,3]thiazol-3-ium 1-dicyanomethylidene-2-[3-(3-dicyanomethylidene-1-oxoindan-2-ylidene)prop-1-enyl]-1*H*-inden-3-olate (13). A filtered solution of tosylate C_3 (0.1 mmol) in CH_2Cl_2 (8 mL) was added to a filtered solution of salt **10** (0.1 mmol) in a mixture of CH_2Cl_2 (6 mL) and dry DMSO (1 mL). After an hour, the precipitate that formed was separated and washed with water, EtOH, and ether to give dye **13** (0.039 g, 48%). UV ($CHCl_3$), λ_{max}/nm (ε): 640 (74000). UV (EtOH), λ_{max}/nm: 660. ^{1}H NMR (DMSO-d_6), δ: 1.30 (t, 6 H, $C\underline{H}_3CH_2N$, J = 7 Hz); 4.40 (q, 4 H, $CH_3C\underline{H}_2N$, J = 7 Hz); 6.40—6.60 (m, 3 H, H_α, H_γ, H_ε); 7.40—8.20 (m, 18 H, H_β, H_δ, H arom.); 8.35 (d, 2 H, $H_{\alpha'}$, $H_{\gamma'}$, J = 12 Hz); 9.15 (t, 1 H, $H_{\beta'}$, J = 12 Hz).

Because dyes **11**—**13** are high-melting (m.p. >260 °C) and nonvolatile salts, they cannot be identified by elemental analysis or mass spectrometry.

3-(Dicyanomethylidene)-2-(ethoxymethylidene)indan-1-one (14). A mixture of 3-(dicyanomethylidene)indan-1-one (**5**) (0.58 g, 3 mmol) and triethyl orthoformate (0.59 g, 4 mmol) was heated in freshly distilled Ac_2O (20 mL) to 60 °C under stirring. After another portion of Ac_2O (10 mL) was added, the solution was heated at 70—80 °C for 1 h and then at 80—90 °C for 1 h, cooled, and concentrated *in vacuo*. The residue was washed with anhydrous ether and the precipitate that formed was filtered off and repeatedly washed with anhydrous ether to give compound **14** (0.61 g, 81%) as a brown powder, m.p. 162—165 °C. Found (%): C, 71.88; H, 3.80; N, 11.64. $C_{15}H_{10}N_2O_2$. Calculated (%): C, 72.00; H, 4.00; N, 11.20. UV ($CHCl_3$), λ_{max}/nm (ε): 285 (25333), 330 sh (8333). UV (EtOH), λ_{max}/nm (ε): 265 (17187), 293 sh (15000), 335 sh (14000). ^{1}H NMR ($CDCl_3$), δ: 1.55 (t, 3 H, $C\underline{H}_3CH_2$, J = 7 Hz); 4.52 (q, 2 H, $C\underline{H}_2CH_3$, J = 7 Hz); 7.70—7.90 (m, 4 H, H arom., CH); 8.70 (m, 1 H, H arom.). MS, m/z: 250 $[M]^+$.

1,3-Bis(dicyanomethylidene)-2-(ethoxymethylidene)indane (15). A mixture of 1,3-bis(dicyanomethylidene)indane (**6**) (0.726 g, 3 mmol) and triethyl orthoformate (0.59 g, 4 mmol) was stirred in freshly distilled Ac_2O (30 mL) at 70—90 °C for 2 h and concentrated *in vacuo*. The oily, nearly black residue was triturated with anhydrous ether. The precipitate that formed was separated and washed with ether to give compound **15** (0.7 g, 79%), m.p. 166—168 °C. Found (%): C, 72.35; H, 3.10; N, 18.52. $C_{18}H_{10}N_4O$. Calculated (%): C, 72.48; H, 3.35; N, 18.79. UV ($CHCl_3$), λ_{max}/nm (ε): 300 (27316), 350 sh (17383), 400 sh (13139). ^{1}H NMR ($CDCl_3$), δ: 1.50 (t, 3 H, $C\underline{H}_3CH_2O$, J = 7 Hz); 4.60 (q, 2 H, $CH_3C\underline{H}_2O$, J = 7 Hz); 7.75 (m, 2 H, H arom.); 8.50 (s, 1 H, CH); 8.65 (m, 2 H, H arom.). MS, m/z: 298 $[M]^+$.

3-(Dicyanomethylidene)-2-(dimethylaminomethylidene)indan-1-one (16). A mixture of compound **14** (0.2 g, 0.8 mmol) and a solution of dimethylamine (0.8 mL) in benzene (1 : 1) was stirred in a mixture of dry benzene (12 mL) and dry acetonitrile (2 mL) for 1 h. The precipitate was separated, washed with anhydrous ether, and recrystallized from EtOH to give product **16** (0.16 g, 80%) as a light brown powder, m.p. 186—188 °C. Found (%): C, 71.98; H, 4.40; N, 16.55. $C_{15}H_{11}N_3O$. Calculated (%): C, 72.29; H, 4.41; N, 16.87. UV (EtOH), λ_{max}/nm (ε): 275 (16750), 300 sh (15845), 315 (16750), 370 sh (9054), 450 (8148). UV ($CHCl_3$), λ_{max}/nm (ε): 250 sh (10197), 285 (19920), 302 (18023), 320 (19446), 370 sh (9960), 450 (8300). ^{1}H NMR ($CDCl_3$), δ: 3.50, 3.60 (both s, 3 H each, NMe_2); 7.60—7.70 (m, 3 H, H arom.); 8.35 (s, 1 H, CH); 8.55 (m, 1 H, H arom.). MS, m/z: 249 $[M]^+$.

1,3-Bis(dicyanomethylidene)-2-(hydroxymethylidene)indane (17). A solution of compound **15** (0.5 g) in 96% EtOH (25 mL) was refluxed for 1 h. On cooling, product **17** (0.3 g) was separated, m.p. >250 °C. Found (%): C, 70.95; H, 2.46; N, 20.44. $C_{16}H_6N_4O$. Calculated (%): C, 71.11; H, 2.22; N, 20.74. UV ($CHCl_3$), λ_{max}/nm (ε): 294 (5245), 350 sh (4320), 400 sh (3471). ^{1}H NMR (DMSO-d_6), δ: 7.80—8.40 (m, 4 H, H arom.); 8.50 (s, 1 H, CH); 13.00 (br.s, 1 H, OH). MS, m/z: 270 $[M]^+$.

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