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# ALTERNATIVE REACTIVITY OF BENZIMIDAZOLIUM YLIDS

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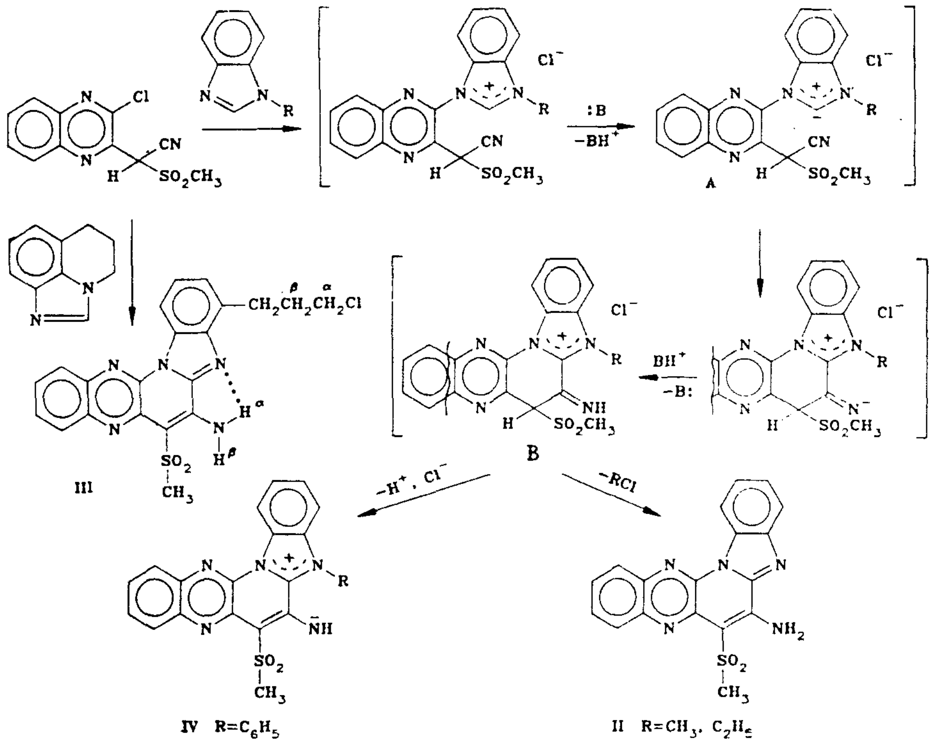
The reaction of α-sulfonyl[2-(3-chloro)quinoxalylacetonitrile (I) with benzimidazole leads ultimately to derivatives of 6-amino-7-sulfonylbenzimidazo[1',2':1,6]pyrido[2,3-b]quinoxaline [1]. A similar reaction with 1-substituted benzimidazoles has so far not been studied. We have found that the course of that reaction is determined by the nature of the substituent on the nitrogen of the benzimidazole nucleus.

In our opinion, the reaction proceeds by the following scheme:



Quaternization of the benzimidazole nucleus facilitates deprotonation of position 2 to a salt by the action of excess benzimidazole in the reaction medium to form ylid (A). Nucleophilic attack by the nitrile carbanion gives the cyclic intermediate B, the subsequent fate of which depends on the nature of the substituent on nitrogen. In the case of 1-alkylbenzimidazoles

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(where R = $CH_3$, $C_2H_5$) the benzimidazole ring undergoes dealkylation. The resulting compound (II) is in all respects analogous to 6-amino-7-methylsulfonylbenzimidazo[1',2':1,6]pyrido[2,3-b]quinoxaline described in [1]. Here the alkyl group is eliminated as the corresponding alkyl chloride. This is unequivocally demonstrated by the formation of (III) by the reaction of chloro derivatives of (I) with 5,6-dihydroimidazo[i,j]quinoline.

With 1-phenylbenzimidazole there is formed the cyclic mesomeric betaine structure (IV) (cf. similar compounds in [2]). The retention of the phenyl substituent in the molecule, just as the detachment of the alkyl, corresponds to the thermal stability of the quaternary azolium salts [3].

Compounds (II-IV) were synthesized by boiling $\alpha$-methylsulfonyl[2-(3-chloro)quinoxalyl]acetonitrile (I) with a twofold excess of the respective 1-substituted benzimidazole in c.p. dimethylformamide.

**6-Amino-7-methylsulfonylbenzimidazo[1',2':1,6]pyrido-[2,3-b]quinoxaline (II).** Mp >300°C. Yield 84% (R = $CH_3$) and 67% (R = $C_2H_5$). ??

**4-(1-Chloro ???** Mp 229°C. PMR spectrum ($CDCl_3$, TMS): 2.35 (m, $\beta$-$CH_2$), 3.33 (t, $\gamma$-$CH_2$), 3.63 (t, $\alpha$-$CH_2$), 3.74 (s, $CH_3$), 6.99 (s, NH), 7.4-7.7 (m, 2- and 3-H), 7.74-7.88 (m, 9- and 12-H), 8.13-8.27 (m, 10- and 11-H), 8.47 (s, NH), 9.01 ppm (d.d, J = 8.1 and 1.1 Hz, 1-H). Yield 61%.

**5-Phenyl-7-methylsulfonylbenzimidazolium[1',2';1,6]-pyrido[2,3-b]quinoxaline-6-imidate (IV).** Mp >30-0°. PMR spectrum ($CF_3COOD$, TMS): 3.72 (s, $CH_3$), 7.59 (d.d, J = 7.8 and 1.3 Hz, 4-H), 7.94-8.78 (m, 2-, 3-H, $C_6H_5$, 9-, 10-, 11-, 12H), 9.83 ppm (d.d, J = 8.1 and 1.2 Hz, 1-H). Yield 78%.

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# NEW INDOLINE SPIROPYRANS WITH $\pi$-ACCEPTOR SUBSTITUENTS IN THE 8' POSITION

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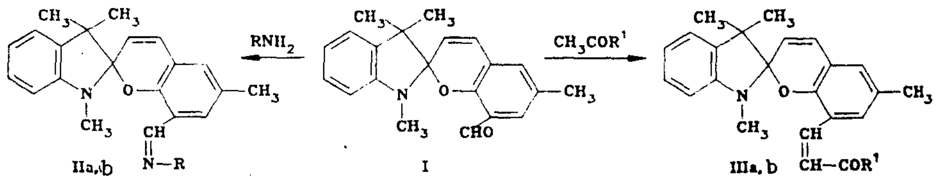
541.14'651

An important factor that determines the use of spiropyrans is the presence of the long-wave absorption of the photoinduced form, traditionally obtained by introducing a nitro group into the molecule [1]. It was of interest to use other $\pi$-acceptor substituents together with a lengthened photomeric cyanine conjugation chain.



II a R=4-$CH_3OC_6H_4$, b R=$CH_2Ph$; III a R'=$CH_3$, b R'=Ph

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