2-Alkylimino-4-(2-Hydroxyaryl)-1,3-Dithiols Derivatives

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Novel 2-alkylimino-4-(2-hydroxyaryl)-1,3-dithiols have been synthesized by the reaction of the corresponding 4-(2-hydroxyaryl)-2-(N,N-dialkylamino)-1,3-dithiol-2-ylium salts with primary alkyl amines. The imines were obtained as a 1:1 mixture of syn/anti isomers. These derivatives were characterized by NMR and MS spectrometry, UV-Vis and IR spectroscopy.

Keywords: amines, 1,3-dithiolium salts, dithiols, imines

The development of new organic superconductors still remains a major topic in the field of molecular conductors [1]. The discovery of the metallic conductivity in a tetrathiafulvalene-tetracyanoquinodimethane complex [2] has stimulated a great deal of work on the synthesis of a wide variety of tetrathiafulvalene (TTF) analogs [3, 4]. For many years, all the organic *ð*-electron donors with conductivity properties were limited to the tetrachalcogenafulvalenes compounds [5, 6]. Recently, the non-tetrachalcogenafulvalenes containing a 1,3-dithiol-2-ylidene based *ð*-donor unity proved to give superconducting salts [7]. For these reasons this type of compounds represent an important resource for the material chemistry with deep implications in the daily life and even in the educational process [8, 9]. Recent reports highlighted the TTFs ability to act as donor groups in intramolecular charge-transfer complexes [10, 11]. Thus, various acceptor units have been investigated, nitrogen and sulfur containing cations receiving a great deal of attention [12-22]. Of special interest for conducting materials are charge-transfer [23-30] and push-pull [31-45] compounds. Recent studies on (1,3-dithiolium-2-yl)phenolates systems revealed that 1,3-dithiolium cations can act as acceptor groups in intramolecular charge-transfer processes [26,30,46,47]. Moreover, this type of compounds has been found to exhibit biological activity, in a particular case, against gram-positive and gram-negative bacteria [48-58]. Dithiolium systems are known for their reactivity at the C(2) - position towards nucleophiles [28].

These considerations indicated that introduction of an imino group at the 2 - position of 1,3-dithiolim ring will be of interest since the *syn-anti* isomerism of conjugated imines is a weakly energetic process and consequently the 2-imino-1,3-dithiol derivatives will adopt the best geometry to fit the strongest stabilizing interaction [59]. We are reporting here the synthesis of novel 2-alkylimino-4-(2-hydroxyaryl)-1,3-dithiols by the reaction of the corresponding 1,3-dithiolium salts with aliphatic primary amines.

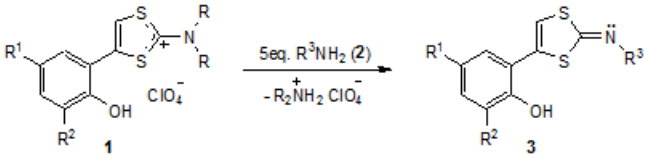
Experimental part

Analysis methods

Melting points were obtained on a *KSPI* melting-point meter. IR spectra were recorded on a Bruker Tensor 27 instrument. NMR spectra were recorded on a Bruker 500 MHz spectrometer. Chemical shifts are reported in ppm downfield from TMS. UV-Vis spectra were recorded on a Varian BioChem 100 spectrophotometer. Mass spectra were recorded on a Thermo Scientific ISQ LT instrument. Elemental analyses (C, H, N and S) were conducted using a CE440 Elemental Analyser; the results were found to be in good agreement (±0.24%) with the calculated values.

Synthesis

The synthetic pathway for the synthesis of 2-alkylimino-1,3-dithiols **3a-d** is described in scheme 1:



1, 3	R^1	R^2	R^3
a	Br	H	$-CH_2CH_2CH_3$
b	CH_3	Br	$-CH_2CH_2CH_3$
c	Br	H	$-CH_2CH(CH_3)CH_3$
d	CH_3	H	$-CH_2CH(CH_3)CH_3$

Scheme 1. Synthesis of 2-alkylimino-1,3-dithiols **3a-d**

4-(5-Bromo-2-hydroxyphenyl)-2-i-butylimino-1,3-dithiol (**3c**)

General Procedure

To a suspension of 4-(5-bromo-2-hydroxyphenyl)-2-(*N*-piperidinyl)-1,3-dithiol-2-ylium perchlorate (**1c**, 1g, 2.2mmol) in ethanol (20mL) *i*-butylamine (1mL, 10mmol) was added. The reaction mixture that became yellow was stirred for 3h at room temperature until a clear homogenous solution was obtained. This was poured into water and acidified with hydrochloric acid to *p*H=1. The resulting precipitate was filtered, washed with water and dried off. Recrystallization from *i*-PrOH (50mL) gave pale yellow crystals. Yield 0.42g (56%); mp 239-240 dec. ^{1}H NMR (DMSO-d_6, selected data for one isomer): δ = 1.02 (6H, m, $2CH_3$), 2.10 (1H, m, CH), 3.32 (2H, m, CH_2), 7.06 ($1H_{ar}$, d, H-3, ^3J=8.7Hz), 7.44 ($1H_{ar}$, m, H-4), 7.71 ($1H_{ar}$, m, H-6), 7.93 (1H, s, H-5), 11.51 (1H, s, OH) ppm. ^{13}C NMR (DMSO-d_6, selected data for one isomer): δ = 20.5, 27.6, 59.8, 111.2, 118.0, 119.1, 119.8, 121.1, 130.7, 130.9, 133.8, 153.7 ppm. FT-IR (ATR): ν = 3334, 2717, 1557, 1405, 1285, 809, 775, 619, 575, 459 cm^{-1}. MS (EI): *m/z* = 343 (28%, M^+ for $C_{13}H_{14}{}^{79}BrNOS_2$), 300 (100%).

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4-(5-Bromo-2-hydroxyphenyl)-2-n-propylimino-1,3-dithiol (**3a**)

Pale yellow solid; 0.4g (58%); mp 215-216 dec. ^{1}H NMR (DMSO-d_6, selected data for one isomer): δ = 0.99 (3H, t, CH_3), 1.76 (2H, m, CH_2), 3.44 (2H, m, CH_2), 7.05 (1H$_{ar}$, d, H-3, ^3J=8.8Hz), 7.44 (1H$_{ar}$, m, H-4), 7.73 (1H$_{ar}$, m, H-6), 7.93 (1H, s, H-5), 11.52 (1H, s, OH) ppm. ^{13}C NMR (DMSO-d_6, selected data for one isomer): δ = 11.8, 21.0, 54.2, 111.3, 118.2, 119.0, 120.0, 120.8, 130.8, 131.1, 133.8, 153.7 ppm. FT-IR (ATR): ν = 3334, 2699, 1559, 1410, 1288, 1258, 809, 574, 464 cm^{-1}. MS (EI): *m/z* = 329 (48%, M$^+$ for $C_{12}H_{12}{}^{79}BrNOS_2$), 181 (100%).

4-(3-Bromo-2-hydroxy-5-methylphenyl)-2-n-propylimino-1,3-dithiol (**3b**)

Pale yellow solid; 0.48g (52%); mp 192-193 dec. ^{1}H NMR (DMSO-d_6, selected data for one isomer): δ = 0.99 (3H, t, CH_3), 1.76 (2H, m, CH_2), 2.26 (3H, s, CH_3), 3.43 (2H, m, CH_2), 7.41 (1H$_{ar}$, m, H-4), 7.48 (1H$_{ar}$, m, H-6), 7.82 (1H, s, H-5), 10.27 (1H, s, OH) ppm. ^{13}C NMR (DMSO-d_6, selected data for one isomer): δ = 11.9, 20.0, 21.1, 54.3, 113.4, 118.4, 121.1, 121.6, 129.0, 131.9, 132.1, 135.0, 148.3 ppm. FT-IR (ATR): ν= 3333, 2962, 2761, 1561, 1431, 1266, 1198, 849, 754, 709, 663 cm^{-1}. MS (EI): *m/z* = 343 (25%, M$^+$ for $C_{13}H_{14}{}^{79}BrNOS_2$), 195 (100%).

4-(2-Hydroxy-5-methylphenyl)-2-i-butylimino-1,3-dithiol (**3d**)

Pale yellow solid; 0.3g (51%); mp 132-133. ^{1}H NMR (DMSO-d_6, selected data for one isomer): δ = 1.01 (6H, m, 2CH_3), 2.12 (1H, m, CH), 2.24 (3H, s, CH_3), 3.31 (2H, m, CH_2), 6.97 (1H$_{ar}$, d, H-3, ^3J=8.6Hz), 7.21 (1H$_{ar}$, m, H-4), 7.48 (1H$_{ar}$, m, H-6), 7.90 (1H, s, H-5), 11.42 (1H, s, OH) ppm. ^{13}C NMR (DMSO-d_6, selected data for one isomer): δ = 19.8, 20.4, 27.5, 59.6, 118.2, 120.8, 122.1, 128.8, 129.2, 130.6, 130.8, 136.4, 151.2 ppm. FT-IR (ATR): ν = 3328, 2701, 1559, 1421, 1280, 819, 737 cm^{-1}. MS (EI): *m/z* = 279 (45%, M$^+$ for $C_{14}H_{17}NOS_2$), 235 (100%).

Results and discussions

The synthesis of 2-arylimino-1,3-dithioles has been previously accomplished by refluxing the 1,3-dithiolium cation with aromatic amines [60]. Following a modified experimental procedure, 1,3-dithiol-2-imines **3a-d** have been synthesized by the reaction of primary amines with 1,3-dithiolium perchlorates **1a-d** in ethanol at room temperature. A convenient method for the synthesis of 2-(*N,N*-dialkylamino)-1,3-dithiol-2-ylium salts of type **1** is represented by the cyclization of the corresponding *N,N*-dialkylamino carbodithioates [61, 62]. The reactions of *a*-bromophenones with salts of dithiocarbamic acid, readily available from the reaction of secondary amine with carbon disulfide [63], represent an accessible way to various substituted phenacyl carbodithioates.

1,3-Dithiol-2-imines **3a-d** have been obtained as pale yellow solids in moderate yields (51-58%). The structure of imines **3** has been proved by analytical and spectral data. The ^{1}H NMR spectra indicate the disappearance of the signals corresponding to the hydrogen atoms from the secondary amine moiety. Also, new signals are recorded in the aliphatic area corresponding to the new *n*-propyl and *i*-butyl substituents.^{13}C NMR spectra indicate the disappearance of the characteristic signal of positively charged C-2 atom in 1,3-dithiolium cation and of secondary amine carbon atoms connected at this position. Both NMR spectra indicated the presence of a mixture of two isomers in a 1:1 ratio. This is the result of relative *syn* or *anti* orientation of alkyl substituent at the nitrogen atom towards the aryl substituent at the 4-position of 1,3-dithiol ring. The IR spectra revealed the disappearance of the broad absorption band corresponding to the perchlorate anion (ca. 1050cm^{-1}) and the presence of new, strong absorption bands at 1405-1431cm^{-1}, corresponding to the imino bond. The mass spectra also confirm the formation of 2-alkylimines **3** providing the following molecular ions (*M*$^+$, a.m.u.): 329 ($C_{12}H_{12}{}^{79}BrNOS_2$) for **3a**, 343 ($C_{13}H_{14}{}^{79}BrNOS_2$) for **3b**, 343 ($C_{13}H_{14}{}^{79}BrNOS_2$) for **3c** and 279 ($C_{14}H_{17}NOS_2$) for **3d**.

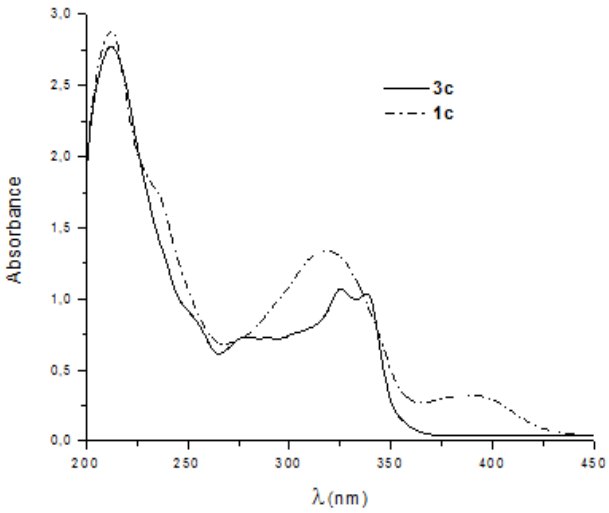
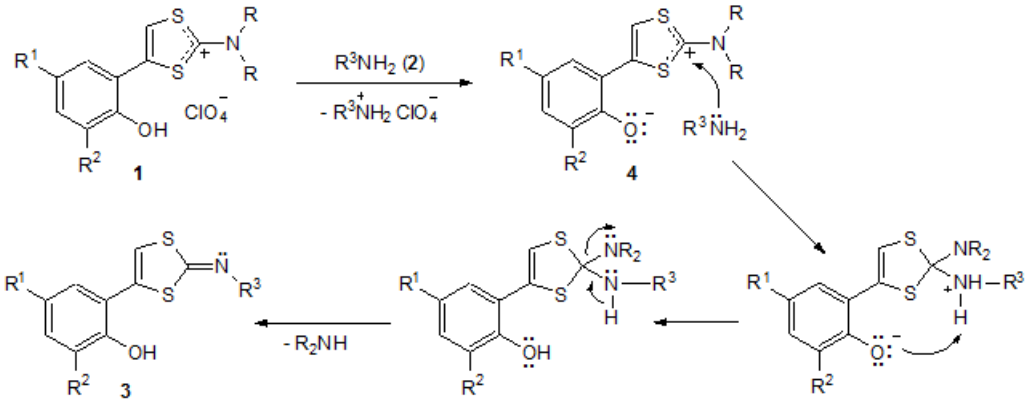


Fig. 1. UV-Vis absorption spectra of 4-(5-bromo-2-hydroxyphenyl)-2-(*N*-piperidinyl)-1,3-dithiol-2-ylium perchlorate (**1c**) and 4-(5-bromo-2-hydroxyphenyl)-2-*i*-butylimino-1,3-dithiol (**3c**) in ethanol

The presence of both *syn* and *anti* isomers has been also confirmed by UV-Vis spectroscopy.

The comparative analysis of the UV-Vis absorption spectra of 4-(5-bromo-2-hydroxyphenyl)-5-methyl-2-(*N*-piperidinyl)-1,3-dithiol-2-ylium perchlorate (**1c**) and 4-(5-bromo-2-hydroxyphenyl)-2-*i*-butylimino-1,3-dithiol **(3c)** (fig. 1) indicate the disappearance of the n →π* transition band from 318 nm in **1c** and a bathochromic shift of π →



Scheme 2. A tentative mechanism for the formation of 2-imino-1,3-dithiols **3**

π^* transition band from 238 nm to 255 nm in imine **3c**. The most interesting feature of the UV-Vis spectrum of **3c** is represented by the two $n \rightarrow \pi^*$ absorption bands of imino group at 325 and 340 nm. These belong to the *syn* and *anti* isomers of imine. According with the literature data the longest wavelength correspond to the *syn* isomer [64].

A mechanistic rationalization of 2-imino-1,3-ditiols **3** formation is presented in Scheme 2. It has been previously reported [27] that under weak basic conditions 1,3-dithiolium salts of type **1** are converted to the corresponding mesoionic phenolates of type **4**. By using an excess of primary amine the first step of this reactions should be the elimination of the ammonium perchlorate and formation of mesoionic compounds that undergo nucleophilic addition of primary amine to the electron deficient C-2 carbon atom. An intramolecular hydrogen transfer followed by secondary amine elimination provides 2-alkylimino-1,3-dithiol derivatives **3**.

Conclusions

The synthesis of several of 2-alkylimino-4-(2-hydroxyaryl)-1,3-dithiol derivatives has been accomplished by the reaction of the corresponding 1,3-dithiol-2-ylium perchlorates with *n*-propylamine and *i*-butylamine. 2-Alkylimino-1,3-dithiols have been obtained as a mixture of *syn* and *anti* isomers through an addition-elimination reaction mechanism. The structure of new synthesized compounds has been proved by analytical and spectral (NMR, IR, UV-Vis, MS) data.

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