ISSN 1070-4280, Russian Journal of Organic Chemistry, 2014, Vol. 50, No. 7, pp. 973–985. © Pleiades Publishing, Ltd., 2014.
Original Russian Text © S.A. Konovalova, A.P. Avdeenko, I.L. Marchenko, 2014, published in Zhurnal Organicheskoi Khimii, 2014, Vol. 50, No. 7, pp. 991–1003.

# Reaction of Some N-Substituted 1,4-Benzoquinone Imines with Sodium Arenesulfinates

**S. A. Konovalova, A. P. Avdeenko, and I. L. Marchenko**

*Donbass State Engineering Academy, ul. Shkadinova 72, Kramatorsk, 84313 Ukraine*
*e-mail: chimist@dgma.donetsk.ua*

Received December 4, 2013

**Abstract**—*N*-Arylsulfonyl, *N*-aroyl, and *N*-[arylsulfonylimino(phenyl)methyl] derivatives of 1,4-benzoquinone imine reacted with sodium arenesulfinates to give 1,4-, 1,6-, and 6,1-addition products which were formed according to two concurrent paths: direct nucleophilic addition of arenesulfinate anion to neutral quinone imine molecule and radical ion addition of arenesulfinate radical to radical anion derived from quinone imine.

**DOI:** 10.1134/S1070428014070082

We previously studied in detail reactions of *N*-aryl-2,6-dialkyl [1], *N*-aroyl, *N*-arylsulfonyl, *N*-[arylsulfonylimino(phenyl)methyl]-2,6(3,5, or 2,5)-dialkyl [2–5], and *N*-aryl(alkyl, trifluoromethyl)sulfonyl [6] derivatives of 1,4-benzoquinone imine with sodium arenesulfinates. Depending on the initial quinone imine structure, 1,4-, 6,3-, 1,6-, or 6,1-addition products were obtained. It was found that the reaction direction is determined not only by the nature of substituent on the nitrogen atom of quinone imine but also by the nature and position of substituents in the quinoid ring [4, 5]. 1,4-Benzoquinone imines having two alkyl substituents in the quinoid ring were mainly studied. We expected that reduction of the number of substituents in the quinoid ring should favor more accurate estimation of the effect of the N-substituent on the reaction direction. Therefore, the goal of the present work was to compare the reactivities of *N*-aroyl, *N*-arylsulfonyl, and *N*-[arylsulfonylimino-(phenyl)methyl] 1,4-benzoquinone imine derivatives containing no substituents in the quinoid ring or substituted at the 2- and/or 3-position toward sodium arenesulfinates.

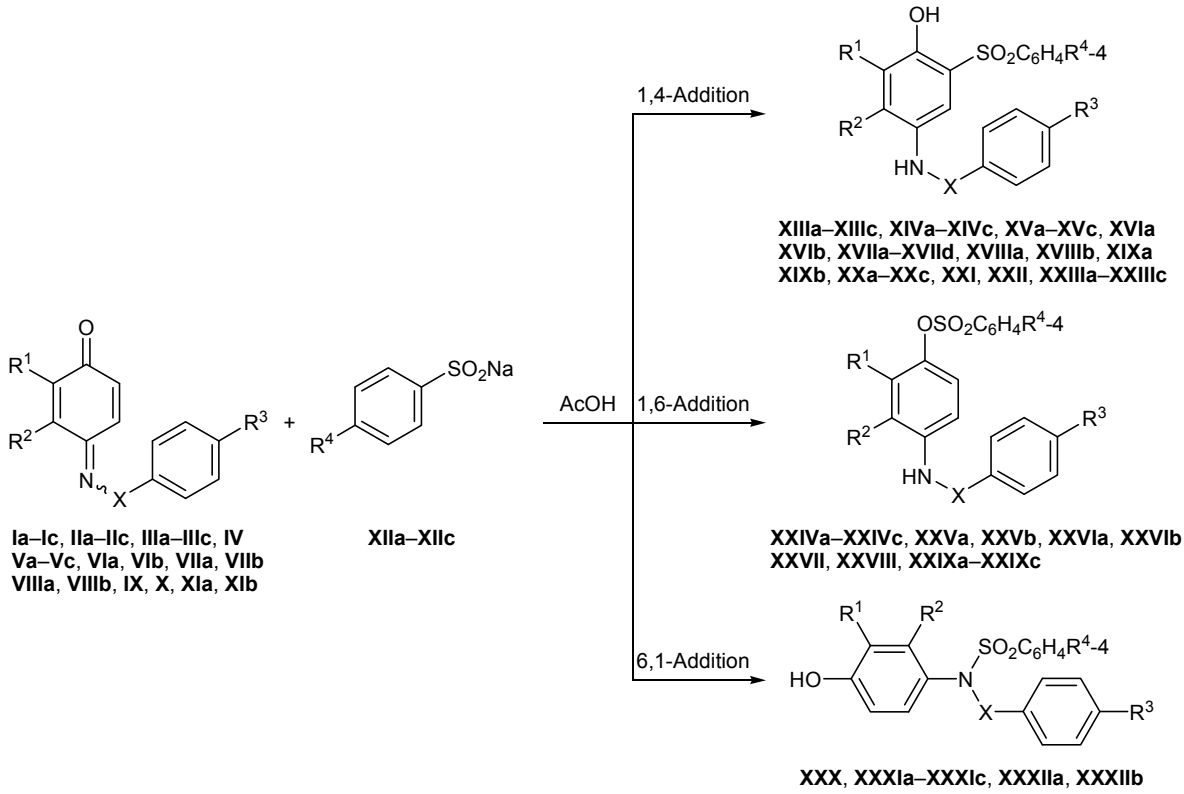
The reactions were carried out using excess sodium arenesulfinate (1:2) in acetic acid. The aryl groups in the quinone imine and arenesulfinate components were mainly *p*-methylphenyl and *p*-methoxyphenyl to facilitate signal assignment in the $^1$H NMR spectra. In order to detect all products, we analyzed by $^1$H NMR the precipitate before and after recrystallization and the product isolated from the mother liquor obtained after recrystallization. We thus succeeded in separating isomeric products and detecting compounds whose amount did not exceed a few percent.

The results of the reactions of *N*-arylsulfonyl (**Ia**–**Ic**, **IIa**–**IIc**, **IIIa**–**IIIc**, **IV**), *N*-aroyl (**Va**–**Vc**, **VIa**, **VIb**, **VIIa**, **VIIb**), and *N*-[arylsulfonylimino(phenyl)-methyl]-1,4-benzoquinone imines (**VIIIa**, **VIIIb**, **IX**, **X**, **XIa**, **XIb**) with sodium arenesulfinates **XIIa**–**XIIc** are shown in Scheme 1, and the product compositions (excluding reduced forms of quinone imines) are given in Table 1.

Compounds **Ia**–**Ic**, **IIa**–**IIc**, **IIIa**–**IIIc**, and **IV** reacted mostly according to the 1,4-addition pattern, yielding amides **XIIIa**–**XIIIc**, **XIVa**–**XIVc**, **XVa**–**XVc**, **XVIa**, and **XVIb**. Only the reaction of quinone imine **IV** having no substituents in the quinoid ring with sodium *p*-methoxybenzenesulfinate gave a mixture of 1,4- and 6,1-addition products **XVIb** and **XXX**.

The reactions of alkyl-substituted *N*-aroyl derivatives **Va**–**Vc**, **VIa**, **VIb**, **VIIa**, and **VIIb** with sodium arenesulfinates **XIIa**–**XIIc** led to the formation of mixtures of 1,4- (**XVIIa**–**XVIId**, **XVIIIa**, **XVIIIb**, **XIXa**, **XIXb**), 1,6- (**XXIVa**–**XXIVc**, **XXVa**, **XXVb**, **XXVIa**, **XXVIb**), and 6,1-addition products (**XXXIa**–**XXXIc**, **XXXIIa**, **XXXIIb**) (Table 1). Completely regioselective 1,4-addition was observed only in the reaction of **Vb** with sodium *p*-chlorobenzenesulfinate (**XIIc**). 1,6-Addition products **XXVIa** and **XXVIb** were formed as the major ones only from quinone imines **VIIa** and **VIIb**. In all other cases, the corresponding 1,4-addition products predominated.

**Scheme 1.**




**I**, $R^1 = R^2 = Me$, $X = SO_2$, $R^3 = Me$ (**a**), MeO (**b**), Cl (**c**); **II**, $R^1 = Me$, $R^2 = H$, $X = SO_2$, $R^3 = Me$ (**a**), MeO (**b**), Cl (**c**); **III**, $R^1 = H$, $R^2 = Me$, $X = SO_2$, $R^3 = Me$ (**a**), MeO (**b**), Cl (**c**); **IV**, $R^1 = R^2 = H$, $R^3 = Me$, $X = SO_2$; **V**, $R^1 = R^2 = Me$, $X = CO$, $R^3 = Me$ (**a**), MeO (**b**), Cl (**c**); **VI**, $R^1 = Me$, $R^2 = H$, $X = CO$, $R^3 = MeO$ (**a**), Br (**b**); **VII**, $R^1 = H$, $R^2 = Me$, $X = CO$, $R^3 = Me$ (**a**), Cl (**b**); **VIII**, $R^1 = R^2 = Me$, $R^3 = H$, $X = C{=}NSO_2Ar$, Ar = 4-$MeC_6H_4$ (**a**), 4-$MeOC_6H_4$ (**b**); **IX**, $R^1 = Me$, $R^2 = R^3 = H$, $X = C{=}NSO_2C_6H_4Me$-4; X, $R^1 = R^3 = H$, $R^2 = Me$, $X = C{=}NSO_2C_6H_4Me$-4; **XI**, $R^1 = R^2 = R^3 = H$, $X = C{=}NSO_2Ar$, Ar = 4-$MeC_6H_4$ (**a**), 4-$ClC_6H_4$ (**b**); **XII**, $R^4 = Me$ (**a**), MeO (**b**), Cl (**c**); **XIII**, $R^1 = R^2 = Me$, $X = SO_2$, $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Cl$ (**b**); $R^3 = Cl$, $R^4 = MeO$ (**c**); **XIV**, $R^1 = Me$, $R^2 = H$, $X = SO_2$: $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Cl$ (**b**); $R^3 = Cl$, $R^4 = MeO$ (**c**); **XV**, $R^1 = H$, $R^2 = Me$, $X = SO_2$: $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Cl$ (**b**); $R^3 = Cl$, $R^4 = MeO$ (**c**); **XVI**, $R^1 = R^2 = H$, $X = SO_2$; $R^3 = R^4 = Me$ (**a**); $R^3 = Me$, $R^4 = MeO$ (**b**); **XVII**, $R^1 = R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Me$ (**b**); $R^3 = MeO$, $R^4 = Cl$ (**c**); $R^3 = Cl$, $R^4 = MeO$ (**d**); **XVIII**, $R^1 = Me$, $R^2 = H$, $X = CO$; $R^3 = MeO$, $R^4 = Me$ (**a**); $R^3 = Br$, $R^4 = MeO$ (**b**); **XIX**, $R^1 = H$, $R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**), $R^3 = Cl$, $R^4 = Me$ (**b**); **XX**, $R^1 = R^2 = Me$, $R^3 = H$, $X = C{=}NSO_2Ar$; Ar = 4-$MeC_6H_4$, $R^4 = MeO$ (**a**); Ar = 4-$MeOC_6H_4$, $R^4 = Me$ (**b**); Ar = 4-$MeOC_6H_4$, $R^4 = MeO$ (**c**); **XXI**, $R^1 = Me$, $R^2 = R^3 = H$, $R^4 = MeO$, $X = C{=}NSO_2C_6H_4Me$-4; **XXII**, $R^1 = R^3 = H$, $R^2 = Me$, $R^4 = MeO$, $X = C{=}NSO_2C_6H_4Me$-4; **XXIII**, $R^1 = R^2 = H$, $R^3 = H$, $X = C{=}NSO_2Ar$; Ar = 4-$MeC_6H_4$, $R^4 = MeO$ (**a**); Ar = 4-$ClC_6H_4$, $R^4 = Me$ (**b**); Ar = 4-$ClC_6H_4$, $R^4 = MeO$ (**c**); **XXIV**, $R^1 = R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Me$ (**b**); $R^3 = Cl$, $R^4 = MeO$ (**c**); **XXV**, $R^1 = Me$, $R^2 = H$, $X = CO$; $R^3 = MeO$, $R^4 = Me$ (**a**), $R^3 = Br$, $R^4 = MeO$ (**b**); **XXVI**, $R^1 = H$, $R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = Cl$, $R^4 = Me$ (**b**); **XXVII**, $R^1 = Me$, $R^2 = H$, $R^3 = H$, $R^4 = MeO$, $X = C{=}NSO_2Ar$, Ar = 4-$MeC_6H_4$; **XXVIII**, $R^1 = R^3 = H$, $R^2 = Me$, $R^4 = MeO$, $X = C{=}NSO_2C_6H_4Me$-4; **XXIX**, $R^1 = R^2 = R^3 = H$, $X = C{=}NSO_2Ar$: Ar = 4-$MeC_6H_4$, $R^4 = MeO$ (**a**); Ar = 4-$ClC_6H_4$, $R^4 = Me$ (**b**); Ar = 4-$ClC_6H_4$, $R^4 = MeO$ (**c**); **XXX**, $R^1 = R^2 = H$, $R^3 = Me$, $R^4 = MeO$, $X = SO_2$; **XXXI**, $R^1 = R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = MeO$, $R^4 = Me$ (**b**); $R^3 = Cl$, $R^4 = MeO$ (**c**); **XXXII**, $R^1 = H$, $R^2 = Me$, $X = CO$; $R^3 = Me$, $R^4 = MeO$ (**a**); $R^3 = Cl$, $R^4 = Me$ (**b**).

Signals in the $^1$H NMR spectra of the addition products were assigned taking into account our previous data [4–6]. In the $^1$H NMR spectra of 1,4-addition products **XIIIa**–**XIIIc**, the 6-H proton in the aminophenol fragment resonated as a singlet at δ 7.08–7.18 ppm, the 2-H and 6-H protons of **XIVa**–**XIVc** gave doublets at δ 7.13–7.22 and 7.38–7.45, and the 3-H and 6-H signals of **XVa**–**XVc** were doublets in the regions δ 6.68–6.80 and 7.22–7.46 ppm. The spectra of **XVIa** and **XVIb** characteristically showed a doublet ($^4J$) from 2-H at δ 7.66–7.69 ppm, a doublet of doublets from 6-H at δ 7.29–7.34 ppm, and a doublet ($^3J$) from 5-H at δ 6.89 ppm.

The signal from 6-H in the aminophenol fragment of **XVIIa**–**XVIId** was a singlet at δ 7.67–7.72 ppm, and compounds **XIXa** and **XIXb** displayed two sin-

glets from 3-H and 6-H at δ 6.78–6.80 and 7.82–7.83 ppm, respectively, which unambiguously confirmed their structure as 1,4-addition products. Mixtures of 1,6- and 6,1-addition products **XXVIa**/**XXXIIa** and **XXVIb**/**XXXIIb** showed in the spectra two sets of signals from three protons in the aminophenol fragment, which were assigned on the basis of the data in [4–6]. In the $^1$H NMR spectra of 1,6-addition products, signals from protons in the aminophenol fragment appeared in a weaker field relative to the corresponding signals of 6,1-adducts. The spectra of **XXVIa** and **XXVIb** contained a singlet from the NH proton (δ 9.83 and 9.99 ppm, respectively), two doublets from 2-H and 5-H (δ 7.00–7.03 and 7.56–7.58 ppm), and a doublet of doublets from 6-H (δ 6.79–6.85 ppm). The signals belonging to **XXXIIa** and **XXXIIb** were observed in a stronger field: OH, δ 9.37–9.58 ppm, s; 3-H, δ 6.53–6.54 ppm, d; 6-H, δ 7.10–7.11 ppm, d; 6-H, δ 6.55–6.58 ppm, d.d; these data characterize compounds **XXXIIa** and **XXXIIb** as 6,1-addition products.

Thus, as might be expected, the direction of the addition of sodium arenesulfinates changes in going from *N*-arylsulfonyl (X = $SO_2$) to *N*-aroyl derivatives (X = CO): 1,4-addition path predominates for the former, while *N*-aroyl derivatives **Va**–**Vc**, **VIa**, **VIb**, **VIIa**, and **VIIb** react along three possible pathways, 1,4-, 1,6-, and 6,1-addition, due to variation of the substituent on the nitrogen atom (Table 1).

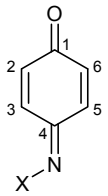
According to the results of quantum chemical calculations, ArCO and $ArSO_2$ groups differently affect the double bonds in the quinoid ring. Their effect can be estimated by the energies of donor–acceptor interactions in the N=C–C=C–C=O bond sequence, i.e., the following interactions: $E_1$ = π($C^1$=O)→π*($C^2$=$C^3$) + π($C^2$=$C^3$)→π*($C^4$=N) and $E_2$ = π($C^4$=N)→π*($C^2$=$C^3$) + π($C^2$=$C^3$)→π*($C^1$=O). These interactions are fairly strong, and they are responsible for the displacement of electron density toward the nitrogen ($E_1$) or oxygen atom ($E_2$), which determines overall displacement of electron density in the quinone imine molecule (Δ$E$; Table 2). The calculations showed that the electron density in the *N*-benzoyl derivative (X = PhCO) is displaced toward the oxygen atom, and in the *N*-phenylsulfonyl derivative (X = $PhSO_2$), in the opposite direction, i.e., to the nitrogen atom. As a result, polarization of the quinone molecule strongly changes, which is reflected in the charges on the nitrogen and oxygen atoms (Table 2). Thus, the predominant formation of 1,4-addition products from *N*-arylsulfonyl derivatives is related to displacement of electron density in the quinoid ring to the nitrogen atom and increase of the electrophilicity of $C^2$ and $C^6$.

**Table 1.** Product ratios in the reactions of 1,4-benzoquinone imines **I**–**XI** with sodium arenesulfinates **XII** (see Scheme 1)

| Quinone imine no. | Sodium arenesulfinate no. | Fraction of addition products, % | | |
|---|---|---|---|---|
| | | 1,4 | 1,6 | 6,1 |
| **Ia** | **XIIb** | 100 | – | – |
| **Ib** | **XIIc** | 100 | – | – |
| **Ic** | **XIIb** | 100 | – | – |
| **Va** | **XIIb** | 77 | 15 | 8 |
| **Vb** | **XIIa** | 80 | 13 | 7 |
| **Vb** | **XIIc** | 100 | – | – |
| **Vc** | **XIIb** | 64 | 25 | 11 |
| **VIIIa** | **XIIb** | 100 | – | – |
| **VIIIb** | **XIIa** | 100 | – | – |
| **VIIIb** | **XIIb** | 100 | – | – |
| **IIa** | **XIIb** | 100 | – | – |
| **IIb** | **XIIc** | 100 | – | – |
| **IIc** | **XIIb** | 100 | – | – |
| **VIa** | **XIIa** | 86 | 14 | – |
| **VIb** | **XIIb** | 79 | 21 | – |
| **IX** | **XIIb** | 58 | 42 | – |
| **IIIa** | **XIIb** | 100 | – | – |
| **IIIb** | **XIIc** | 100 | – | – |
| **IIIc** | **XIIb** | 100 | – | – |
| **VIIa** | **XIIb** | 40 | 45 | 15 |
| **VIIb** | **XIIa** | 32 | 52 | 16 |
| **X** | **XIIb** | 60 | 40 | – |
| **IV** | **XIIa** | 100 | – | – |
| **IV** | **XIIb** | 91 | – | 09 |
| **XIa** | **XIIb** | 52 | 48 | – |
| **XIb** | **XIIa** | 55 | 45 | – |
| **XIb** | **XIIb** | 56 | 44 | – |

The reactions with sodium arenesulfinates were carried out in acetic acid; therefore, the reacting species should be arenesulfinate anion rather than free arenesulfinic acid since the latter is a stronger acid than acetic (p$K_a$ 1.5 and 4.75 for benzenesulfinic and acetic acids, respectively) [7]. We believe that the most probable mechanism of the reaction of *N*-arylsulfonyl-1,4-benzoquinone imines **I**–**IV** with sodium arenesulfinates is direct nucleophilic attack by arenesulfinate anion on neutral quinone imine molecule.

**Table 2.** Energies of donor–acceptor interactions and charges on the oxygen and nitrogen atoms in the molecules of *N*-phenylsulfonyl, *N*-benzoyl, and *N*-[phenylsulfonylimino(phenyl)methyl] 1,4-benzoquinone imine derivatives



| Parameter | X = PhCO | X = $PhSO_2$ | X = $PhSO_2C(Ph)$ |
|---|---|---|---|
| | Energy, kJ/mol | | |
| $E_1$ | 103.01 | 112.51 | 105.90 |
| $E_2$ | 111.96 | 108.37 | 110.21 |
| $\Delta E = E_1 - E_2$ | –8.95 | 4.14 | –4.31 |
| | Charge | | |
| $Z_1$(N) | –0.4332 | –0.6422 | –0.4024 |
| $Z_2$(O) | –0.5148 | –0.5062 | –0.5133 |
| $\Delta Z = Z_1 - Z_2$ | 0.0814 | –0.1360 | +0.1109 |

The reactivity of the $C^2$ and $C^6$ atoms in the quinoid ring of *N*-aroyl derivatives **V**–**VII** toward nucleophiles is considerably reduced due to displacement of electron density to the oxygen atom. Moreover, *N*-aroyl derivatives are characterized by higher redox potentials than *N*-arylsulfonyl analogs [8], which favors generation of radical anions from the former via single-electron transfer process.

We previously presumed that 1,6- and 6,1-additions follow a radical ion mechanism involving initial abstraction of an electron from arenesulfinic acid anion with formation of fairly stable semiquinone-like radical anion [1]; the latter was detected by ESR spectroscopy [3].

Thus, change of the substituent nature on the nitrogen atom in going from *N*-arylsulfonyl- to *N*-aroyl-1,4-benzoquinone imines is accompanied by change of polarization of the quinoid ring, considerable increase of redox potential, and hence increase of electron-acceptor properties of the quinoid ring, which reduces the contribution of direct nucleophilic addition and increases the contribution of radical anion path. The overall yield of the addition products formed according to the radical anion mechanism (1,6- and 6,1-addition) in the reactions with *N*-aroyl-1,4-benzoquinone imines is 20–36, 14–21, and 60–68% for 2,3-dimethyl- (**Va**–**Vc**), 2-methyl- (**VIa**, **VIb**), and 3-methyl-substituted derivatives (**VIIa**, **VIIb**), respectively (Scheme 1, Table 1).

The formation of 6,1-addition product **XXX** from *N*-arylsulfonyl derivative **IV** may be rationalized by facile generation of radical anion species in the system quinone imine **IV**–sodium 4-methoxybenzenesulfinate (**XIIb**). The lack of methyl groups in the quinoid ring of **IV** is responsible for its highest redox potential among quinone imines **I**–**IV** and correspondingly minimum energy of the lowest antibonding orbital ($E_{\pi^*}$). The presence in the *para* position of **XIIb** of a strong π-donating group (OMe) increases the energy of the lone electron pair on the sulfur atom ($E_{n\sigma S}$). Therefore, the reactant couple **IV**–**XIIb** is characterized by the smallest energy gap ($\Delta E$) between the LUMO of quinone imine and HOMO of arenesulfinate anion, which favors single-electron transfer, i.e., oxidation of arenesulfinate anion with quinone imine and generation of radical anion.

Among *N*-aroyl derivatives **V**–**VII**, direct nucleophilic addition was not accompanied by single electron transfer only in the reaction of quinone imine **Vb**, which is the weakest electron acceptor (its low redox potential is determined by the presence of two methyl groups in the quinoid ring), with sodium 4-chlorobenzenesulfinate (**XIIc**) as the weakest nucleophile (Scheme 1, Table 1). Reduction of the number of methyl groups in the quinoid ring in going from 2,3-dimethyl (**Va**–**Vc**) to 3-methyl derivatives (**VIIa**, **VIIb**) increases the redox potential and hence electron-acceptor properties of quinone imines, which favors radical anion addition path leading to 1,6- and 6,1-addition products (20–36 and 60–68%, respectively).

The presence of a methyl group in position *2* of the quinoid ring in **VIa** and **VIb** provides additional stabilization of the radical center on the quinone carbonyl carbon atom, and their reactions with arenesulfinates follow only 1,6-addition pattern. Localization of the radical center on the nitrogen atom is less favorable.

Thus, increase of the electron-acceptor power of quinone imines as the number of electron-donating substituent in the quinoid ring decreases and increase of the electron-donor power of the *para* substituent in arenesulfinate change the reaction mechanism, so that direct nucleophilic 1,4-addition may be accompanied by concurrent radical anion 1,6- and 6,1-additions.

On the one hand, quinone imines **VIIIa**, **VIIIb**, **IX**, **X**, **XIa**, and **XIb** are structural analogs of *N*-aroyl derivatives [9]; on the other hand, their redox potentials occupy an intermediate place between those of

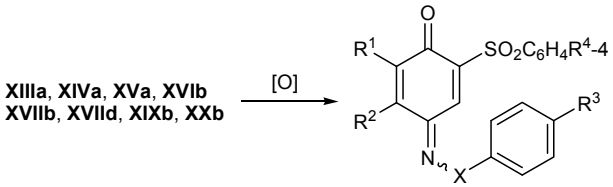
*N*-arylsulfonyl and *N*-aroyl derivatives [8]. Likewise, polarization of the quinoid ring in *N*-[phenylsulfonylimino(phenyl)methyl]-1,4-benzoquinone imine may be estimated as intermediate between *N*-arylsulfonyl and *N*-aroyl derivatives (Table 2). On the basis of these considerations, the reactions of quinone imines **VIII**–**XI** with sodium arenesulfinates **XII**, were expected to give 1,6- and 6,1-addition products according to the radical anion mechanism, but their fraction should be lower than in the reactions with *N*-aroyl analogs **V**–**VII**. In fact, 2,3-dimethyl-substituted quinone imines **VIIIa** and **VIIIb** reacted with sodium arenesulfinates **XIIa** and **XIIb** completely regioselectively with exclusive formation of 1,4-addition products **XXa**–**XXc**, whereas 2(3)-methyl and unsubstituted derivatives **XIa**, **XIb**, **IX**, and **X** gave rise to mixtures of 1,4- (**XXI**, **XXII**, **XXIIIa**–**XXIIIc**) and 1,6-addition products (**XXVII**, **XXVIII**, **XXIXa**–**XXIXc**; Scheme 1, Table 1).

By analogy with *N*-aroyl derivatives **V**–**VII**, the presence of two methyl groups in the quinoid ring and of σ- (Me) and π-donor (MeO) groups in the *para* position of the **Ar**$SO_2$N=C(Ph) group of **VIIIa** and **VIIIb** reduces their redox potential, thus hampering generation of radical anion and hence 1,6-addition. As the number of methyl groups in the quinoid ring decreases (in going to quinone imines **XIa**, **XIb**, **IX**, and **X**), the radical anion mechanism becomes possible, and 1,6-addition products are formed together with 1,4-adducts. No products of addition to the nitrogen atom are formed because of considerable steric hindrances created by the bulky Ar$SO_2$N=C(Ph) substituent.

It should be noted that identification of addition products obtained from quinone imines **V**–**VII** and **IX**–**XI** was fairly difficult, for in some cases the product mixtures contained up to four compounds (including reduced forms of the initial quinone imines), and the $^1$H NMR spectra in the region δ 6.00–8.00 ppm were very complex. Many products were isolated as pure substances. In the other cases, the product ratio was determined from the intensity ratio of the singlet signals from the MeO and Me protons in the arenesulfonyl fragment, taking into account that their positions were different for the 1,4-, 6,1- and 1,6-addition products. For instance, the chemical shifts of the methoxy protons in the arylsulfonyl group were as follows, δ, ppm: 1,6-addition, 3.87–3.89 (DMSO-$d_6$), 3.94 (acetone-$d_6$); 1,4-addition, 3.83–3.85 (DMSO-$d_6$), 3.90–3.93 (acetone-$d_6$); 6,1-addition, 3.77–3.78 (DMSO-$d_6$), 3.85–3.86 (acetone-$d_6$).

The structure of 1,4-addition products **XIIIa**, **XIVa**, **XVa**, **XVIb**, **XVIIb**, **XVIId**, **XIXb**, and **XXb** was additionally confirmed by their oxidation with lead tetraacetate in glacial acetic acid to the corresponding quinone imines **XXXIII**–**XXXVI**, **XXXVIIa**, **XXXVIIb**, **XXXVIII**, and **XXXIX** (Scheme 2), for the $^1$H NMR spectra of the latter were more informative. The $^1$H NMR spectra of quinone imines **XXXIII** and **XXXV** contained a very downfield singlet from the 6-H proton (δ 9.03 ppm), and in the spectra of **XXXIV** and **XXXVI** we observed two sets of signals from the *Z* and *E* isomers, which unambiguously indicated that the hydrogen atoms are attached to $C^2$ and $C^6$ of the quinoid ring. In the spectra of **XXXVII**–**XXXIX**, the 6-H signal was located at δ 7.57–7.76 ppm.

**Scheme 2.**




**XXXIII**–**XXXVI**, **XXXVIIa**
**XXXVIIb**, **XXXVIII**, **XXXIX**

**XXXIII**, $R^1 = R^2 = R^3$ = Me, $R^4$ = MeO, X = $SO_2$; **XXXIV**, $R^1 = R^3$ = Me, $R^2$ = H, $R^4$ = MeO, X = $SO_2$; **XXXV**, $R^1$ = H, $R^2 = R^3$ = Me, $R^4$ = MeO, X = $SO_2$; **XXXVI**, $R^1 = R^2$ = H, $R^3$ = Me, $R^4$ = MeO, X = $SO_2$; **XXXVII**, $R^1 = R^2$ = Me, X = CO; $R^3$ = MeO, $R^4$ = Me (**a**); $R^3$ = Cl, $R^4$ = MeO (**b**); **XXXVIII**, $R^1$ = H, $R^2 = R^4$ = Me, $R^3$ = Cl, X = CO; **XXXIX**, $R^1 = R^2 = R^4$ = Me, $R^3$ = H, X = C=N$SO_2C_6H_4$OMe-4.

In most cases, the reaction mixtures obtained from quinone imines **V**–**XI** contained their reduction products whose concentration was estimated at 6–12% for **VIII**–**XI** and 8–16% for **V**–**VII**. This is consistent with increase of the redox potential in going from *N*-arylsulfonyl to *N*-[arylsulfonylimino(phenyl)methyl] and then to *N*-aroyl derivatives.

In summary, the results of the present study showed that the addition of sodium arenesulfinates to *N*-substituted 1,4-benzoquinone imines follows two concurrent paths, nucleophilic addition of arenesulfinate anion at the *ortho* position with respect to the quinoid carbonyl carbon atom of neutral quinone imine and addition of radical anion generated by oxidation of arenesulfinate anion with quinone imine to the oxygen or nitrogen atom. The contribution of the single-electron transfer process increases in parallel with the redox potential of quinone imine and energy of the lone electron pair on the sulfur atom in arenesulfinate.

## EXPERIMENTAL

The $^1$H NMR spectra were measured on a Varian VXR-300 spectrometer at 300 MHz relative to tetramethylsilane. Thin-layer chromatography was performed on Silufol UV-254 plates using benzene–hexane (10:1) as eluent; spots were visualized under UV light.

Quantum chemical calculations were performed using GAUSSIAN 03 software package [10]. The molecular structures were calculated in terms of the density functional theory with B3LYP functional [11–16] and standard 6-31+G(*d*) basis set [17, 18]. Conjugative and hyperconjugative interactions were analyzed in terms of the natural bond orbital (NBO) theory [19] using NBO 5.0 program [20]. Compounds **I**–**XI** were synthesized by oxidation of the corresponding 4-aminophenols in glacial acetic acid with sodium dichromate (**I**–**IV**) [21] or lead tetraacetate (**V**–**XI**) [22] and were purified by recrystallization from glacial acetic acid. Sodium arenesulfinates **XIIa**–**XIIc** were prepared as described in [23].

The properties of previously described compounds **Ia** [24], **IIa**, **IIIa** [25], **IV** [26], **Va** [27], **VIa**, **VIb**, **VIIa**, **VIIb** [28], **IX**, **XIa**, and **XIb** [9] were consistent with published data.

***N*-(2,3-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-4-methoxybenzenesulfonamide (Ib).** Yield 84%, mp 100–101°C. $^1$H NMR spectrum ($CDCl_3$),* δ, ppm: 2.05 br.s (3H, 3-Me), 2.06 br.s (3H, 2-Me), 3.87 s (3H, MeO), 6.63 d (1H, 5-H, *J* = 10.2 Hz), 7.02 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.93 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz), 8.14 d (1H, 6-H, *J* = 10.2 Hz). Found, %: N 4.50, 4.72; S 10.21, 10.41. $C_{15}H_{15}NO_4S$. Calculated, %: N 4.59; S 10.50.

**4-Chloro-*N*-(2,3-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzenesulfonamide (Ic).** Yield 79%, mp 138–139°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.05 br.s (3H, 3-Me), 2.06 br.s (3H, 2-Me), 6.62 d (1H, 5-H, *J* = 10.2 Hz), 7.56 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.96 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 8.15 d (1H, 6-H, *J* = 10.2 Hz). Found, %: N 4.60, 4.68; S 10.21, 10.41. $C_{14}H_{12}ClNO_3S$. Calculated, %: N 4.59; S 10.50.

**4-Methoxy-*N*-(3-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzenesulfonamide (IIb).** Yield 79%, mp 75–76°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: *E* isomer: 2.06 d (3H, 3-Me), 3.88 s (3H, MeO), 6.68 d (1H, 5-H, *J* = 10.2 Hz), 6.82 q (1H, 2-H), 7.03 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.92 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz), 8.14–8.17 d.d (1H, 6-H, *J* = 2.4, 10.2 Hz); *Z* isomer: 2.15 d (3H, 3-Me), 6.69 d (1H, 5-H, *J* = 10.2 Hz), 6.90–6.93 d.d (1H, 6-H, *J* = 2.4, 10.2 Hz), 7.03 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.92 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz), 8.01 q (1H, 2-H). Found, %: N 4.62, 4.87; S 11.16, 11.34. $C_{14}H_{13}NO_4S$. Calculated, %: N 4.81; S 11.01.

**4-Chloro-*N*-(3-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzenesulfonamide (IIc).** Yield 64%, mp 89–90°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: *E* isomer: 2.05 d (3H, 3-Me), 3.87 s (3H, MeO), 6.68 d (1H, 5-H, *J* = 10.2 Hz), 6.81 q (1H, 2-H), 7.56 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.96 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 8.14–8.17 d.d (1H, 6-H, *J* = 2.4, 10.2 Hz); *Z* isomer: 2.14 d (3H, 3-Me), 6.70 d (1H, 5-H, *J* = 10.2 Hz), 6.91–6.94 d.d (1H, 6-H, *J* = 2.4, 10.2 Hz), 7.55 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.95 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 8.02 q (1H, 2-H). Found, %: N 4.58, 4.75; S 10.58, 10.96. $C_{13}H_{10}ClNO_3S$. Calculated, %: N 4.74; S 10.84.

**4-Methoxy-*N*-(2-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzenesulfonamide (IIIb).** Yield 69%, mp 138–139°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.06 d (3H, 2-Me), 3.88 s (3H, MeO), 6.60–6.64 d.d (1H, 5-H, *J* = 1.5, 9.0 Hz), 6.57 q (1H, 3-H), 8.18 d (1H, 6-H, *J* = 9.0 Hz), 7.03 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.93 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz). Found, %: N 4.61, 4.88; S 10.84, 11.26. $C_{14}H_{13}NO_4S$. Calculated, %: N 4.81; S 11.01.

**4-Chloro-*N*-(2-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzenesulfonamide (IIIc).** Yield 68%, mp 140–141°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.06 d (3H, 2-Me), 6.62–6.66 d.d (1H, 5-H), 6.57 br.s (1H, 3-H), 7.56 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.96 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 8.13 d (1H, 6-H, *J* = 10.2 Hz). Found, %: N 4.70, 4.92; S 11.03, 11.16. $C_{13}H_{10}ClNO_3S$. Calculated, %: N 4.74; S 10.84.

**4-Methoxy-*N*-(3-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzamide (VIa).** Yield 59%, mp 100–101°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.12 d (3H, 3-Me), 3.89 s (3H, MeO), 6.65 d (1H, 5-H, *J* = 10.2 Hz), 7.03–7.07 d.d (1H, 6-H, *J* = 2.4, 10.2 Hz), 6.95 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.89 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 7.94 q (1H, 2-H). Found, %: N 5.60, 5.74. $C_{15}H_{13}NO_3$. Calculated, %: N 5.49.

***N*-(2,3-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-4-methoxybenzamide (Vb).** Yield 42%,

* Hereinafter, unprimed locants refer to the quinoid ring, primed locants refer to the aromatic ring of the substituent on the quinone imine nitrogen atom, and double-primed locants refer to the aromatic ring formerly belonging to arenesulfinate.

mp 99–101°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.09 d (3H, 3-Me, $J$ = 1.2 Hz), 2.28 d (3H, 2-Me, $J$ = 1.2 Hz), 3.88 s (3H, MeO), 6.47 d (1H, 5-H, $J$ = 10.2 Hz), 6.83 d (1H, 6-H, $J$ = 10.2 Hz), 6.95 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.83 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz). Found, %: N 5.05, 5.29. $C_{16}H_{15}NO_3$. Calculated, %: N 5.20.

**4-Chloro-*N*-(2,3-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)benzamide (Vc).** Yield 51%, mp 130–131°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.10 d (3H, 3-Me, $J$ = 1.2 Hz), 2.27 d (3H, 2-Me, $J$ = 1.2 Hz), 6.49 d (1H, 5-H, $J$ = 10.2 Hz), 6.82 d (1H, 6-H, $J$ = 10.2 Hz), 7.46 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.83 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz). Found, %: N 4.89, 5.04. $C_{15}H_{12}ClNO_2$. Calculated, %: N 5.12.

***N*-(2,3-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-*N*′-(4-methylphenylsulfonyl)benzenecarboximidamide (VIIIa).** Yield 86%, mp 146–147°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.04 br.s (3H, 3-Me), 2.05 br.s (3H, 2-Me), 2.42 s (3H, **Me**$C_6H_4$), 6.66 d (1H, 5-H, $J$ = 10.2 Hz), 7.32 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.42–7.85 m (5H, Ph), 6.96 d (1H, 6-H, $J$ = 10.2 Hz), 7.81 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz). Found, %: N 7.28, 7.44; S 7.88, 8.27. $C_{20}H_{20}N_2O_3S$. Calculated, %: N 7.14; S 8.17.

***N*-(2,3-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-*N*′-(4-methoxyphenylsulfonyl)benzenecarboximidamide (VIIIb).** Yield 85%, mp 150.5–152°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.03 br.s (3H, 3-Me), 2.04 br.s (3H, 2-Me), 3.87 s (3H, MeO), 6.65 d (1H, 5-H, $J$ = 10.2 Hz), 7.01 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.42–7.85 m (5H, Ph), 6.95 d (1H, 6-H, $J$ = 10.2 Hz), 7.93 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz). Found, %: N 6.59, 6.96; S 8.14, 7.95. $C_{22}H_{20}N_2O_4S$. Calculated, %: N 6.86; S 7.85.

***N*-(2-Methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-*N*′-(4-methylphenylsulfonyl)benzenecarboximidamide (X).** Yield 67%, mp 129–131°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.04 br.s (3H, 2-Me), 2.42 s (3H, **Me**$C_6H_4$), 6.66 d (1H, 5-H, $J$ = 10.2 Hz), 6.75 q (1H, 3-H), 7.31 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.41–7.86 m (5H, Ph), 6.95–6.98 d.d (1H, 6-H, $J$ = 2.4, 10.2 Hz), 7.80 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz). Found, %: N 7.51, 7.76; S 8.12, 8.41. $C_{22}H_{23}NO_6S_2$. Calculated, %: N 7.40; S 8.47.

**General procedure for the reactions of quinone imines I–XI with sodium arenesulfinates XIIa–XIIc.** A solution of 2 mmol of quinone imine **I**–**XI** in 20 mL of glacial acetic acid was heated to the boiling point, 4 mmol of the corresponding sodium arenesulfinate **XIIa**–**XIIc** was added in one portion, and the mixture was stirred for several minutes until it turned colorless. The mixture was cooled, and water was added until complete precipitation. The colorless precipitate was filtered off and washed with water (sample **1**). A part of product **1** was recrystallized from glacial acetic acid to obtain sample **2**. The mother liquor obtained after recrystallization was diluted with water, and the precipitate was filtered off (sample **3**). All three samples were analyzed by $^1$H NMR spectroscopy.

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]-4-methylbenzenesulfonamide (XIIIa).** Yield 67%, mp 195–196°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.11 s (3H, 3-Me), 2.13 s (3H, 2-Me), 2.45 s (3H, **Me**$C_6H_4$), 3.93 s (3H, MeO), 7.15 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.18 s (1H, 6-H), 7.35 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.53 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.77 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 8.39 br.s (1H, NH), 9.22 s (1H, OH). Found, %: N 2.91, 3.10; S 14.00, 14.22. $C_{22}H_{23}NO_6S_2$. Calculated, %: N 3.03; S 13.89.

***N*-[5-(4-Chlorophenylsulfonyl)-4-hydroxy-2,3-dimethylphenyl]-4-methoxybenzenesulfonamide (XIIIb).** Yield 81%, mp 218–220°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.01 s (3H, 3-Me), 2.08 s (3H, 2-Me), 3.85 s (3H, MeO), 7.12 s (1H, 6-H), 7.13 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.57 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 7.69 s (4H, $ClC_6H_4$), 9.40 s (1H, NH), 9.64 s (1H, OH). Found, %: N 2.82, 2.95; S 13.12, 13.48. $C_{21}H_{20}ClNO_6S_2$. Calculated, %: N 2.91; S 13.31.

**4-Chloro-*N*-[4-hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]benzenesulfonamide (XIIIc).** Yield 80%, mp 203–204°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.03 s (3H, 3-Me), 2.10 s (3H, 2-Me), 3.84 s (3H, MeO), 7.08 s (1H, 6-H), 7.12 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.61 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.66 s (4H, $ClC_6H_4$), 9.46 s (1H, NH), 9.67 s (1H, OH). Found, %: N 2.76, 2.88; S 12.90, 13.18. $C_{21}H_{20}ClNO_6S_2$. Calculated, %: N 2.91; S 13.31.

***N*-[4-Hydroxy-3-(4-methoxyphenylsulfonyl)-5-methylphenyl]-4-methylbenzenesulfonamide (XIVa).** Yield 79%, mp 161–162°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.14 s (3H, 5-Me), 2.37 s (3H, **Me**$C_6H_4$), 3.93 s (3H, MeO), 7.15 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.22 d (1H, 6-H, $J$ = 2.4 Hz), 7.28 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.43 d (1H, 2-H, $J$ = 2.4 Hz), 7.57 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.84 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 8.89 s (1H, NH),

9.11 br.s (1H, OH). Found, %: N 3.04, 3.20; S 14.29, 14.67. $C_{21}H_{21}NO_6S_2$. Calculated, %: N 3.13; S 14.33.

***N*-[3-(4-Chlorophenylsulfonyl)-4-hydroxy-5-methylphenyl]-4-methoxybenzenesulfonamide (XIVb).** Yield 84%, mp 177–178°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.05 s (3H, 5-Me), 3.80 s (3H, MeO), 7.11 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.14 d (1H, 6-H, $J$ = 2.1 Hz), 7.45 d (1H, 2-H, $J$ = 2.1 Hz), 7.60 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 7.70 d (2H, 3″-H, 5″-H, $J$ = 8.4 Hz), 7.90 d (2H, 2″-H, 6″-H, $J$ = 8.4 Hz), 9.66 br.s (1H, NH), 9.99 s (1H, OH). Found, %: N 2.78, 2.94; S 13.27, 13.60. $C_{20}H_{18}ClNO_6S_2$. Calculated, %: N 2.99; S 13.70.

**4-Chloro-*N*-[4-hydroxy-3-(4-methoxyphenylsulfonyl)-5-methylphenyl]benzenesulfonamide (XIVc).** Yield 87%, mp 152–153°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.08 s (3H, 5-Me), 3.84 s (3H, MeO), 7.12 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.13 d (1H, 6-H, $J$ = 2.4 Hz), 7.38 d (1H, 2-H, $J$ = 2.4 Hz), 7.68 s (4H, $ClC_6H_4$), 7.74 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 9.61 br.s (1H, NH), 10.18 s (1H, OH). Found, %: N 2.93, 3.14; S 13.45, 13.73. $C_{20}H_{18}ClNO_6S_2$. Calculated, %: N 2.99; S 13.70.

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2-methylphenyl]-4-methylbenzenesulfonamide (XVa).** Yield 86%, mp 177–177.5°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.13 s (3H, 2-Me), 2.45 s (3H, **Me**$C_6H_4$), 3.91 s (3H, MeO), 6.80 s (1H, 3-H), 7.12 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.38 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.46 s (1H, 6-H), 7.57 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.80 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 8.35 s (1H, NH), 9.46 s (1H, OH). Found, %: N 3.10, 3.28; S 13.96, 14.24. $C_{21}H_{21}NO_6S_2$. Calculated, %: N 3.13; S 14.33.

***N*-[5-(4-Chlorophenylsulfonyl)-4-hydroxy-2-methylphenyl]-4-methoxybenzenesulfonamide (XVb).** Yield 83%, mp 212–214°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.04 s (3H, 2-Me), 3.85 s (3H, MeO), 6.67 s (1H, 3-H), 10.84 br.s (1H, OH), 9.36 br.s (1H, NH), 7.28 s (1H, 6-H), 7.13 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.58 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 7.67 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.73 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz). Found, %: N 3.11, 3.17; S 13.68, 14.06. $C_{20}H_{18}ClNO_6S_2$. Calculated, %: N 2.99; S 13.70.

**4-Chloro-*N*-[4-hydroxy-5-(4-methoxyphenylsulfonyl)-2-methylphenyl]benzenesulfonamide (XVc).** Yield 77%, mp 186–187°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.06 s (3H, 2-Me), 3.84 s (3H, MeO), 6.68 s (1H, 3-H), 7.11 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.22 s (1H, 6-H), 7.64 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.69 s (4H, $ClC_6H_4$), 9.57 br.s (1H, NH), 10.79 br.s (1H, OH). Found, %: N 2.86, 3.04; S 13.50, 13.91. $C_{20}H_{18}ClNO_6S_2$. Calculated, %: N 2.99; S 13.70.

***N*-[4-Hydroxy-3-(4-methylphenylsulfonyl)-phenyl]-4-methylbenzenesulfonamide (XVIa).** Yield 83%, mp 185–187°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.42 s (3H, 4″-Me), 2.39 s (3H, 4′-Me), 6.89 d (1H, 5-H, $J$ = 8.4 Hz), 7.31–7.34 d.d (1H, 6-H, $J$ = 3.0, 8.4 Hz), 7.32 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.41 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.59 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 7.69 d (1H, 2-H, $J$ = 3.0 Hz), 7.76 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 8.90 br.s (1H, NH), 9.48 s (1H, OH). Found, %: N 3.30, 3.17; S 15.72, 15.81. $C_{20}H_{19}NO_5S_2$. Calculated, %: N 3.35; S 15.36.

***N*-[4-Hydroxy-3-(4-methoxyphenylsulfonyl)-phenyl]-4-methylbenzenesulfonamide (XVIb).** Yield 70%, mp 148– 149°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.39 s (3H, 4′-Me), 3.91 s (3H, MeO), 6.89 d (1H, 5-H, $J$ = 8.7 Hz), 7.10 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.29–7.32 d.d (1H, 6-H, $J$ = 2.4, 8.7 Hz), 7.32 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.59 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 7.66 d (1H, 2-H, $J$ = 2.4 Hz), 7.83 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 8.89 br.s (1H, NH), 9.45 s (1H, OH). Found, %: N 3.31, 3.28; S 14.76, 14.78. $C_{20}H_{19}NO_6S_2$. Calculated, %: N 3.23; S 14.79.

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]-4-methylbenzamide (XVIIa).** Yield 55%, mp 205.5–207°C. $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.21 s (6H, 2-Me, 3-Me), 2.41 s (3H, 4′-Me), 3.90 s (3H, MeO), 7.14 d (2H, 3″-H, 5″-H, $J$ = 8.4 Hz), 7.33 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.72 s (1H, 6-H), 7.92 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.95 d (2H, 2″-H, 6″-H, $J$ = 8.4 Hz), 9.18 br.s (1H, NH), 9.30 br.s (1H, OH). Found, %: N 3.08, 3.25; S 7.26, 7.50. $C_{23}H_{23}NO_5S$. Calculated, %: N 3.29; S 7.54.

***N*-[4-Hydroxy-2,3-dimethyl-5-(4-methylphenylsulfonyl)phenyl]-4-methoxybenzamide (XVIIb).** Yield 59%, mp 225–227°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.10 s (6H, 2-Me, 3-Me), 2.38 s (3H, 4″-Me), 3.84 s (3H, MeO), 7.07 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.41 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.67 s (1H, 6-H), 7.81 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 7.99 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 9.51 s (1H, NH), 9.92 s (1H, OH). Found, %: N 3.18, 3.33; S 7.18, 7.60. $C_{23}H_{23}NO_5S$. Calculated, %: N 3.29; S 7.54.

***N*-[5-(4-Chlorophenylsulfonyl)-4-hydroxy-2,3-dimethylphenyl]-4-methoxybenzamide (XVIIc).** Yield 76%, mp 231–132°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.11 s (6H, 2-Me, 3-Me), 3.84 s (3H, MeO), 7.06 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.68 d (2H, 3″-H,

5″-H, $J$ = 8.7 Hz), 7.69 s (1H, 6-H), 7.91 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.97 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz), 9.44 br.s (1H, NH), 9.85 br.s (1H, OH). Found, %: N 3.14, 3.30; S 7.35, 7.46. $C_{22}H_{20}ClNO_5S$. Calculated, %: N 3.14; S 7.19.

**4-Chloro-*N*-[4-hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]benzamide (XVIId).** Yield 38%, mp 214–215°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.11 s (6H, 2-Me, 3-Me), 3.84 s (3H, MeO), 7.12 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.61 d (2H, 3′-H, 5′-H, $J$ = 8,4 Hz), 7.66 s (1H, 6-H), 7.86 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 8.01 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 9.49 br.s (1H, NH), 10.10 s (1H, OH). Found, %: N 3.16, 3.36; S 6.89, 7.24. $C_{22}H_{20}ClNO_5S$. Calculated, %: N 3.14; S 7.19.

***N*-[4-Hydroxy-3-methyl-5-(4-methylphenylsulfonyl)phenyl]-4-methoxybenzamide (XVIIIa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.15 s (3H, 3-Me), 2.38 s (3H, 4″-Me), 3.85 s (3H, MeO), 7.06 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.41 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.82 d (1H, 2-H, $J$ = 2.4 Hz), 7.97 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 7.80 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 8.23 d (1H, 6-H, $J$ = 2.4 Hz), 9.38 s (1H, NH), 10.12 s (1H, OH).

**4-Bromo-*N*-[4-hydroxy-3-(4-methoxyphenylsulfonyl)-5-methylphenyl]benzamide (XVIIIb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.16 s (3H, 5-Me), 3.83 s (3H, MeO), 7.12 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.76 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.81 br.s (1H, 6-H), 7.86 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.93 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 8.22 d (1H, 2-H, $J$ = 2.1 Hz), 9.47 s (1H, NH), 10.35 s (1H, OH).

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2-methylphenyl]-4-methylbenzamide (XIXa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.17 s (3H, 2-Me), 2.38 s (3H, 4′-Me), 3.83 s (3H, MeO), 6.78 s (1H, 3-H), 7.32 d (2H, 3″-H, 5″-H, $J$ = 8.4 Hz), 7.33 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.82 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.83 s (1H, 6-H), 7.86 d (2H, 2″-H, 6″-H, $J$ = 8.4 Hz), 9.79 s (1H, NH), 10.60 br.s (1H, OH).

**4-Chloro-*N*-[4-hydroxy-2-methyl-5-(4-methylphenylsulfonyl)phenyl]benzamide (XIXb).** Yield 15%, mp 197–199°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.17 s (3H, 2-Me), 2.38 s (3H, 4″-Me), 6.80 s (1H, 3-H), 7.40 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.62 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.82 s (1H, 6-H), 7.79 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 8.01 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 9.96 s (1H, NH), 10.64 s (1H, OH). Found, %: N 3.50, 3.28; S 7.59, 7.89. $C_{21}H_{18}ClNO_4S$. Calculated, %: N 3.37; S 7.71.

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]-*N*′-(4-methylphenylsulfonyl)-benzenecarboximidamide (XXa).** Yield 64%, mp 190–191°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.04 s (3H, 3-Me), 2.07 s (3H, 2-Me), 2.32 s (3H, 4′-Me), 3.83 s (3H, MeO), 7.09 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.20 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.39 s (1H, 6-H), 7.45–7.69 m (5H, Ph), 7.54 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.80 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 9.56 br.s (1H, NH), 10.21 br.s (1H, OH). Found, %: N 4.64, 4.86; S 11.18, 11.54. $C_{29}H_{28}N_2O_6S_2$. Calculated, %: N 4.96; S 11.36.

***N*-(4-Hydroxy-2,3-dimethyl-5-(4-methylphenylsulfonyl)phenyl)-*N*′-(4-methoxyphenylsulfonyl)benzenecarboximidamide (XXb).** Yield 56%, mp 192–193°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.03 s (3H, 3-Me), 2.07 s (3H, 2-Me), 2.38 s (3H, 4″-Me), 3.78 s (3H, MeO), 6.92 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.36 d (2H, 3″-H, 5″-H, $J$ = 7.8 Hz), 7.38 s (1H, 6-H), 7.40–7.64 m (5H, Ph), 7.55 d (2H, 2″-H, 6″-H, $J$ = 7.8 Hz), 7.70 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 9.53 br.s (1H, NH), 10.22 br.s (1H, OH). Found, %: N 4.81, 5.03; S 11.05, 11.40. $C_{29}H_{28}N_2O_6S_2$. Calculated, %: N 4.96; S 11.36.

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2,3-dimethylphenyl]-*N*′-(4-methoxyphenylsulfonyl)-benzenecarboximidamide (XXc).** Yield 69%, mp 169–171°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.04 s (3H, 3-Me), 2.07 s (3H, 2-Me), 7.43 s (1H, 6-H), 3.83 s (3H, 4″-OMe), 3.78 s (3H, 4′-OMe), 6.93 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.08 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.32–7.58 m (5H, Ph), 7.56 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.78 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 9.58 br.s (1H, NH), 10.19 br.s (1H, OH). Found, %: N 4.67, 4.91; S 11.05, 11.40. $C_{29}H_{28}N_2O_7S_2$. Calculated, %: N 4.82; S 11.04.

***N*-[4-Hydroxy-3-(4-methoxyphenylsulfonyl)-5-methylphenyl]-*N*′-(4-methylphenylsulfonyl)carboximidamide (XXI).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.02 s (3H, 5-Me), 2.35 s (3H, 4′-Me), 3.84 s (3H, MeO), 7.09 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.32 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.48–7.60 m (5H, Ph), 7.65 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.75 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.77 br.s (1H, 6-H), 8.10 br.s (1H, 2-H), 9.72 br.s (1H, NH), 10.56 br.s (1H, OH).

***N*-[4-Hydroxy-5-(4-methoxyphenylsulfonyl)-2-methylphenyl]-*N*′-(4-methylphenylsulfonyl)benzenecarboximidamide (XXII).** Yield 41%, mp 244–246°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.15 s (3H, 2-Me), 2.32 s (3H, 4′-Me), 3.83 s (3H, MeO),

6.74 s (1H, 3-H), 7.07 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.21 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.34–7.61 m (5H, Ph), 7.62 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.75 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.87 s (1H, 6-H), 10.15 s (1H, NH), 10.80 s (1H, OH). Found, %: N 5.16, 5.28; S 10.21, 10.41. $C_{28}H_{26}N_2O_6S_2$. Calculated, %: N 5.09; S 11.65.

***N*-[4-Hydroxy-3-(4-methoxyphenylsulfonyl)-phenyl]-*N*′-(4-methylphenylsulfonyl)benzenecarboximidamide (XXIIIa).** Yield 34%, mp 124–126°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.34 s (3H, 4′-Me), 3.83 s (3H, MeO), 6.82 d (1H, 5-H, $J$ = 9.0 Hz), 7.08 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.30 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.50 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.56–7.68 m (5H, Ph), 7.58–7.61 d.d (1H, 6-H, $J$ = 2.4, 8.7 Hz), 7.73 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 8.30 br.s (1H, 2-H), 10.56 s (1H, NH), 10.80 s (1H, OH). Found, %: N 4.99, 5.23; S 12.12, 12.34. $C_{27}H_{24}N_2O_6S_2$. Calculated, %: N 5.22; S 11.95.

***N*′-(4-Chlorophenylsulfonyl)-*N*-[4-hydroxy-3-(4-methylphenylsulfonyl)phenyl]benzenecarboximidamide (XXIIIb).** Yield 30%, mp 254–255°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.38 s (3H, 4″-Me), 6.84 d (1H, 5-H, $J$ = 8.7 Hz), 7.37 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.50–7.67 m (5H, Ph), 7.51 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 7.55 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.57–7.60 d.d (1H, 6-H, $J$ = 2.4, 8.7 Hz), 7.73 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 8.31 br.s (1H, 2-H), 10.69 s (1H, NH), 10.83 s (1H, OH). Found, %: N 4.89, 5.10; S 12.15, 11.99. $C_{26}H_{21}ClN_2O_5S_2$. Calculated, %: N 5.18; S 11.85.

***N*′-(4-Chlorophenylsulfonyl)-*N*-[4-hydroxy-3-(4-methoxyphenylsulfonyl)phenyl]benzenecarboximidamide (XXIIIc).** Yield 37%, mp 213–215°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 3.84 s (3H, MeO), 6.84 d (1H, 5-H, $J$ = 9.0 Hz), 7.08 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.47–7.62 m (5H, Ph), 7.55 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.57–7.60 d.d (1H, 6-H, $J$ = 2.1, 9.0 Hz), 7.73 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.73 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 8.28 br.s (1H, 2-H), 10.65 s (1H, NH), 10.78 s (1H, OH). Found, %: N 5.05, 5.18; S 11.06, 11.34. $C_{26}H_{21}ClN_2O_6S_2$. Calculated, %: N 5.03; S 11.51.

**2,3-Dimethyl-4-(4-methylbenzamido)phenyl 4-methoxybenzenesulfonate (XXIVa).** $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.17 s (3H, 2-Me), 2.23 s (3H, 3-Me), 2.45 s (3H, 4′-Me), 3.94 s (3H, MeO), 6.80 d (1H, 6-H, $J$ = 9.0 Hz), 6.97 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.06 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.38 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 7.86 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.18 d (1H, 5-H, $J$ = 9.0 Hz), 9.14 s (1H, NH).

**4-(4-Methoxybenzamido)-2,3-dimethylphenyl 4-methylbenzenesulfonate (XXIVb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.00 s (3H, 2-Me), 2.07 s (3H, 3-Me), 2.45 s (3H, 4″-Me), 3.83 s (3H, MeO), 6.80 d (1H, 6-H, $J$ = 8.7 Hz), 7.05 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.14 d (1H, 5-H, $J$ = 8.7 Hz), 7.51 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.80 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz), 7.95 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 9.84 s (1H, NH).

**4-(4-Chlorobenzamido)-2,3-dimethylphenyl 4-methoxybenzenesulfonate (XXIVc).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.01 s (3H, 2-Me), 2.08 s (3H, 3-Me), 3.89 s (3H, MeO), 6.81 d (1H, 6-H, $J$ = 8.4 Hz), 7.16 d (1H, 5-H, $J$ = 8.7 Hz), 7.21 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.61 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.83 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.99 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz), 10.08 s (1H, NH).

**4-(4-Methoxybenzamido)-2-methylphenyl 4-methylbenzenesulfonate (XXVa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.05 s (3H, 2-Me), 2.45 s (3H, 4″-Me), 3.82 s (3H, MeO), 7.02 d (1H, 6-H, $J$ = 8.4 Hz), 7.06 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.12 d (2H, 3″-H, 5″-H, $J$ = 7.8 Hz), 7.55 d (2H, 2″-H, 6″-H, $J$ = 7.8 Hz), 7.56–7.59 d.d (1H, 5-H, $J$ = 2.1, 8.1 Hz), 7.70 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz), 7.91 d (1H, 3-H, $J$ = 2.1 Hz), 9.62 s (1H, NH).

**4-(4-Bromobenzamido)-2-methylphenyl 4-methoxybenzenesulfonate (XXVb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.04 s (3H, 2-Me), 3.88 s (3H, MeO), 7.04 d (1H, 6-H, $J$ = 8.7 Hz), 7.12 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.49 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.59–7.62 d.d (1H, 5-H, $J$ = 1.8, 8.7 Hz), 7.75 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 7.82 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 7.83 d (1H, 3-H, $J$ = 1.8 Hz), 9.89 s (1H, OH).

**3-Methyl-4-(4-methylbenzamido)phenyl 4-methoxybenzenesulfonate (XXVIa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.18 s (3H, 3-Me), 2.37 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.79–6.82 d.d (1H, 6-H, $J$ = 2.7, 8.7 Hz), 7.00 d (1H, 2-H, $J$ = 2.7 Hz), 7.11 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.18 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.80 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.88 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 7.58 d (1H, 5-H, $J$ = 8.7 Hz), 9.83 s (1H, NH).

**4-(4-Chlorobenzamido)-3-methylphenyl 4-methylbenzenesulfonate (XXVIb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.18 s (3H, 3-Me), 2.38 s (3H, 4″-Me), 6.82–6.85 d.d (1H, 6-H, $J$ = 2.7, 8.4 Hz), 7.03 d (1H, 2-H, $J$ = 2.7 Hz), 7.49 d (2H, 3″-H, 5″-H,

*J* = 8.1 Hz), 7.56 d (1H, 5-H, *J* = 8.4 Hz), 7.60 d (2H, 3′-H, 5′-H, *J* = 8.7 Hz), 7.78 d (2H, 2″-H, 6″-H, *J* = 8.1 Hz), 7.98 d (2H, 2′-H, 6′-H, *J* = 8.7 Hz), 9.99 s (1H, NH).

**2-Methyl-4-[*N*′-(4-methylphenylsulfonyl)benzenecarboximidamido]phenyl 4-methoxybenzenesulfonate (XXVII).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 1.83 s (3H, 2-Me), 2.36 s (3H, 4′-Me), 3.89 s (3H, MeO), 6.92 d (1H, 6-H, *J* = 9.0 Hz), 7.19 d (2H, 3″-H, 5″-H, *J* = 8.7 Hz), 7.30 d (2H, 3′-H, 5′-H, *J* = 8.1 Hz), 7.45–7.69 m (5H, Ph), 7.46–7.49 d.d (1H, 5-H, *J* = 3.6, 9.0 Hz), 7,53 d (1H, 3-H, *J* = 3.6 Hz), 7.65 d (2H, 2′-H, 6′-H, *J* = 8.1 Hz), 7.78 d (2H, 2″-H, 6″-H, *J* = 8.7 Hz), 9.98 s (1H, NH).

**3-Methyl-4-[*N*′-(4-methylphenylsulfonyl)benzenecarboximidamido]phenyl 4-methoxybenzenesulfonate (XXVIII).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.13 s (3H, 3-Me), 2.32 s (3H, 4′-Me), 3.87 s (3H, MeO), 6.78–6.81 d.d (1H, 6-H, *J* = 2.7, 8.1 Hz), 7.02 d (2H, 3″-H, 5″-H, *J* = 8.7 Hz), 7.10 d (1H, 2-H, *J* = 2.7 Hz), 7.15 d (2H, 3′-H, 5′-H, *J* = 8.1 Hz), 7.40–7.66 m (5H, Ph), 7.58 d (1H, 5-H, *J* = 8.1 Hz), 7.75 d (2H, 2′-H, 6′-H, *J* = 8.1 Hz), 7.79 d (2H, 2″-H, 6″-H, *J* = 8.7 Hz), 9.97 s (1H, NH).

**4-[*N*′-(4-Methylphenylsulfonyl)benzenecarboximidamido]phenyl 4-methoxybenzenesulfonate (XXIXa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.36 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.96 d (2H, 2-H, 6-H, *J* = 8.7 Hz), 7.17 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.27 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz), 7.43–7.63 m (5H, Ph), 7.43 d (2H, 3″-H, 5″-H, *J* = 9.0 Hz), 7.60 d (2H, 3-H, 5-H, *J* = 8.7 Hz), 7.73 d (2H, 2″-H, 6″-H, *J* = 9.0 Hz), 10.61 s (1H, NH).

**4-[*N*′-(4-Chlorophenylsulfonyl)benzenecarboximidamido]phenyl 4-methylbenzenesulfonate (XXIXb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 2.44 s (3H, 4″-Me), 6.99 d (2H, 2-H, 6-H, *J* = 8.4 Hz), 7.45–7.56 m (5H, Ph), 7.48 d (2H, 3″-H, 5″-H, *J* = 8.7 Hz), 7.60 (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.62 d (2H, 3-H, 5-H, *J* = 8.4 Hz), 7.73 d (2H, 2″-H, 6″-H, *J* = 8.7 Hz), 7.74 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 10.74 s (1H, NH).

**4-[*N*′-(4-Chlorophenylsulfonyl)benzenecarboximidamido]phenyl 4-methoxybenzenesulfonate (XXIXc).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 3.88 s (3H, MeO), 6.98 d (2H, 2-H, 6-H, *J* = 9.0 Hz), 7.08 d (2H, 3″-H, 5″-H, *J* = 9.0 Hz), 7.17 d (2H, 3′-H, 5′-H, *J* = 9.0 Hz), 7.43–7.62 m (5H, Ph), 7.61 d (2H, 3-H, 5-H, *J* = 9.0 Hz), 7.71 d (2H, 2′-H, 6′-H, *J* = 9.0 Hz), 7.77 d (2H, 2″-H, 6″-H, *J* = 9.0 Hz), 10.73 s (1H, NH).

***N*-(4-Hydroxyphenyl)-*N*-(4-methoxyphenylsulfonyl)-4-methylbenzenesulfonamide (XXX).** $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.38 s (3H, 4′-Me), 3.85 s (3H, MeO), 6.70 d (2H, 2-H, 6-H, *J* = 8.4 Hz), 6.97 d (2H, 3-H, 5-H, *J* = 8.4 Hz), 7.06 d (2H, 3″-H, 5″-H, *J* = 8.4 Hz), 7.37 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.50 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 7.86 d (2H, 2″-H, 6″-H, *J* = 8.4 Hz), 9.08 br.s (1H, OH).

***N*-(4-Hydroxy-2,3-dimethylphenyl)-*N*-(4-methoxyphenylsulfonyl)-4-methylbenzamide (XXXIa).** $^1$H NMR spectrum (acetone-$d_6$), δ, ppm: 2.12 s (3H, 2-Me), 2.15 s (3H, 3-Me), 2.38 s (3H, 4′-Me), 3.86 s (3H, MeO), 6.70 d (1H, 6-H, *J* = 8.7 Hz), 6.87 d (1H, 5-H, *J* = 8.7 Hz), 7.02 d (2H, 3″-H, 5″-H, *J* = 9.0 Hz), 7.26 d (2H, 3′-H, 5′-H, *J* = 8.4 Hz), 7.51 d (2H, 2′-H, 6′-H, *J* = 8.4 Hz), 7.82 d (2H, 2″-H, 6″-H, *J* = 9.0 Hz), 8.97 br.s (1H, OH).

***N*-(4-Hydroxy-2,3-dimethylphenyl)-4-methoxy-*N*-(4-methylphenylsulfonyl)benzamide (XXXIb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 1.95 s (3H, 2-Me), 1.99 s (3H, 3-Me), 2.43 s (3H, 4″-Me), 3.70 s (3H, MeO), 6.62 d (1H, 6-H, *J* = 8.4 Hz), 6.77 d (1H, 5-H, *J* = 8.4 Hz), 7.15 d (2H, 3′-H, 5′-H, *J* = 8.7 Hz), 7.27 d (2H, 3″-H, 5″-H, *J* = 8.1 Hz), 7.55 d (2H, 2″-H, 6″-H, *J* = 8.1 Hz), 7.86 d (2H, 2′-H, 6′-H, *J* = 8.7 Hz), 9.36 s (1H, OH).

**4-Chloro-*N*-(4-hydroxy-2,3-dimethylphenyl)-*N*-(4-methoxyphenylsulfonyl)benzamide (XXXIc).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 1.95 s (3H, 2-Me), 2.00 s (3H, 3-Me), 3.77 s (3H, MeO), 6.60 d (1H, 6-H, *J* = 8.7 Hz), 6.81 d (1H, 5-H, *J* = 8.4 Hz), 6.88 d (2H, 3″-H, 5″-H, *J* = 9.0 Hz), 7.22 d (2H, 3′-H, 5′-H, *J* = 8.7 Hz), 7.49 d (2H, 2″-H, 6″-H, *J* = 9.0 Hz), 7.89 d (2H, 2′-H, 6′-H, *J* = 8.7 Hz), 9.52 s (1H, OH).

***N*-(4-Hydroxy-2-methylphenyl)-*N*-(4-methoxyphenylsulfonyl)-4-methylbenzamide (XXXIIa).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 1.95 s (3H, 2-Me), 2.40 s (3H, 4′-Me), 3.77 s (3H, MeO), 6.53 d (1H, 3-H, *J* = 2.4 Hz), 6.55–6.58 d.d (1H, 5-H, *J* = 2.4, 8.7 Hz), 6.88 d (2H, 3″-H, 5″-H, *J* = 9.0 Hz), 7.04 d (2H, 3′-H, 5′-H, *J* = 7.8 Hz), 7.11 d (1H, 6-H, *J* = 8.7 Hz), 7.53 d (2H, 2′-H, 6′-H, *J* = 7.8 Hz), 7,78 d (2H, 2″-H, 6″-H, *J* = 9.0 Hz), 9.37 br.s (1H, OH).

**4-Chloro-*N*-(4-hydroxy-2-methylphenyl)-*N*-(4-methylphenylsulfonyl)benzamide (XXXIIb).** $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 1.93 s (3H, 2-Me), 2.40 s (3H, 4″-Me), 6.54 d (1H, 3-H, *J* = 2.4 Hz), 6.55–6.58 d.d (1H, 6-H, *J* = 2.4, 8.7 Hz), 7.10 d (1H, 5-H, *J* = 8.4 Hz), 7.10 d (2H, 3″-H, 5″-H, *J* = 8.1 Hz), 7.46 d (2H, 2″-H, 6″-H, *J* = 8.1 Hz),

7.57 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.85 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 9.58 br.s (1H, OH).

Quinone imines **XXXIII**–**XXXVI**, **XXXVIIa**, **XXXVIIb**, **XXXVIII**, and **XXXIX** were synthesized by oxidation of compounds **XIIIa**, **XIVa**, **XVa**, **XVIb**, **XVIIb**, **XVIId**, **XIXb**, and **XXb**, respectively, with lead tetraacetate in glacial acetic acid [22]. The products were recrystallized from glacial acetic acid.

***N*-[5-(4-Methoxyphenylsulfonyl)-2,3-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene]-4-methylbenzenesulfonamide (XXXIII).** Yield 75%, mp 229.5–230°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.00 br.s (3H, 3-Me), 2.03 br.s (3H, 2-Me), 2.48 s (3H, 4′-Me), 3.88 s (3H, MeO), 7.02 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.39 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.92 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 8.04 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 9.03 s (1H, 6-H). Found, %: N 2.96, 3.10; S 14.40, 14.44. $C_{22}H_{21}NO_6S_2$. Calculated, %: N 3.05; S 13.96.

***N*-[5-(4-Methoxyphenylsulfonyl)-3-methyl-4-oxocyclohexa-2,5-dien-1-ylidene]-4-methylbenzenesulfonamide (XXXIV).** Yield 70%, mp 157–158°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: *E* isomer: 2.09 br.s (3H, 3-Me), 2.49 s (3H, 4′-Me), 3.88 s (3H, MeO), 7.01 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.40 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.82 d (1H, 6-H, $J$ = 2.4 Hz), 7.90 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.99 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 8.01 q (1H, 2-H, $J$ = 2.4 Hz); *Z* isomer: 2.01 br.s (3H, 3-Me), 2.49 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.85 q (1H, 2-H, $J$ = 2.4 Hz), 7.01 d (2H, 3″-H, 5″-H, $J$ = 8.7 Hz), 7.40 d (2H, 3′-H, 5′-H, $J$ = 7.8 Hz), 7.90 d (2H, 2′-H, 6′-H, $J$ = 7.8 Hz), 7.99 d (2H, 2″-H, 6″-H, $J$ = 8.7 Hz), 9.04 d (1H, 6-H, $J$ = 2.4 Hz). Found, %: N 2.21, 3.30; S 14.56, 14.87. $C_{21}H_{19}NO_6S_2$. Calculated, %: N 3.14; S 14.33.

***N*-[5-(4-Methoxyphenylsulfonyl)-2-methyl-4-oxocyclohexa-2,5-dien-1-ylidene]-4-methylbenzenesulfonamide (XXXV).** Yield 70%, mp 210–211°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.05 d (3H, 2-Me, $J$ = 1.2 Hz), 2.48 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.47 q (1H, 3-H, $J$ = 1.2 Hz), 7.01 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.40 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.92 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 8.02 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 9.03 s (1H, 6-H). Found, %: N 3.00, 3.11; S 14.17, 14.63. $C_{21}H_{19}NO_6S_2$. Calculated, %: N 3.14; S 14.39.

***N*-[3-(4-Methoxyphenylsulfonyl)-4-oxocyclohexa-2,5-dien-1-ylidene]-4-methylbenzenesulfonamide (XXXVI).** Yield 78%, mp 166–167°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: *E* isomer: 2.49 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.61 d (1H, 5-H, $J$ = 10.2 Hz), 7.01 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.41 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.90 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.99 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 8.19–8.22 d.d (1H, 6-H, $J$ = 3.0, 10.2 Hz), 7.87 d (1H, 2-H, $J$ = 3 Hz); *Z* isomer: 2.49 s (3H, 4′-Me), 3.88 s (3H, MeO), 6.61 d (1H, 5-H, $J$ = 10.2 Hz), 7.01–7.04 d.d (1H, 6-H, $J$ = 3.0, 10.2 Hz), 7.01 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.41 d (2H, 3′-H, 5′-H, $J$ = 8.1 Hz), 7.90 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.99 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz), 9.07 d (1H, 2-H, $J$ = 3 Hz). Found, %: N 3.32, 3.18; S 15.12, 14.84. $C_{20}H_{17}NO_6S_2$. Calculated, %: N 3.25; S 14.86.

***N*-[2,3-Dimethyl-5-(4-methylphenylsulfonyl)-4-oxocyclohexa-2,5-dien-1-ylidene]-4-methoxybenzamide (XXXVIIa).** Yield 68%, mp 167–168°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.02 br.s (3H, 3-Me), 2.27 br.s (3H, 2-Me), 2.42 s (3H, 4″-Me), 3.91 s (3H, MeO), 6.99 d (2H, 3′-H, 5′-H, $J$ = 9.0 Hz), 7.32 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.75 s (1H, 6-H), 7.84 d (2H, 2′-H, 6′-H, $J$ = 9.0 Hz), 7.90 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz). Found, %: N 3.23, 3.41; S 8.04, 7.76. $C_{23}H_{21}NO_5S$. Calculated, %: N 3.31; S 7.57.

**4-Chloro-*N*-[5-(4-methoxyphenylsulfonyl)-2,3-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene]benzamide (XXXVIIb).** Yield 81%, mp 172–174°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2,04 br.s (3H, 3-Me), 2.26 br.s (3H, 2-Me), 3.87 s (3H, MeO), 6.98 d (2H, 3″-H, 5″-H, $J$ = 9.0 Hz), 7.71 s (1H, 6-H), 7.84 d (2H, 2′-H, 6′-H, $J$ = 8.1 Hz), 7.96 d (2H, 2″-H, 6″-H, $J$ = 9.0 Hz). Found, %: N 3.15, 3.26; S 7.08, 6.84. $C_{22}H_{18}ClNO_5S$. Calculated, %: N 3.16; S 7.22.

**4-Chloro-*N*-[2-methyl-5-(4-methylphenylsulfonyl)-4-oxocyclohexa-2,5-dien-1-ylidene]benzamide (XXXVIII).** Yield 63%, mp 192–193°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.28 br.s (3H, 2-Me), 2.43 s (3H, 4″-Me), 6.53 q (1H, 3-H, $J$ = 1.8 Hz), 7.33 d (2H, 3″-H, 5″-H, $J$ = 8.1 Hz), 7.51 d (2H, 3′-H, 5′-H, $J$ = 8.4 Hz), 7.76 s (1H, 6-H), 7.84 d (2H, 2′-H, 6′-H, $J$ = 8.4 Hz), 7.90 d (2H, 2″-H, 6″-H, $J$ = 8.1 Hz). Found, %: N 3.43, 3.51; S 7.94, 8.11. $C_{21}H_{16}ClNO_4S$. Calculated, %: N 3.38; S 7.75.

***N*-[2,3-Dimethyl-5-(4-methylphenylsulfonyl)-4-oxocyclohexa-2,5-dien-1-ylidene)-*N*′-(4-methoxyphenylsulfonyl)benzenecarboximidamide (XXXIX).** Yield 73%, mp 186–187°C. $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 2.05 br.s (3H, 3-Me), 2.27 br.s (3H, 2-Me), 2.41 s (3H, 4″-Me), 3.88 s (3H, MeO), 7.00 d (2H, 3′-H, 5′-H, $J$ = 8.7 Hz), 7.33 d (2H, 3″-H, 5″-H, $J$ = 7.8 Hz), 7.44–7.78 m (5H, Ph), 7.57 s (1H, 6-H),

7.90 d (2H, 2″-H, 6″-H, $J$ = 7.8 Hz), 7.93 d (2H, 2′-H, 6′-H, $J$ = 8.7 Hz). Found, %: N 5.20, 5.29; S 10.81, 11.19. $C_{29}H_{26}N_2O_6S_2$. Calculated, %: N 4.98; S 11.40.

The authors are grateful to the staff of the X-Ray Diffraction Analysis and Crystal Chemistry Department, Institute for Single Crystals of the National Academy of Sciences of Ukraine and of the Ukrainian–American Laboratory of Quantum Chemistry (Kharkiv, Ukraine–Jackson, USA) for their help in performing quantum chemical calculations).

## REFERENCES

1. Burmistrov, K.S., Nichvoloda, V.M., Markov, V.I., and Romanchenko, V.A., *Zh. Org. Khim*., 1986, vol. 22, p. 1306.
2. Nichvoloda, V.M., Burmistrov, K.S., and Markov, V.I., *Zh. Org. Khim*., 1985, vol. 21, p. 1069.
3. Nichvoloda, V.M., Alaev, Yu.N., Luk'yanenko, L.V., Markov, V.I., and Burmistrov, K.S., *Zh. Org. Khim*., 1986, vol. 22, p. 1111; Avdeenko, A.P. and Koshechko, V.G., *Zh. Obshch. Khim*., 1974, vol. 44, p. 1459.
4. Avdeenko, A.P., Konovalova, S.A., Roman'kov, D.A., Burmistrov, K.S., Nichvoloda, V.M., Shishkin, O.V., Zubatyuk, R.I., and Palamarchuk, G.V., *Russ. J. Org. Chem*., 2009, vol. 45, p. 48.
5. Avdeenko, A.P., Konovalova, S.A., Roman'kov, D.A., Ludchenko, O.N., and Marchenko, I.L., *Russ. J. Org. Chem*., 2009, vol. 45, p. 383.
6. Avdeenko, A.P., Konovalova, S.A., Mikhailichenko, O.N., Shelyazhenko, S.V., Pirozhenko, V.V., and Yagupol'skii, L.M., *Russ. J. Org. Chem*., 2012, vol. 48, p. 221.
7. *Spravochnik khimika* (Chemist's Handbok), Nikol'skii, B.P., Ed., Leningrad: Khimiya, 1964, 2nd ed., vol. 2, p. 352.
8. Burmistrov, K.S., Toropin, N.V., Vakulenko, A.V., Yurchenko, A.G., and Markov, V.I., *Vopr. Khim. Khim. Tekhnol*., 2000, no. 2, p. 30.
9. Avdeenko, A.P., Pirozhenko, V.V., Yagupol'skii, L.M., and Marchenko, I.L., *Russ. J. Org. Chem*., 2001, vol. 37, p. 991.
10. Frisch, M.J., Trucks, G.W., Schlegel, H.B., Scuseria, G.E., Robb, M.A., Cheeseman, J.R., Montgomery, J.A., Jr., Vreven, T., Kudin, K.N., Burant, J.C., Millam, J.M., Iyengar, S.S., Tomasi, J., Barone, V., Mennucci, B., Cossi, M., Scalmani, G., Rega, N., Petersson, G.A., Nakatsuji, H., Hada, M., Ehara, M., Toyota, K., Fukuda, R., Hasegawa, J., Ishida, M., Nakajima, T., Honda, Y., Kitao, O., Nakai, H., Klene, M., Li, X., Knox, J.E., Hratchian, H.P., Cross, J.B., Bakken, V., Adamo, C., Jaramillo, J., Gomperts, R., Stratmann, R.E., Yazyev, O., Austin, A.J., Cammi, R., Pomelli, C., Ochterski, J.W., Ayala, P.Y., Morokuma, K., Voth, G.A., Salvador, P., Dannenberg, J.J., Zakrzewski, V.G., Dapprich, S., Daniels, A.D., Strain, M.C., Farkas, O., Malick, D.K., Rabuck, A.D., Raghavachari, K., Foresman, J.B., Ortiz, J.V., Cui, Q., Baboul, A.G., Clifford, S., Cioslowski, J., Stefanov, B.B., Liu, G., Liashenko, A., Piskorz, P., Komaromi, I., Martin, R.L., Fox, D.J., Keith, T., Al-Laham, M.A., Peng, C.Y., Nanayakkara, A., Challacombe, M., Gill, P.M.W., Johnson, B., Chen, W., Wong, M.W., Gonzalez, C., and Pople, J.A., *Gaussian 03, Revision C.01*, Wallingford CT: Gaussian, 2004.
11. Becke, A.D., *Phys. Rev. A*, 1988, vol. 38, p. 3098.
12. Becke, A.D., *J. Chem. Phys*., 1993, vol. 98, p. 5648.
13. Lee, C., Yang, W., and Parr, R.G., *Phys. Rev. B*, 1988, vol. 37, p. 785.
14. Volko, S.H., Wilk, L., and Nusair, M., *Can. J. Phys*., 1980, vol. 58, p. 1200.
15. Hohenberg, R. and Kohn, W., *Phys. Rev.*, 1964, vol. 136, p. B864.
16. Parr, R.G. and Yang, W., *Density Functional Theory of Atoms and Molecules*, New York: Oxford Univ., 1989.
17. Rassolov, V.A., Pople, J.A., Ratner, M.A., and Windus, T.L., *J. Chem. Phys*., 1998, vol. 109, p. 1223.
18. Rassolov, V.A., Ratner, M.A., Pople, J.A., Redfern, P.C., and Curtiss, L.A., *J. Comput. Chem*., 2001, vol. 22, p. 976.
19. Schleyer, P.v.R., Schreiner, P.R., Allinger, N.L., Clark, T., Gasteiger, J., Kollman, P., and Schaefer, H.F. III, *Encyclopedia of Computational Chemistry*, Chichester: Wiley, 1998, p. 1792.
20. Glendening, E.D., Badenhoop, J.K., Reed, A.E., Carpenter, J.E., Bohmann, J.A., Morales, C.M., and Weinhold, F., *NBO 5.0*, Madison: Theoretical Chemistry Institute, Univ. of Wisconsin, 2001.
21. Burmistrov, S.I. and Titov, E.A., *Zh. Obshch. Khim*., 1952, vol. 22, p. 999.
22. Avdeenko, A.P., Burmistrov, K.S., Dubina, V.L., and Skripets, V.I., *Ukr. Khim. Zh*., 1980, vol. 46, p. 1081.
23. Whitmore, F.C. and Hamilton, F.H., *Org. Synth*., 1922, vol. 2, p. 89.
24. Avdeenko, A.P. and Konovalova, S.A., *Russ. J. Org. Chem*., 2006, vol. 42, p. 669.
25. Avdeenko, A.P. and Konovalova, S.A., *Russ. J. Org. Chem*., 2006, vol. 42, p. 349.
26. Pirozhenko, V.V., Burmistrov, K.S., Belov, V.V., and Nichvoloda, V.N., *Ukr. Khim. Zh*., 1992, vol. 58, p. 68.
27. Avdeenko, A.P., Konovalova, S.A., and Ludchenko, O.N., *Russ. J. Org. Chem*., 2009, vol. 45, 1799.
28. Avdeenko, A.P., Konovalova, S.A., and Ludchenko, O.N., *Russ. J. Org. Chem*., 2010, vol. 46, p. 1629.