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Synthesis and Structure of *N*-Alkyl(aryl)aminocarbonyl-1,4-benzoquinone Imines

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Abstract—New *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines were synthesized by reaction of isocyanates with the corresponding substituted 4-aminophenols, and their structure was determined on the basis of ^{1}H and ^{13}C NMR spectra and X-ray diffraction data.

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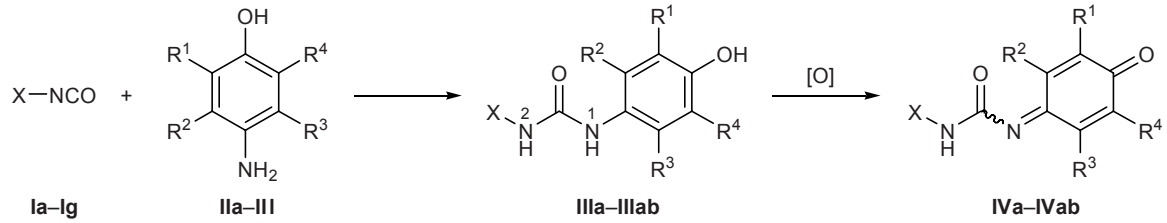
Urea derivatives are quite reactive compounds and are widely used in organic synthesis [1]. They are convenient starting materials for the preparation of various heterocycles [2], including those possessing strong biological activity [3–5]. Broad synthetic potential of ureas makes it possible to obtain new classes of compounds. N-Substituted *p*-quinone imines have been known for more than hundred years [6, 7]. They readily undergo halogenation [7] and hydrohalogenation [8] and react with nucleophiles [9] and alcohols [10]; as a result, numerous derivatives of *p*-quinone imines were synthesized, many of which showed fungicidal, bactericidal [11], and sometimes antitumor activity [12]. A combination of quinone imine and urea fragments in a single molecule could considerably extend synthetic potential of such compounds and give rise to broader spectrum of their properties.

The goal of the present work was to synthesize compounds belonging to a new class of quinone imines, *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines, whose molecules include structural fragments of quinone imines and substituted ureas. It is known that N-substituted ureas can be prepared by reactions of aryl or alkyl isocyanates with various amines. Isocyanates are capable of reacting with both amines and alcohols [5, 13]. The most widely used procedure for the synthesis of N-substituted 1,4-benzoquinone imines is based on the reaction of *p*-aminophenols with the corresponding carboxylic or sulfonic acid chlorides, followed by oxidation of the acylamino derivative thus formed. Taking into account that *p*-aminophenols have two nucleophilic centers, NH_2 and OH groups, their reactions with isocyanates could give addition products at both amino and hydroxy groups. According to Beaver et al. [3], aryl isocyanates reacted with *p*-aminophenols only at the amino group of the latter with formation of *N*-arylaminocarbonyl-4-aminophenols. The products were not subjected to oxidation to obtain the corresponding quinone imines. *N*-(Diphenylaminocarbonyl)-2,6-diisopropyl-1,4-benzoquinone imine was reported in [14].

We synthesized a large series of *N*-alkyl(aryl)aminocarbonyl-4-aminophenols **IIIa**–**IIIab** by reaction of aryl and alkyl isocyanates **Ia**–**Ig** with various substituted *p*-aminophenols **IIa**–**III** in dioxane, and the products were oxidized to *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines **IVa**–**IVab** (Scheme 1). As oxidants we used lead tetraacetate, diacetoxy(phenyl)-λ^3-iodane, and silver(I) oxide; different oxidants were tried with a view to improve the yield and simplify the isolation procedure.

Quinone imines **IVa**–**IVg**, **IVi**, **IVl**, **IVt**–**IVv**, and **IVy** were obtained by oxidation of the corresponding aminophenols with lead tetraacetate in acetic acid, and aminophenols **IIIh**–**IIIk**, **IIIm**, **IIIp**, **IIIr**, and **IIIs** were oxidized with diacetoxy(phenyl)-λ^3-iodane. In these cases, the yields of quinone imines **IV** ranged from 25 to 85%. However, the use of the above oxidants did not allow us to isolate quinone imines **IVn**, **IVw**, **IVx**, and **IVaa**–**IVac**; the latter were

Scheme 1.



I, X = Ph (**a**), 4-MeC_6H_4 (**b**), 4-$MeOC_6H_4$ (**c**), 4-ClC_6H_4 (**d**), *cyclo*-C_6H_{11} (**e**), Bu (**f**), *t*-Bu (**g**); **II**, $R^1 = R^2 = R^3 = R^4 = H$ (**a**), R^1 = Me, $R^2 = R^3 = R^4 = H$ (**b**); $R^1 = R^3 = R^4 = H$, R^2 = Me (**c**); $R^1 = R^2$ = Me, $R^3 = R^4 = H$ (**d**); $R^1 = R^3$ = Me, $R^2 = R^4 = H$ (**e**); $R^1 = R^4$ = Me, $R^2 = R^3 = H$ (**f**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**g**); $R^1 = R^4$ = *i*-Pr, $R^2 = R^3 = H$ (**h**); $R^1 = R^4$ = *t*-Bu, $R^2 = R^3 = H$ (**i**); R^1 = *i*-Pr, $R^2 = R^4$ = H, R^3 = Me (**j**); R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**k**); $R^1 = R^4$ = Cl, $R^2 = R^3 = H$ (**l**); **III**, **IV**, X = Ph, $R^1 = R^2 = R^3 = R^4 = H$ (**a**), R^1 = Me, $R^2 = R^3 = R^4 = H$ (**b**); $R^1 = R^3 = R^4 = H$, R^2 = Me (**c**); $R^1 = R^2$ = Me, $R^3 = R^4 = H$ (**d**); $R^1 = R^3$ = Me, $R^2 = R^4 = H$ (**e**); $R^1 = R^4$ = Me, $R^2 = R^3 = H$ (**f**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**g**); $R^1 = R^4$ = *i*-Pr, $R^2 = R^3 = H$ (**h**); $R^1 = R^4$ = *t*-Bu, $R^2 = R^3 = H$ (**i**); R^1 = *i*-Pr, $R^2 = R^4$ = H, R^3 = Me (**j**); R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**k**); $R^1 = R^4$ = Cl, $R^2 = R^3 = H$ (**l**); X = 4-MeC_6H_4, $R^1 = R^3 = R^4 = H$, R^2 = Me (**m**); $R^1 = R^4$ = Me, $R^2 = R^3 = H$ (**n**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**o**); R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**p**); X = 4-$MeOC_6H_4$, $R^1 = R^4$ = Me, $R^2 = R^3 = H$ (**q**); R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**r**); X = 4-ClC_6H_4, R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**s**); X = *cyclo*-C_6H_{11}, $R^1 = R^3 = R^4 = H$, R^2 = Me (**t**); $R^1 = R^3$ = Me, $R^2 = R^4 = H$ (**u**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**v**); X = Bu, $R^1 = R^2 = R^3 = R^4 = H$ (**w**); $R^1 = R^4$ = Me, $R^2 = R^3 = H$ (**x**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**y**); R^1 = Me, $R^2 = R^4 = H$, R^3 = *i*-Pr (**z**); X = *t*-Bu, R^1 = Me, $R^2 = R^3 = R^4 = H$ (**aa**); $R^1 = R^4 = H$, $R^2 = R^3$ = Me (**ab**).

obtained by oxidation of the corresponding aminophenols with silver(I) oxide in acetone, benzene, or methylene chloride. As a rule, these quinone imines were very unstable, and their yield was as poor as 8 to 20%. We failed to isolate quinone imine **IVx**, for it underwent fast hydrolysis to 2,6-dimethyl-1,4-benzoquinone. Quinone imine **IVx** was detected in the reaction mixture by TLC.

The structure of the isolated compounds (**IIIa**–**IIIk**, **IIIn**, **IIIu**, **IVa**–**IVw**, and **IVy**–**IVab**) was proved by their elemental analyses and IR and ^{1}H NMR spectra. The IR spectra of aminophenols **IIIa**–**IIIi** contained absorption bands in the regions 3200–3350, 3380–3430, and 1580–1665 cm^{-1} due to stretching vibrations of NH, OH, and C=O groups, respectively. Quinone imines **IVa**–**IVw** and **IVy**–**IVab** displayed NH absorption bands at 3220–3300 cm^{-1}, carbonyl bands at 1620–1650 (quinoid) and 1680–1730 cm^{-1} (HNCONH), and C=N stretching vibration band at 1570–1610 cm^{-1}.

In the ^{1}H NMR spectra of aminophenols **IIIa**–**IIIe**, **IIIg**, **IIIj**–**IIIm**, **IIIo**–**IIIw**, and **IIIy**–**IIIab**, signals from the hydroxy proton appeared in the region δ 8.45–9.09 ppm, the N^2H proton resonated at δ 8.52–8.73 ppm, and the N^1H signal was located at δ 7.39–8.31 ppm. The spectra of *N*-arylaminocarbonyl-2,6-dialkyl-4-aminophenols **IIIf**, **IIIh**, **IIIi**, and **IIIx** were characterized by upfield shift of the OH signal (δ 6.64–7.88 ppm) due to effect of substituents in positions *2* and *6*. Signals from the NH protons of aminophenols **IIIf**, **IIIh**, and **IIIi** also appeared in a stronger field (δ 8.21–8.32 and 8.44–8.52 ppm for N^1H and N^2H, respectively) relative to the corresponding signals of the other aminophenols. *N*-Arylaminocarbonyl-1,4-benzoquinone imines **IVa**–**IVs** displayed in the ^{1}H NMR spectra signals from the NH proton as a broadened singlet in the region δ 7.11–7.45 ppm. The NH signal of *N*-alkylaminocarbonyl-1,4-benzoquinone imines **IVt**–**IVw** and **IVy**–**IVab** was located in a stronger field, at δ 4.98–5.34 ppm.

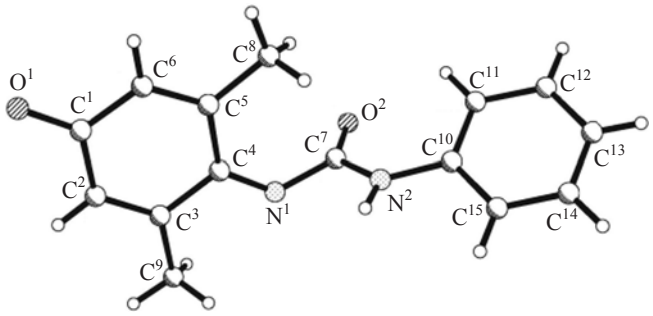
N-Alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines are analogs of *N*-aroyl-1,4-benzoquinone imines whose properties and structural specificity were studied in sufficient detail [15, 16]. We previously found that *N*-aroyl-1,4-benzoquinone imines are characterized by the lowest barrier to *Z*,*E* isomerization about the C=N bond among known *p*-quinone imines [16]. In the ^{1}H NMR spectra of *N*-aroyl-2,6-dimethyl-1,4-benzoquinone imines, protons in the quinoid ring appear as one broadened singlet even at room temperature [17]. The 3-H and 5-H protons in the quinoid ring of *N*-arylaminocarbonyl-2,6-dimethyl-1,4-benzoquinone imines **IVf**, **IVh**, and **IVi** gave fairly narrow singlets relatively distant from each other. These findings suggest that the barrier to *Z*,*E* isomerization about the C=N bond in *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines is higher than that for the corresponding *N*-aroyl derivatives.

As we showed previously, considerable reduction of the barrier to *Z*,*E* isomerization of *N*-aroyl-1,4-benzoquinone imines as compared to other *p*-benzoquinone imines is related to nonplanar structure of these compounds where π-electron systems of the

quinoid and aroyl fragments are mutually orthogonal. In keeping with the results of quantum-chemical calculations, interaction between the unshared electron pair on the nitrogen atom and π*-orbital of the C=O bond in such structure could reduce the isomerization barrier to a considerable extent [16]. The X-ray diffraction data for *N*-(4-chlorobenzoyl)-2,6-di-*tert*-butyl-1,4-benzoquinone imine confirmed almost orthogonal orientation of the *p*-chlorophenyl and quinoid ring planes (the corresponding dihedral angle is 86.1°); this means that the molecular conformation is optimal for conjugation between π electrons of the carbonyl group and unshared electron pair on the nitrogen atom ($n_{\sigma N}$–$\pi^*_{C=O}$ interaction) [15]. Thus, higher barrier to *Z,E* isomerization about the C=N bond in *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imine may result from different mutual orientation of the quinoid ring and C=O bond in the urea fragment.

The structure of *N*-phenylaminocarbonyl-3,5-dimethyl-1,4-benzoquinone imine (**IVg**) was unambiguously determined by X-ray analysis (see figure). The C^1–$C^6C^8C^9O^1$ quinoid fragment is planar within 0.02 Å. The N^1 atom deviates from that plane by 0.15 Å [the torsion angle $C^2C^3C^4N^1$ is 176.6(4)°]. Conjugation between the phenylaminocarbonyl and quinoid fragments is disrupted due to rotation about the C^7–N^1 bond [torsion angle $C^4N^1C^7N^2$ 96.4(5)°], which is induced by steric repulsion between these fragments. This follows from the presence of shortened intramolecular contacts $H^{8B}\cdots C^7$ 2.56 Å (the sum of the corresponding van der Waals radii is 2.87 Å [18]), $H^{8C}\cdots C^7$ 2.67 Å (2.87 Å), $C^8\cdots C^7$ 2.86 Å (3.42 Å), and $H^{8C}\cdots N^2$ 2.64 Å (2.66 Å). In addition, the bond angles at the C^4 atom are considerably distorted [$C^5C^4N^1$ 127.0(3)°, $C^3C^4N^1$ 114.3(4)°]. The aminocarbonyl fragment somewhat deviates from the C^{10}–C^{15} benzene ring plane: the torsion angle $C^7N^2C^{10}C^{11}$ is 25.2(7)°. Molecules **IVg** in crystal give rise to infinite chains along the (010) crystallographic axis via intermolecular hydrogen bonds N^2–$H^{2A}\cdots O^{2'}$ (H···O 2.10 Å, ∠NHO 156°).

As might be expected, the structure of the XC(O)N= fragment in *N*-arylaminocarbonyl-1,4-benzoquinone imines differs from the structure of the corresponding fragment in *N*-aroyl derivatives. The dihedral angle between the C=O bond and quinoid ring plane is 63.3°; i.e., it is considerably smaller than in *N*-aroyl-1,4-benzoquinone imines. As a result, the $n_{\sigma N}$–$\pi^*_{C=O}$ interaction in molecules **IV** is much weaker. It should also be noted that conjugation between the π-C=O orbital and π-system of the aryl substituent



Structure of the molecule of 1-(2,6-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (**IVg**) according to the X-ray diffraction data.

exists in *N*-aroyl-1,4-benzoquinone imines, and the C=O bond always lies in the plane of the aryl group. In going to *N*-arylaminocarbonyl-1,4-benzoquinone imines, such conjugation becomes impossible, for the C=O and aryl groups are separated by NH group.

Thus we have synthesized new *N*-alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines whose properties should differ from those of other N-substituted *p*-quinone imines, and their steric structure was determined.

EXPERIMENTAL

The ^{1}H NMR spectra were recorded on a Varian VXR-300 spectrometer (300 MHz) from solutions in DMSO-d_6 (aminophenols **IIIa**–**IIIk**, **IIIn**, **IIIu**, and **IIIx**) or $CDCl_3$ (quinone imines **IVa**–**IVw** and **IVy**–**IVab**). The purity of quinone imines **IV** was checked by TLC on Silufol UV-254 plates. Spots were applied from solutions in chloroform, chloroform–ethanol (10 : 1) was used as eluent, and the chromatograms were developed by UV irradiation.

The X-ray diffraction data for compound **IVg** were acquired on an Xcalibur-3 diffractometer (MoK_α irradiation, CCD detector, graphite monochromator, ω-scanning, $2\theta_{max}$ = 51°). Rhombic crystals, $C_{15}H_{14}N_2O_2$, with the following unit cell parameters (at 293 K): a = 6.552(4), b = 8.438(3), c = 24.213(6) Å; V = 1338.5(10) Å^3; M_r = 254.28; Z = 4; space group $P2_12_12_1$; d_{calc} = 1.262 g/cm^3; μ(MoK_α) = 0.085 mm^{-1}; F(000) = 536. Intensities of 4340 reflections were measured (2035 independent reflections with R_{int} = 0.046). The structure was solved by the direct method using SHELXTL software package [19]. The positions of hydrogen atoms were determined by the difference synthesis of electron density and were refined using the riding model ($U_{iso} = nU_{eq}$ for the non-hydrogen atom linked to the given hydrogen atom; n = 1.5 for

methyl groups, and n = 1.2 for the other hydrogen atoms). The structure was refined with respect to F^2 by the full-matrix least-squares procedure in anisotropic approximation for non-hydrogen atoms to wR_2 = 0.166 (4172 reflections) and R_1 = 0.068 [1380 reflections with $F > 4\sigma(F)$, S = 1.059]. The complete set of crystallographic data was deposited to the Cambridge Crystallographic Data Centre (12 Union Road, Cambgidge, CB2 1EZ, UK; *e-mail:* deposit@ccdc.cam.ac.uk); entry no. CCDC 675401.

Alkyl and aryl isocyanates Ia–Ig (*general procedure***).** A 2-l flask was charged with 4 mol of tolylene diisocyanate, and 1 mol of the corresponding alkyl- or arylamine was added under stirring. The mixture was heated to the boiling point, and the resulting alkyl or aryl isocyanate was distilled off until its amount reached that theoretically possible. The remaining hot mixture was quickly poured into a large metal container (strong foaming with subsequent solidification was possible). The product was additionally purified by vacuum distillation. The properties of isocyanates **Ia**–**Ig** thus obtained were in agreement with published data [20].

N*-Alkyl(aryl)aminocarbonyl-4-aminophenols IIIa–IIIab (general procedure***).** A suspension of 0.01 mol of 4-aminophenol **IIa**–**III** in 50–55 ml of dioxane was heated to 100°C, the hot bath was removed, an equimolar amount of isocyanate **Ia**–**Ig** was slowly added under stirring, and the hot mixture was quickly filtered. After cooling, the precipitate was filtered off and recrystallized from dioxane.

1-(4-Hydroxyphenyl)-3-phenylurea (IIIa). Yield 39%, mp 224–226°C. ^{1}H NMR spectrum, δ, ppm: 6.68 d (2H, 3-H, 5-H, J = 8.7 Hz), 6.91–7.44 m (5H, Ph), 7.22 d (2H, 2-H, 6-H, J = 8.4 Hz), 8.31 s (1H, N^{1}H), 8.52 s (1H, N^{2}H), 9.06 br.s (1H, OH). Found, %: N 11.98, 12.17. $C_{13}H_{12}N_2O_2$. Calculated, %: N 12.27.

1-(4-Hydroxy-3-methylphenyl)-3-phenylurea (IIIb). Yield 74%, mp 233–236°C. ^{1}H NMR spectrum, δ, ppm: 2.10 s (3H, CH_3), 6.68 d (1H, 5-H, J = 8.4 Hz), 6.91–7.44 m (5H, Ph), 7.02–7.05 d.d (1H, 6-H, J = 1.8 Hz), 7.13 br.s (1H, 2-H), 8.25 s (1H, N^{1}H), 8.53 s (1H, N^{2}H), 8.94 s (1H, OH). Found, %: N 11.34, 11.45. $C_{14}H_{14}N_2O_2$. Calculated, %: N 11.56.

1-(4-Hydroxy-2-methylphenyl)-3-phenylurea (IIIc). Yield 41%, mp 225–228°C. ^{1}H NMR spectrum, δ, ppm: 2.14 s (3H, CH_3), 6.54–6.56 d.d (1H, 5-H, ^4J = 1.7, ^3J = 8.7 Hz), 7.33 d (1H, 6-H), 6.59 d (1H, 3-H), 7.67 s (1H, N^{1}H), 6.90–7.45 m (5H, Ph), 8.73 s (1H, N^{2}H), 9.07 s (1H, OH). $C_{14}H_{14}N_2O_2$. Found, %: N 11.49, 11.63. Calculated, %: N 11.56.

1-(4-Hydroxy-2,3-dimethylphenyl)-3-phenylurea (IIId). Yield 31%, mp 233–235°C. ^{1}H NMR spectrum, δ, ppm: 2.09 s (3H, 2-CH_3), 2.07 s (3H, 3-CH_3), 6.61 d (1H, 5-H, J = 8.1 Hz), 7.06 d (1H, 6-H), 6.89–7.44 m (5H, Ph), 7.69 s (1H, N^{1}H), 8.65 s (1H, N^{2}H), 9.03 s (1H, OH). Found, %: N 10.61, 10.67. $C_{15}H_{16}N_2O_2$. Calculated, %: N 10.93.

1-(4-Hydroxy-2,5-dimethylphenyl)-3-phenylurea (IIIe). Yield 64%, mp 252–254°C. ^{1}H NMR spectrum, δ, ppm: 2.06 s (3H, 2-CH_3), 2.10 s (3H, 5-CH_3), 6.58 s (1H, 3-H), 7.23 s (1H, 6-H), 7.61 s (1H, N^{1}H), 6.90–7.44 m (5H, Ph), 8.70 s (1H, N^{2}H), 8.92 s (1H, OH). Found, %: N 10.74, 10.42. $C_{15}H_{16}N_2O_2$. Calculated, %: N 10.93.

1-(4-Hydroxy-3,5-dimethylphenyl)-3-phenylurea (IIIf). Yield 50%, mp 234–236°C. ^{1}H NMR spectrum, δ, ppm: 2.14 s (6H, CH_3), 6.91–7.44 m (5H, Ph), 6.98 s (2H, 2-H, 6-H), 7.88 s (1H, OH), 8.21 s (1H, N^{1}H), 8.52 s (1H, N^{2}H). Found, %: N 10.95, 11.06. $C_{15}H_{16}N_2O_2$. Calculated, %: N 10.93.

1-(4-Hydroxy-2,6-dimethylphenyl)-3-phenylurea (IIIg). Yield 65%, mp 240–241°C. ^{1}H NMR spectrum, δ, ppm: 2.11 s (6H, CH_3), 6.47 s (2H, 3-H, 5-H), 6.88–7.44 m (5H, Ph), 7.39 s (1H, N^{1}H), 8.60 s (1H, N^{2}H), 9.09 s (1H, OH). Found, %: N 10.70, 10.84. $C_{15}H_{16}N_2O_2$. Calculated, %: N 10.93.

1-(4-Hydroxy-3,5-diisopropylphenyl)-3-phenylurea (IIIh). Yield 29%, mp 205–206°C. ^{1}H NMR spectrum, δ, ppm: 1.14 d (12H, CH_3), 3.24–3.33 m (2H, CH), 6.91–7.44 m (5H, Ph), 7.04 s (2H, 2-H, 6-H), 7.73 s (1H, OH), 8.32 s (1H, N^{1}H), 8.46 s (1H, N^{2}H). Found, %: N 8.92, 9.05. $C_{19}H_{24}N_2O_2$. Calculated, %: N 8.97.

1-(4-Hydroxy-3,5-di-*tert*-butylphenyl)-3-phenylurea (IIIi). Yield 25%, mp 211–214°C. ^{1}H NMR spectrum, δ, ppm: 1.38 s (18H, *t*-Bu), 6.64 s (1H, OH), 6.91–7.44 m (5H, Ph), 7.20 s (2H, 2-H, 6-H), 8.31 s (1H, N^{1}H), 8.44 s (1H, N^{2}H). Found, %: N 8.19, 8.39. $C_{21}H_{28}N_2O_2$. Calculated, %: N 8.23.

1-(4-Hydroxy-5-isopropyl-2-methylphenyl)-3-phenylurea (IIIj). Yield 53%, mp 222–223°C. ^{1}H NMR spectrum, δ, ppm: 1.13 d (6H, *i*-Pr), 2.10 s (3H, CH_3), 3.09–3.18 m (1H, CH), 6.58 s (1H, 3-H), 7.27 s (1H, 6-H), 6.90–7.44 m (5H, Ph), 7.64 s (1H, N^{2}H), 8.71 s (1H, N^{1}H), 8.94 s (1H, OH). Found, %: N 9.88, 9.94. $C_{17}H_{20}N_2O_2$. Calculated, %: N 9.85.

1-(4-Hydroxy-2-isopropyl-5-methylphenyl)-3-phenylurea (IIIk). Yield 70%, mp 199–202°C. ^{1}H NMR spectrum, δ, ppm: 1.12 d (6H, *i*-Pr), 2.06 s (3H, CH_3), 2.99–3.08 m (1H, CH), 6.67 s (1H, 3-H), 6.89–7.44 m (5H, Ph), 7.08 s (1H, 6-H), 7.61 s (1H, N^2H), 8.68 s (1H, N^1H), 9.02 s (1H, OH). Found, %: N 9.87, 9.98. $C_{17}H_{20}N_2O_2$. Calculated, %: N 9.85.

1-(2,6-Dichloro-4-hydroxyphenyl)-3-phenylurea (IIIl). Yield 63%, mp 242–243°C. Found, %: N 9.41, 9.63. $C_{13}H_{10}Cl_2N_2O_2$. Calculated, %: N 9.43.

1-(4-Hydroxy-2-methylphenyl)-3-(4-methylphenyl)urea (IIIm). Yield 57%, mp 217–219°C. Found, %: N 10.95, 11.10. $C_{15}H_{12}N_2O_2$. Calculated, %: N 10.93.

1-(4-Hydroxy-3,5-dimethylphenyl)-3-(4-methylphenyl)urea (IIIn). Yield 52%, mp 234–236°C. ^{1}H NMR spectrum, δ, ppm: 2.14 s (6H, 3-CH_3, 5-CH_3), 2.23 s (3H, C**H**$_3$C$_6$H$_4$), 6.97 s (2H, 2-H, 6-H), 7.05–7.31 d.d (4H, C_6H_4, J = 7.8 Hz), 7.85 s (1H, OH), 8.14 s (1H, N^1H), 8.38 s (1H, N^2H). Found, %: N 10.38, 10.48. $C_{16}H_{18}N_2O_2$. Calculated, %: N 10.36.

1-(4-Hydroxy-2,6-dimethylphenyl)-3-(4-methylphenyl)urea (IIIo). Yield 64%, mp 255–257°C. Found, %: N 10.32, 10.49. $C_{16}H_{18}N_2O_2$. Calculated, %: N 10.36.

1-(4-Hydroxy-2-isopropyl-5-methylphenyl)-3-(4-methylphenyl)urea (IIIp). Yield 24%, mp 222–225°C. Found, %: N 9.38, 9.57. $C_{18}H_{22}N_2O_2$. Calculated, %: N 9.39.

1-(4-Hydroxy-2,6-dimethylphenyl)-3-(4-methoxyphenyl)urea (IIIq). Yield 33%, mp 253–256°C. Found, %: N 9.62, 9.85. $C_{16}H_{18}N_2O_3$. Calculated, %: N 9.78.

1-(4-Hydroxy-2-isopropyl-5-methylphenyl)-3-(4-methoxyphenyl)urea (IIIr). Yield 26%, mp 250–252°C. Found, %: N 8.73, 8.90. $C_{18}H_{22}N_2O_3$. Calculated, %: N 8.91.

1-(4-Chlorophenyl)-3-(4-hydroxy-2-isopropyl-5-methylphenyl)urea (IIIs). Yield 21%, mp 257–259°C. Found, %: N 8.74, 8.95. $C_{17}H_{19}ClN_2O_2$. Calculated, %: N 8.79.

1-Cyclohexyl-3-(4-hydroxy-2-methylphenyl)urea (IIIt). Yield 27%, mp 183–185°C. Found, %: N 11.37, 11.47. $C_{14}H_{20}N_2O_2$. Calculated, %: N 11.28.

1-Cyclohexyl-3-(4-hydroxy-2,5-dimethylphenyl)-urea (IIIu). Yield 27%, mp 189–193°C. ^{1}H NMR spectrum, δ, ppm: 1.07–1.81 m (11H, C_6H_{11}), 2.02 s (6H, CH_3), 6.05 d (1H, N^1H, J = 7.5 Hz), 6.52 s (1H, N^2H), 7.21 s (2H, 3-H, 6-H), 8.78 s (1H, OH). Found, %: N 10.58, 10.63. $C_{15}H_{22}N_2O_2$. Calculated, %: N 10.68.

1-Cyclohexyl-3-(4-hydroxy-2,6-dimethylphenyl)-urea (IIIv). Yield 60%, mp 195–197°C. Found, %: N 10.69, 10.83. $C_{15}H_{22}N_2O_2$. Calculated, %: N 10.68.

1-Butyl-3-(4-hydroxyphenyl)urea (IIIw). Yield 25%, mp 189–191°C. Found, %: N 13.48, 13.71. $C_{11}H_{16}N_2O_2$. Calculated, %: N 13.45.

1-Butyl-3-(4-hydroxy-3,5-dimethylphenyl)urea (IIIx). Yield 89%, mp 196–198°C. Found, %: N 11.66, 11.87. $C_{13}H_{20}N_2O_2$. Calculated, %: N 11.85.

1-Butyl-3-(4-hydroxy-2,6-dimethylphenyl)urea (IIIy). Yield 74%, mp 192–194°C. Found, %: N 11.89, 12.08. $C_{13}H_{20}N_2O_2$. Calculated, %: N 11.85.

1-Butyl-3-(4-hydroxy-2-isopropyl-5-methylphenyl)urea (IIIz). Yield 51%, mp 183–187°C. Found, %: N 10.34, 10.57. $C_{15}H_{24}N_2O_2$. Calculated, %: N 10.60.

1-*tert*-Butyl-3-(4-hydroxy-3-methylphenyl)urea (IIIaa). Yield 65%, mp 170–175°C. Found, %: N 12.51, 12.93. $C_{12}H_{18}N_2O_2$. Calculated, %: N 12.60.

1-*tert*-Butyl-3-(4-hydroxy-2,6-dimethylphenyl)-urea (IIIab). Yield 67%, mp 177–180°C. Found, %: N 11.80, 11.99. $C_{13}H_{20}N_2O_2$. Calculated, %: N 11.85.

***N*-Alkyl(aryl)aminocarbonyl-1,4-benzoquinone imines IVa–IVab (*general procedure*).** *a*. A suspension of 0.01 mol of aminophenol **IIIa**–**IIIg**, **IIIi**, **IIIl**, **IIIt**–**IIIv**, or **IIIy** in a small amount of glacial acetic acid (2–3 ml) was cooled using an ice bath, 0.011 mol of lead tetraacetate was added, and the mixture was stirred for 30 min until a bright orange crystalline material separated. Several drops of ethylene glycol were added, the mixture was stirred for 5 min, and the precipitate was filtered off and recrystallized from benzene. If the product failed to separate from the reaction mixture, it was precipitated by addition of hexane.

b. Diacetoxy(phenyl)-λ^3-iodane, 0.011 mol, was added to a cold suspension of 0.01 mol of aminophenol **IIIh**–**IIIk**, **IIIm**, **IIIp**, **IIIr**, or **IIIs** in a small amount of glacial acetic acid (2–3 ml), and the mixture was stirred for 30 until a bright orange crystalline material separated. The precipitate was filtered off and recrystallized from benzene. If the product failed to separate from the reaction mixture, it was precipitated by addition of hexane.

c. A suspension of 1.2 mmol of aminophenol **IIIn**, **IIIw**, or **IIIz**–**IIIab** in 20 ml of benzene, acetone, or methylene chloride was cooled to 10–15°C, 1.6 mmol

of silver(I) oxide was added under stirring using a magnetic stirrer, and the mixture was stirred until it turned bright yellow. The mixture was filtered through a layer of cotton wool, the filtrate was evaporated under reduced pressure at room temperature, and the residue was recrystallized from hexane.

1-(4-Oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVa). Yield 15%, mp 140–143°C. ^{1}H NMR spectrum, δ, ppm: 6.57–6.61 d.d (1H, 5-H, ^4J = 2.4, ^3J = 9.9 Hz), 6.70–6.73 d.d (1H, 3-H, J = 9.9 Hz), 7.10–7.14 d.d (1H, 6-H, ^4J = 2.4 Hz), 7.17–7.57 m (5H, Ph), 7.44–7.48 d.d (1H, 2-H), 7.28 br.s (1H, NH). Found, %: N 12.39, 12.55. $C_{13}H_{10}N_2O_2$. Calculated, %: N 12.38.

1-(3-Methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVb). Yield 44%, mp 129–132°C. ^{1}H NMR spectrum, δ, ppm: *Z* isomer (41%): 2.04 d (3H, CH_3), 6.69 d (1H, 5-H, J = 9.9 Hz), 7.03–7.07 d.d (1H, 6-H, ^4J = 2.7 Hz), 7.14–7.58 m (5H, Ph), 7.21 br.s (1H, NH), 7.24 br.s (1H, 2-H); *E* isomer (59%): 2.09 d (3H, CH_3), 6.57 d (1H, 5-H, J = 10.5 Hz), 6.93 br.s (1H, 2-H, J = 2.7 Hz), 7.14 br.s (1H, NH), 7.14–7.58 m (5H, Ph), 7.36–7.39 d.d (1H, 6-H). Found, %: N 11.68, 11.88. $C_{14}H_{12}N_2O_2$. Calculated, %: N 11.66.

1-(2-Methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVc). Yield 61%, mp 157–159°C. ^{1}H NMR spectrum, δ, ppm: 2.23 s (3H, CH_3), 6.50–6.54 d.d (1H, 5-H, ^3J = 10.5, ^4J = 2.4 Hz), 6.57 q (1H, 3-H), 7.14 br.s (1H, NH), 7.14–7.58 m (5H, Ph), 7.32 d (1H, 6-H). Found, %: N 11.53, 11.85. $C_{14}H_{12}N_2O_2$. Calculated, %: N 11.66.

1-(2,3-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVd). Yield 43%, mp 133–135°C. ^{1}H NMR spectrum, δ, ppm: 2.04 s (3H, 3-CH_3), 2.16 s (3H, 2-CH_3), 6.51 d (1H, 5-H, J = 10.2 Hz), 7.12–7.56 m (5H, Ph), 7.22 d (1H, 6-H), 7.24 br.s (1H, NH). Found, %: N 10.81, 10.96. $C_{15}H_{14}N_2O_2$. Calculated, %: N 11.02.

1-(2,5-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVe). Yield 52%, mp 144–146°C. ^{1}H NMR spectrum, δ, ppm: 2.00 d (3H, 2-CH_3, ^4J = 1.2 Hz), 2.18 br.s (3H, 5-CH_3), 6.54 d (1H, 3-H), 7.08 br.s (1H, NH), 7.09 br.s (1H, 6-H), 7.14–7.58 m (5H, Ph). Found, %: N 10.72, 10.97. $C_{15}H_{14}N_2O_2$. Calculated, %: N 11.02.

1-(3,5-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVf). Yield 59%, mp 148–150°C. ^{1}H NMR spectrum, δ, ppm: 2.03 s (3H, 3-CH_3), 2.08 s (3H, 5-CH_3), 6.85 br.s (1H, 2-H), 7.13–7.58 m (5H, Ph), 7.17 br.s (1H, NH), 7.22 br.s (1H, 6-H). Found, %: N 11.05, 11.32. $C_{15}H_{14}N_2O_2$. Calculated, %: N 11.02.

1-(2,6-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVg). Yield 52%, mp 155–156°C. ^{1}H NMR spectrum, δ, ppm: 2.22 s (6H, CH_3), 6.41 s (2H, 3-H, 5-H), 7.09 br.s (1H, NH), 7.13–7.53 m (5H, Ph). Found, %: N 11.01, 11.13. $C_{15}H_{14}N_2O_2$. Calculated, %: N 11.02.

1-(3,5-Diisopropyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVh). Yield 23%, mp 114–116°C. ^{1}H NMR spectrum, δ, ppm: 1.13 d (12H, CH_3, J = 6.3 Hz), 3.03–3.19 m (2H, CH), 6.79 s (1H, 6-H), 7.13 s (1H, 2-H), 7.13–7.59 m (5H, Ph), 7.32 br.s (1H, NH). Found, %: N 9.05, 9.32. $C_{19}H_{22}N_2O_2$. Calculated, %: N 9.02.

1-(3,5-Di-*tert*-butyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVi). Yield 35%, mp 122–126°C. ^{1}H NMR spectrum, δ, ppm: 1.27 (18H, *t*-Bu, J = 9.3 Hz), 6.82 br.s (1H, 6-H), 7.13–7.57 m (5H, Ph), 7.15 br.s (1H, 2-H), 7.17 br.s (1H, NH). Found, %: N 8.19, 8.35. $C_{21}H_{26}N_2O_2$. Calculated, %: N 8.28.

1-(5-Isopropyl-2-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVj). Yield 38%, mp 132–134°C. ^{1}H NMR spectrum, δ, ppm: 1.08 d [6H, (C**H**$_3$)$_2$CH, J = 6.9 Hz], 2.16 s (3H, 2-CH_3), 2.97–3.06 m [1H, (CH$_3$)$_2$C**H**], 6.51 s (1H, 3-H), 6.99 s (1H, 6-H), 7.13–7.58 m (5H, Ph), 7.27 br.s (1H, NH). Found, %: N 9.93, 10.16. $C_{17}H_{18}N_2O_2$. Calculated, %: N 9.92.

1-(2-Isopropyl-5-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVk). Yield 32%, mp 123–125°C. ^{1}H NMR spectrum, δ, ppm: 1.19 d [6H, (C**H**$_3$)$_2$CH, J = 6.9 Hz], 2.00 d (3H, 5-CH_3, ^4J = 1.2 Hz), 3.25–3.34 m [1H, (CH$_3$)$_2$C**H**], 6.50 s (1H, 3-H), 7.09 br.s (1H, 6-H), 7.11 br.s (1H, NH), 7.13–7.58 m (5H, Ph). Found, %: N 10.05, 10.31. $C_{17}H_{18}N_2O_2$. Calculated, %: N 9.92.

1-(2,6-Dichloro-4-oxocyclohexa-2,5-dien-1-ylidene)-3-phenylurea (IVl). Yield 56%, mp 174–175°C. ^{1}H NMR spectrum, δ, ppm: 6.86 s (2H, 3-H, 5-H), 7.14–7.38 m (5H, Ph), 7.45 s (1H, NH). Found, %: N 9.41, 9.54. $C_{13}H_8Cl_2N_2O_2$. Calculated, %: N 9.49.

1-(2-Methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methylphenyl)urea (IVm). Yield 12%, mp 119–120°C. ^{1}H NMR spectrum, δ, ppm: 2.20 s (3H, 2-CH_3), 2.34 s (3H, 4-CH_3), 6.48–6.52 d.d (1H, 5-H, ^4J = 1.8, ^3J = 12 Hz), 6.55 br.s (1H, 3-H), 7.09 br.s (1H, NH), 7.17–7.43 d.d (4H, C_6H_4, J = 8.1 Hz), 7.31 d (1H,

6-H). Found, %: N 11.08, 11.19. $C_{15}H_{14}N_2O_2$. Calculated, %: N 11.02.

1-(3,5-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methylphenyl)urea (IVn). Yield 78%, mp 163–165°C. ^{1}H NMR spectrum, δ, ppm: 2.02 s (3H, 5-CH_3), 2.06 s (1H, 3-CH_3), 2.33 s (3H, C**H**$_3$C$_6$H$_4$), 6.83 br.s (1H, 6-H), 7.15–7.44 d.d (4H, C_6H_4, J = 7.8 Hz), 7.16 br.s (1H, 2-H), 7.26 br.s (1H, NH). Found, %: N 10.37, 10.52. $C_{16}H_{16}N_2O_2$. Calculated, %: N 10.44.

1-(2,6-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methylphenyl)urea (IVo). Yield 8%, mp 158–160°C. ^{1}H NMR spectrum, δ, ppm: 2.20 s (6H, CH_3), 2.33 s (3H, C**H**$_3$C$_6$H$_4$), 6.38 s (2H, 3-H, 5-H), 7.15–7.40 d.d (4H, C_6H_4, J = 7.5 Hz), 7.21 br.s (1H, NH). Found, %: N 10.47, 10.64. $C_{16}H_{16}N_2O_2$. Calculated, %: N 10.44.

1-(2-Isopropyl-5-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methylphenyl)urea (IVp). Yield 8%, mp 127–129°C. ^{1}H NMR spectrum, δ, ppm: 1.20 d [6H, (C**H**$_3$)$_2$CH, J = 6.6 Hz], 2.00 d (3H, 5-CH_3, ^4J = 1.5 Hz), 2.35 s (3H, C**H**$_3$C$_6$H$_4$), 3.26–3.35 m [1H, (CH$_3$)$_2$C**H**], 6.51 s (1H, 3-H), 6.91 br.s (1H, NH), 7.10 br.s (1H, 6-H), 7.18–7.45 d.d (4H, C_6H_4, J = 8.1 Hz). Found, %: N 9.34, 9.55. $C_{18}H_{20}N_2O_2$. Calculated, %: N 9.45.

1-(3,5-Dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methoxyphenyl)urea (IVq). Yield 11%, mp 139–141°C. ^{1}H NMR spectrum, δ, ppm: 2.03 s (3H, 5-CH_3), 2.06 s (1H, 3-CH_3), 3.81 s (3H, MeO), 6.85 br.s (1H, 6-H), 6.90–7.47 d.d (4H, C_6H_4, J = 9.3 Hz), 7.04 br.s (1H, NH), 7.21 br.s (1H, 2-H). Found, %: N 9.80, 9.93. $C_{16}H_{16}N_2O_3$. Calculated, %: N 9.85.

1-(2-Isopropyl-5-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)-3-(4-methoxyphenyl)urea (IVr). Yield 10%, mp 146–148°C. ^{1}H NMR spectrum, δ, ppm: 1.20 d [6H, (C**H**$_3$)$_2$CH, J = 6.9 Hz], 2.01 d (3H, CH_3, ^4J = 1.2 Hz), 3.26–3.35 m [1H, (CH$_3$)$_2$C**H**], 3.82 s (3H, MeO), 6.51 s (1H, 3-H), 7.11 br.s (1H, 6-H), 6.92–7.48 d.d (4H, C_6H_4, J = 9 Hz), 7.37 br.s (1H, NH). Found, %: N 8.82, 8.95. $C_{18}H_{20}N_2O_3$. Calculated, %: N 8.97.

1-(4-Chlorophenyl)-3-(2-isopropyl-5-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVs). Yield 13%, mp 139–141°C. ^{1}H NMR spectrum, δ, ppm: 1.20 d [6H, (C**H**$_3$)$_2$CH, J = 6.9 Hz], 2.01 d (3H, CH_3, ^4J = 1.2 Hz), 3.24–3.33 m [1H, (CH$_3$)$_2$C**H**], 6.52 s (1H, 3-H), 7.01 br.s (1H, NH), 7.08 br.s (1H, 6-H), 7.34–7.53 d.d (4H, C_6H_4, J = 8.7 Hz). Found, %: N 8.61, 8.80. $C_{17}H_{17}ClN_2O_2$. Calculated, %: N 8.84.

1-Cyclohexyl-3-(2-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVt). Yield 20%, mp 118–120°C. ^{1}H NMR spectrum, δ, ppm: 1.17–3.85 m (11H, C_6H_{11}), 2.18 br.s (3H, CH_3), 5.17 d (1H, NH, J = 7.8 Hz), 6.47–6.50 d.d (1H, 5-H, ^4J = 2.1, ^3J = 9.6 Hz), 7.13 d (1H, 6-H), 6.51 q (1H, 3-H). Found, %: N 11.34, 11.55. $C_{14}H_{18}N_2O_2$. Calculated, %: N 11.37.

1-Cyclohexyl-3-(2,5-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVu). Yield 18%, mp 113–116°C. ^{1}H NMR spectrum, δ, ppm: 1.06–3.81 m (11H, C_6H_{11}), 1.99 d (3H, 5-CH_3, ^4J = 1.8 Hz), 2.14 d (3H, 2-CH_3, ^4J = 1.5 Hz), 5.15 d (1H, NH, J = 8.1 Hz), 6.49 d (1H, 3-H), 6.59 d (1H, 6-H). Found, %: N 10.69, 10.87. $C_{15}H_{20}N_2O_2$. Calculated, %: N 10.76.

1-Cyclohexyl-3-(2,6-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVv). Yield 63%, mp 115–117°C. ^{1}H NMR spectrum, δ, ppm: 1.12–3.79 m (11H, C_6H_{11}), 2.20 s (6H, CH_3), 5.00 d (1H, NH, J = 7.8 Hz), 6.38 s (2H, 3-H, 5-H). Found, %: N 10.54, 10.73. $C_{15}H_{20}N_2O_2$. Calculated, %: N 10.76.

1-Butyl-3-(4-oxocyclohexa-2,5-dien-1-ylidene)-urea (IVw). Yield 35%, mp 110–112°C. ^{1}H NMR spectrum, δ, ppm: 0.94–3.41 m (9H, Bu), 5.32 q (1H, NH), 6.55–6.58 d.d (1H, 5-H, ^4J = 2.1, ^3J = 10.2 Hz), 6.65–6.68 d.d (1H, 3-H), 7.05–7.09 d.d (1H, 6-H, ^4J = 2.1 Hz), 7.25–7.28 d.d (1H, 2-H). Found, %: N 13.46, 13.62. $C_{11}H_{14}N_2O_2$. Calculated, %: N 13.58.

1-Butyl-3-(2,6-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVy). Yield 35%, mp 110–112°C. ^{1}H NMR spectrum, δ, ppm: 0.90–3.40 m (9H, Bu), 2.19 s (6H, CH_3), 5.17 q (1H, NH), 6.38 s (2H, 3-H, 5-H). Found, %: N 12.06, 12.28. $C_{13}H_{18}N_2O_2$. Calculated, %: N 11.96.

1-Butyl-3-(2-isopropyl-5-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVz). Yield 18%, mp 113–115°C. ^{1}H NMR spectrum, δ, ppm: 0.95–3.43 m (9H, Bu), 1.16 d [6H, (C**H**$_3$)$_2$CH, J = 6.9 Hz], 1.20 d (3H, CH_3, ^4J = 1.2 Hz), 3.21–3.30 m [1H, (CH$_3$)$_2$C**H**], 5.14 q (1H, NH), 6.47 s (1H, 3-H), 6.94 br.s (1H, 6-H). Found, %: N 10.55, 10.71. $C_{15}H_{22}N_2O_2$. Calculated, %: N 10.68.

1-*tert*-Butyl-3-(3-methyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVaa). Yield 12%, mp 138–142°C. ^{1}H NMR spectrum, δ, ppm: *E* isomer (42%): 1.44 s (9H, *t*-Bu), 2.06 s (3H, CH_3), 5.14 br.s (1H, NH), 6.55 d (1H, 5-H, J = 10.5 Hz), 6.85 br.s (1H, 2-H, J =

2.7 Hz), 7.18–7.21 d.d (1H, 6-H); *Z* isomer (58%): 1.45 s (9H, *t*-Bu), 2.04 s (3H, CH_3), 5.14 q (1H, NH), 6.65 d (1H, 5-H, J = 9.9 Hz), 6.96–7.00 d.d (1H, 6-H, ^4J = 2.7 Hz), 7.06 br.s (1H, 2-H). Found, %: N 12.68, 12.80. $C_{12}H_{16}N_2O_2$. Calculated, %: N 12.72.

1-*tert*-Butyl-3-(2,6-dimethyl-4-oxocyclohexa-2,5-dien-1-ylidene)urea (IVab). Yield 15%, mp 153–155°C. ^{1}H NMR spectrum, δ, ppm: 1.44 s (9H, *t*-Bu), 2.20 s (6H, CH_3), 4.96 q (1H, NH), 6.37 s (2H, 3-H, 5-H). Found, %: N 11.85, 11.99. $C_{13}H_{18}N_2O_2$. Calculated, %: N 11.96.

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