Russian Journal of Organic Chemistry, Vol. 40, No. 6, 2004, pp. 879–883. Translated from Zhurnal Organicheskoi Khimii, Vol. 40, No. 6, 2004, pp. 917–921.
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Dedicated to Full Member of the Russian Academy of Sciences O.N. Chupakhin on his 70th Anniversary

Synthesis of 1-Substituted 3-Alkyl-1,2,3-triazol-3-ium-5-olates

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Received March 10, 2004

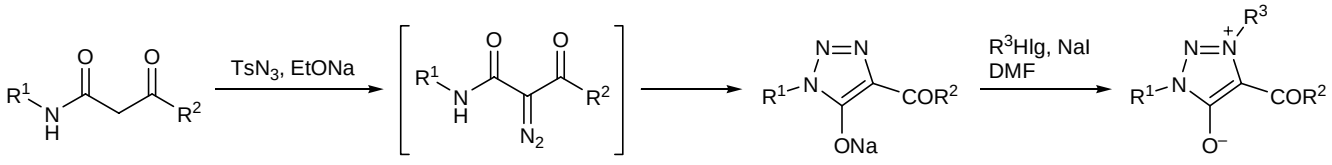
Abstract—Alkylation of 1-substituted sodium 1,2,3-triazol-5-olates with halogen derivatives occurs at the nitrogen atom in position *3* of the heteroring to give zwitterionic 1,2,3-triazol-3-ium-5-olates.

1,2,3-Triazole derivatives exhibit various kinds of physiological and biological activity [1], specifically antitumor [2], antiviral [3], antiphlogistic, etc. [4]. Zwitterionic fused 1,2,3-triazoles were shown to act as immunosuppressants [5]. Search for new derivatives possessing biological and interesting technical properties has been continued in the recent years. We previously showed that alkylation of sodium 1-aryl- and 1-arylmethyleneamino-1,2,3-triazol-5-olates with alkyl halides gives zwitterionic 3-alkyl-1,2,3-triazol-3-ium-5-olates [6] and [1,2,3]triazolo[1,5-*a*]pyrazinium-3-olates [7]. While extending these studies, the present communication reports on the synthesis of 1-alkyl-3-aryl-1,2,3-triazol-3-ium-4-olates.

1-Substituted sodium 1,2,3-triazol-5-olates **IIa**–**IIe** were synthesized by diazo transfer reaction [8] with malonamides **Ia**–**Ie**. The corresponding diazo compounds were not isolated, and they underwent intramolecular cyclization by the action of sodium ethoxide. The cyclic structure of the products follows mainly from the absence in their IR spectra of characteristic diazo group absorption at 2100 cm^{-1}. In addition, similar substituents at the N^1 atom and carboxamide moiety in position *4* in compounds **IIa**–**IIc** give different signals in the ^1H NMR spectra. In the spectra of **IId** and **IIe** we observed a signal at δ ~9 ppm from the CH=N proton, which is typical of cyclic structure [9].

The alkylation of sodium 1-phenyl-4-phenylcarbamoyl-1,2,3-triazol-5-olate (**IIa**) with chloromethyloxirane in DMF at 70–80°C in the presence of sodium iodide as catalyst afforded a single product in a good yield (79%). According to the data of elemental analysis and IR, NMR, and mass spectra, the alkylation occurred with conservation of the oxirane ring. In the ^{13}C NMR spectrum of **IIIa**, the C^5 signal appeared

Scheme 1.



Ia–Ie **IIa–IIe** **IIIa–IIIaa**

I, **II**, R^1 = Ph, R^2 = PhNH (**a**); R^1 = 4-MeC_6H_4, R^2 = 4-MeC_6H_4NH (**b**); R^1 = 4-$MeOC_6H_4$, R^2 = 4-$MeOC_6H_4NH$ (**c**); R^1 = PhCH=N, R^2 = EtO (**d**); R^1 = 4-ClC_6H_4CH=N, R^2 = EtO (**e**); **III**, R^1 = Ph, R^2 = PhNH, R^3 = oxiranylmethyl (**a**), Me (**b**), Et (**c**), Bu (**d**), $PhCH_2$ (**e**), 4-$MeOC_6H_4COCH_2$ (**f**), $PhCOCH_2$ (**g**), $MeCOCH_2$ (**h**), $EtOCOCH_2$ (**i**), 4-piperidinylmethyl (**j**); R^1 = 4-MeC_6H_4, R^2 = 4-MeC_6H_4NH, R^3 = Me (**k**), Et (**l**), $NCCH_2$ (**m**), $PhCH_2$ (**n**), $PhCOCH_2$ (**o**), $MeCOCH_2$ (**p**), 4-piperidinylmethyl (**q**); R^1 = 4-$MeOC_6H_4$, R^2 = 4-$MeOC_6H_4NH$, R^3 = Me (**r**), Et (**s**), $NCCH_2$ (**t**), $PhCH_2$ (**u**), 4-$MeOC_6H_4COCH_2$ (**v**), $MeCOCH_2$ (**w**), $EtOCOCH_2$ (**x**), 4-piperidinylethyl (**y**); R^1 = PhCH=N, R^2 = EtO, R^3 = $PhCH_2$ (**z**); R^1 = 4-ClC_6H_4CH=N, R^2 = EtO, R^3 = $PhCH_2$ (**aa**).

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as a singlet at δ_C 156.1 ppm, and the C^4 signal was a triplet at δ_C 111.3 ppm with a coupling constant of 1.2 Hz. These data indicate that the alkylation involves the N^3 atom. Otherwise, i.e., in the case of alkylation at N^2, no such coupling would be observed, while O-alkylation product should be characterized by a triplet signal from C^5 and singlet from C^4. We also found that alkylation of triazolates **IIa**–**IIe** with various halogen derivatives, such as alkyl chlorides, phenacyl halides, and chloroacetic acid derivatives, also results in formation of a single product. Analysis of the chemical shifts of the methylene and methyl protons in the ^{1}H NMR spectra of compounds **IIIb**–**IIIaa** allowed us to assign zwitterionic structure to the products.

EXPERIMENTAL

The progress of reactions and the purity of products were monitored by TLC on Silufol UV-254 plates using the following solvent systems: chloroform, chloroform–ethanol (9:1, 15:1, 20:1), and ethyl acetate–hexane (1.5:2, 1:2). The IR spectra were measured in KBr on a UR-20 spectrometer. The NMR spectra were recorded from solutions in DMSO-d_6–CCl_4 on Bruker WM-250 (250 MHz for ^{1}H) and Bruker DRX-500 (500 MHz for ^{1}H and 125 MHz for ^{13}C) spectrometers using tetramethylsilane as internal reference. The mass spectra (electron impact, 70 eV) were obtained on Varian MAT-311A and Finnigan MAT-8200 instruments with direct sample admission into the ion source. The solvents were dried and purified by standard procedures.

General procedure for the synthesis of sodium 1-aryl-1,2,3-triazol-5-olates IIa–IIe. To a solution of 0.01 mol of malonamide **Ia**–**Ie** in 50 ml of a sodium ethoxide solution prepared from 0.23 g (0.01 mol) of metallic sodium we added 1.97 ml (0.01 mol) of *p*-toluenesulfonyl azide. The mixture was stirred for 2 h and evaporated under reduced pressure, 200 ml of water was added to the residue, undissolved *p*-toluenesulfonamide was filtered off, the filtrate was evaporated to dryness under reduced pressure, and the residue was dried over P_2O_5.

Sodium 1-phenyl-4-phenylcarbamoyl-1,2,3-triazol-5-olate (IIa). Yield 2.11 g (70%), mp >250°C. ^{1}H NMR spectrum, δ, ppm: 10.67 s (1H, NH), 8.07 d.d (2H, H_{arom}, J = 7.5, 1.2 Hz), 7.70 d.d (1H, H_{arom}, J = 4.8, 1.7 Hz), 7.63 d.d (2H, H_{arom}, J = 7.5, 1.1 Hz), 7.31–7.22 m (4H, H_{arom}), 6.99–6.94 m (1H, H_{arom}). Found, %: N 18.80. $C_{15}H_{12}N_4NaO_2$. Calculated, %: N 18.54.

Sodium 1-*p*-tolyl-4-(*p*-tolylcarbamoyl)-1,2,3-triazol-5-olate (IIb). Yield 2.41 g (73%), mp >250°C. ^{1}H NMR spectrum, δ, ppm: 10.12 s (1H, NH), 7.69 d (2H, H_{arom}, J = 8.6 Hz), 7.66 d (2H, H_{arom}, J = 8.6 Hz), 7.39 d (2H, H_{arom}, J = 8.2 Hz), 7.15 d (2H, H_{arom}, J = 8.2 Hz), 2.39 s (3H, CH_3), 2.28 s (3H, CH_3). Found, %: N 16.81. $C_{17}H_{15}N_4NaO_2$. Calculated, %: N 16.96.

Sodium 1-*p*-methoxyphenyl-4-(*p*-methoxyphenylcarbamoyl)-1,2,3-triazol-5-olate (IIc). Yield 2.62 g (77%), mp >250°C. ^{1}H NMR spectrum, δ, ppm: 10.39 s (1H, NH), 7.92 d (2H, H_{arom}, J = 9.1 Hz), 7.56 d (2H, H_{arom}, J = 9.1 Hz), 7.01 d (2H, H_{arom}, J = 9.1 Hz), 6.87 d (2H, H_{arom}, J = 9.1 Hz), 3.78 s (3H, CH_3), 3.73 s (3H, CH_3). Found, %: N 15.70. $C_{17}H_{15}N_4NaO_4$. Calculated, %: N 15.46.

Sodium 1-benzylideneamino-4-ethoxycarbonyl-1,2,3-triazol-5-olate (IId). Yield 1.80 g (69%), mp >250°C. ^{1}H NMR spectrum, δ, ppm: 9.36 s (1H, N=CH), 7.44–7.88 m (5H, H_{arom}), 4.28 q (2H, OCH_2, J = 7.0 Hz), 1.32 t (3H, CH_3, J = 7.0 Hz). Found, %: N 20.20. $C_{12}H_{11}N_4NaO_3$. Calculated, %: N 19.85.

Sodium 1-(4-chlorobenzylideneamino)-4-ethoxycarbonyl-1,2,3-triazol-5-olate (IIe). Yield 1.62 g (58%), mp >250°C. ^{1}H NMR spectrum, δ, ppm: 9.46 s (1H, N=CH), 7.14 d (2H, H_{arom}), 7.88 d (2H, H_{arom}), 4.32 q (2H, OCH_2, J = 7.0 Hz), 1.28 t (3H, Me, J = 7.0 Hz). Found, %: N 17.70. $C_{12}H_{10}ClN_4NaO_3$. Calculated, %: N 17.69.

General procedure for the synthesis of 3-alkyl-1,2,3-triazol-3-ium-5-olates IIIa–IIIz. To a suspension of 1 mmol of sodium salt **IIa**–**IIe** in 1 ml of DMF we added 3 mmol of alkyl halide and 1.5 mg (0.01 mmol) of sodium iodide, and the mixture was heated for 3 h at 100°C. The mixture was cooled to room temperature and diluted with 50 ml of water, and the precipitate was filtered off and recrystallized from alcohol.

3-Oxiranylmethyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIa). Yield 2.80 g (79%), mp 162°C. ^{1}H NMR spectrum, δ, ppm: 10.48 s (1H, NH), 8.00 d (2H, H_{arom}, J = 7.6 Hz), 7.21–7.66 m (7H, H_{arom}), 7.08 d.d (1H, H_{arom}), 5.70 d (1H, CH, J = 5.8 Hz), 5.04 d.d (1H, NCH, J = 3.3, 12.8 Hz), 4.64 d.d (1H, NCH, J = 8.2, 12.8 Hz), 4.25–4.28 m (1H, CH), 3.50–3.68 m (1H, CH). ^{13}C NMR spectrum, δ_C, ppm: 156.1 s (C^5), 155.6 d (C^{12}, J = 1.4 Hz), 137.7 t (C^6, J = 7.7 Hz), 134.6 t (C^{13}, J = 8.3 Hz),

129.4 d (C^7, C^{11}, J = 160.4 Hz), 129.1 d (C^{14}, C^{18}, J = 158.7 Hz), 128.7 d (C^9, J = 156.0 Hz), 124.0 d (C^{16}, J = 159.9 Hz), 121.3 d (C^8, C^{10}, J = 163.7 Hz), 119.4 d (C^{15}, C^{17}, J = 159.5 Hz), 111.3 t (C^4, J = 1.2 Hz), 68.2 d (C^2, J = 147.8 Hz), 56.9 t (C^1, J = 146.0 Hz), 46.9 t (C^3, J = 151.0 Hz). Found, %: N 16.8. $C_{18}H_{16}N_4O_3$. Calculated, %: N 16.66.

3-Methyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIb). Yield 0.22 g (76%), mp 150°C. ^{1}H NMR spectrum, δ, ppm: 10.36 s (1H, NH), 7.98 d (2H, H_{arom}, J = 7.5 Hz), 7.63 d (2H, H_{arom}, J = 7.5 Hz), 7.57 t (2H, H_{arom}, J = 7.5 Hz), 7.46 t (1H, H_{arom}, J = 7.5 Hz), 7.33 t (2H, H_{arom}, J = 7.5 Hz), 7.08 t (1H, H_{arom}, J = 7.5 Hz), 4.41 s (3H, CH_3). Found, %: N 18.81. $C_{16}H_{14}N_4O_2$. Calculated, %: N 19.04.

3-Ethyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIc). Yield 0.25 g (82%), mp 140°C. ^{1}H NMR spectrum, δ, ppm: 10.46 s (1H, NH), 8.00 d (2H, H_{arom}, J = 7.5 Hz), 7.65–7.55 m (4H, H_{arom}), 7.46 t (1H, H_{arom}, J = 7.4 Hz), 7.33 t (2H, H_{arom}, J = 7.5 Hz), 7.08 t (1H, H_{arom}, J = 7.4 Hz), 4.86 q (2H, CH_2, J = 7.2 Hz), 1.61 t (3H, CH_3, J = 7.2 Hz). Found, %: N 18.18. $C_{17}H_{16}N_4O_2$. Calculated, %: N 18.17.

3-Butyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIId). Yield 0.27 g (79%), mp 110°C. IR spectrum: ν 1680 cm^{-1} (C=O). ^{1}H NMR spectrum, δ, ppm: 10.47 s (1H, NH), 7.98 d.d (2H, H_{arom}, J = 7.5, 1.3 Hz), 7.65–7.55 m (4H, H_{arom}), 7.46 t (1H, H_{arom}, J = 7.4 Hz), 7.33 d (2H, H_{arom}, J = 7.5 Hz), 7.08 t (1H, H_{arom}, J = 7.4 Hz), 4.82 t (2H, CH_2, J = 7.2 Hz), 1.98 p (2H, CH_2, J = 7.4 Hz), 1.48–1.42 m (2H, CH_2), 1.00 t (3H, CH_3, J = 7.3 Hz). Found, %: N16.81. $C_{19}H_{20}N_4O_2$. Calculated, %: N 16.65.

3-Benzyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIe). Yield 0.27 g (73%), mp 140°C. ^{1}H NMR spectrum, δ, ppm: 10.49 s (1H, NH), 7.97 d (2H, H_{arom}, J = 8.5 Hz), 7.64–7.60 m (4H, H_{arom}), 7.54–7.48 m (3H, H_{arom}), 7.41–7.35 m (5H, H_{arom}), 7.13 t (1H, H_{arom}, J = 7.4 Hz), 6.07 s (2H, CH_2). Found, %: N 15.18. $C_{22}H_{18}N_4O_2$. Calculated, %: N 15.13.

3-*p*-Methoxyphenacyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIf). Yield 0.21 g (50%), mp 180°C. ^{1}H NMR spectrum, δ, ppm: 10.30 s (1H, NH), 8.09 d.d (2H, H_{arom}, J = 5.0, 1.9 Hz), 8.02 d.d (2H, H_{arom}, J = 7.4, 1.3 Hz), 7.65 t (2H, H_{arom}, J = 7.4 Hz), 7.56 t (3H, H_{arom}, J = 7.5 Hz), 7.33 t (2H, H_{arom}, J = 7.5 Hz), 7.16 d (2H, H_{arom}, J = 7.5 Hz), 7.10 t (1H, H_{arom}, J = 7.4 Hz), 6.45 s (2H, CH_2), 3.90 s (3H, CH_3). Found, %: N 13.08. $C_{24}H_{20}N_4O_2$. Calculated, %: N 13.08.

3-Phenacyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIg). Yield 0.27 g (67%), mp 184°C. ^{1}H NMR spectrum, δ, ppm: 10.28 s (1H, NH), 8.12 d (2H, H_{arom}, J = 7.1 Hz), 8.03–8.00 m (2H, H_{arom}), 7.77 t (1H, H_{arom}, J = 7.4 Hz), 7.65 t (4H, H_{arom}, J = 7.5 Hz), 7.58–7.53 m (3H, H_{arom}), 7.33 t (2H, H_{arom}, J = 7.5 Hz), 7.10 t (2H, H_{arom}, J = 7.4 Hz), 6.51 s (2H, CH_2). Mass spectrum: m/z 398 $[M]^+$. Found, %: N 13.89. $C_{23}H_{18}N_4O_2$. Calculated, %: N 14.06.

3-Acetonyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIh). Yield 0.23 g (67%), mp 184°C. ^{1}H NMR spectrum, δ, ppm: 10.23 s (1H, NH), 8.00 d (2H, H_{arom}, J = 7.3 Hz), 7.59 t (4H, H_{arom}, J = 7.0 Hz), 7.48 t (1H, H_{arom}, J = 7.3 Hz), 7.33 d (2H, H_{arom}, J = 7.3 Hz), 7.08 t (1H, H_{arom}, J = 7.3 Hz), 5.74 s (2H, CH_2), 2.37 s (3H, CH_3). Found, %: N 16.78. $C_{18}H_{16}N_4O_2$. Calculated, %: N 16.66.

3-Ethoxycarbonylmethyl-1-phenyl-4-phenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIi). Yield 0.17 g (46%), mp 142°C. ^{1}H NMR spectrum, δ, ppm: 10.24 s (1H, NH), 7.98 d (2H, H_{arom}, J = 7.7 Hz), 7.67–7.61 m (4H, H_{arom}), 7.55 t (1H, H_{arom}, J = 7.5 Hz), 7.38 t (2H, H_{arom}, J = 7.7 Hz), 7.14 t (1H, H_{arom}, J = 7.3 Hz), 5.70 s (2H, CH_2), 4.24 q (2H, CH_2, J = 7.0 Hz), 1.25 t (3H, CH_3, J = 7.0 Hz). Mass spectrum: m/z 366 $[M]^+$. Found, %: N 15.15. $C_{19}H_{18}N_4O_2$. Calculated, %: N 15.29.

1-Phenyl-4-phenylcarbamoyl-3-(4-piperidinylmethyl)-1*H*-1,2,3-triazol-3-ium-5-olate (IIIj). Yield 0.22 g (56%), mp 208°C. IR spectrum: ν 1690 cm^{-1} (C=O). ^{1}H NMR spectrum, δ, ppm: 10.47 s (1H, NH), 10.17 br.s (1H, NH), 7.97 d.d (2H, H_{arom}, J = 7.4, 1.2 Hz), 7.68–7.52 m (5H, H_{arom}), 7.42–7.37 m (2H, H_{arom}), 7.15 d.d (1H, H_{arom}, J = 7.4, 1.1 Hz), 5.26 t (2H, CH_2, J = 5.8 Hz), 3.76–3.40 m (4H, CH_2), 3.02 br.s (2H, CH_2), 1.83–1.42 m (6H, CH_2). Found, %: N 15.80. $C_{22}H_{25}N_5O_2$. Calculated, %: N 16.37.

3-Methyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIk). Yield 0.25 g (75%), mp 218°C. ^{1}H NMR spectrum, δ, ppm: 10.28 s (1H, NH), 7.84 d (2H, H_{arom}, J = 8.4 Hz), 7.50 d (2H, H_{arom}, J = 8.5 Hz), 7.35 d (2H, H_{arom}, J = 8.4 Hz), 7.12 d (2H, H_{arom}, J = 8.5 Hz), 4.38 s (3H, CH_3), 2.41 s (3H, CH_3), 2.31 s (3H, CH_3). Found, %: N 17.48. $C_{18}H_{18}N_4O_2$. Calculated, %: N 17.38.

3-Ethyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIl). Yield 0.27 g (81%),

mp 160°C. ^{1}H NMR spectrum, δ, ppm: 10.30 s (1H, NH), 7.82 d (2H, H_{arom}, J = 8.5 Hz), 7.50 d (2H, H_{arom}, J = 8.5 Hz), 7.36 d (2H, H_{arom}, J = 8.5 Hz), 7.12 d (2H, H_{arom}, J = 8.5 Hz), 4.92 q (2H, CH_2, J = 7.0 Hz), 2.41 s (3H, CH_3), 2.31 s (3H, CH_3), 1.59 t (3H, CH_3, J = 7.0 Hz). Found, %: N 16.80. $C_{19}H_{20}N_4O_2$. Calculated, %: N 16.66.

3-Cyanomethyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIm). Yield 0.31 g (56%), mp 205°C. ^{1}H NMR spectrum, δ, ppm: 10.15 s (1H, NH), 7.85 d (2H, H_{arom}, J = 8.24 Hz), 7.54 d (2H, H_{arom}, J = 8.24 Hz), 7.38 d (2H, H_{arom}, J = 8.24 Hz), 7.15 d (2H, H_{arom}, J = 8.24 Hz), 6.09 s (2H, CH_2). Found, %: N 20.10. $C_{19}H_{17}N_5O_2$. Calculated, %: N 20.18.

3-Benzyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIn). Yield 0.31 g (79%), mp 170°C. ^{1}H NMR spectrum, δ, ppm: 10.43 s (1H, NH), 7.84 d (2H, H_{arom}, J = 8.47 Hz), 7.52 d (2H, H_{arom}, J = 8.43 Hz), 7.47 d (2H, H_{arom}, J = 8.31 Hz), 7.42–7.35 m (4H, H_{arom}), 7.17 d (2H, H_{arom}, J = 8.21 Hz), 2.39 s (3H, CH_3), 2.28 s (3H, CH_3). Found, %: N 14.25. $C_{24}H_{22}N_4O_2$. Calculated, %: N 14.06.

3-Phenacyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIo). Yield 0.30 g (71%), mp 195°C. ^{1}H NMR spectrum, δ, ppm: 10.21 s (1H, NH), 8.09 d (2H, H_{arom}, J = 8.55 Hz), 7.90 d (2H, H_{arom}, J = 8.54 Hz), 7.73 t (1H, H_{arom}, J = 7.42 Hz), 7.60 t (2H, H_{arom}, J = 7.94 Hz), 7.45–7.35 m (4H, H_{arom}), 7.06 d (2H, H_{arom}, J = 8.25 Hz), 6.39 s (2H, CH_2), 3.25 s (3H, CH_3), 3.28 s (3H, CH_3). Found, %: N 13.15. $C_{25}H_{22}N_4O_3$. Calculated, %: N 13.14.

3-Acetonyl-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIp). Yield 0.26 g (71%), mp 208°C. IR spectrum: ν 1680 cm^{-1} (C=O). ^{1}H NMR spectrum, δ, ppm: 10.16 s (1H, NH), 7.86 d (2H, H_{arom}, J = 8.54 Hz), 7.47 d (2H, H_{arom}, J = 8.24 Hz), 7.36 d (2H, H_{arom}, J = 8.24 Hz), 7.61 d (2H, H_{arom}, J = 8.24 Hz), 5.70 s (2H, CH_2), 2.42 s (3H, CH_3), 2.35 s (3H, CH_3), 2.29 s (3H, CH_3). Found, %: N 15.55. $C_{20}H_{20}N_4O_3$. Calculated, %: N 15.37.

3-(4-Piperidinylmethyl)-1-*p*-tolyl-4-*p*-tolylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIq). Yield 0.33 g (78%), mp 255°C. ^{1}H NMR spectrum, δ, ppm: 10.6 s (1H, NH), 7.85 d (2H, H_{arom}, J = 8.5 Hz), 7.52 d (2H, H_{arom}, J = 8.5 Hz), 7.36 d (2H, H_{arom}, J = 8.24 Hz), 7.14 d (2H, H_{arom}, J = 8.55 Hz), 5.23 t (2H, CH_2, J = 5.50 Hz), 3.78–3.49 m (2H, CH_2), 3.24–2.86 m (6H, CH_2), 2.43 s (3H, CH_3), 2.32 s (3H, CH_3), 1.89–1.56 m (4H, CH_2). Mass spectrum, m/z (I_{rel}, %): 419 (8) $[M]^+$. Found, %: N 16.78. $C_{24}H_{29}N_4O_2$. Calculated, %: N 16.69.

1-*p*-Methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-3-methyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIr). Yield 0.28 g (79%), mp 162°C. ^{1}H NMR spectrum, δ, ppm: 10.18 s (1H, NH), 7.87 d (2H, H_{arom}, J = 8.5 Hz), 7.47 d (2H, H_{arom}, J = 8.4 Hz), 7.35 d (2H, H_{arom}, J = 8.4 Hz), 7.60 d (2H, H_{arom}, J = 8.5 Hz), 4.38 s (3H, CH_3), 2.42 s (3H, CH_3), 3.85 s (3H, CH_3), 3.79 s (3H, CH_3). Found, %: N 15.80. $C_{18}H_{18}N_4O_4$. Calculated, %: N 15.81.

3-Ethyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIs). Yield 0.29 g (80%), mp 192°C. ^{1}H NMR spectrum, δ, ppm: 10.34 s (1H, NH), 7.85 d (2H, H_{arom}, J = 9.1 Hz), 7.53 d (2H, H_{arom}, J = 8.8 Hz), 7.07 d (2H, H_{arom}, J = 9.1 Hz), 6.87 d (2H, H_{arom}, J = 8.8 Hz), 4.92 q (2H, CH_2, J = 7.0 Hz), 3.85 s (3H, CH_3), 3.76 s (3H, CH_3), 1.59 t (3H, CH_3, J = 7.0 Hz). Mass spectrum: m/z 368 $[M]^+$. Found, %: N 15.50. $C_{19}H_{20}N_4O_4$. Calculated, %: N 15.21.

3-Cyanomethyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIt). Yield 0.29 g (80%), mp 180°C. ^{1}H NMR spectrum, δ, ppm: 10.41 s (1H, NH), 7.86 d (2H, H_{arom}, J = 9.0 Hz), 7.53 d (2H, H_{arom}, J = 8.8 Hz), 7.08 d (2H, H_{arom}, J = 9.0 Hz), 6.86 d (2H, H_{arom}, J = 8.8 Hz), 5.99 s (2H, CH_2), 3.85 s (3H, CH_3), 3.77 s (3H, CH_3). Mass spectrum: m/z 379 $[M]^+$. Found, %: N 18.34. $C_{19}H_{17}N_5O_4$. Calculated, %: N 18.46.

3-Benzyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIu). Yield 0.32 g (75%), mp 154°C. IR spectrum: ν 1680 cm^{-1} (C=O). ^{1}H NMR spectrum, δ, ppm: 10.34 s (1H, NH), 7.85 d (2H, H_{arom}, J = 9.1 Hz), 7.56–7.32 m (7H, H_{arom}), 7.05 d (2H, H_{arom}, J = 9.1 Hz), 6.86 d (2H, H_{arom}, J = 8.9 Hz), 6.03 s (2H, CH_2), 3.84 s (3H, CH_3), 3.76 s (3H, CH_3). Mass spectrum: m/z 431 $[M + 1]$. Found, %: N 13.00. $C_{24}H_{22}N_4O_4$. Calculated, %: N 13.02.

3-*p*-Methoxyphenacyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIv). Yield 0.24 g (48%), mp 220°C. ^{1}H NMR spectrum, δ, ppm: 10.17 s (1H, NH), 8.06 d (2H, H_{arom}, J = 8.8 Hz), 7.92 d (2H, H_{arom}, J = 9.1 Hz), 7.47 d (2H, H_{arom}, J = 9.1 Hz), 7.40 d (2H, H_{arom}, J = 7.0 Hz), 7.07 d (4H, H_{arom}, J = 7.0 Hz), 6.81 d (2H, H_{arom}, J = 8.8 Hz), 6.32 s (2H, CH_2), 3.91 s (3H, CH_3), 3.86 s

(3H, CH_3), 3.74 s (3H, CH_3). Found, %: N 11.23. $C_{26}H_{24}N_4O_6$. Calculated, %: N 11.47.

3-Acetonyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIw). Yield 0.30 g (75%), mp 186°C. ^{1}H NMR spectrum, δ, ppm: 10.11 s (1H, NH), 7.87 d (2H, H_{arom}, J = 9.1 Hz), 7.50 d (2H, H_{arom}, J = 8.8 Hz), 7.07 d (2H, H_{arom}, J = 9.1 Hz), 6.85 d (2H, H_{arom}, J = 8.8 Hz), 5.68 s (2H, CH_2), 3.85 s (3H, CH_3), 3.76 s (3H, CH_3), 2.35 s (3H, CH_3). Found, %: N 14.18. $C_{20}H_{20}N_4O_5$. Calculated, %: N 14.13.

3-Ethoxycarbonylmethyl-1-*p*-methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIx). Yield 0.35 g (82%), mp 168°C. ^{1}H NMR spectrum, δ, ppm: 10.10 s (1H, NH), 7.89 d (2H, H_{arom}, J = 9.1 Hz), 7.52 d (2H, H_{arom}, J = 9.1 Hz), 7.08 d (2H, H_{arom}, J = 9.1 Hz), 6.85 d (2H, H_{arom}, J = 9.1 Hz), 5.57 s (2H, CH_2), 4.26 q (2H, CH_2, J = 7.0 Hz), 3.85 s (3H, CH_3), 3.76 s (3H, CH_3), 1.31 t (3H, CH_3, J = 7.0 Hz). Found, %: N 13.12. $C_{21}H_{22}N_4O_6$. Calculated, %: N 13.14.

1-*p*-Methoxyphenyl-4-*p*-methoxyphenylcarbamoyl-3-(4-piperidinylethyl)-1*H*-1,2,3-triazol-3-ium-5-olate (IIIy). Yield 0.34 g (75%), mp 240°C. ^{1}H NMR spectrum, δ, ppm: 10.32 s (1H, NH), 7.86 d (2H, H_{arom}, J = 9.16 Hz), 7.56 d (2H, H_{arom}, J = 9.16 Hz), 7.09 d (2H, H_{arom}, J = 9.15 Hz), 6.89 d (2H, H_{arom}, J = 8.85 Hz), 5.22 t (2H, CH_2, J = 5.51 Hz), 3.87 s (3H, CH_3), 3.78 s (3H, CH_3), 4.00–3.53 m (2H, CH_2), 3.19–2.83 m (4H, CH_2), 2.10–1.41 m (6H, CH_2). Found, %: N 15.82. $C_{24}H_{29}N_5O_4$. Calculated, %: N 15.51.

3-Benzyl-1-benzylideneamino-4-ethoxycarbonyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIz). Yield 0.19 g (55%), mp 98–100°C. ^{1}H NMR spectrum, δ, ppm: 9.68 s (1H, N=CH), 7.91–7.39 m (10H, H_{arom}), 5.88 s (2H, CH_2), 4.24 d (2H, OCH_2, J = 6.0 Hz), 1.23 t (3H, CH_3, J = 6.0 Hz). Found, %: N 15.63. $C_{19}H_{18}N_4O_3$. Calculated, %: N 15.99.

3-Benzyl-3-(4-chlorobenzylideneamino)-4-ethoxycarbonyl-1*H*-1,2,3-triazol-3-ium-5-olate (IIIaa). Yield 0.25 g (65%), mp 136–138°C. ^{1}H NMR spectrum, δ, ppm: 9.68 s (1H, N=CH), 7.94–7.38 m (9H, H_{arom}), 5.88 s (2H, CH_2), 4.28 d (2H, OCH_2, J = 6.8 Hz), 1.23 t (3H, CH_3, J = 6.8 Hz). Found, %: Cl 9.40; N 14.18. $C_{19}H_{17}ClN_4O_3$. Calculated, %: Cl 9.21; N 14.56.

This study was performed under financial support by the Russian Foundation for Basic Research (project no. 04-03-96116r2004Ural_a) and by CRDF (grant nos. RC1-2393-EK-02 and REC-005).

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