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The Reaction of Aroyl-Substituted Heterocyclic Ketene Aminals with Aryl Azides

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ABSTRACT: *The aroyl-substituted heterocyclic ketene aminals* **1** *or* **2** *reacted with p-chlorophenyl azide (***3a***) to give the polysubstituted 1,2,3-triazoles* **4** *or* **5,** *as well as the fused heterocycles* **6** *or* **7.** *Compounds* **1** *and* **2** *reacted with p-nitrophenyl azide (***3b***) much faster, and polysubstituted 1,2,3-triazoles* **8** *or* **9** *were obtained as sole products.* © 2000 John Wiley & Sons, Inc. Heteroatom Chem 11:387–391, 2000

INTRODUCTION

As important intermediates, heterocyclic ketene aminals have shown great potential for the synthesis of a wide variety of new heterocycles and fused heterocycles. Thus, the synthesis and reactions of heterocyclic ketene aminals have attracted much attention [1]. The reactions of heterocyclic ketene aminals with 1,3-dipolar reagents, such as azides [2–7], nitrile imines [8,9], benzonitrile oxide [10,11] or its precursor [12], have been reported. To continue our studies, we report here the results of the reactions of heteroatom aroyl-substituted ketene aminals with aryl azides.

RESULTS AND DISCUSSION

The heterocyclic aroyl groups substituted in ketene aminals used in our reactions are furoyl, thiophenoyl and picolinoyl groups, as well as the *p*-fluorobenzoyl group. Heterocyclic aroyl-substituted ketene aminals **1** and **2** were prepared by the reaction of aroyl ketene-*S,S*-diacetals with diamines according to the literature procedure [13].

Compounds **1** or **2** reacted with *p*-chlorophenyl azide (**3a**) in dioxane at reflux temperature to give the polysubstituted 1,2,3-triazoles **4** or **5**, as well as the fused heterocycles **6** or **7**. (Scheme 1) However, in the case of picolinoyl-substituted heterocyclic ketene aminals **1c** or **2c**, only 1,2,3-triazoles **4c** or **5c** were obtained. When **1** or **2** was reacted with *p*-nitrophenyl azide (**3b**), the rate of reaction was much faster and took place easily at room temperature. The sole products obtained were polysubstituted 1,2,3-triazoles **8** or **9** (Scheme 1). The above results are similar to those of the reactions of benzoyl-substituted heterocyclic ketene aminals with phenyl azides. The polysubstituted 1,2,3-triazoles are formed by the nucleophilic attack of the α-carbon of the ketene aminal on the terminal nitrogen atom of the azide. Then, through the cyclocondensation and aromatization sequences, the fused heterocycles resulted by a 1,3-dipolar addition at first, and then through a Dimroth rearrangement and deamination of chloroaniline [7].

The reaction conditions and the yields of products are listed in Table 1.

The structures of products **4–9** were confirmed by microanalytical data and mass spectra and also by IR and ^{1}H NMR spectra.

1-Methyl-2-[(2-furoyl)methylene]hexahydropyririmidine (**10**) also reacted with **3b** at room temperature to afford the 1,2,3-triazoles **11** (Scheme 2).

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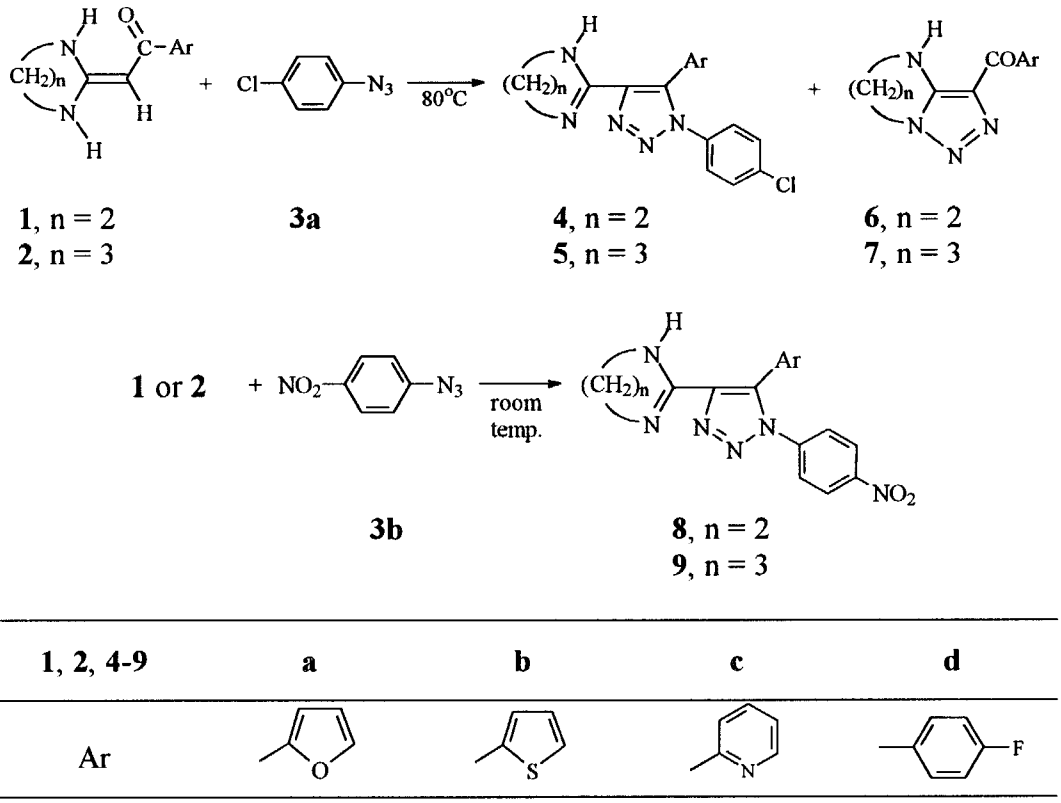


SCHEME 1

TABLE 1. Reaction Conditions and Product Yields

Reactant	*Temp* (°C)	*Time* (*h*)	*Product, Yield* (%)
1a 3a	80	16	**4a** 44; **6a** 23
1b 3a	80	16	**4b** 37; **6b** 25
1c 3a	80	12	**4c** 80
1d 3a	80	16	**4d** 46; **6d** 15
2a 3a	80	12	**5a** 49; **7a** 18
2b 3a	80	12	**5b** 46; **7b** 8
2c 3a	80	12	**5c** 72
2d 3a	80	12	**5d** 37; **7d** 21
1a 3b	r.t.	24	**8a** 57
1b 3b	r.t.	24	**8b** 57
1d 3b	r.t.	24	**8d** 81
2a 3b	r.t.	24	**9a** 63
2d 3b	r.t.	24	**9d** 85

However, when both protons of the heterocyclic ketene aminals substituted by methyl group were used to react with **3b**, only a mixed product that was difficult to separate resulted.

EXPERIMENT

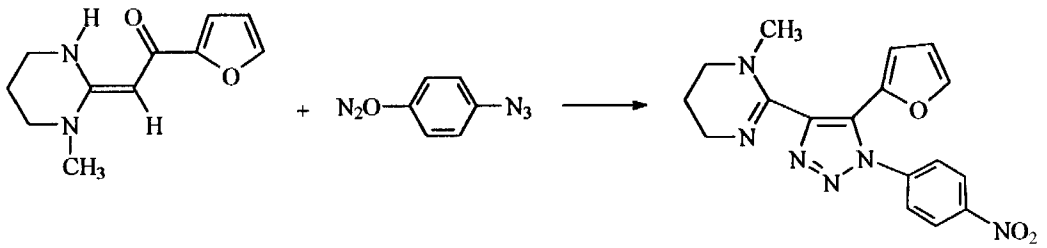
Melting points were uncorrected. ^{1}H NMR spectra were recorded on a Varian Unity 200 spectrometer. IR spectra were recorded on a Perkin-Elmer 782 spectrometer. Mass spectra were obtained on an AEI MS-50 instrument. Elemental Analyses were carried out by the Analytical Laboratory of the Institute.

Preparation of the Ketene Aminals **1** *and* **2**

1 and **2** were prepared by the reaction of heterocyclic aroyl ketene-*S,S*-diacetals with appropriate diamines according to the literature procedure [13].

2-[(2-Furoyl)methylene]imidazolidine. **1a**: Yield 76%; m.p. 184–186°C; IR (KBr): ν 3190, 1603, 1570, 1530, 1480 cm^{-1}; ^{1}H NMR (DMSO-D$_6$): δ 9.01 (s, 1H, NH), 7.64–7.67 (m, 1H), 7.51 (s, 1H, NH), 6.74–6.78 (m, 1H), 6.50–6.55 (m, 1H), 5.18 (s, 1H), 3.50–3.60 (m, 4H); MS: *m/z* 178 [M$^+$] (100), 161 (10), 150 (30). Anal. calcd for $C_9H_{10}N_2O_2$: C, 60.66; H, 5.66; N, 15.72. Found: C, 60.31; H, 5.47; N, 15.61.

2-[(2-Picolinoyl)methylene]imidazolidine. **1c**: Yield 65%; mp 222–223°C; IR (KBr): ν 3184, 1603, 1549, 1505, 1479 cm^{-1}; ^{1}H NMR (DMSO-D$_6$): δ 9.55 (s, 1H, NH), 8.55 (d, 1H), 8.11 (d, 1H), 7.77 (t, 1H), 7.28 (t, 1H), 6.10 (s, 1H), 5.14 (s, 1H, NH), 3.78 (t, 2H), 3.56 (t, 2H); MS: m/z 189 [M$^+$] (23), 161 (16),



10 **3b** **11**

SCHEME 2

111 (100). Anal. calcd for $C_{10}H_{11}N_3O$: C, 63.47; H, 5.86; N, 22.21. Found: C, 63.47; H, 5.92; N, 22.21.

2-[(p-Fluorobenzoyl)methylene]imidazolidine. **1d**: Yield 87%; m.p. 225–226.5°C; IR (KBr): *v* 3280, 1600, 1585, 1550, 1485 cm^{-1}; ^1H NMR (DMSO-D_6): δ 9.25 (s, 1H, NH), 7.76–7.80 (m, 2H), 7.43 (s, 1H, NH), 7.19 (t, 2H), 5.25 (s, 1H), 3.60 (t, 2H), 3.47 (t, 2H); MS: m/z 206 [M^+] (83), 205 (100), 177 (21), 149 (19). Anal. calcd for $C_{11}H_{11}FN_2O$: C, 64.06; H, 5.38; N, 13.59. Found: C, 64.00; H, 5.53; N, 13.31.

2-[(2-Furoyl)methylene]hexahydropyrimidine. **2a**: Yield 81%; m.p. 180–181°C; IR (KBr): *v* 3250, 1620, 1605, 1593, 1520 cm^{-1}; ^1H NMR (DMSO-D_6): δ 10.68 (s, 1H, NH), 7.59–7.61 (m, 1H), 7.50 (br, 1H, NH), 6.63–6.66 (m, 1H), 6.46–6.50 (m, 1H), 5.02 (s, 1H), 3.50–3.56 (m, 4H), 1.81 (quin, 2H); MS: *m/z* 192 [M^+] (100), 175 (11), 163 (20), 138 (60). Anal. calcd for $C_{10}H_{12}N_2O_2$: C, 62.48; H, 6.29; N, 14.58. Found: C, 62.76; H, 6.34; N, 14.79.

2-[(2-Picolinoyl)methylene]hexahydropyrimidine. **2c**: Yield 90%; m.p. 212–213°C; IR (KBr): *v* 3262, 1624, 1599, 1568, 1533 cm^{-1}; ^1H NMR (DMSO-D_6): δ 11.22 (s, 1H, NH), 8.47 (d, 1H), 8.06 (d, 1H), 7.73 (t, 1H), 7.22 (t, 1H), 5.85 (s, 1H), 5.77 (s, 1H, NH), 3.30 (t, 4H), 1.91 (quin, 2H); MS: *m/z* 203 [M^+] (30), 174 (20), 125 (100). Anal. calcd for $C_{11}H_{13}N_3O$: C, 65.01; H, 6.45; N, 20.68. Found: C, 65.14; H, 6.70; N, 20.57.

2-[(p-Fluorobenzoyl)methylene]hexahydropyrimidine. **2d**: Yield 81%; m.p. 222–224°C; IR (KBr): *v* 3240, 1611, 1590, 1522, 1495 cm^{-1}; ^1H NMR (DMSO-D_6): δ 11.07 (s, 1H, NH), 7.68–7.73 (m, 2H), 7.41 (s, 1H, NH), 7.16 (t, 2H), 5.05 (s, 1H), 3.27 (t, 4H), 1.84 (quin, 2H); MS: *m/z* 220 [M^+] (63), 191 (33), 125 (100). Anal. calcd for $C_{12}H_{13}FN_2O$: C, 65.44; H, 5.95; N, 12.72. Found: C, 65.22; H, 5.99; N, 12.71.

1-Methyl-2-[(2-furoyl)methylene]hexahydropyrimidine. **10**: Yield 57%; m.p. 129.5–130.5°C; IR (KBr): *v* 3240, 1605, 1582, 1560, 1485 cm^{-1}; ^1H NMR (DMSO-D_6): δ 11.14 (s, 1H, NH), 7.64–7.66 (m, 1H), 6.74–6.77 (m, 1H), 6.50–6.52 (m, 1H), 5.13 (s, 1H), 3.37 (t, 2H), 3.27 (t, 2H), 2.93 (s, 3H), 1.90 (quin, 2H); MS: *m/z* 206 [M^+] (100), 189 (27), 177 (62), 149 (44). Anal. calcd for $C_{11}H_{14}N_2O_2$: C, 64.06; H, 6.84; N, 13.59. Found: C, 64.54; H, 6.54; N, 13.42.

General Procedure for the Reaction of **1** *or* **2** *with* **3a**. A mixture of **1** or **2** (1 mmol), *p*-chlorophenyl azide (**3a**) (1.2 mmol) and 15 mL of 1.4-dioxane was stirred at 80°C for a period of time (see Table 1). The reaction was monitored by TLC. After the starting material had disappeared, the solvent was removed and the residue was purified by use of a basic aluminum oxide column and gradient elution with petroleum ether (60–90°C)/ethyl acetate. The products were thus separated and recrystallized from petroleum ether (60–90°C)-ethyl acetate.

1-(p-Chlorophenyl)-4-(2-imidazolinyl)-5-(2-furyl)-1,2,3-triazole. **4a**: A white crystal, m.p. 130–131°C; IR (KBr): *v* 3260 (NH), 1620, 1548, 1500 cm^{-1}; ^1H NMR ($CDCl_3$): δ 7.71 (d, 1H), 7.26–7.49 (m, 5H), 6.50–6.52 (m, 1H), 5.20 (br, 1H, NH), 3.82 (s, 4H); MS: m/z 313 [M^+] (90), 312 (61), 285 (100), 250 (45). Anal. calcd for $C_{15}H_{12}ClN_5O$: C, 57.42; H, 3.86; N, 22.32. Found: C, 57.29; H, 3.82; N, 22.24.

1-(p-Chlorophenyl)-4-(2-imidazolinyl)-5-(2-thienyl)-1,2,3-triazole. **4b**: A white crystal, m.p. 193–194°C; IR (KBr): *v* 3360 (NH), 1625, 1600, 1498 cm^{-1}; ^1H-NMR ($CDCl_3$): δ 7.61–7.64 (m, 1H), 7.26–7.46 (m, 5H), 7.04–7.08 (m, 1H), 5.30 (br, 1H, NH), 3.78 (s, 4H); MS: *m/z* 329 [M^+] (32), 328 (100), 300 (41). Anal. calcd for $C_{15}H_{12}ClN_5S$: C, 54.62; H, 3.67; N, 21.24. Found: C, 54.52; H, 3.77; N, 21.62.

1-(p-Chlorophenyl)-4-(2-imidazolinyl)-5-(2-pyridyl)-1,2,3-triazole. **4c**: A white crystal, m.p. 179–180°C; IR (KBr): *v* 3280 (NH), 1630, 1569, 1495 cm^{-1}; ^1H-NMR ($CDCl_3$): δ 8.53–8.57 (m, 1H), 7.73–7.88 (m, 2H), 7.21–7.38 (m, 5H), 5.57 (br, 1H, NH), 3.75 (s,

4H); MS: m/z 324 [M^+] (36), 323 (100), 295 (88). Anal. calcd for $C_{16}H_{13}Cl\ N_6$: C, 59.17; H, 4.03; N, 25.88. Found: C, 59.33; H, 4.16; N, 25.99.

1-(p-Chlorophenyl)-4-(2-imidazolinyl)-5-(p-fluorophenyl)-1,2,3-triazole. **4d**: A white crystal, m.p. 208–209°C; IR (KBr): *v* 3358 (NH), 1612, 1512, 1493 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 7.02–7.41 (m, 8H), 5.18 (br, 1H, NH), 3.73 (s, 4H); MS: *m/z* 341 [M^+] (26), 340 (100), 312 (61). Anal. calcd for $C_{17}H_{13}ClFN_5$: C, 59.74; H, 3.83; N, 20.49. Found: C, 59.15; H, 4.01; N, 20.15.

1-(p-Chlorophenyl)-4-(2-tetrahydropyrimidinyl)-5-(2-furyl)-1,2,3-triazole. **5a**: A white crystal, m.p. 118–119°C; IR (KBr): *v* 3410 (NH), 1628, 1550, 1500 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 7.27–7.46 (m, 6H), 6.45–6.48 (m, 1H), 5.38 (br, 1H, NH), 3.54 (t, 4H, J = 6.0 Hz), 1.90 (quin, 2H, J = 6.0 Hz), MS: *m/z* 327 [M^+] (99), 299 (100), 270 (77). Anal. calcd for $C_{16}H_{14}ClN_5O$: C, 58.63; H, 4.30; N, 21.37. Found: C, 58.64; H, 4.16; N, 21.36.

1-(p-Chlorophenyl)-4-(2-tetrahydropyrimidinyl)-5-(2-thienyl)-1,2,3-triazole. **5b**: A white crystal, m.p. 167–168°C; IR (KBr): *v* 3420 (NH), 1620, 1600, 1530, 1495 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 7.25–7.44 (m, 6H), 6.95–7.00 (m, 1H), 5.18 (br, 1H, NH), 3.48 (t, 4H, J = 6.0 Hz), 1.86 (quin, 2H, J = 6.0 Hz); MS: *m/z* 343 [M^+] (34), 342 (100), 314 (31). Anal. calcd for $C_{16}H_{14}ClN_5S$: C, 55.89; H, 4.10; N, 20.37. Found: C, 55.63; H, 4.23; N, 20.23.

1-(p-Chlorophenyl)-4-(2-tetrahydropyrimidinyl)-5-(2-pyridyl)-1,2,3-triazole. **5c**: A white crystal, m.p. 202–203°C; IR (KBr): *v* 3370 (NH), 1627, 1572, 1511, 1498 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 8.52 (d, 1H), 7.67–7.80 (m, 2H), 7.19–7.35 (m, 5H), 5.78 (br, 1H, NH), 3.44 (t, 4H, J = 6.0 Hz), 1.83 (quin, 2H, J = 6.0 Hz); MS: *m/z* 338 [M^+] (27), 337 (100), 309 (45). Anal. calcd for $C_{17}H_{15}ClN_6$: C, 60.26; H, 4.46; N, 24.81. Found: C, 60.14; H, 4.49; N, 24.83.

1-(p-Chlorophenyl)-4-(2-tetrahydropyrimidinyl)-5-(p-fluorophenyl)-1,2,3-triazole. **5d**: A white crystal, m.p. 176–177°C; IR (KBr): *v* 3400 (NH), 1615, 1518, 1495 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 6.90–7.38 (m, 8H), 3.41 (t, 4H, J = 5.6 Hz), 1.83 (quin, 2H, J = 5.6 Hz); MS: *m/z* 355 [M^+] (34), 354 (100), 326 (84), 292 (27). Anal. calcd for $C_{18}H_{15}ClFN_5$: C, 60.76; H, 4.25; N, 19.69. Found: C, 60.56; H, 4.34; N, 19.30.

3-(2-Furoyl)-5,6-dihydro-4H-imidazo[1,2-c][1,2,3]triazole. **6a**: A white crystal, m.p. 198–199°C; IR (KBr): *v* 3370 (NH), 1625, 1592, 1470 cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 7.92–8.02 (m, 2H), 7.42 (s, 1H, NH), 6.72–6.75 (m, 1H), 4.43, 4.23 (A_2B_2, 4H, J = 9.2 Hz); MS: *m/z* 204 [M^+] (35), 176 (32), 148 (21), 93 (100). Anal. calcd for $C_9H_8N_4O_2$: C, 52.94; H, 3.95; N, 27.44. Found: C, 52.54; H, 3.97; N, 26.97.

3-(2-Thiophenoyl)-5,6-dihydro-4H-imidazo[1,2-c][1,2,3]triazole. **6b**: A white crystal, m.p. 209–210°C; IR (KBr): *v* 3310 (NH), 1625, 1570, 1517 cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.49–8.52 (m, 1H), 7.97–8.00 (m, 1H), 7.44 (s, 1H, NH), 7.27 (t, 1H), 4.44, 4.24 (A_2B_2, 4H, J = 9.2 Hz); MS: *m/z* 220 [M^+] (34), 192 (31), 109 (100). Anal. calcd for $C_9H_8N_4OS$: C, 49.08; H, 3.66; N, 25.44. Found: C, 48.61; H, 3.77; N, 25.10.

3-(p-Fluorobenzoyl)-5,6-dihydro-4H-imidazo[1,2-c][1,2,3]triazole. **6d**: A white crystal, m.p. 214–215°C; IR (KBr): *v* 3335 (NH), 1628, 1600, 1568, 1505 cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.40–8.47 (m, 2H), 7.49 (s, 1H, NH), 7.32–7.41 (m, 2H), 4.46, 4.24 (A_2B_2, 4H, J = 9.2 Hz); MS: *m/z* 232 [M^+] (19), 204 (40), 121 (100). Anal. calcd for $C_{11}H_9FN_4O$: C, 56.89; H, 3.91; N, 24.13. Found: C, 56.94; H, 4.15; N, 24.11.

3-(2-Furoyl)-4,5,6,7-tetrahydro[1,2,3]triazolo-[1,5-a]pyrimidine. **7a**: A white crystal, m.p. 151–152°C; IR (KBr): *v* 3355 (NH), 1630, 1595, 1552 cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 7.92–8.00 (m, 2H), 7.46 (s, 1H, NH), 6.71–6.74 (m, 1H), 4.29 (t, 2H, J = 6.0 Hz), 3.32 (t, 2H, J = 6.0 Hz), 2.03 (quin, 2H, J = 6.0 Hz); MS: *m/z* 218 [M^+] (100), 190 (28), 134 (40). Anal. calcd for $C_{10}H_{10}N_4O_2$: C, 55.04; H, 4.62; N, 25.68. Found: C, 55.04; H, 4.63; N, 25.48.

3-(p-Fluorobenzoyl)-4,5,6,7-tetrahydro[1,2,3]triazolo[1,5-a]pyrimidine. **7b**: A white crystal, m.p. 163–164°C; IR (KBr): *v* 3445 (NH), 1638, 1600, 1520 cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.41–8.49 (m, 2H), 7.62 (s, 1H, NH), 7.36(t, 2H), 4.31 (t, 2H, J = 6.0 Hz), 3.37 (t, 2H, J = 6.0 Hz), 2.05 (quin, 2H, J = 6.0 Hz); MS: *m/z* 246 [M^+] (100), 217 (79), 190 (29). Anal. calcd for $C_{12}H_{11}FN_4O$: C, 58.53; H, 4.50; N, 22.75. Found: C, 58.19; H, 4.65; N, 22.81.

General Procedure for the Reaction of **1** *or* **2** *with* **3b**

A mixture of **1** or **2** (1 mmol) and **3b** (1.1 mmol) in 1,4-dioxane (15 mL) was stirred at ambient temperature for 24 hours. After removal of solvent the residue was purified by use of a basic aluminum oxide column and gradient elution with petroleum ether (60–90°C)/ethyl acetate. The products were thus separated and recrystallized from petroleum ether (60–90°C)-ethyl acetate.

1-(p-Nitrophenyl)-5-(2-furyl)-4-(2-imidazolinyl)-1,2,3-triazole. **8a**: A yellow crystal, m.p. 183–184°C;

IR (KBr): *v* 3210 (NH), 1617, 1596, 1512, 1349 (NO_2) cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.42 (d, 2H, J = 9.0 Hz), 7.84–7.85 (m, 1H), 7.79 (d, 2H, J = 9.0 Hz), 7.67–7.68 (m, 1H), 6.63–6.66 (m, 1H), 3.65 (s, 4H); MS: *m/z* 324 [M^+] (79), 296 (100), 249 (52). Anal. calcd for $C_{15}H_{12}N_6O_3$: C, 55.55; H, 3.73; N, 25.92. Found: C, 55.20; H, 3.89; N, 25.79.

1-(p-Nitrophenyl)-4-(2-imidazolinyl)-5-(2-thienyl)-1,2,3-triazole. **8b**: A pale yellow crystal, m.p. 182–184°C; IR (KBr): *v* 3260 (NH), 1605, 1560, 1515, 1349 (NO_2) cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.38 (d, 2H, J = 9.2 Hz), 7.78 (d, 2H, J = 9.2 Hz), 7.73–7.76 (m, 1H), 7.60–7.63 (m, 1H), 7.09–7.13 (m, 1H), 3.60 (s, 4H); MS: *m/z* 340 [M^+] (31), 339 (100), 311 (16), 265 (29). Anal. calcd for $C_{15}H_{12}N_6O_2S$: C, 52.93; H, 3.56; N, 24.69. Found: C, 52.60; H, 3.79; N, 24.39.

1-(p-Nitrophenyl)-4-(2-imidazolinyl)-5-(p-fluorophenyl)-1,2,3-triazole. **8d**: A pale yellow crystal, m.p. 175–176°C; IR (KBr): *v* 3332 (NH), 1630, 1610, 1590, 1510, 1342 (NO_2) cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 8.28 (d, 2H, J = 8.6 Hz), 7.48 (d, 2H, J = 8.6 Hz), 7.35–7.45 (m, 2H), 7.11 (t, 2H), 4.65 (br, 1H, NH), 3.74 (s, 4H); MS: *m/z* 352 [M^+] (30), 351 (100), 323 (25), 277 (36). Anal. calcd for $C_{17}H_{13}FN_6O_2$: C, 57.95; H, 3.72; N, 23.86. Found: C, 57.95; H, 3.89; N, 23.85.

1-(p-Nitrophenyl)-4-(tetrahydropyrimidinyl)-5-(2-furyl)-1,2,3-triazole. **9a**: A pale yellow crystal, m.p. 149–150°C; IR (KBr): *v* 3425 (NH), 1630, 1596, 1520, 1345 (NO_2) cm^{-1}; ^{1}H-NMR (DMSO-D_6): δ 8.42 (d, 2H, J = 9.2 Hz), 7.72 (d, 2H, J = 9.2 Hz), 7.68–7.71 (m, 1H), 7.48 (d, 1H), 6.61–6.64 (m, 1H), 3.38 (t, 4H, J = 6.0 Hz), 1.73 (quin, 2H, J = 6.0 Hz); MS: *m/z* 338 [M^+] (88), 310 (100), 281 (39), 263 (63). Anal. calcd for $C_{16}H_{14}N_6O_3$: C, 56.80; H, 4.17; N, 24.84. Found: C, 56.86; H, 4.07; N, 24.74.

1-(p-Nitrophenyl)-5-(p-fluorophenyl)-4-(2-tetrahydropyrimidinyl)-1,2,3-triazole. **9d**: A yellow crystal, m.p. 117–118°C; IR (KBr): *v* 3365 (NH), 1625, 1592, 1515, 1340 (NO_2) cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 8.25 (d, 2H, J = 9.0 Hz), 7.47 (d, 2H, J = 9.2 Hz), 7.01–7.40 (m, 4H), 3.42 (t, 4H, J = 6.0 Hz), 1.83 (quin, 2H, J = 6.0 Hz); MS: *m/z* 366 [M^+] (32), 365 (100), 337 (22). Anal. calcd for $C_{18}H_{15}FN_6O_2$: C, 59.01; H, 4.13; N, 22.94. Found: C, 58.65; H, 4.22; N, 22.68.

1-(p-Nitrophenyl)-4-(N-methyl-2-tetrahydropyrimidinyl)-5-(2-Furyl)-1,2,3-triazole. **11**: Yield 51%; a yellow crystal, m.p. 137–138°C; IR (KBr): *v* 1596, 1582, 1520, 1345 (NO_2) cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ 8.36 (d, 2H, J = 8.6 Hz), 7.63 (d, 2H, J = 8.6 Hz), 7.38–7.41 (m, 1H), 7.03–7.06 (m, 1H), 6.50–6.53 (m, 1H), 3.58 (t, 2H, J = 6.0 Hz), 3.35 (t, 2H, J = 6.0 Hz), 2.85 (s, 3H), 2.02 (quin, 2H, J = 6.0 Hz); MS: *m/z* 352 [M^+] (100), 323 (48), 295 (80.6). Anal. calcd for $C_{17}H_{16}N_6O_3$: C, 57.95; H, 4.58; N, 23.85. Found: C, 57.97; H, 4.58; N, 23.84.

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