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Microwave-Assisted Synthesis of Some 1,2,4-Triazol-5-one Derivatives

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ABSTRACT: *A series of 3-alkyl(aryl)-4-(p-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-ones* **2** *were obtained from the reaction of alkyl (aryl) ester ethoxycarbonyl hydrazones* **1** *with p-hydroxy aniline. The reaction of* **1** *with 1,4-diamino benzene (1:1) to afford 3-alkyl(aryl)-4-(p-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-ones* **3**. *The reaction of* **3** *with benzaldehyde gave 3-alkyl(aryl)-4-(4′-benzilidenamino)-4,5-dihydro-1H-1,2,4-triazol-5-ones* **4**. *All of the above reactions occurred under microwave heating and conventional methods. Their structures were confirmed by* ^{1}H *NMR,* ^{13}C *NMR, IR, and elemental analyses.* © 2008 Wiley Periodicals, Inc. Heteroatom Chem 19:38–42, 2008; Published online in Wiley InterScience (www.interscience.wiley.com). DOI 10.1002/hc.20381

INTRODUCTION

Classically, organic synthesis is carried out by conductive heating with a heat source (e.g., an oil bath). This is a comparatively slow and inefficient technique for transferring energy into the system, because it depends on convection currents and the thermal conductivity of the various materials that must be penetrated and results in the temperature of the reaction vessel being higher than that of the reaction mixture.

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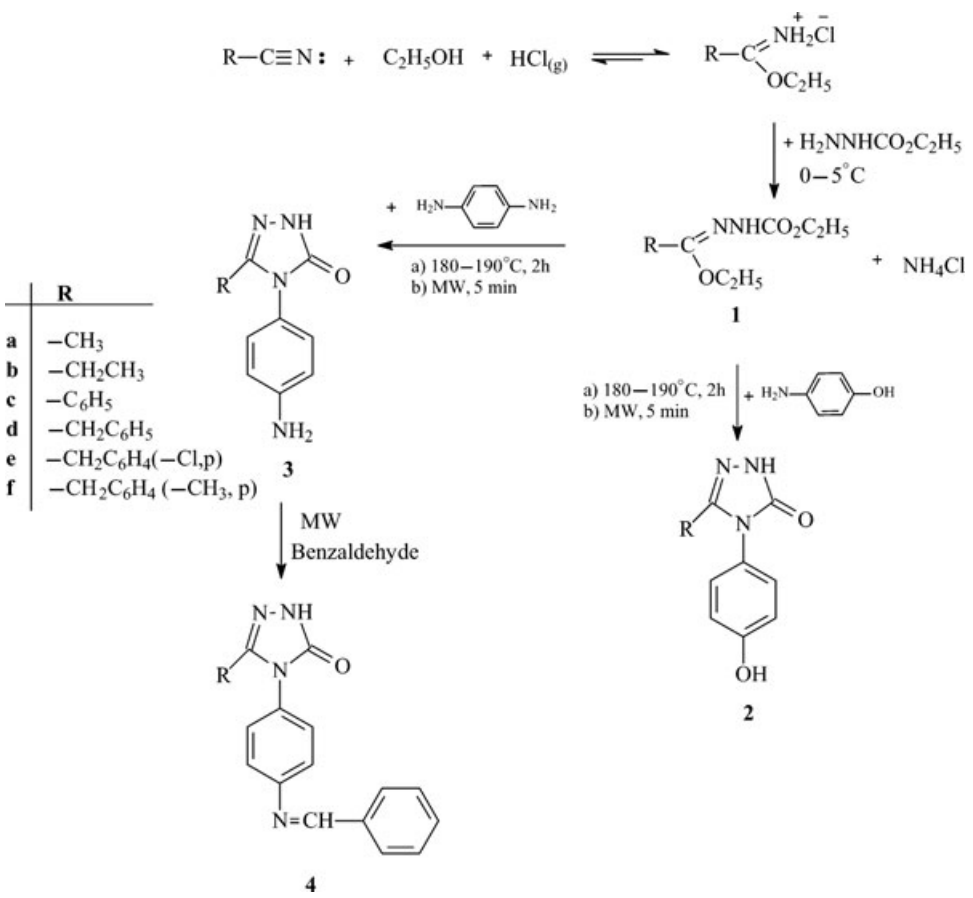
In addition, a temperature gradient can develop within the sample and local overheating can lead to product decomposition. In contrast, microwave heating produces efficient internal heating by direct coupling of microwave energy with the molecules that are present in the reaction mixture. On the other hand, organic chemists saw that microwave heating significantly speeds up chemical reactions from hours or days to minutes.

The increase in interest in this method stems from the realization that microwave-assisted synthesis, apart from many other enabling technologies, actually provides significant practical and economic advantages [1–6].

In recent years, the synthesis of some 1,2,4-triazol-5-one derivatives from ester carbethoxyhydrazones (**1**-type compounds) has been reported [7]. These are usually prepared by conductive heating. These reactions are time consuming. In the previous study, we synthesized some new 1,2,4-triazole-5-one derivatives by the reaction of ester carbethoxyhydrazones (**1**) with 4-aminophenol and 1,4-diaminobenzene under microwave irradiation.

RESULT AND DISCUSSION

Ether carbethoxyhydrazones can be considered as useful intermediates leading to the formation of some heterocycles such as 4-amino-1,2,4-triazole-5-ones, which were used to synthesize 1,2,4-triazole-5-one derivatives that showed pharmacological activities [8–15]. Therefore, treatment of compound **1** with



SCHEME 1

4-aminophenol under microwave heating resulted in the formation of 3-alkyl(aryl)-4-(*p*-hydroxyphenyl)-4,5-dihydro-1*H*-1,2,4-triazol-5-ones **2.** Treatment of compound **1** with 1,4-diaminobenzene (1:1) resulted in formation of 3-alkyl(aryl)-4-(*p*-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-ones **3.** The reaction of **3** with benzaldehyde under microwave heating resulted in the formation imines **4** (Scheme 1).

The IR spectrum of compound **2** showed absorption bands around at 3360 and 3250 due to NH and OH functions, respectively and its ^{1}H NMR spectrum revealed two signals around at 9.80 and 11.50 assigned to OH and NH protons, respectively. The IR spectrum of compound **3** showed absorption bands around at 3420 cm^{-1}, 3330 cm^{-1}, and 1690 cm^{-1} due to NH_2, NH, and C=O, respectively, and its ^{1}H NMR spectrum revealed two signals at 5.40 and 11.46 assigned to NH_2 and NH protons, respectively.

The IR spectrum of compound **4** showed absorption bands around at 3300 cm^{-1}, 1710 cm^{-1},and 1680 cm^{-1} due to NH, C=O, and C=N, respectively, and its ^{1}H NMR spectrum revealed two signals at 5.90 and 11.00 assigned to N=CH and NH protons, respectively.

EXPERIMENTAL

Melting points were measured with a Büchi oil heated melting point apparatus and are uncorrected. IR spectra were recorded for KBr pellets on Perkin-Elmer 1600 FTIR spectrophotometer. The ^{1}H NMR and ^{13}C NMR spectra were determined in DMSO-d_6 at 300 MHz on a Varian NMR spectrometer using TMS as an internal standard. Elemental analyses were carried out on a Carlo Erba 1106 CHN analyzer. The compounds **1a–f** were prepared according to [16–18].

*Synthesis of 3-Alkyl(aryl)-4-(P-hydroxyphenyl)-4, 5-dihydro-1h-1,2,4-triazol-5-ones (**2a–f**)*

General Procedure Conventional Method. A mixture of **1** (0.01 mol) and 4-aminophenol (0.01 mol) was taken in a round bottom flask and heated at 180°C–190°C for 2 h. After completion of the reaction, the solution was cooled. The crude product obtained was purified by using ethanol and identified as compound **2.** These compounds were soluble in common organic solvents such as ethanol,

ethyl acetate, dichloromethane, $CHCl_3$, DMF, and DMSO.

Microwave Method. A mixture of **1** (0.01 mol) and 4-aminophenol (0.01 mol) in water was taken in a flask. The mixture was irradiated in a microwave oven at 175°C, 350 W for 5 min. The above purification methods were also applied to this material.

*3-Methyl-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2a**)*. Prepared from **1a**, yield: 73.2%, mp 267°C–268°C; IR (cm^{-1}): 805.3 (1,4-disubstitue arom.), 1519.0 (C=N), 1673.7 (C=O), 3259.2 (O—H), 3362.6 (N—H); ^{1}H NMR δ (ppm): 1.99 (3H, s, CH_3), 6.86 (A part of AB system, dt, 2H, $J_{1a,1b} = 9.1$ Hz), 7.17 (B part of AB system, dt, 2H, $J_{1a,1b} = 9.1$ Hz), 9.83 (1H, s, OH), 11.53 (1H, s, NH); ^{13}C NMR δ (ppm): 12.07, 115.64, 123.98, 128.43, 144.12, 154.47, 157.35; elemental analyses: Calcd for $C_9H_9N_3O_2$ (191.19): C, 56.54; H, 4.74; N, 21.98; Found: C, 56.31; H, 4.69; N, 22.03.

*3-Ethyl-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2b**)*. Prepared from **1b**, yield: 82.9%, mp 224°C–225°C; IR (cm^{-1}): 804.7 (1,4-disubstitue arom.), 1518.3 (C=N), 1678.1 (C=O), 3246.9 (O—H), 3339.4 (N—H); ^{1}H NMR δ (ppm): 1.02 (3H, t, CH_3), 2.44 (2H, q, CH_2), 6.71 (A part of AB system, dt, 2H, $J_{1a,1b} = 8.9$ Hz), 7.23 (B part of AB system, dt, 2H, $J_{1a,1b} = 8.9$ Hz), 9.69 (1H, s, OH), 11.23 (1H, s, NH); ^{13}C NMR δ (ppm): 9.93, 18.12, 112.61, 124.76, 129.21, 145.22, 153.63, 158.02; elemental analyses: Calcd for $C_{10}H_{11}N_3O_2$ (205.21): C, 58.53; H, 5.40; N, 20.48; Found: C, 58.32; H, 5.53; N, 20.23.

*3-Phenyl-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2c**)*. Prepared from **1c**, yield: 39.2%, mp 295°C–296°C; IR (cm^{-1}): 811.2 (1,4-disubstitue arom.), 1522.1 (C=N), 1673.2 (C=O), 3289.4 (O—H), 3312.1 (N—H); ^{1}H NMR δ (ppm): 6.82 (A part of AB system, dt, 2H, $J_{1a,1b} = 11.9$ Hz), 7.07 (B part of AB system, dt, 2H, $J_{1a,1b} = 11.9$ Hz), 7.38 (5H, m, Ar), 9.81 (1H, s, OH), 12.62 (1H, s, NH); ^{13}C NMR δ (ppm): 117.84, 128.58, 129.05, 129.24, 130.22, 130.87, 131.43, 147.35, 158.71, 159.35; elemental analyses: Calcd for $C_{14}H_{11}N_3O_2$ (253.26): C, 66.40; H, 4.38; N, 16.59; Found: C, 65.94; H, 4.57; N, 16.28.

*3-Benzyl-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2d**)*. It is reported in [19] and prepared from **1d**.

*3-(4-Chlorobenzyl)-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2e**)*. Prepared from **1e**, yield: 86.1%, mp 256°C–257°C; IR (cm^{-1}): 824.2 (1,4-disubstitue arom.), 1519.6 (C=N), 1699.0 (C=O), 3191.6 (O—H), 3298.3 (N—H); ^{1}H NMR δ (ppm): 3.75 (2H, s, CH_2) 6.72–7.23 (8H, m, Ar-H), 9.79 (1H, s, OH), 11.87 (1H, s, NH); ^{13}C NMR δ (ppm): 22.13, 127.67, 129.21, 129.74, 131.43, 131.97, 132.69, 149.12, 159.98, 161.15; elemental analyses: Calcd. for $C_{15}H_{12}ClN_3O_2$ (301.73): C, 59.71; H, 4.01; N, 13.93; Found: C, 58.67; H, 4.05; N, 13.57.

*3-(4-Methylbenzyl)-4-(4-hydroxyphenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**2f**)*. Prepared from **1f**, yield: 62.5%, mp 240°C–241°C; IR (cm^{-1}): 832.9, 803.3 (1,4-disubstitue arom.), 1609.5 (C=N), 1707.3 (C=O), 3174.1 (O—H), 3288.6 (N—H); ^{1}H NMR δ (ppm): 2.20 (3H, s, CH_3), 3.64 (2H, s, CH_2), 6.60–7.20 (8H, m, Ar-H), 9.80 (1H, s, OH), 11.68 (1H, s, NH); ^{13}C NMR δ (ppm): 20.49, 31.32, 115.52, 123.69, 128.24, 128.26, 128.74, 132.08, 135.51, 146.36, 154.59, 157.35; elemental analyses: Calcd for $C_{16}H_{15}N_3O_2$ (281.31): C, 68.31; H, 5.37; N, 14.94; Found: C, 67.88; H, 5.01; N, 14.35.

*Synthesis of 3-Alkyl(aryl)-4-(p-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-ones (**3a–f**)*

General Procedure Conventional Method. A mixture of **1** (0.01 mol) and 4-aminoaniline (0.01 mol) was taken in a round bottom flask and heated at 180°C–190°C for 2 h. After the completion of reaction, the solution was cooled. The crude product obtained was purified by using ethyl acetate and identified as compound **3.** These compounds were soluble in common organic solvents such as ethanol, ethyl acetate, dichloromethane, $CHCl_3$, DMF, and DMSO.

Microwave Method. A mixture of **1** (0.01 mol) and 4-aminoaniline (0.01 mol) in water was taken in a flask. The mixture was irradiated in a microwave oven at 175°C, 350 W for 5 min. The above purification methods were also applied to this material.

*3-Methyl-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3a**)*. Prepared from **1a** yield: 87.0%, mp 241°C–242°C; IR (cm^{-1}): 803.6 (1,4-disubstitue arom.), 1520.8 (C=N), 1687.5 (C=O), 3331.1 (N—H), 3420.0–3422.5 (NH_2); ^{1}H NMR δ (ppm): 1.97 (3H, s, CH_3), 5.39 (2H, s, NH_2), 6.62 (A part of AB system, dd, 2H, $J_{1a,1b} = 8.4$ Hz), 6.96 (B part of AB system, dd, 2H, $J_{1a,1b} = 8.4$ Hz), 11.47 (1H, s, NH); ^{13}C NMR δ (ppm): 12.05, 113.67, 120.61, 127.89, 144.45, 148.93, 154.67; elemental analyses: Calcd for $C_9H_{10}N_4O$ (190.20): C, 56.83; H, 5.30; N, 29.46; Found: C, 56.13; H, 5.01; N, 28.87.

*3-Ethyl-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3b**).* Prepared from **1b**, yield: 77.0%, mp 197°C–198°C; IR (cm^{-1}): 804.9 (1,4-disubstitue arom.), 1521.3 (C=N), 1686.2 (C=O), 3346.1 (N—H), 3430.5–3432.4 (NH_2); ^{1}H NMR δ (ppm): 1.00 (3H, t, CH_3), 2.31 (2H, q, CH_2), 5.39 (2H, s, NH_2), 6.61 (A part of AB system, dt, 2H, $J_{1a,1b}$ = 8.6 Hz), 6.95 (B part of AB system, dt, 2H, $J_{1a,1b}$ = 8.6 Hz), 11.46 (1H, s, NH); ^{13}C NMR δ (ppm): 9.79, 19.11, 113.69, 120.48, 128.06, 148.45, 148.99, 154.87; elemental analyses: Calcd for $C_{10}H_{12}N_4O$ (204.23): C, 58.81; H, 5.92; N, 27.43; Found: C, 58.33; H, 5.89; N, 26.99.

*3-Phenyl-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3c**).* Prepared from **1c**, yield: 67.5%, mp 267°C–268°C; IR (cm^{-1}): 803.2 (1,4-disubstitue arom.), 1509.1 (C=N), 1698.2 (C=O), 3332.1 (N—H), 3412.4–3422.6 (NH_2); ^{1}H-NMR δ (ppm): 5.05 (2H, s, NH_2), 6.95 (A part of AB system, dd, 2H, $J_{1a,1b}$ = 10.2 Hz), 7.22 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 10.2 Hz), 7.36 (5H, m, Ar), 12.62 (1H, s, NH); ^{13}C NMR δ (ppm): 116.54, 127.98, 128.06, 129.13, 129.99, 130.67, 132.03, 146.41, 158.61, 159.98; elemental analyses: Calcd for $C_{14}H_{12}N_4O$ (252.27): C, 66.65; H, 4.79; N, 22.21; Found: C, 66.38; H, 4.82; N, 21.02.

*3-Benzyl-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3d**).* Prepared from **1d** yield: 54.1%, mp 236°C–237°C; IR (cm^{-1}): 808.7, 828.5 (1,4-disubstitue arom.), 1520.9 (C=N), 1702.9 (C=O), 3358.4 (N—H), 3440.1–3443.6 (NH_2); ^{1}H NMR δ (ppm): 3.71 (2H, s, CH_2), 5.38 (2H, s, NH_2), 6.54 (A part of AB system, dd, 2H, $J_{1a,1b}$ = 8.4 Hz), 6.77 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 8.4 Hz), 7.11 (5H, m, Ar), 11.61 (1H, s, NH); ^{13}C NMR δ (ppm): 31.71, 113.55, 120.28, 126.50, 128.16, 128.21, 128.41, 135.38, 146.53, 148.96, 154.81; elemental analyses: Calcd (%) for $C_{15}H_{14}N_4O$ (266.30): C, 67.65; H, 5.30; N, 21.04; Found: C, 67.49; H, 5.82; N, 20.57.

*3-(4-Chlorobenzyl)-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3e**).* Prepared from **1e**, yield: 72.2%, mp 205°C–206°C; IR (cm^{-1}): 805.6, 821.3 (1,4-disubstitue arom.), 1534.6 (C=N), 1698.9 (C=O), 3344.5 (N—H), 3439.5–3442.4 (NH_2); ^{1}H NMR δ (ppm): 3.65 (2H, s, CH_2), 5.42 (2H, s, NH_2), 6.63–7.20 (8H, m, Ar), 11.78 (1H, s, NH); ^{13}C NMR δ (ppm): 33.51, 112.54, 121.29, 127.21, 127.99, 128.13, 131.42, 136.23, 145.54, 148.56, 155.69; elemental analyses: Calcd for $C_{15}H_{13}ClN_4O$ (300.74): C, 59.91; H, 4.36; N, 18.63; Found: C, 59.78; H, 4.61; N, 18.08.

*3-(4-Methylbenzyl)-4-(4-aminophenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**3f**).* Prepared from **1f**, yield: 67.1%, mp 227°C–228°C; IR (cm^{-1}): 829.4 (1,4-disubstitue arom.), 1519.7 (C=N), 1701.7 (C=O), 3363.4 (N—H), 3450.6–3456.0 (NH_2); ^{1}H NMR δ (ppm): 2.31 (3H, s, CH_3), 3.65 (2H, s, CH_2), 5.37 (2H, s, NH_2), 6.53 (A part of AB system, dd, 2H, $J_{1a,1b}$ = 8.7 Hz), 6.78 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 8.7 Hz), 6.86 (A part of AB system, dd, 2H, $J_{1a,1b}$ = 8.1 Hz), 7.03 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 8.1 Hz), 11.57 (1H, s, NH); ^{13}C NMR δ (ppm): 21.30, 32.04, 114.30, 121.07, 128.98, 129.04, 129.52, 133.06, 136.26, 147.41, 149.72, 155.55; elemental analyses: Calcd for $C_{16}H_{16}N_4O$ (280.32): C, 68.55; H, 5.75; N, 19.99; Found: C, 68.20; H, 5.60; N, 19.08.

*Synthesis of 3-Alkyl(aryl)-4-(4′-benzilidenamino)-4,5-dihydro-1h-1,2,4-triazol-5-ones (**4a–f**)*

General Procedure Conventional Method. A mixture of **3** (0.01 mol) and benzaldehyde (0.01 mol) was taken in a round bottom flask and heated at 180°C–190°C for 2 h. After completion of the reaction, the solution was cooled. The crude product obtained was purified by using ethanol and identified as compound **4.** These compounds were soluble in common organic solvents such as ethanol, ethyl acetate, dichloromethane, $CHCl_3$, DMF, and DMSO.

Microwave Method. A mixture of **3** (0.01 mol) and benzaldehyde (0.01 mol) in ethanol was taken in a flask. The mixture was irradiated in a microwave oven at 175°C, 350 W for 5 min. The above purification methods were also applied to this material.

*3-Methyl-4-(4-{[(1E)-phenylmethylene]amino}-phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4a**).* Prepared from **3a,** yield: 51.8%, mp 204°C–205°C; IR (cm^{-1}): 833.6 (1,4-disubstitue arom.), 1549.8–1660.7 (C=N), 1716.4 (C=O), 3307.6 (N—H); ^{1}H NMR δ (ppm): 2.03 (3H, s, CH_3), 7.31 (A part of AB system, dd, 2H, $J_{1a,1b}$ = 7.7 Hz), 7.71 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 7.7 Hz), 10.2 (1H, s, CH), 11.6 (1H, s, NH); ^{13}C NMR δ (ppm): 12.18, 119.28, 121.98, 123.57, 125.12, 127.46, 129.13, 135.19, 139.21, 143.86, 154.25, 168.46; elemental analyses: Calcd for $C_{16}H_{14}N_4O$ (278.31): C, 69.05; H, 5.07; N, 20.13; Found: C, 69.22; H, 4.89; N, 20.15.

*3-Ethyl-4-(4-{[(1E)-phenylmethylene]amino}phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4b**).* Prepared from **3b**, yield: 27.8%, mp 163°C–164°C; IR (cm^{-1}): 828.5 (1,4-disubstitue arom.), 1546.7–1676.7 (C=N), 1701.7 (C=O), 3313.2 (N—H); ^{1}H NMR δ (ppm): 1.13 (3H, t, CH_3), 2.27 (2H, q, CH_2), 7.26 (A

part of AB system, dd, 2H, $J_{1a,1b}$ = 8.3 Hz), 7.55 (B part of AB system, dd, 2H, $J_{1a,1b}$ = 8.3 Hz), 10.91 (1H, s, CH), 12.19 (1H, s, NH); ^{13}C NMR δ (ppm): 9.83, 20.4, 117.68, 122.41, 123.01, 123.88, 127.02, 129.85, 136.13, 138.43, 143.29, 153.43, 166.21; elemental analyses: Calcd for $C_{17}H_{16}N_4O$ (292.34): C, 69.85; H, 5.52; N, 19.17; found: C, 69.23; H, 5.50; N, 19.04.

*3-Phenyl-4-(4-{[(1E)-phenylmethylene]amino}phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4c**).* Prepared from **3c**, yield: 29.4%, mp 257°C–258°C; IR (cm^{-1}): 809.5 (1,4-disubstitue arom.), 1551.4–1626.6 (C=N), 1713.5 (C=O), 3321.4 (N—H); ^{1}H NMR δ (ppm): 7.06–7.64 (14H, m, Ar), 10.65 (1H, s, CH), 11.98 (1H, s, NH); ^{13}C NMR δ (ppm): 116.06, 116.69, 123.51, 123.97, 125.76, 126.48, 127.09, 128.40, 129.35, 130,68, 135.33, 138.98, 144.42, 150.13, 162.38; elemental analyses: Calcd for $C_{21}H_{16}N_4O$ (340.38): C, 74.10; H, 4.74; N, 16.46; Found: C, 73.75; H, 4.50; N, 16.51.

*3-Benzyl-4-(4-{[(1E)-phenylmethylene]amino}phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4d**).* Prepared from **3d**, yield: 26.6%, mp 107°C–108°C; IR (cm^{-1}): 832.1 (1,4-disubstitue arom.), 1570.2–1629.4 (C=N), 1701.6 (C=O), 3321.4 (N—H); ^{1}H NMR δ (ppm): 3.63 (2H, s, CH_2), 7.20–7.57 (14H, m, Ar), 10.13 (1H, s, CH), 12.23 (1H, s, NH); ^{13}C NMR δ (ppm) 35.76, 115.16, 116.61, 122.34, 123.36, 123.87, 126.04, 127.43, 128.12, 128.75, 130.02, 134.42, 139.38, 145.23, 151.13, 160.18; elemental analyses: Calcd for $C_{22}H_{18}N_4O$ (354.40): C, 74.56; H, 5.12; N, 15.81; found: C, 74.76; H, 5.10; N, 15.54.

*3-(4-Chlorobenzyl)-4-(4-{[(1E)-phenylmethylene]amino}phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4e**).* Prepared from **3e**, yield: 34.2%, mp 253°C–254°C; IR (cm^{-1}): 818.1 (1,4-disubstitue arom.), 1516.9–1647.5 (C=N), 1708.8 (C=O), 3302.1 (N—H); ^{1}H NMR δ (ppm): 3.79 (2H, s, CH_2), 7.04–7.65 (13H, m, Ar), 10.14 (1H, s, CH), 11.75 (1H, s, NH); ^{13}C NMR δ (ppm): 31.08, 116.23, 119.08, 126.21, 127.04, 127.83, 128.02, 130.38, 131.20, 132.43, 134.03, 139.35, 140.25, 145.70, 154.39, 168.46; elemental analyses: Calcd for $C_{22}H_{17}ClN_4O$ (388.85): C, 67.95; H, 4.41; N, 14.41; Found: C, 67.91; H, 4.53; N, 14.58.

*3-(4-Methylbenzyl)-4-(4-{[(1E)-phenylmethylene]amino}phenyl)-4,5-dihydro-1H-1,2,4-triazol-5-one (**4f**).* Prepared from **3f**, yield: 29.2%, mp 237°C; IR (cm^{-1}): 820.2 (1,4-disubstitue arom.), 1508.8–1668.5 (C=N), 1718.5 (C=O), 3310.4 (N—H); ^{1}H NMR δ (ppm): 2.25 (3H, s, CH_3), 3.48 (2H, s, CH_2), 7.10–7.73 (13H, m, Ar), 10.37 (1H, s, CH), 11.88 (1H, s, NH); ^{13}C NMR δ (ppm) 20.15, 33.15, 117.63, 119.18, 125.69, 127.22, 127.80, 129.15, 130.41, 131.44, 132.09, 133.77, 139.64, 141.28, 145.68, 155.64, 167.51; elemental analyses: Calcd for $C_{23}H_{20}N_4O$ (368.43): C, 74.98; H, 5.47; N, 15.21; Found: C, 74.67; H, 5.23; N, 15.59.

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