

Microwave-Assisted Synthesis of Novel 2,4-Dihydro-5-[4-(trifluoromethyl)phenyl]-3*H*-1,2,4-triazol-3-ones and Potentiometric Determination of Their pK_a in Nonaqueous Solvents

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The novel 4-amino- or 4-aryl-substituted 2,4-dihydro-5-[4-(trifluoromethyl)phenyl]-3*H*-1,2,4-triazol-3-ones **3a–3g** were synthesized by reaction of *N*-(ethoxycarbonyl)-4-(trifluoromethyl)benzenehydrazonic acid ethyl ester (**2**) and primary amines or hydrazine by microwave irradiation. Compounds **3a–3g** were potentiometrically titrated with tetrabutylammonium hydroxide (Bu₄NOH) in four nonaqueous solvents, *i.e.*, ¹PrOH, ¹BuOH, MeCN, and *N,N*-dimethylformamide (DMF). Also half-neutralization potential values and the corresponding pK_a values were determined in all cases.

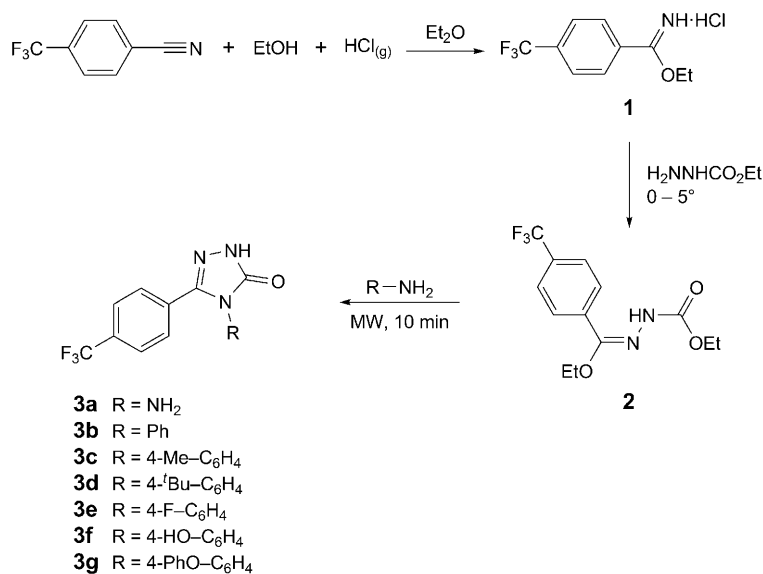
Introduction. – Triazol compounds have been found to be associated with diverse pharmacological activities such as antibacterial, antifungal, anticancer, and anticonvulsant [1–6]. In recent years, the synthesis of some 3*H*-1,2,4-triazol-3-one derivatives from arenehydrazonic esters (compounds of type **2**) has been reported [7]. However, most of the conventional syntheses of such compounds are time-consuming and lack high selectivity. On the other hand, in recent years, microwave-assisted reactions have attracted much research interest because of the simplicity in operation, enhanced reaction rates, and great selectivity [8]. Thus, microwave irradiation, which has become a powerful synthetic tool for the rapid synthesis of a variety of compounds, is used to enhance the rates of classical organic reactions [9]. In this study, we synthesized some new 3*H*-1,2,4-triazol-3-one derivatives containing trifluoromethyl groups by the reaction of **2** and primary amines under microwave irradiation for 10 min. We have already previously synthesized triazole compounds by using the microwave-irradiation method [10–12].

It is known that 2,4-dihydro-3*H*-1,2,4-triazol-3-ones are weakly acidic [13][14]. The acidity of a compound in a given medium is influenced by both the electronic effects of the substituents and the polarity of the solvent.

Results and Discussion. – The first step in our synthesis was the formation of the hydrochloride of 4-(trifluoromethyl)benzenimidic acid ethyl ester (**1**; *Scheme*). Passing HCl gas through solutions of 4-(trifluoromethyl)benzonitrile in dry EtOH, followed by precipitation with Et₂O, gave **1**. Treatment of **1** with ethyl hydrazinecarboxylate resulted in the formation of **2**. Such compounds can be considered as useful intermediates leading to the formation of heterocycles such as 3*H*-1,2,4-triazol-3-ones. Therefore, treatment of compound **2** with primary amines or hydrazine under microwave heating resulted in the formation of 3*H*-1,2,4-triazol-3-ones **3a–3g**.

(Scheme). The reactions were performed in dry EtOH. The reaction time was as short as 10 min, and **3a–3g** were isolated in up to 91% yield. Their structure was confirmed by IR and ^1H - and ^{13}C -NMR spectroscopy and elemental analysis.

Scheme. Pathway for the Formation of the 3H-1,2,4-Triazol-3-ones



In the IR spectrum (KBr) of compound **1**, the characteristic NH_2^+ absorption bands appeared at 2977, 2920, and 850 cm^{-1} and the $\text{C}=\text{N}$ band at 1647 cm^{-1} . The IR spectrum (KBr) of **2** showed the $\text{C}=\text{O}$ band at 1698 cm^{-1} and the $\text{C}=\text{N}$ band at 1613 cm^{-1} . The IR spectra (KBr) of **3a–3g** were characterized by absorption bands at 3160, 1699, and 1602 cm^{-1} attributable to NH, $\text{C}=\text{O}$, and $\text{C}=\text{N}$ functions, respectively. In addition, observation of NH and aromatic H-atoms, as well as the lack of EtO signals in the ^1H -NMR spectra of **3a–3g** and the triazolone $\text{C}=\text{O}$ signal at $\delta(\text{C})$ ca. 155, a $\text{C}=\text{N}$ signal at $\delta(\text{C})$ ca. 150, as well as the CF_3 signal in average at $\delta(\text{C})$ 122 in the ^{13}C -NMR spectra supported the proposed structures. Moreover, the elemental analyses of **3a–3g** showed good agreement with the calculated values.

All compounds were potentiometrically titrated with Bu_4NOH in four nonaqueous solvents, namely $^i\text{PrOH}$, $^t\text{BuOH}$, MeCN, and DMF. The mV values read in each titration were plotted against 0.05M Bu_4NOH in $^i\text{PrOH}$ (in ml) yielding the potentiometric titration curves for all 3H-1,2,4-triazol-3-ones (see Fig. 1 for **3d**). From these titration curves, the half-neutralization potential (HNP) values were determined (see Fig. 2 for **3d**), and the corresponding $\text{p}K_a$ values were calculated (see Table). Also the derived potentiometric titration curves were plotted (Figs. 3–5 for **3d**).

The pH of the weak acids are given by the $\text{pH} = \text{p}K_a + \log[\text{A}^-]/[\text{HA}]$, i.e., $\text{pH} = \text{p}K_a$ at $[\text{A}^-] = [\text{HA}]$ at the half-neutralization point. Therefore, the pH values can be regarded as $\text{p}K_a$ at the half-neutralization points (HNP). When the dielectric permittivity of solvents is taken into consideration, the acidic sequence can be

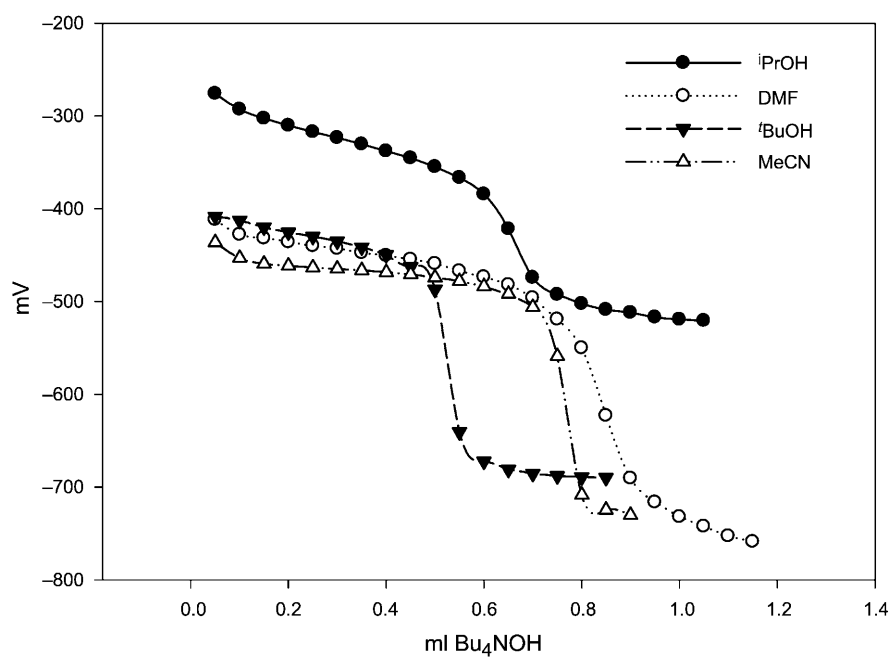


Fig. 1. *mV* vs. *ml Bu₄NOH* Potentiometric titration curves of 0.001M solutions of compound **3d** titrated with 0.05M *Bu₄NOH* at 25°

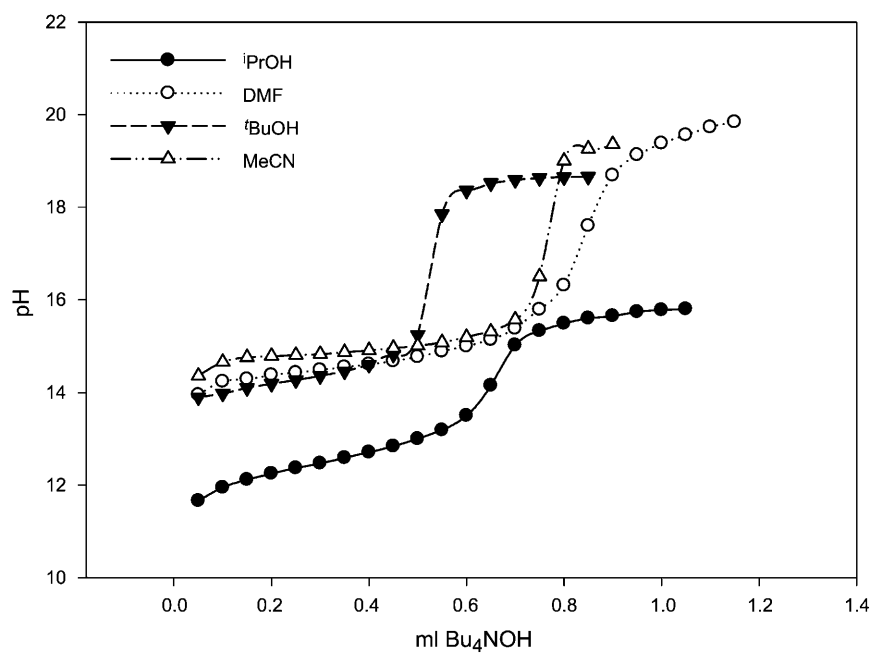
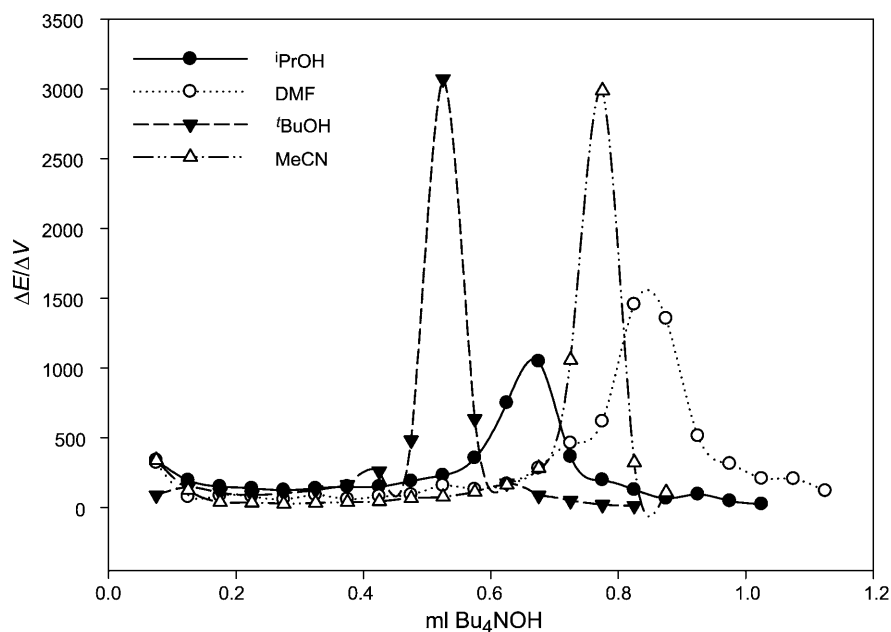


Fig. 2. *pH* vs. *ml Bu₄NOH* Potentiometric titration curves of 0.001M solutions of **3d** titrated with 0.05M *Bu₄NOH* at 25°

Table. Half-Neutralization-Potential (HNP) Values and Corresponding pK_a Values of **3a–3g**

	ⁱ PrOH		DMF		^t BuOH		MeCN	
	pK_a	HNP [mV]	pK_a	HNP [mV]	pK_a	HNP [mV]	pK_a	HNP [mV]
3a	10.68	–212.3	15.18	–484.0	12.69	–337.4	15.22	–487.0
3b	12.49	–324.9	15.03	–474.9	14.48	–442.5	14.89	–467.1
3c	12.64	–334.2	14.40	–437.7	14.36	–435.6	14.99	–472.9
3d	12.55	–328.0	14.63	–451.8	14.30	–431.7	14.90	–467.8
3e	12.36	–316.9	14.59	–448.0	14.84	–463.9	14.06	–417.8
3f	12.05	–298.9	14.84	–456.2	9.85	–166.6	8.67	–98.1
3g	11.21	–247.7	14.27	–421.6	9.68	–233.9	15.98	–514.8

Fig. 3. $\Delta E/\Delta V$ vs. ml Bu_4NOH Potentiometric titration curves of 0.001M solutions of compound **3d** titrated with 0.05M Bu_4NOH at 25°

expected as follows: DMF ($\epsilon = 36.7$) > MeCN ($\epsilon = 36$) > ⁱPrOH ($\epsilon = 19.4$) > ^tBuOH ($\epsilon = 12$). In ⁱPrOH, all these solvents show the strongest acidic properties. The degree to which a pure solvent (SH) ionizes is represented by its autoprotolysis constant, K_{SH} . For the above reaction, the constant is defined by $K_{SH} = [H_2S^+] \cdot [S^-]$. The importance of the autoprotolysis constant in titrations lies in its effect on the completeness of a titration reaction. As it is well known, the acidity of a compound depends on several factors. The most important ones are solvent effect and molecular structure. The Table shows that the HNP values and the corresponding pK_a values obtained from potentiometric titrations depend on the type of nonaqueous solvents used and the molecular structure of the compound.

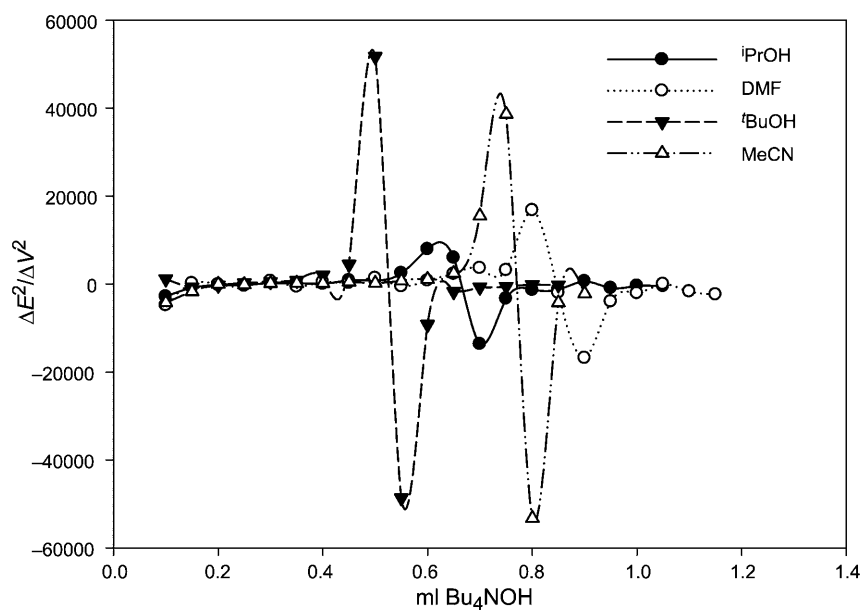


Fig. 4. $\Delta E^2/\Delta V^2$ vs. ml Bu_4NOH Potentiometric titration curves of 0.001M solutions of compound **3d** titrated with 0.05M Bu_4NOH at 25°

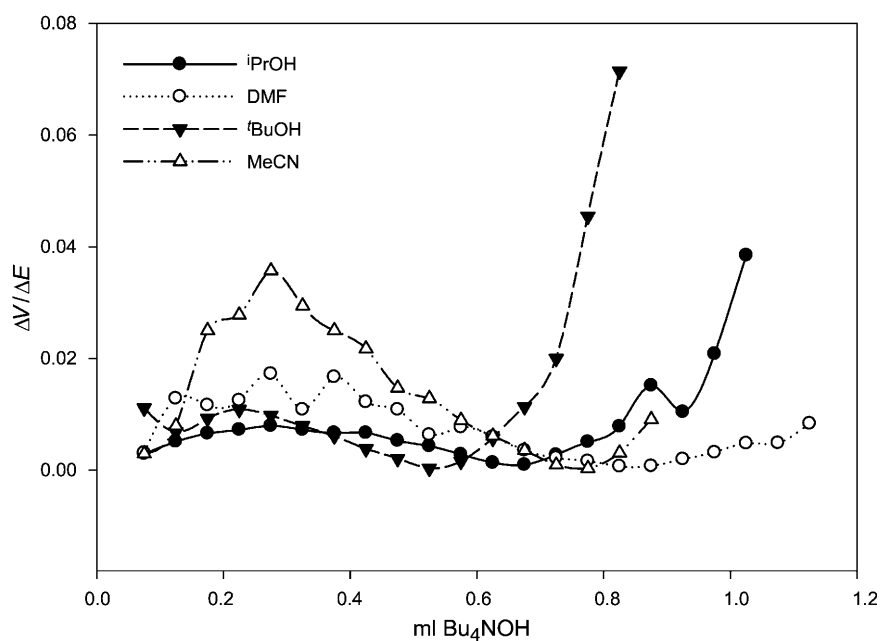


Fig. 5. $\Delta V/\Delta E$ vs. ml Bu_4NOH Potentiometric titration curves of 0.001M solutions of compound **3d** titrated with 0.05M Bu_4NOH at 25°

As seen in the *Table*, the acidic sequence for compounds **3a**, **3c**, and **3d** is ${}^i\text{PrOH} > {}^t\text{BuOH} > \text{DMF} > \text{MeCN}$; for compound **3b**, it is ${}^i\text{PrOH} > {}^t\text{BuOH} > \text{MeCN} > \text{DMF}$; for compound **3e**, it is ${}^i\text{PrOH} > \text{MeCN} > \text{DMF} > {}^t\text{BuOH}$ for compound **3f**, it is $\text{MeCN} > {}^t\text{BuOH} > {}^i\text{PrOH} > \text{DMF}$; and finally for **3g**, it is ${}^t\text{BuOH} > {}^i\text{PrOH} > \text{DMF} > \text{MeCN}$. In another words, **3a–3e** show the strongest acidic properties in ${}^i\text{PrOH}$, and **3f** and **3g** show the strongest acidic properties in MeCN and ${}^t\text{BuOH}$, respectively. On the other hand, the order of the weakest acidic properties is as follows: **3a**, **3c**, **3d**, and **3g** in MeCN, **3b** and **3f** in DMF, and **3e** in ${}^t\text{BuOH}$. This situation may be attributed to H-bonding between the formed anions and the solvent molecules in the amphiprotic neutral solvents.

Experimental Part

General. M.p.: Büchi melting-point apparatus, in open capillaries; uncorrected. IR Spectra: Perkin–Elmer-100 FT-IR spectrophotometer; KBr pellets; $\tilde{\nu}$ in cm^{-1} . ${}^1\text{H}$ - and ${}^{13}\text{C}$ -NMR Spectra: Varian-200 spectrometer; in $(\text{D}_6)\text{DMSO}$; chemical shifts δ in ppm rel. to Me_4Si as internal standard and coupling constants J in Hz. Elemental analyses: Carlo-Erba-1106-CHN analyzer; the measured percentages were in agreement ($\pm 0.4\%$) with the calculated ones.

Microwave Experiments. A monomode CEM-Discover microwave apparatus was used in the standard configuration as delivered, including its software. All experiments were carried out in microwave process vials (30 ml) with control of the temp. by an IR sensor. The temp. was computer-monitored and maintained constant by a discrete modulation of delivered microwave power. After completion of the reaction, the vial was cooled to 60° by air-jet cooling (see also the *General Procedure* below).

Potentiometric Titrations. For the potentiometric titrations, an Orion-720A pH/ion meter, equipped with a combined pH electrode (in gold) and indicator electrode, were used (Fig. 6). Also, a magnetic stirrer, a semi-microburette, and a 25 ml beaker were used in titrations. Before potentiometric titrations, the pH meter was calibrated according to the instructions supplied by the manufacturers of the pH meter. During the titrations, the titrant was added in increments of 0.05 ml after each stable reading, and mV

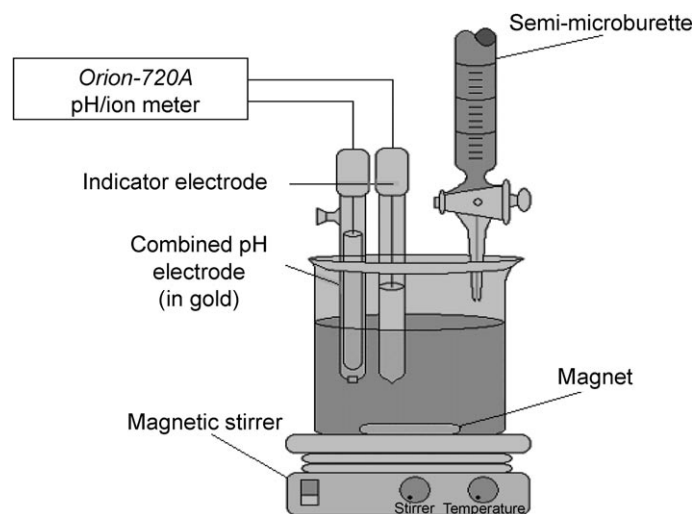


Fig. 6. Potentiometric-titration cell

values were recorded. The necessary chemicals were supplied by *Fluka* and *Merck*. After purifications, $^i\text{PrOH}$ was used to prepare 0.05N Bu_4NOH by dilution of 0.1N Bu_4NOH . The 0.05M Bu_4NOH in $^i\text{PrOH}$, which is widely used in the titration of acids, was used as titrant, and the mV values were recorded with the pH meter. Finally, the HNP values were determined by plotting the mV/ml (Bu_4NOH) graphics.

4-(Trifluoromethyl)benzenimidic Acid Ethyl Ester Hydrochloride (1:1) (1). To an ice-cooled soln. of the 4-(trifluoromethyl)benzonitrile (1 mol) in dry EtOH (1.1 mol), dry HCl gas was added until 1.1 mol had been absorbed. The resulting soln. was then allowed to stand at 0° in the refrigerator for 12 h, after which cold Et_2O was added. The precipitated crystals were filtered off immediately, washed with cold Et_2O , and dried in a dessicator: 87% of **1**. White solid. M.p. 131° . IR: 2977, 2920, 850 (NH_2^+), 1647 ($\text{C}=\text{N}$), 850 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 1.39 (*t*, $^3J = 7.1$, 3 H); 4.32 (*q*, $^3J = 7.1$, 2 H); 6.65 (br. *s*, 2 H); 7.53–7.64 (*m*, 4 H). $^{13}\text{C-NMR}$: 14.02; 68.12; 114.33 (CF_3); 126.16; 128.04; 129.04; 136.60; 163.88 ($\text{C}=\text{N}$). Anal. calc. for $\text{C}_{10}\text{H}_{11}\text{ClF}_3\text{NO}$: C 47.35, H 4.37, N 5.52; found: C 47.21, H 4.30, N 5.44.

***N*-(Ethoxycarbonyl)-4-(trifluoromethyl)benzenehydrazonic Acid Ethyl Ester (2).** In an ice bath, **1** (0.01 mol) was dissolved in dry EtOH (50 ml), and ethyl hydrazinecarboxylate (0.01 mol) in dry EtOH (20 ml) was then added. After stirring for 6 h at 0° , the mixture was filtered to remove NH_4Cl which separated from the soln., and the filtrate was concentrated at $35-40^\circ$. The solid residue, after drying in a dessicator, was recrystallized from petroleum ether: pure **2** (79%). White solid. M.p. 70° . IR: 3257 (NH), 1698 ($\text{C}=\text{O}$), 1613 ($\text{C}=\text{N}$), 847 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 1.19 (*t*, $^3J = 6.8$, 3 H); 1.29 (*t*, $^3J = 7.3$, 3 H); 4.09 (*q*, $^3J = 6.8$, 2 H); 4.51 (*q*, $^3J = 7.3$, 2 H); 7.94 (*d*-like, *AA'* of *AA'XX'*, $^3J = 8.2$, 2 arom. H); 8.33 (*d*-like, *XX'* of *AA'XX'*, $^3J = 8.2$, 2 arom. H); 8.44 (br. *s*, 1 H). $^{13}\text{C-NMR}$: 14.16; 15.23; 62.14; 63.10; 115.39 (CF_3); 126.51; 127.94; 128.88; 134.87; 153.82 ($\text{C}=\text{O}$); 157.01 ($\text{C}=\text{N}$). Anal. calc. for $\text{C}_{13}\text{H}_{15}\text{F}_3\text{N}_2\text{O}_5$: C 51.32, H 4.97, N 9.21; found: C 51.31, H 4.90, N 9.28.

3H-1,2,4-Triazol-3-ones 3: General Procedure. A mixture of **2** (0.01 mol) and amino compound (0.01 mol) in EtOH (25 ml) was microwave-irradiated in closed vessels with pressure control at 130° for 10 min (hold time) at 300 W maximum power. After the completion of the reaction (TLC monitoring ($\text{AcOEt}/\text{hexane } 3:1$)), the crude product was recrystallized from EtOH: pure **3**.

4-Amino-2,4-dihydro-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3a): Yield 84%. M.p. $253-254^\circ$. IR: 3342, 3320 (NH_2), 3194 (NH), 1698 ($\text{C}=\text{O}$), 1601 ($\text{C}=\text{N}$), 833 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 5.49 (*s*, 2 H); 7.87 (*d*-like, *AA'* of *AA'XX'*, $^3J = 8.4$, 2 arom. H); 8.24 (*d*-like, *XX'* of *AA'XX'*, $^3J = 8.4$, 2 arom. H); 12.08 (*s*, 1 H). $^{13}\text{C-NMR}$: 120.13 (CF_3); 124.97; 128.62; 132.75; 134.13; 146.29 ($\text{C}=\text{N}$); 156.44 ($\text{C}=\text{O}$). Anal. calc. for $\text{C}_9\text{H}_7\text{F}_3\text{N}_4\text{O}$: C 44.27, H 2.89, N 22.95; found: C 44.21, H 2.84, N 22.93.

2,4-Dihydro-4-phenyl-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3b): Yield 81%. M.p. $213-214^\circ$. IR: 3156 (NH), 1694 ($\text{C}=\text{O}$), 1620 ($\text{C}=\text{N}$), 837 (1,4-disubstituted benzene ring), 695, 750 (monosubstituted benzene ring). $^1\text{H-NMR}$: 7.24–7.58 (*m*, 9 H); 10.35 (*s*, 1 H). $^{13}\text{C-NMR}$: 119.41 (CF_3); 121.22; 122.34; 124.75; 125.48; 126.50; 127.83; 128.65; 135.21; 148.04 ($\text{C}=\text{N}$); 153.82 ($\text{C}=\text{O}$). Anal. calc. for $\text{C}_{15}\text{H}_{10}\text{F}_3\text{N}_3\text{O}$: C 59.02, H 3.30, N 13.77; found: C 59.09, H 3.27, N 13.81.

2,4-Dihydro-4-(4-methylphenyl)-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3c): Yield 87%. M.p. $193-194^\circ$. IR: 3160 (NH), 1699 ($\text{C}=\text{O}$), 1620 ($\text{C}=\text{N}$), 835, 847 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 2.31 (*s*, 3 H); 7.12–7.74 (*m*, 8 H); 10.42 (*s*, 1 H). $^{13}\text{C-NMR}$: 21.80; 120.13 (CF_3); 121.87; 122.56; 123.05; 125.22; 126.13; 127.22; 129.05; 134.12; 147.84 ($\text{C}=\text{N}$); 154.76 ($\text{C}=\text{O}$). Anal. calc. for $\text{C}_{16}\text{H}_{12}\text{F}_3\text{N}_3\text{O}$: C 60.19, H 3.79, N 13.15; found: C 60.17, H 3.77, N 13.19.

4-[4-(tert-Butyl)phenyl]-2,4-dihydro-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3d): Yield 91%. M.p. $219-220^\circ$. IR: 3162 (NH), 1698 ($\text{C}=\text{O}$), 1621 ($\text{C}=\text{N}$), 833, 849 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 1.30 (*s*, 9 H); 7.21–7.77 (*m*, 8 H); 12.35 (*s*, 1 H). $^{13}\text{C-NMR}$: 31.70; 35.18; 126.18 (CF_3); 126.26; 126.91; 127.84; 128.94; 130.05; 131.42; 131.69; 142.85; 150.02 ($\text{C}=\text{N}$); 155.23 ($\text{C}=\text{O}$). Anal. calc. for $\text{C}_{19}\text{H}_{18}\text{F}_3\text{N}_3\text{O}$: C 63.15, H 5.02, N 11.63; found: C 63.17, H 5.04, N 11.60.

4-(4-Fluorophenyl)-2,4-dihydro-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3e): Yield 86%. M.p. $205-206^\circ$. IR: 3171 (NH), 1691 ($\text{C}=\text{O}$), 1621 ($\text{C}=\text{N}$), 833, 843 (1,4-disubstituted benzene ring). $^1\text{H-NMR}$: 7.33–7.78 (*m*, 8 H); 12.39 (*s*, 1 H). $^{13}\text{C-NMR}$: 124.21 (CF_3); 125.14; 126.21; 127.34; 128.37; 129.13; 131.87; 133.10; 143.23; 149.92 ($\text{C}=\text{N}$); 155.14 ($\text{C}=\text{O}$). Anal. calc. for $\text{C}_{15}\text{H}_9\text{F}_4\text{N}_3\text{O}$: C 55.73, H 2.81, N 13.00; found: C 55.70, H 2.84, N 13.02.

2,4-Dihydro-4-(4-hydroxyphenyl)-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3f): Yield 83%. M.p. 175–176°. IR: 3339 (OH), 3174 (NH), 1697 (C=O), 1614 (C=N), 824, 845 (1,4-disubstituted benzene ring). ¹H-NMR: 6.37–7.49 (*m*, 8 H); 8.36 (*br. s*, 1 H); 12.23 (*s*, 1 H). ¹³C-NMR: 123.14 (CF₃); 125.32; 125.98; 126.67; 128.21; 129.61; 131.66; 132.73; 146.21 (C=N); 147.23; 156.21 (C=O). Anal. calc. for C₁₅H₁₀F₃N₃O₂: C 56.08, H 3.14, N 13.08; found: C 56.02, H 3.14, N 13.10.

2,4-Dihydro-4-(4-phenoxyphenyl)-5-[4-(trifluoromethyl)phenyl]-3H-1,2,4-triazol-3-one (3g): Yield 85%. M.p. 183–184°. IR: 3195 (NH), 1683 (C=O), 1602 (C=N), 837, 852 (1,4-disubstituted benzene ring), 688, 749 (monosubstituted benzene ring). ¹H-NMR: 6.94–8.02 (*m*, 13 H); 11.97 (*s*, 1 H). ¹³C-NMR: 122.76 (CF₃); 123.14; 124.22; 125.03; 125.87; 126.58; 127.21; 128.56; 129.44; 130.37; 131.62; 145.14 (C=N); 146.44; 147.33; 155.53 (C=O). Anal. calc. for C₂₁H₁₄F₃N₃O₂: C 63.48, H 3.55, N 10.58; found: C 63.42, H 3.54, N 10.51.

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