Synthesis of 1,4-Disubstituted 1,2,3-Triazoles from Aromatic α-Bromoketones, Sodium Azide and Terminal Acetylenes *via* Cu/Cu(OTf)$_2$-catalyzed Click Reaction under Microwave Irradiation

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Reaction of aromatic α-bromoketones, sodium azide and aromatic or aliphatic terminal acetylenes in the presence of Cu/Cu(OTf)$_2$ following the classical method (aqueous acetonitrile at room temperature) and under microwave irradiation (H_2O at 85 °C) leads to 1,4-disubstituted 1,2,3-triazoles as the final products after simple filtration.

Key words: Click Reaction, Copper/Copper Triflate, Cycloaddition, α-Bromoketones, 1,4-Disubstituted 1,2,3-Triazoles, Three-Component Reaction, Microwave Irradiation

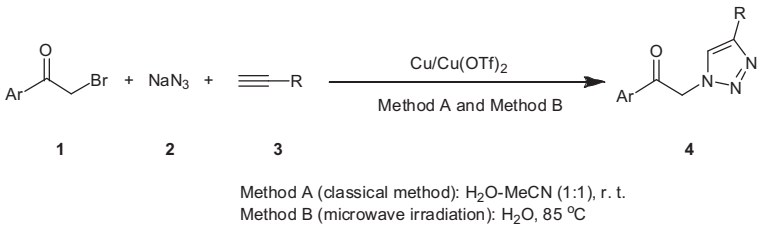
Introduction

Multi-component reactions (MCRs) are one of the most important reactions in combinatorial chemistry [1]. In these reactions, compounds can be synthesized from three or more compounds reacting in a few steps and usually in a one-pot operation.

1,2,3-Triazoles have a wide range of biologically relevant properties such as anti-bacterial [2], anti-allergic [3] and anti-HIV [4] activity, and because of these properties they are targets for drug discovery [5]. In 1963, Huisgen and his co-workers described a systematic study on 1,3-dipolar cycloaddition between azides and terminal alkynes [6]. This uncatalyzed cycloaddition reaction was improved in 2002 by Meldal [7] and Sharpless [8] *via* the use of copper(I) which can improve the rates and regioselectivity of the azide-alkyne cycloaddition reaction. Thus the Huisgen 1,3-dipolar cycloaddition to triazoles can be performed under copper catalysis in a click reaction and is then known as copper-catalyzed azide-alkyne cycloaddition (CuAAC) [9]. The CuAAC produces only 1,4-disubstituted triazoles with excellent yields and purities.

Some of 1,4-disubstituted 1,2,3-triazoles have been synthesized using the click chemistry between α-azidoketones and terminal acetylenes, and the products have shown modest Src kinase inhibitory activity [10]. They are also important compounds for their cytotoxicity in A549 (lung cancer) [11], HT-29 (colon cancer) [11] and He La (cervial cancer) [11]. In this paper we wish to report a simple method for the synthesis of these compounds using click reaction conditions (Fig. 1).

α-Azidoketones cannot be easily prepared by the reaction of sodium azide and α-bromoketones, because isolation and purification of some α-azidoketones is difficult [12]. Therefore, *in situ* generation of α-azidoketones from the reaction of α-bromoketones with sodium azide is very important. The cycloaddition reaction between α-azidoketones and terminal acetylenes has been reported using various methods, including (1) $CuSO_4 \cdot 5H_2O$/sodium ascorbate, PEG-H_2O [10]; (2) CuI, H_2O or H_2O-acetone [11]; (3) $CuSO_4$/sodium ascorbate, PEG-H_2O [12]; (4) CuI, PEG-H_2O [13]; (5) $CuSO_4$/sodium ascorbate, *t*-BuOH-H_2O [14]; and (6) P_4VPy-CuI, H_2O [15] systems.



Scheme 1. Synthesis of 1,4-disubstituted 1,2,3-triazoles.

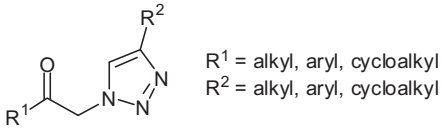


Fig. 1. Structure of 1,4-disubstituted 1,2,3-triazoles.

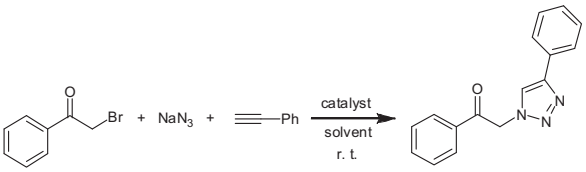
In continuation of our studies of multi-component reactions [16 – 21] and running organic reactions under microwave irradiation [22 – 24], herein we wish to report a facile synthesis of 1,4-disubstituted 1,2,3-triazoles involving the three-component, one-pot condensation of α-bromoketones, sodium azide, and terminal alkynes using the Cu/Cu(OTf)$_2$ as a catalyst. We investigated these procedures both by the classical method and by microwave irradiation (Scheme 1).

Results and Discussion

To optimize the cycloaddition reaction in the classical method, the reaction of 2-bromoacetophenone with sodium azide and phenyl acetylene as an aromatic terminal alkyne was used as a model reaction. In initial attempts, we examined the effect of various Cu(I) catalysts such as Cu/Cu(OAc)$_2$, Cu/CuSO$_4$ and Cu/Cu(OTf)$_2$ in various solvents such as H$_2$O, MeOH-H$_2$O, EtOH-H$_2$O, DMSO-H$_2$O, *t*-BuOH-H$_2$O, MeCN-H$_2$O, and acetone-H$_2$O at room temperature (Table 1, entries 1 – 11).

In this reaction, the Cu(I) catalyst was prepared *in situ* by the comproportionation of Cu(0) and Cu(II), and Cu/Cu(OTf)$_2$ in aqueous acetonitrile was chosen as an effective catalyst (Table 1, entry 4). The advantage of this catalyst is the availability of Cu(0) and Cu(OTf)$_2$ powder and the power of this mixture to produce 1,4-disubstituted 1,2,3-triazoles in high yields and short reaction times. The use of Cu(OTf)$_2$ in the absence of Cu powder gives the product in 24 h with 78 % yield (Table 1, entry 10).

Table 1. Cu(I)-catalyzed 1,3-dipolar cycloaddition of 2-bromo acetophenone with sodium azide and phenyl acetylene under classical conditions[a].

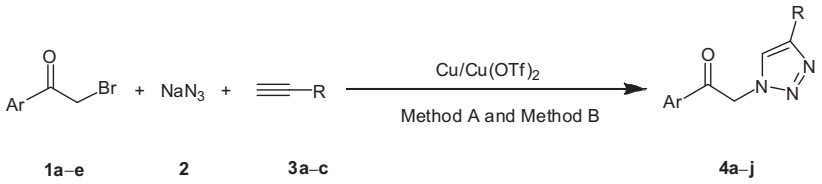


Entry	Catalyst[b]	Solvent	Time (h)	Yield (%)[c]
1	Cu/Cu(OTf)$_2$	DMSO-H$_2$O	2.83	60
2	Cu/Cu(OTf)$_2$	acetone-H$_2$O	6.5	52
3	Cu/Cu(OTf)$_2$	*t*-BuOH-H$_2$O	2.83	40
4	Cu/Cu(OTf)$_2$	MeCN-H$_2$O	2.66	91
5	Cu/Cu(OTf)$_2$	EtOH-H$_2$O	4	70
6	Cu/Cu(OTf)$_2$	MeOH-H$_2$O	5.5	60
7	Cu/Cu(OAc)$_2$	*t*-BuOH-H$_2$O	2.66	72
8	Cu/Cu(OAc)$_2$	MeCN-H$_2$O	2.5	82
9	Cu/CuSO$_4$	MeCN-H$_2$O	2.83	77
10	Cu(OTf)$_2$	MeCN-H$_2$O	24	78
11	Cu/Cu(OTf)$_2$	H$_2$O	13	45

[a] All reactions were performed on a 0.5 mmol scale in 1.5 mL of solvent at room temperature; [b] 5 mol-% of Cu/Cu(II); [c] yields are given for isolated products.

Various aromatic α-bromoketones were examined with the classical method to obtain 1,4-disubstituted 1,2,3-triazoles. In this reaction, we investigated the effects of phenylacetylene and other terminal acetylenes with hydroxyl as a functional group in the reaction of α-bromoketones and NaN$_3$. The results with various terminal acetylenes have demonstrated the hydroxyl group tolerance of this protocol (Table 2, entries 4 – 10).

The use of microwave irradiation in this reaction instead of the conventional method was also investigated. The model reaction was examined in various solvents and at different temperatures with controlled microwave powers (200 – 700 W). The best result was obtained in H$_2$O as a solvent at 85 °C and 700 W. As



Method A (classical method): H_2O-MeCN (1:1), r. t.
Method B (microwave irradiation): H_2O, 85 °C

Table 2. Synthesis of 1,4-disubstituted 1,2,3-triazoles **4**.

Entry	Ar (**1a–e**)	R (**3a–c**)	Product (**4a–j**)	Method A[a]		Method B[b]	
				Time (h)	Yield (%)[c]	Time (min)	Yield (%)[c]
1	Ph (**1a**)	Ph (**3a**)	**4a**	2.66	91	10	92
2	4-Me-C_6H_4 (**1b**)	Ph (**3a**)	**4b**	1.83	92	10	95
3	4-Cl-C_6H_4 (**1c**)	Ph (**3a**)	**4c**	1.33	88	8	90
4	Ph (**1a**)	CH_2OH (**3b**)	**4d**	1.16	84	8	90
5	4-Cl-C_6H_4 (**1c**)	$C(CH_3)_2OH$ (**3c**)	**4e**	1.5	80	11	85
6	4-Me-C_6H_4 (**1b**)	CH_2OH (**3b**)	**4f**	1.25	89	9	92
7	4-Me-C_6H_4 (**1b**)	$C(CH_3)_2OH$ (**3c**)	**4g**	1.5	82	12	87
8	4-Br-C_6H_4 (**1d**)	CH_2OH (**3b**)	**4h**	2	80	9	90
9	4-Br-C_6H_4 (**1d**)	$C(CH_3)_2OH$ (**3c**)	**4i**	2.16	78	14	85
10	4-Ph-C_6H_4 (**1e**)	CH_2OH (**3b**)	**4j**	2	72	10	85

[a] Under classical heating in aqueous acetonitrile at room temperature; [b] under microwave irradiation in water at 85 °C; [c] yields of pure products.

can be seen from Table 2, the yields of the products and the reaction times under microwave irradiation are more acceptable than those for conventional methods. Another advantage of this reaction under microwave irradiation is the use of H_2O as a "green solvent" instead of aqueous acetonitrile in the classical method. Using H_2O as a solvent in 13 h under the classical method produced triazole **4a** only in 45 % yield (Table 1, entry 11).

The progress of the reaction was monitored by TLC, IR and ^{1}H NMR. Disappearance of the azide absorption of the α-azidoketone (2100 cm^{-1}) and creation of the triazole (1223 cm^{-1}) and carbonyl (1701 cm^{-1}) absorptions in the IR spectra indicated the formation of 1,4-disubstituted 1,2,3-triazoles, and the ^{1}H NMR spectrum displayed a singlet at $\delta = 8.54$ ppm for triazole H-5.

Conclusion

In summary, we have developed mild reaction conditions for the regioselective synthesis of new derivatives of 1,4-disubstituted 1,2,3-triazoles. Reaction of aromatic α-bromoketones, sodium azide and aromatic or aliphatic terminal acetylenes in the presence of $Cu/Cu(OTf)_2$ under the conventional method (aqueous acetonitrile at room temperature) or under microwave irradiation (H_2O at 85 °C) gave the corresponding 1,4-disubstituted 1,2,3-triazoles. In this reaction the products could be separated by a simple filtration. High yields, short reaction times, regioselective synthesis and mild reaction conditions in the classical method make this method very simple and useful for the prepration of 1,4-disubstituted 1,2,3-triazoles. However, this reaction was more facile and much faster, when it was performed under microwave irradiation. Because of homogeneous heating throughout the reaction media by microwave irradiation, the reactions were completed in shorter reaction time (8 – 14 min) and in higher yields (85 – 95 %). Because H_2O can be used as a solvent under microwave irradiation, we can claim that this one-pot, three-component protocol has all qualities and superiorities expected from click chemistry, which was first fully described by Sharpless *et al.* in 2001 [25]. In comparison with other methods which needed long reaction times (8 – 12 h) [11, 12] and gave moderate yields (61 – 86 %) [11, 13], this method is completed in about one to three hours giving good yields of products (72 – 92 %) under the conventional method and in 8 – 14 minutes giving excellent

yields of products (85 – 95 %) under microvawe irradiation. The pure products are obtained by simple filtration.

Experimental Section

Melting points were measured using a capillary tube method with a Barnstead Electrothermal 9200 apparatus. FTIR spectra were recorded using KBr discs on an FTIR Bruker Tensor 27 instrument. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AQS-Avance spectrometer at 500 and 125 MHz, respectively, using TMS as an internal standard. Mass spectra were documented on an Agilent Technology (HP) spectrometer operating at an ionization potential of 70 eV, and all reactions were carried out in an Ethos MR microwave reactor.

General procedure for the synthesis of 1,4-disubstituted 1,2,3-triazoles under the classical method (Method A)

To a stirred mixture of the α-bromoketone (1 mmol) and sodium azide (1.5 mmol) in aqueous acetonitrile (5 mL), Cu (10 mol-%), $Cu(OTf)_2$ (10 mol-%) and terminal acetylene (1.2 mmol) were added at room temperature, and the mixture was stirred for the reaction time indicated in Table 2 (the progress of reaction was monitored by TLC). After the completion of the reaction, the reaction mixture was diluted with water and filtered to collect the product. The product was washed with cold water and 0.2 M HCl (10 mL). For further purification, the products should be washed with 10 mL of ether (Table 2, condition A).

General procedure for the synthesis of 1,4-disubstituted 1,2,3-triazoles under microwave irradiation (Method B)

α-Bromoketone (1 mmol), sodium azide (1.5 mmol), terminal acetylene (1.25 mmol), Cu (10 mol-%) and $Cu(OTf)_2$ (10 mol-%) were added to water (7 mL) in a reaction vessel of Ethos MR, and the vessel was capped. The reaction mixture was irradiated for 8 – 14 min at 85 °C (the progress of the reaction was monitored by TLC). After the completion of the reaction, the reaction mixture was cooled to room temperature, diluted with water and filtered to collect the product. The product was washed with cold water and 0.2 M HCl (10 mL). For further purification, the products should be washed with 10 mL of ether (Table 2, condition B).

*1-Phenyl-2-(4-phenyl-[1,2,3]triazol-1-yl)ethanone (**4a**) [11 – 13, 26]*

M. p. 169 – 171 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.54 (s, 1H, CH), 8.12 – 7.34 (m, 10H, Ar), 6.28 (s, 2H, CH_2) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 193.05, 147.20, 135.16, 135.00, 131.60, 129.88, 129.83, 129.100, 128.76, 126.06, 123.95, 56.89 ppm. – IR (KBr): ν = 1701, 1223 cm^{-1}.

*2-(4-Phenyl-[1,2,3]triazol-1-yl)-1-p-tolyl-ethanone (**4b**) [11, 13, 26]*

M. p. 157 – 158 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.52 (s, 1H, CH), 8.00 – 7.99 (m, 2H, Ar), 7.88 – 7.87 (m, 2H, Ar), 7.47 – 7.33 (m, 5H, Ar), 6.21 (s, 2H, CH_2), 2.41 (s, 3H, CH_3) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.48, 147.18, 145.76, 132.51, 131.64, 130.40, 129.80, 129.18, 128.72, 126.02, 123.95, 56.73, 22.15 ppm. – IR (KBr): ν = 1695, 1232 cm^{-1}.

*1-(4-Chlorophenyl)-2-(4-phenyl-[1,2,3]triazol-1-yl)ethanone (**4c**) [11 – 13, 26]*

M. p. 108 – 111 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.52 (s, 1H, CH), 8.12 – 7.87 (m, 4H, Ar), 7.71 – 7.34 (m, 5H, Ar), 6.27 (s, 2H, CH_2) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.25, 147.19, 140.08, 133.68, 131.57, 131.03, 130.02, 129.84, 128.77, 126.02, 123.89, 56.87 ppm. – IR (KBr): ν = 1710, 1230 cm^{-1}.

*2-(4-Hydroxymethyl-[1,2,3]triazol-1-yl)-1-phenyl-ethanone (**4d**) [11, 26]*

M. p. 122 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.14 (s, 1H, CH), 8.06 – 8.05 (m, 2H, Ar), 7.75 – 7.72 (m, 1H, Ar), 7.62 – 7.59 (m, 2H, Ar), 6.23 (s, 2H, CH_2), 5.39 (s, 1H, OH), 4.66 (s, 2H, CH_2) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.58, 135.16, 134.88, 129.86, 129.04, 57.31, 55.89 ppm. – IR (KBr): ν = 3389, 1699, 1229 cm^{-1}.

*1-(4-Chlorophenyl)-2-[4-(1-hydroxy-1-methylethyl)-[1,2,3]triazol-1-yl]ethanone (**4e**) [27]*

M. p. 149 – 152 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.04 (m, 2H, Ar), 7.81 (s, 1H, CH), 7.66 – 7.65 (m, 2H, Ar), 6.12 (s, 2H, CH_2), 5.44 (brs, 1H, OH), 1.51 (s, 6H, $2CH_3$) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.52, 139.93, 133.80, 130.92, 129.94, 56.8, 31.86 ppm. – IR (KBr): ν = 3350, 1702, 1231 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 279.4 (5) $[M]^+$, 139 (100), 111 (91), 43 (85).

*2-(4-Hydroxymethyl-[1,2,3]triazol-1-yl)-1-p-tolyl-ethanone (**4f**)*

M. p. 184 – 185 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.04 (s, 1H, CH), 7.96 – 7.95 (m, 2H, Ar), 7.41 – 7.40 (m, 2H, Ar), 6.13 (s, 2H, CH_2), 5.27 (s, 1H, OH), 4.58 (s, 2H, CH_2), 2.41 (s, 3H, CH_3) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.40, 145.66, 132.56, 130.37, 129.13, 56.70, 55.98,

22.13 ppm. – IR (KBr): ν = 3420, 1696, 1235 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 231.3 (3) [M]$^+$, 202 (31), 169 (37), 119 (100), 91 (96), 65 (39). – $C_{12}H_{13}N_3O_2$: calcd. C 62.33, H 5.67, N 18.17; found C 62.46, H 5.75, N 18.07.

*2-[4-(1-Hydroxy-1-methylethyl)-[1,2,3]triazol-1-yl]-1-p-tolyl-ethanone (**4g**)*

M. p. 155 – 158 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.14 (s, 1H, CH), 7.93 – 7.37 (m, 4H, Ar), 6.08 (s, 2H, CH_2), 5.5 (s, 1H, OH), 2.37 (s, 3H, CH_3), 1.52 (s, 6H, $2CH_3$) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.30, 145.60, 132.89, 132.59, 130.97, 130.34, 129.09, 57.01, 31.99, 22.13 ppm. – IR (KBr): ν = 3353, 1689, 1233 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 259.3 (3) [M]$^+$, 244 (64), 119 (100), 105 (48), 91 (92). – $C_{14}H_{17}N_3O_2$: calcd. C 64.85, H 6.61, N 16.21; found C 64.78, H 6.66, N 16.31.

*1-(4-Bromophenyl)-2-(4-hydroxymethyl-[1,2,3]triazol-1-yl)ethanone (**4h**)*

M. p. 159 – 160 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 7.99 (m, 2H, Ar), 7.94 (s, 1H, CH), 7.82 (m, 2H, Ar), 6.15 (s, 2H, CH_2), 5.25 (s, 1H, OH), 4.57 (s, 2H, CH_2) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.49, 148.50, 134.05, 132.92, 131.01, 129.21, 125.35, 56.60, 55.94 ppm. – IR (KBr): ν = 3347, 1696, 1229 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 296.2 (4) [M]$^+$, 268 (20), 266 (20), 183 (100), 155 (42), 76 (26). – $C_{11}H_{10}BrN_3O_2$: calcd. C 44.62, H 3.40, N 14.19; found C 44.54, H 3.47, N 14.23.

*1-(4-Bromophenyl)-2-[4-(1-hydroxy-1-methylethyl)-[1,2,3]triazol-1-yl]ethanone (**4i**)*

M. p. 173 – 175 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 7.99 – 7.97 (m, 2H, Ar), 7.84 (s, 1H, CH), 7.82 – 7.81 (m, 2H, Ar), 6.10 (s, 2H, CH_2), 5.2 (s, 1H, OH), 1.48 (s, 6H, $2CH_3$) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.52, 141.78, 134.13, 132.90, 131.00, 130.99, 129.15, 56.53, 50.84, 31.65 ppm. – IR (KBr): ν = 3301, 1702, 1228 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 324.2 (5) [M]$^+$, 310 (66), 308 (66), 183 (100), 155 (51), 112 (53), 94 (63). – $C_{13}H_{14}BrN_3O_2$: calcd. C 48.17, H 4.35, N 12.96; found C 48.11, H 4.27, N 12.83.

*1-Biphenyl-4-yl-2-(4-hydroxymethyl-[1,2,3]triazole-1-yl)ethanone (**4j**)*

M. p. 171 – 172 °C. – ^{1}H NMR (500 MHz, DMSO): δ = 8.15 (brs, 2H, Ar), 7.91 (brs, 3H, Ar), 7.79 – 7.46 (m, 5H, Ar), 6.18 (s, 2H, CH_2), 5.23 (s, 1H, OH), 4.58 (s, 2H, CH_2) ppm. – ^{13}C NMR (125 MHz, DMSO): δ = 192.67, 146.30, 139.52, 133.87, 130.01, 129.78, 129.49, 127.94, 125.35, 56.61, 55.98 ppm. – IR (KBr): ν = 3288, 1689, 1232 cm^{-1}. – MS (EI, 70 eV): m/z (%) = 293.3 (6) [M]$^+$, 198 (29), 181 (100), 169 (71), 152 (96). – $C_{17}H_{15}N_3O_2$: calcd. C 69.61, H 5.15, N 14.33; found C 69.72, H 5.09, N 14.48.

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