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Synthesis of N-2-aryl-substituted 1,2,3-triazoles mediated by magnetic and recoverable $CuFe_2O_4$ nanoparticles

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Abstract An efficient and economic system for the synthesis of N-2-aryl-substituted 1,2,3-triazoles in the presence of $CuFe_2O_4$ was developed. The corresponding products can be obtained in good to excellent yields. It is interesting to note that the catalyst could be reused for five consecutive trials without significant decreases in its activity.

Keywords Triazoles · Chalcone · $CuFe_2O_4$ nanoparticles

Introduction

1,2,3-triazoles are the core structure in a large number of natural products and biologically and pharmaceutically active molecules [1–9], thus developing an efficient method for the synthesis of 1,2,3-triazoles have aroused great attention in organic synthesis. At present, most of the methods for their synthesis are mainly limited to the N-1 position [10–20]. However, there are only a few methods (Scheme 1) for the functionalization of triazoles at the N-2 position [21–28]. The general procedure for the synthesis of N-2-substituted 1,2,3-triazoles is from the reaction of α-hydroxyketones with hydrazine [29], but the starting materials are complex. Therefore, versatile and practical methods for the synthesis of N-2-substituted 1,2,3-triazoles are still desirable.

In recent years, magnetic nanoparticles have been widely used in organic transformations because of their environmentally benign, cost-effective, ease of separation with an external magnet, and reusability properties [30–34]. Processes

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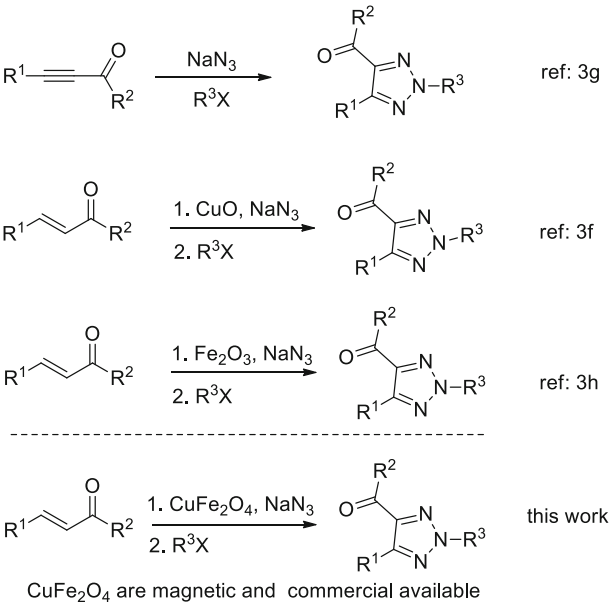


Scheme 1 Background of synthesis of N-2-substituted 1,2,3-triazoles

such as C–S [35, 36] and C–O [37, 38] bond formation and click reaction [39, 40], as well as others [41–48], catalyzed by magnetic $CuFe_2O_4$ nanoparticles have been well described. In continuation of our studies on green chemistry [41, 49–51], herein we present an efficient and practical method for the synthesis of N-2-substituted 1,2,3-triazoles mediated by $CuFe_2O_4$ nanoparticles.

Experimental section

NMR spectra were recorded at 500 MHz for protons on Bruker Advance III HD 500 MHz spectrometers. ^{1}H NMR chemical shifts (d) are given in ppm relative to TMS ($d = 0.0$). Chemical shifts for ^{13}C NMR spectra are reported in parts per million (ppm) from tetramethylsilane with the solvent as the internal standard. Data Reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet), integration, coupling constant, and identification. All major chemicals and solvents were obtained from commercial sources and used without further purification. $CuFe_2O_4$ were purchased directly from Sigma-Aldrich.

General procedure

A 25-mL Schlenk tube equipped with a magnetic stirring bar was charged with 3-(4-chlorophenyl)-1-phenylprop-2-en-1-one (0.20 mmol), sodium azide (0.20 mmol), DMSO (1 mL), and catalyst $CuFe_2O_4$ (1 equiv.). The tube was sealed and heated at

80 °C for 24 h. Then 2-F-$C_6H_4NO_2$ (0.20 mmol) was added to the mixture, and the reaction continued at 80 °C for 5 h. After completion of the reaction, the catalyst was separated from the reaction mixture with an external magnet, and the reaction mixture was extracted with ethyl acetate, dried with anhydrous Na_2SO_4, and concentrated under vacuum. The residue was chromatographed on a silica gel column eluted with a mixture of petroleum ether and ethyl acetate to give pure products.

After catalyst was washed successively with diethyl ether and dried in an oven at 60 °C for 4 h, it was reused directly for the next time without further purification. The recovered catalyst was recovered and reused for five consecutive trials without loss of its activity.

*(2-(2-nitrophenyl)-5-phenyl-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2a)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.12 (d, J = 7.5 Hz, 2H), 8.07 (d, J = 8.0 Hz, 1H), 8.01–7.88 (m, 3H), 7.80 (t, J = 7.7 Hz, 1H), 7.66 (t, J = 7.5 Hz, 2H), 7.55 (t, J = 7.7 Hz, 2H), 7.51–7.43 (m, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.52, 151.37, 144.12, 143.84, 136.76, 133.85, 133.06, 132.00, 130.56, 129.75, 129.74, 128.90, 128.73, 128.54, 128.53, 126.23, 125.80, 125.10.

*(5-(4-chlorophenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2b)*** white solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.02 (d, J = 7.0 Hz, 2H), 7.94 (d, J = 8.5 Hz, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.80 (d, J = 8.5 Hz, 2H), 7.69 (t, J = 7.8 Hz, 1H), 7.56 (dd, J = 8.0 Hz, 7.5 Hz, 2H), 7.45 (t, J = 7.5 Hz, 2H), 7.35 (d, J = 8.5 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.35, 150.42, 144.01, 143.85, 136.67, 135.88, 133.94, 133.11, 131.91, 130.57, 130.31, 129.92, 128.79, 128.56, 127.26, 125.80, 125.14.

*(2-(2-nitrophenyl)-5-(4-nitrophenyl)-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2c)*** white solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.24 (d, J = 9.0 Hz, 2H), 8.13–8.01 (m, 4H), 7.96 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.91 (dd, J = 8.1 Hz, 1.1 Hz, 1H), 7.74 (td, J = 7.5 Hz, 1.0 Hz, 1H), 7.65–7.56 (m, 2H), 7.47 (t, J = 7.5 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.03, 149.31, 148.43, 144.47, 143.94, 136.41, 135.09, 134.17, 133.25, 131.81, 130.60, 130.32, 129.95, 128.65, 125.94, 125.27, 123.70.

*(5-(4-fluorophenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2d)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.13–8.06 (m, 2H), 8.01 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.96–7.86 (m, 3H), 7.76 (td, J = 8.0 Hz, 1.0 Hz, 1H), 7.62 (td, J = 7.5, 3.4 Hz, 2H), 7.52 (t, J = 8.0 Hz, 2H), 7.18–7.10 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.39, 164.68, 162.69, 150.57, 143.90, 143.86, 136.75, 133.85, 133.05, 131.07, 131.01, 130.54, 129.83, 128.53, 125.75, 125.10, 124.94, 124.91, 115.68, 115.51.

*(2-(2-nitrophenyl)-5-(p-tolyl)-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2e)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.12–8.05 (m, 2H), 8.03 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.92 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.83–7.73 (m, 3H), 7.65–7.58 (m, 2H), 7.55–7.47 (m, 2H), 7.24 (s, 2H), 2.40 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.55, 151.44, 144.02, 143.83, 139.84, 136.86, 133.75, 132.99, 132.02, 130.54, 129.63, 129.25, 128.79, 128.50, 125.85, 125.73, 125.05, 21.43.

(5-(3,4-dimethylphenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl)(phenyl)methanone ***(2f)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.17–8.05 (m, 2H), 8.02 (dt, J = 23.2, 11.6 Hz, 1H), 7.92 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.75 (tt, J = 12.1, 6.1 Hz, 1H), 7.62 (ddd, J = 13.0, 9.4, 6.7 Hz, 4H), 7.50 (dd, J = 19.2, 11.6 Hz, 2H), 7.18 (t, J = 12.6 Hz, 1H), 2.31 (d, J = 2.0 Hz, 6H). ^{13}C NMR (125 MHz, $CDCl_3$) δ 187.56, 151.56, 144.08, 138.52, 136.91, 136.79, 133.69, 132.97, 132.10, 130.51, 129.85, 129.79, 129.59, 128.47, 126.39, 126.18, 125.82, 125.05, 77.27, 77.02, 76.76, 19.78, 19.71.

*(4-bromophenyl)(5-(4-fluorophenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl) methanone****(2g)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.97 (ddd, J = 17.1, 12.7, 5.9 Hz, 6H), 7.78 (t, J = 7.5 Hz, 1H), 7.65 (dd, J = 14.8, 8.3 Hz, 3H), 7.15 (t, J = 8.0 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.11, 164.76, 162.77, 150.82, 143.83, 143.50, 135.52, 133.09, 131.98, 131.89, 131.80, 131.16, 131.10, 129.94, 129.27, 125.68, 125.13, 124.78, 115.71, 115.53. HRMS (ESI) calculated for $C_{21}H_{12}BrFN_4O_3$: 488.99661$(M + Na)^+$, Found: 488.99690.

(4-methoxyphenyl)(2-(2-nitrophenyl)-5-(p-tolyl)-2H-1,2,3-triazol-4-yl)methanone ***(2h)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.06 (dd, J = 9.0 Hz, 28.5 Hz, 3H), 7.92 (d, J = 8.0 Hz, 1H), 7.76 (d, J = 8.0 Hz, 3H), 7.61 (t, J = 8.0 Hz, 1H), 7.29–7.19 (m, 2H), 6.99 (d, J = 9.0 Hz, 2H), 3.90 (s, 3H), 2.39 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.06, 164.30, 151.03, 144.37, 143.80, 139.68, 133.01, 132.95, 132.09, 129.74, 129.45, 129.25, 128.65, 125.96, 125.70, 125.01, 113.85, 55.57, 21.41.

*(4-methoxyphenyl)(2-(2-nitrophenyl)-5-(4-nitrophenyl)-2H-1,2,3-triazol-4-yl) methanone****(2i)*** white solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.30 (d, J = 9.0 Hz, 2H), 8.16–8.07 (m, 4H), 8.04 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.98 (dd, J = 8.0 Hz, 1.5 Hz, 1H), 7.81 (td, J = 7.5 Hz, 1.0 Hz, 1H), 7.74–7.64 (m, 1H), 7.02 (d, J = 8.5 Hz, 2H), 3.92 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 185.40, 164.65, 148.93, 148.35, 144.86, 135.22, 133.21, 133.18, 131.86, 130.17, 129.80, 129.26, 125.90, 125.22, 123.71, 114.03, 55.64.

*(5-(4-chlorophenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl)(4-methoxyphenyl) methanone****(2j)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.10 (d, J = 8.0 Hz, 2H), 8.02 (d, J = 8.0 Hz, 1H), 7.93 (d, J = 7.5 Hz, 1H), 7.85 (d, J = 8.5 Hz, 2H), 7.77 (t, J = 7.5 Hz, 1H), 7.63 (t, J = 7.5 Hz, 1H), 7.41 (d, J = 8.0 Hz, 2H), 7.00 (d, J = 8.5 Hz, 2H), 3.90 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 185.77, 164.44, 150.01, 144.38, 143.82, 135.74, 133.07, 131.95, 130.17, 129.74, 129.54, 128.78, 127.39, 125.75, 125.09, 113.92, 55.60.

*(4-chlorophenyl)(5-(4-chlorophenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl) methanone****(2k)*** white solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.11–7.99 (m, 3H), 7.95 (dd, J = 8.0 Hz, 1.0 Hz, 1H), 7.91–7.84 (m, 2H), 7.78 (td, J = 8.0 Hz, 1.5 Hz, 1H), 7.69–7.61 (m, 1H), 7.50 (d, J = 8.5 Hz, 2H), 7.47–7.40 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 185.87, 150.64, 143.67, 140.51, 136.04, 135.03, 133.11,

131.87, 130.37, 130.00, 128.85, 127.14, 125.72, 125.15. HRMS (ESI) calculated for $C_{21}H_{12}Cl_2N_4O_3$: 461.01810 $(M + Na)^+$, Found: 461.01787.

*(5-(4-methoxyphenyl)-2-(2-nitrophenyl)-2H-1,2,3-triazol-4-yl)(4-nitrophenyl) methanone**(2l)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 (d, $J = 9.0$ Hz, 2H), 8.23 (d, $J = 8.5$ Hz, 2H), 8.04 (d, $J = 8.0$ Hz, 1H), 7.98–7.88 (m, 3H), 7.78 (t, $J = 9.0$, Hz, 1H), 7.65 (t, $J = 7.5$ Hz, 1H), 7.01 (d, $J = 9.0$ Hz, 2H), 3.88 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 185.54, 161.15, 152.04, 150.43, 143.78, 142.89, 141.91, 133.06, 131.69, 131.39, 130.69, 129.97, 125.56, 125.09, 123.50, 120.72, 114.00, 77.28, 77.02, 76.77, 55.39. HRMS (ESI) calculated for $C_{22}H_{15}N_5O_6$: 468.09125 $(M + Na)^+$, Found: 468.09145.

(4-methoxyphenyl)(2-(2-nitrophenyl)-5-phenyl-2H-1,2,3-triazol-4-yl)methanone ***(2m)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.16–8.09 (m, 2H), 8.06 (dd, $J = 8.0$ Hz, 1.5 Hz, 1H), 7.94 (dd, $J = 8.5$ Hz, 1.5 Hz, 1H), 7.83–7.75 (m, 3H), 7.64 (td, $J = 7.5$ Hz, 1.5 Hz, 1H), 7.26 (d, $J = 8.0$ Hz, 2H), 7.04–6.98 (m, 2H), 3.93 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.00, 164.34, 150.93, 144.45, 143.79, 133.04, 132.04, 129.59, 128.82, 128.75, 128.54, 125.75, 125.05, 113.87, 55.60.

(4-chlorophenyl)(2-(2-nitrophenyl)-5-phenyl-2H-1,2,3-triazol-4-yl)methanone ***(2n)*** yellow solid,^{1}H NMR (500 MHz, $CDCl_3$) δ 8.15–8.03 (m, 3H), 8.02–7.87 (m, 3H), 7.80 (td, $J = 8.0$ Hz, 1.5 Hz, 1H), 7.72–7.63 (m, 1H), 7.59–7.41 (m, 5H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.01, 151.58, 143.72, 140.39, 135.09, 133.08, 131.91, 131.86, 129.85, 128.96, 128.86, 128.58, 128.54, 125.71, 125.10.

(4-bromophenyl)(2-(2-nitrophenyl)-5-phenyl-2H-1,2,3-triazol-4-yl)methanone ***(2o)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.98–7.93 (m, 1H), 7.92–7.87 (m, 2H), 7.87–7.76 (m, 3H), 7.69 (td, $J = 7.5$ Hz, 1.0 Hz, 1H), 7.62–7.51 (m, 3H), 7.43–7.33 (m, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.20, 151.61, 143.80, 143.69, 135.53, 133.09, 131.99, 131.86, 129.87, 129.24, 128.98, 128.59, 128.55, 125.72, 125.11.

*(4-nitrophenyl)(2-(2-nitrophenyl)-5-phenyl-2H-1,2,3-triazol-4-yl)methanone**(2p)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.29 (d, $J = 4.0$ Hz, 2H), 8.18 (d, $J = 14.0$ Hz, 2H), 7.98 (d, $J = 8.0$ Hz, 1H), 7.92–7.80 (m, 3H), 7.72 (t, $J = 8.0$ Hz, 1H), 7.56 (dt, $J = 15.2$, 7.9 Hz, 1H), 7.47–7.34 (m, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 185.39, 152.18, 150.48, 143.82, 143.18, 141.67, 133.16, 131.69, 131.43, 130.15, 129.16, 128.58, 128.34, 125.67, 125.16, 123.56.

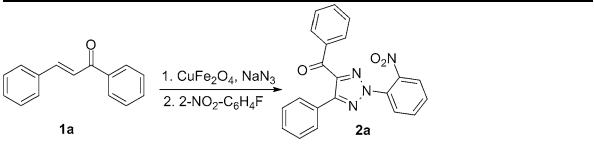
*(2-benzyl-5-phenyl-2H-1,2,3-triazol-4-yl)(phenyl)methanone**(2q)*** yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.96 (d, $J = 7.6$ Hz, 2H), 7.72 (d, $J = 6.9$ Hz, 2H), 7.48 (t, $J = 7.3$ Hz, 1H), 7.45–7.17 (m, 10H), 5.58 (s, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 187.90, 150.07, 142.33, 137.35, 134.56, 133.36, 130.51, 129.70, 129.14, 128.93, 128.79, 128.66, 128.39, 128.33, 128.23, 59.30.

Results and discussion

In order to optimize the reaction conditions, we chose chalcone as a model substrate. In our initial experiment, the effect of various solvents and temperatures were examined. When reactions were conducted in dimethyl sulfoxide (Table 1, entry 2), a high yield was obtained. The use of *N*, *N*-dimethylformamide (Table 1, entry 1) as the solvent led to slower reaction. Poor yields of the desired products were obtained when reactions were conducted in dioxane (Table 1, entry 3) and THF (Table 1, entry 4). When toluene (Table 1, entry 5), CH_3CN (Table 1, entry 6), and PhCl (Table 1, entry 7) were used, only trace amounts of the desired products were isolated. To our disappointment, when reaction temperature was elevated from 80 to 100 °C, there was no great improvement on the yield (Table 1, entry 8). But the yield was slightly decreased when the reaction was conducted at 60 °C (Table 1, entry 9). Furthermore, neither increased nor decreased loading of catalyst $CuFe_2O_4$ gave any obvious improvement in the yields. (Table 1, entry 10–11).

After completion of the search for the optimized reaction conditions, we chose a variety of structurally diverse chalcones with wide range of functional groups to understand the scope and generality of this reaction (Table 2). It was found that the nature of the substituent and its position in the aromatic ring (R^1 or R^2) showed some impact on the yields of the desired products. It is interesting to note that chalcones with electron-withdrawing groups showed slightly higher yield than those with electron-donating groups. But most reactions proceeded smoothly to afford the corresponding N-2-aryl-substituted 1,2,3-triazoles in good to excellent yields. In addition to 1-fluoro-2-nitrobenzene, we also tried alkyl halide such as benzyl chloride, and to our delight, a high yield of the corresponding product was obtained in 87 % yield (Scheme 2). All products were fully characterized based on the

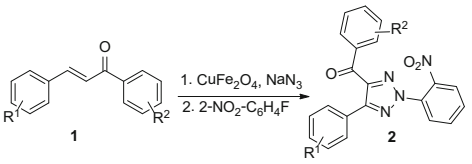
Table 1 Optimization of the reaction conditions



Entry	$CuFe_2O_4$ (equiv)	*T* (°C)	Solvent	Yield (%)
1	1	80	DMF	79
2	1	80	DMSO	90
3	1	80	Dioxane	23
4	1	80	THF	16
5	1	80	Toluene	Trace
6	1	80	CH_3CN	Trace
7	1	80	PhCl	Trace
8	1	100	DMSO	89
9	1	60	DMSO	82
10	0.5	80	DMSO	76
11	1.5	80	DMSO	88

Reaction conditions: **1a** (0.2 mmol), NaN_3 (0.2 mmol), in designated solvent (1 mL) for 24 h under air, then 2-NO_2-C_6H_4F (0.2 mmol) was added to the mixture and the reaction continued for 5 h. Isolated yields

Table 2 Substrate scope of N-2-aryl-substituted 1,2,3-triazoles



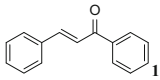
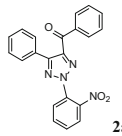
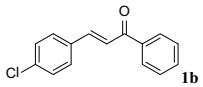
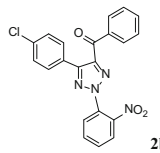
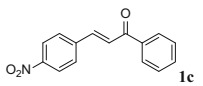
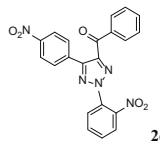
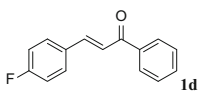
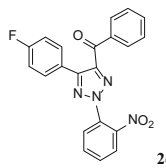
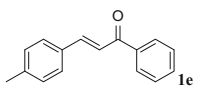
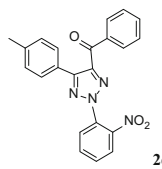
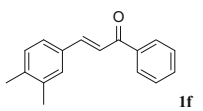
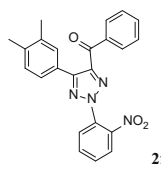
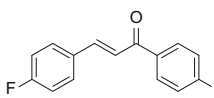
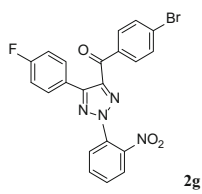
Entry	1	2	Yield (%)[a]
1	 **1a**	 **2a**	90
2	 **1b**	 **2b**	94
3	 **1c**	 **2c**	92
4	 **1d**	 **2d**	85
5	 **1e**	 **2e**	78
6	 **1f**	 **2f**	80
7	 **1g**	 **2g**	82

Table 2 continued

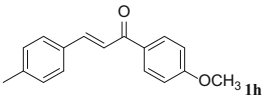
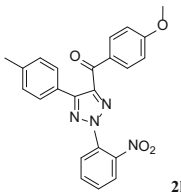
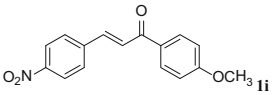
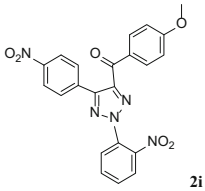
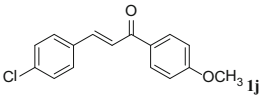
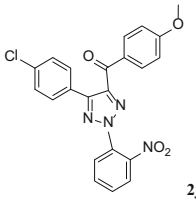
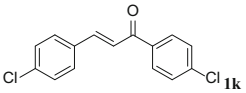
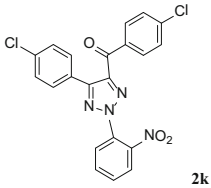
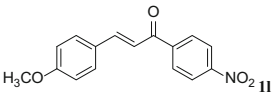
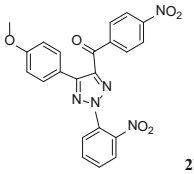
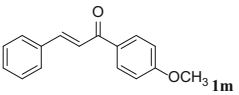
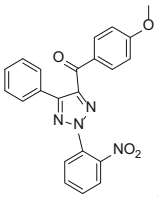
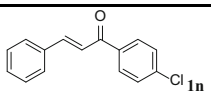
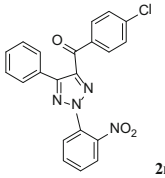
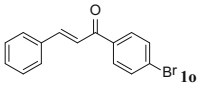
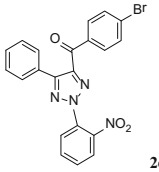
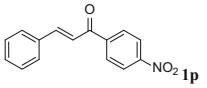
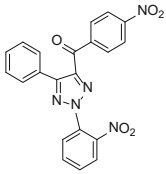
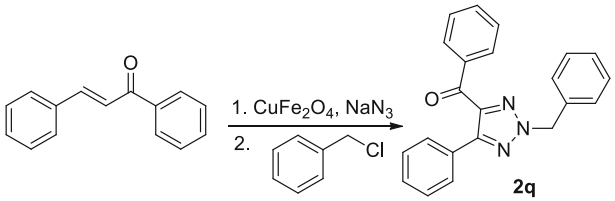
Entry	1	2	Yield (%)[a]
8	 **1h**	 **2h**	70
9	 **1i**	 **2i**	81
10	 **1j**	 **2j**	86
11	 **1k**	 **2k**	84
12	 **1l**	 **2l**	93
13	 **1m**	 **2m**	75

Table 2 continued

Entry	1	2	Yield (%)[a]
14	 **1n**	 **2n**	85
15	 **1o**	 **2o**	81
16	 **1p**	 **2p**	82

Reaction conditions: **1** (0.2 mmol), NaN_3 (0.2 mmol), $CuFe_2O_4$ (1 equiv) in the DMSO (1 mL), at 80 °C for 24 h, under air, then 2-NO_2-C_6H_4F (0.2 mmol) was added to the mixture, and the reaction continued for 5 h



Scheme 2 Synthesis of **2q**

spectroscopic data. To put the structure beyond any ambiguity, a single crystal of **2m** was cultivated by slow evaporation of solvent from its solution in a mixture of ethyl acetate and n-hexane, and its X-ray molecular structure (CCDC 1407347) was determined (Fig. 1).

To screen the recyclability of the $CuFe_2O_4$, a more practical method was applied to the reaction using chalcone as substrate under present reaction conditions (Table 3). After separation of the product from the reaction mixture and the recovery of $CuFe_2O_4$ catalyst with an external magnet, fresh starting materials were charged into the reaction system after the catalyst was washed successively with diethyl ether and dried in an oven at 60 °C for 4 h. The reactions still proceeded well, and high yields were obtained at the end of the reaction. The catalyst $CuFe_2O_4$

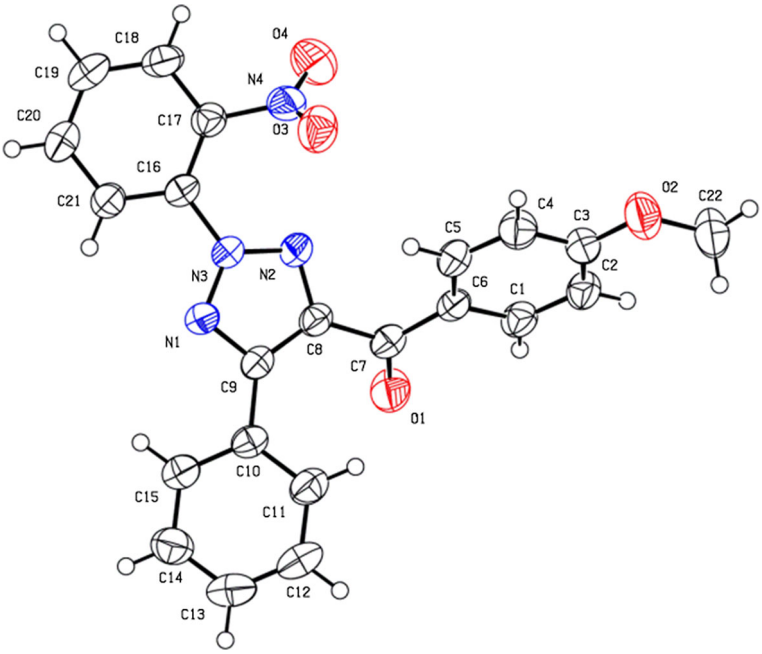


Fig. 1 X-ray crystal structures of **2 m**

Table 3 Recovery and reuse of $CuFe_2O_4$

Entry	Yield[a] (%)
1	90
2	91
3	89
4	89
5	87

Reaction conditions: **1a** (0.2 mmol), NaN_3 (0.2 mmol), $CuFe_2O_4$ (1equiv) in DMSO (1 mL), at 80 °C for 24 h, under air, then 2-NO_2-C_6H_4F(0.2 mmol) was added to the mixture, and the reaction continued for 5 h

can be recycled five times without significant losses of its catalytic activity. The $CuFe_2O_4$ nanoparticles were also characterized by SEM spectrum (Fig. 2). The SEM images analysis of the recovered $CuFe_2O_4$ particles revealed that the morphology of the catalyst remains unchanged, even after five cycles.

Next, the reaction of chalcone with sodium azide was tested in the presence of one equivalent of 2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPO) as a radical scavenger. No significant difference was observed in the yield (Scheme 3), ruling out the presence of radicals during the reaction.

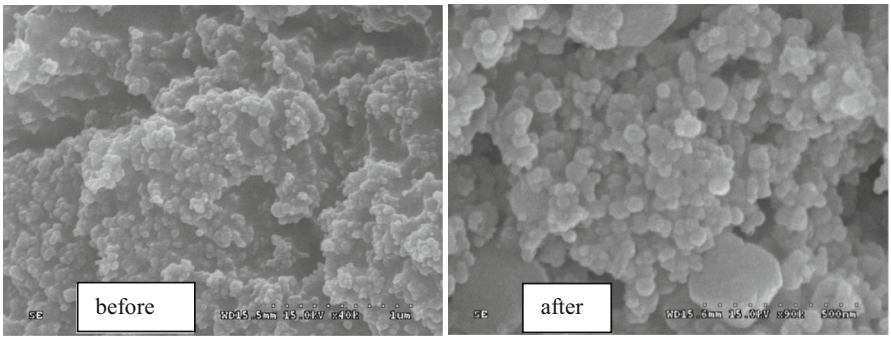
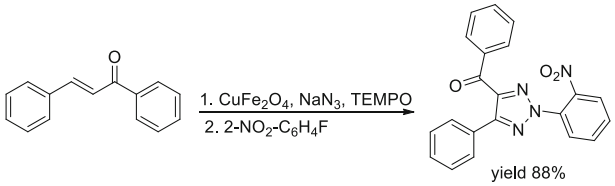
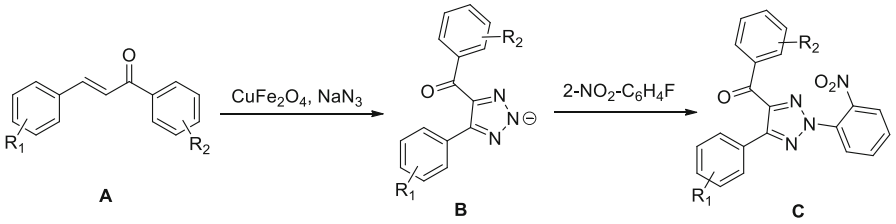


Fig. 2 SEM images of $CuFe_2O_4$ nanoparticles before and after



Scheme 3 Reaction in the presence of TEMPO under the optimized reaction conditions



Scheme 4 Proposed mechanism for this reaction

Although the mechanism for this reaction was not clear, according to the literature[3f] and our experiment, a plausible mechanism was proposed (Scheme 4). Firstly, intermediate **B** was produced from the reaction of chalcones **A** with sodium azide in the presence of $CuFe_2O_4$, then nucleophilic reaction of intermediate **B** with aryl halide occurred to provide the final product **C**.

Conclusions

In conclusion, we have developed an efficient and economic system for the synthesis of N-2-aryl-substituted 1,2,3-triazoles in the presence of $CuFe_2O_4$. The reaction could generate the corresponding products in good to excellent yields under the present reaction conditions. Furthermore, the catalyst could be recovered and

recycled with an external magnet and reused for five consecutive trials without significant decreases in its activity. To explore more effective and economic system for organic synthesis is being done in our laboratory.

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